Size-sensitive phenomena
in finite Yukawa-balls

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1. Introduction

Size effects are fascinating phenomena found in nature. In a classical view one expects to find smaller parts of the same matter to physically behave exactly like larger ones. However, greek philosophers like Democritus already assumed that there has to be a smallest part that cannot be divided further, the “uncutable” atom. If large matter composed of innumerable atoms exhibits phase transitions, colors, electromagnetism and other physical phenomena while the smallest part of it is virtually indestructible without having any of those properties, there has to be a transition region in which the collective phenomena of bulk matter establish. Modern physics confirmed these qualitative assumptions with the detection of atoms in the 19th and 20th century and the discovery of small atomic clusters in the 1980s. In the transition region, where clusters consist of less than a million atoms, the mentioned physical phenomena start to form but differ from the bulk material due to different structural properties.

The aim of my research is to study similar size-dependent effects in clusters made of micron-sized particles in a plasma environment, the so called Yukawa-balls. They belong to the wide family of dusty plasmas which emerged as a research topic in the 1980s as well. In a dusty plasma, macroscopic particles are added to the mix of atoms, ions and electrons that make up a normal plasma. Natural dusty plasmas can be found as very large, weakly coupled clouds in space \[1, 2\]. Famous examples are the dust tails of comets, the rings of Saturn and star formation regions in general. First man-made dusty plasmas were generated unintentionally in the process of industrial plasma etching \[3–5\]. It was found that particles in the nano- and micrometer size range charge up negatively in the plasma environment enabling them to be levitated in the sheath region.

While investigations in the field of applied physics aimed to avoid the destructive effect of dust in plasma etching devices, other groups recognized the potential of plasma crystals for fundamental research. Following theoretical predictions \[6, 7\], the first plasma crystals were generated 1994 in laboratory plasmas \[8–10\]. It was found that these systems are strongly-coupled and thus are able to form solid-like and liquid-like structures \[11–15\]. With diameters of several microns the dust particles are much heavier than other plasma components. This determines their dynamical timescale to be in the millisecond range, which is rather slow compared to the microsecond and nanosecond timescales of ions and electrons respectively. Additionally, the repulsive forces of the negatively charged dust grains result in large interparticle distance making the systems optically thin. Both properties, the slow dynamics and the beneficial optical properties, allow us to investigate dusty plasmas via video-microscopy. This unique combination of a very good experimental accessibility and strong coupling is what makes dusty plasmas so interesting for fundamental research.
1. Introduction

Dusty plasmas exhibit a variety of interesting phenomena connected to the number of constituting particles. Bulk properties of plasma crystals can be studied in extended dust clouds consisting of several hundred thousands of particles or more. In the laboratory, large dust clouds exhibit unique characteristics like a crystalline body centered cubic (bcc) or face centered cubic (fcc) lattice structure \cite{16,18}, a central dust-free region called the void \cite{19,20}, vortex motions \cite{21,22} and self-excited dust density waves \cite{23,26}, to name a few.

For single or very few particles the dynamic is determined mainly by the confining forces in the plasma environment \cite{27,30}. As more particles are added, 3D structures and collective behavior start to establish determined by the interplay between the confinement and the particle interaction. Yukawa-balls are 3D particle systems that can be found in the transition zone between single dust grains and extended dust clouds. They consist of 10 to 10,000 particles arranged on nested concentric shells featuring hexagonal and pentagonal order. In the past 15 years since the first Yukawa-balls were formed using vertical and radial electric forces of the plasma sheath \cite{31}, many experiments and simulations have been performed to investigate the properties of those systems. General objects of investigation are the ground-state structure and their energy \cite{32,41}, the mode-driven dynamics \cite{42,49}, phase-transitions \cite{50,54} and the influence of streaming ions \cite{55,58}. It was found that the intermediate size-region populated by Yukawa-balls is characterized by the onset of collective phenomena under the influence of plasma forces making it a rich object of investigation. Especially the impact of the number of constituting particles on the dynamical and structural properties is still an open research topic and poses the central question of this thesis. To investigate this, an experimental setup is needed in which very small clusters as well as larger ones can be confined, manipulated, observed and evaluated. With the construction of my advanced stereoscopic setup that will be presented in this thesis, I was able to address the following open questions that were experimentally not accessible before:

- How can phase transitions be quantified? Or more specific, what is an appropriate melting criterion for finite strongly-coupled systems?
- What impact does the number of particles in the cluster have on the phase transition?
- Which dynamical behavior do medium-sized clusters with particle numbers between 100 and 1000 exhibit?
- How does the screening affect the dynamics of large clusters with more than 1000 particles?

In the following chapters I will give answers to these questions while following the overall aim, the understanding of size effects in finite Yukawa-balls.
2. Physical Background

To form a dusty plasma, two very obvious components are needed: dust particles and a plasma environment. In order to understand the interplay between them one needs to contemplate the properties of the individual constituents first. Therefore, this chapter will start with a brief introduction to plasma physics, particularly on the topic of capacitively-coupled, low-pressure, high-frequency discharges. In the following, the consequences of adding micron-sized particles to a plasma system are shown and the properties of a dust-plasma-ensemble will be discussed.

2.1. Characteristics of low temperature plasmas

In general, a plasma is a gaseous, quasi-neutral system of multiple charged species as first described by Irving Langmuir [59]. A typical low-temperature plasma consists of negative electrons, positive ions and, due to the low degree of ionization, neutral atoms. The charge-carrying species are not in thermal equilibrium: While the ions are roughly at room temperature, the electrons have an energy \( E = k_B T \) of the order of the ionization energy of the supply gas, i.e. energies of some electron volts. Thus they are able to ionize neutrals and provide the main drive of low-temperature plasmas.

A single free charge carrier with charge \( Q \) causes a potential \( \Phi_c = Q/(4\pi\varepsilon_0 r) \) at a distance \( r \), known as Coulomb’s law. However, in a plasma environment the coexistence of many charge carriers generates a shielding effect. A local charge produces an opposing net space charge around it provided by the charge carriers in the plasma that weakens the electric field [60]. Effectively, this causes a screened interaction law

\[
\Phi = \frac{Q}{4\pi\varepsilon_0 r} e^{-\frac{r}{\lambda_D}},
\]

the Debye-Hückel [61] or Yukawa [62] potential. Compared to the Coulomb potential it is declining exponentially with the Debye-length \( \lambda_D \) as a characteristic length-scale. The latter can be written for each charge carrier individually as \( \lambda_{D,e/i} = \sqrt{\varepsilon_0 k_B T_{e/i}/(n_{e/i}e^2)} \) and depends on the temperature \( T_{e/i} \) and density \( n_{e/i} \) of the surrounding electrons and ions (indexed with \( e \) and \( i \) respectively). The overall Debye-length is a linearization of the individual components so that \( \lambda_D^{-2} = \lambda_{D,e}^{-2} + \lambda_{D,i}^{-2} \).

Due to the shielding local charge density deviations will be compensated within a Debye-length. Considering discharges larger than several Debye-lengths a global equilibrium of electron and ion densities will form in a plasma, the already mentioned quasi-neutrality. Assuming single-ionized charge carriers one can directly deduce \( n_e \approx n_i \) for their densities. To evaluate on which time scales local charge density deviations are compensated, one
2. Physical Background

needs to consider the thermal velocity \( v_{e/i} = \sqrt{\frac{k_B T_{e/i}}{m_{e/i}}} \). Assuming that a charge carrier needs to travel approximately one Debye-length to compensate charge fluctuations one can derive a screening time \( \tau_{e/i} \approx \lambda_{D,e/i}/v_{e/i} \) and a characteristic plasma frequency

\[
\omega_{P,e/i} = \frac{1}{\tau_{e/i}} = \sqrt{\frac{n_{e/i} e^2}{\varepsilon_0 m_{e/i}}},
\]  

(2.2)

which is of the order of GHz for electrons and of the order of MHz for ions in a typical low temperature plasma with densities of the order of \( 10^{15} \text{ m}^{-3} \). Electrons and ions can follow electric field fluctuations up to their respective plasma frequency. Above that, they react to the time-averaged field.

2.2. Radio frequency discharges

There are two ways of electrically driving low temperature plasmas in the laboratory: Either by using direct (DC) or alternating current (AC) voltages to ionize the supply gas. The method of applying the AC signal to the gas further distinguishes the latter into capacitive and inductively coupled (IC) discharges. Although there have been studies on dusty plasmas in many different plasma sources (see e.g. [15, 63, 64] for DC plasmas, [65–67] for IC plasmas and [68, 69] for a mixed type), the most common way of generating laboratory dusty plasma is the use of capacitive radio-frequency (rf) discharge that will now be presented in more detail.

It features two electrodes inside a vacuum chamber between which an electric field, like in a capacitor, is generated. By applying an rf-voltage of typically 13.56 MHz to the electrodes, the electrons can follow the field oscillation and thus are heated while the ions only see the time-averaged voltage due to their lower plasma frequency \( \omega_{P,i} \). As a consequence, a plasma is generated between the electrodes. Typically, the rf-voltage is sent through a coupling capacitor in series with the electrodes, hence the name capacitively-coupled rf-discharge.

Since a laboratory plasma is bounded, a transition zone between the plasma bulk and the containing walls establishes. The electric wall potential will become negative with respect to the plasma to retain the more mobile electrons in the plasma. The ions are accelerated towards the walls forming a positive space charge sheath where quasi-neutrality is broken. An important criterion for a stable sheath is that the ions must travel with a higher speed than the Bohm velocity \( v_B = \sqrt{\frac{k_B T_e}{m_i}} \). Otherwise the sheath region would expand into the plasma bulk in form of an ion acoustic wave [60]. The ions gain this velocity in the pre-sheath where quasi-neutrality is still given but the electron and ion densities start to decline.

For rf-discharges the applied alternating electric field results in a harmonic displacement of the electrons between the electrodes. The negative sheath potential reflects the electrons on the one hand and accelerates them upon expansion on the other hand. Effectively the electrons gain additional velocity from this so-called wave rider or \( \alpha \)-regime. They dissipate their energy to the neutral gas particles by collisions generating new ion-electron pairs.
2.3. Dusty plasmas

There are other important heating mechanisms which are discussed e.g. in [60] but for the experiments presented in this thesis the \( \alpha \)-regime is the dominant one.

2.3. Dusty plasmas

After introducing pure low temperature plasmas and their sources, what happens to dust particles that are injected into such environments? Usually, the particles gain negative surface charges up to several thousand elementary charges \([71–73]\) due to the sheath forming around them. Thus, the particles act as a third plasma species with number density \( n_d \) and charge number \( Z_d \) that modifies the quasi neutrality condition to
\[
 n_i = n_e + Z_d n_d.
\]

The ensemble of plasma and particles is often referred to as complex plasmas or dusty plasmas. Similar to the electron and ions one can also define a dust plasma frequency
\[
\omega_{\text{P},d} = \sqrt{\frac{n_d Z_d^2 e^2}{\varepsilon_0 m_d}},
\]
which is below 100 Hz for typical experimental parameters with micron-sized particles and marks the range of dust dynamics in the plasma environment.

2.3.1. Particle charging

The charging behavior of particles in a plasma is similar to that of a floating probe \([74–76]\) and can in a simplifying approach be described by the same orbital-motion-limit (OML) theory. In a nutshell, this approach calculates the ion and electron currents onto a spherical body by evaluating the collection cross-section of the charge carriers. Assuming that the sum of all currents vanishes once the floating potential \( \Psi_{\text{fl}} \) is reached and that the particle behaves like a spherical capacitor with radius \( a \) yields the charge
\[
 Z_d = 4 \pi \varepsilon_0 a \Psi_{\text{fl}},
\]
for particles with \( a \ll \lambda_D \). The floating potential typically is of the order of \( \Psi = -2 k_B T_e/e \). As a rule of thumb this can be translated to \( Z_d \approx 1400 \cdot a [\mu\text{m}] \cdot T_e [\text{eV}] \) for typical laboratory parameters. Due to its rather simple assumptions the OML theory can only give a rough estimate of the charge. For a detailed quantitative description one has to consider the following points:

- Collisions are not considered in OML. However, at typical gas pressures around 10 Pa collisions between ions and neutrals have a significant influence \([77]\). Upon a collision an ion transfers energy to a neutral atom resulting in a lower velocity. Therefore, it will be more likely to hit the dust particle thus the overall ion current rises. Once there are multiple collisions due to a high neutral density the current will decline again since the ions are now inhibited from reaching the particle \([78]\).
- OML assumes a Maxwellian velocity distribution for the electrons and ions. While this might be a good assumption in the plasma bulk, many dusty plasmas rely on trapping particles in the sheath region where this is no longer appropriate. In fact
2. Physical Background

the ions flow at least with Bohm velocity which is higher than the thermal speed. Therefore the ion current rather depends on the kinetic energy of the ions and their streaming velocity than their thermal energy and velocity \[79\].

- The high charging of the particles can lead to electron depletion around them, especially when the dust density is rather high \[72, 80\]. The local reduction of the electron density affects the electron current onto the particle lowering the resulting charge.
- An oscillating rf-sheath periodically changes the local electron density which will lead to an alternating charge on particles inside the sheath.
- The fact that the particle charge is obtained by collecting discrete charge carriers leads to a stochastic fluctuation of the overall charge \[71\]. This effect becomes more dominant for smaller particles.
- Electrons from photo or secondary emission affect the electron density as well but are considered rather unimportant for laboratory dusty plasmas.

Aside from these points the OML approach is still a good indicator for experimental dusty plasmas to gain a rough estimate of the particle charge.

2.3.2. Forces on particles in a plasma

A charged particle submerged in a plasma environment can be influenced by many different forces, which will be presented in the following list. A comparison of their respective strength in dependence of the particle radius \(a\) is shown in Fig. 2.1.

![Figure 2.1: Forces on a dust particles at typical plasma parameters, taken from [81].](image)

**Electrical force**

Electrical fields exert a strong force on highly charged dust particles in a laboratory
2.3. Dusty plasmas

plasma environment and are mainly used to confine particles in the discharge. It is defined as the product of particle charge and electrical field and can be described as

\[ \vec{F}_p = Q_d \vec{E} = 4\pi \varepsilon_0 \Psi_{fl} \vec{E}, \]

using the OML particle charge from Eq. 2.4.

Gravitation

The gravitational pull is the dominant force on typical melamine-formaldehyde (MF) particles with diameters larger than roughly 5 \( \mu \)m. Using the particles’ mass density \( \rho_d \) and the gravitational acceleration \( g \) the gravitation force yields for a spherical particle

\[ \vec{F}_g = m_d \vec{g} = \frac{4}{3} \pi a^3 \rho_d \vec{g}, \]

Neutral gas friction

In general a spherical particle moving through a gas with a velocity \( \vec{v}_d \) will experience a friction force

\[ \vec{F}_n = -\frac{4}{3} \pi a^2 m_n v_{th,n} n_a \vec{v}_d, \]

due to a transfer of momentum from the gas molecules with density \( n_n \), mass \( m_n \) and thermal velocity \( v_{th,n} \).

Ion drag

In a plasma, dust particles will interact with ions that are streaming past the particle. Besides the direct drag of the ions collected by the particle there will also be an additional force from ions that are only scattered by the Coulomb interaction. The modeling becomes more difficult than for neutral gas particles. A first ion drag model was proposed by Barnes et al. \[83\], where direct and indirect forces are given by

\[ \vec{F}_{dir} = \frac{\pi a^2 m_i v_{s,i} n_i \vec{u}_i}{m_i v_{s,i}^2} \left( 1 - \frac{2e \Psi_{fl}}{m_i v_{s,i}^2} \right), \]

and

\[ \vec{F}_{Coul} = 2\pi \frac{a^2 e^2 \Psi_{fl} n_i \vec{u}_i}{m_i v_{s,i}^3} \ln \left( \frac{\lambda_D^2 + b_{\pi/2}^2}{b_c^2 + b_{\pi/2}^2} \right), \]

respectively. Here, \( v_s = \sqrt{u_{th,i}^2 + v_{th,i}^2} \) is the mean velocity derived from the ion thermal velocity \( v_{th,i} \) and the ion drift velocity \( u_i \), \( m_i \) is the mass of the ions, \( b_c \) is the minimum collision parameter above which ions are not collected by the particle and \( b_{\pi/2} = ae \Psi_{fl}/m_i v_{th,i}^2 \) is the collision parameter for a deflection angle of 90°. The total ion drag force is the sum of both components. While the qualitative model proposed by Barnes et al. gives good predictions, it has its limitations. Hence, more refined, quantitative models have been proposed by Hutchinson \[84\] and Khrapak et al. \[85\] that shall only be mentioned at this point.

Radiation pressure

Using the momentum of photons it is possible to apply a force on macroscopic particles as well. A focused laser with intensity \( I \) will exert a force

\[ F_{rad} = \frac{\gamma I A_d}{c}, \]
on a particle with geometric cross section \( A_d = \pi a^2 \), depending on the dimensionless material coefficient \( \gamma \) that ranges between \( \gamma = 1 \) for pure photon absorption and \( \gamma = 2 \) for pure photon reflection [29] by the particle.

**Thermophoresis**

The last force to be mentioned here arises due to temperature gradients \( \vec{\nabla} T_n \) in the neutral gas environment. Evaluating the gas kinetics yields a force [80, 87]

\[
\vec{F}_{th} = -\frac{32a^2 k_n \vec{\nabla} T_n}{15 v_{th,n}}
\]

for a gas with thermal conductivity \( k_n \) that pushes the particles against the heat gradient. In laboratory dusty plasmas thermophoresis has become very useful to help counterbalance gravitation [88, 89].

2.4. Yukawa-balls

First (unintentionally) trapped particles have been observed by Selwyn [4, 5] in rf-plasma-etching devices where the dust was produced in-situ by the etching process. In this case the gravitation on the dust was counterbalanced by the sheath electric field while local minima in this field also provided a vertical confinement forming two dimensional dust rings. Following these findings it was possible to produce laboratory dusty plasmas ranging from very small, finite 1D or 2D systems containing less than 100 particles (see e.g. [90-92] or [8, 13, 14] respectively) to extended dust clouds with millions of particles filling the whole plasma bulk [16, 19, 93]. Exemplary images of these different dusty plasma types are shown in Fig. 2.2(a,b,d)

**Figure 2.2**: Different forms of laser-illuminated laboratory dusty plasmas: a) 1D chain seen from above [92]. b) 2D clusters seen from above [94]. c) 3D Yukawa-ball seen from the side. d) Vertical slice through a half of an extended dust cloud.

The object of investigation of this thesis lies between those examples as can be seen in Fig. 2.2(c): Yukawa-balls are finite three-dimensional dust clusters that exhibit a unique set of characteristics [31, 34, 95]. Being strongly coupled, forming crystalline structures while being observable via optical methods are just a few examples of their interesting qualities. This section will present the generation of these systems and their dynamical and structural properties.
2.4. Yukawa-balls

2.4.1. Confinement and Structure

The vertical equilibrium position of trapped Yukawa-balls is determined by the already mentioned balance between sheath electric field and gravitation. This balance can be altered via thermophoresis, an additional upwards pushing force generated by a heated electrode. Thus particles injected into the plasma will levitate above the electrode and the height can be influenced by the temperature of the electrode while the plasma parameters stay the same. To hinder the particles from drifting away horizontally additional forces are needed. These can be provided by placing a glass cuvette on the electrode [31] or via a brass confinement ring that will be presented in Ch. [3]. Both generate an inwards-facing electric field that confines the cluster in its center. By finding the right dimensions of the cuvette or ring combined with the gravitation and sheath force a 3D harmonic trapping potential [32] is generated.

The ground-state energy for such a trapped finite system [96] with particle coordinates \( \vec{r}_i \) and inter-particle distance \( r_{ij} = |\vec{r}_i - \vec{r}_j| \) can be written as

\[
E = \frac{m_d \omega_0^2}{2} \sum_{i=1}^{N} (\vec{r}_i)^2 + \frac{Z^2 e^2}{4 \pi \varepsilon_0} \sum_{i>j}^{N} \frac{e^{-r_{ij}/\lambda_D}}{r_{ij}},
\]

where the first term expresses the particles’ potential energy in the harmonic trap while the second term accounts for the Yukawa interaction between the particles. The energy of such a system of particles with a given charge only depends on the number of particles \( N \), the screening represented by \( \lambda_D \) and their arrangement. In an isotropic 3D confinement, the particle ensemble naturally will form a ball structure in order to minimize its ground-state energy [33, 97]. This ground-state is characterized by a nested concentric shell structure that determines the cluster’s strength, meaning that certain “magical” particle numbers exist where the collective structural stability is higher [33, 50]. However, it is also possible that a metastable configuration is reached in which the cluster stays for a certain time [36, 38, 39, 42, 98]. These not necessarily ball-shaped configurations are either formed initially upon injecting particles into the plasma or due to random inter-shell particle rearrangements if the thermal energy of the particles is high enough.

Another major influence on the cluster’s configuration are the streaming ions. As already discussed in Sec. 2.3.2 ions are moving towards the electrode and exert a drag force on the particles. Additionally, upon streaming around a negatively charged particle the ions are focused directly below it forming a so-called ion wakefield or ion focus [55–57]. The focus point itself is a positive space-charge zone which attracts other particles that produce wakefields again. As a consequence, particles tend to form vertical chains and the clusters are more cylindrical than spherical under the influence of ion wakefields [58]. An interesting property of these wakefields is that they act as a non-reciprocal force [49]. In a binary system a movement of the upper particle will move the ion focus and the lower particle trapped therein. However, the lower particle cannot influence the upper particle accordingly besides the normal Yukawa interaction. Additionally, the wakefield also leads to instabilities in the cluster [59] which affects the overall structure as well.
2.4.2. Crystallization and melting

After discussing the properties of highly ordered Yukawa-balls this section will present the requirements to form a solid-like structure. Generally, a strongly coupled system will form a crystalline structure once its Coulomb energy considerably exceeds its thermal energy. The ratio of these two energies is called the coupling parameter

\[ \Gamma = \frac{Z^2 e^2}{4\pi\varepsilon_0 b_{WS} k_B T_d}, \]  

where \( b_{WS} = \sqrt[3]{3/(4\pi n_d)} \) is the Wigner-Seitz distance that represents the inter-particle distance. The analysis of an infinite three-dimensional one-component plasma done by Ichimaru [100] revealed that a critical coupling parameter of \( \Gamma_{\text{crit}} \approx 168 \) has to be exceeded as a crystallization condition. Since a one-component plasma model neglects shielding effects one needs to modify the coupling parameter accordingly. An estimate given by Ikezi [6] adds a shielding term \( \kappa = b_{WS}/\lambda_D \) forming an effective coupling parameter \( \Gamma_{\text{eff}} = \Gamma \exp(-\kappa) \). Thus, the critical coupling parameters retains its numerical value of \( \Gamma_{\text{eff, crit}} \approx 168 \). However, there has been studies of more complex models that offer a better reproduction of the shielding effect [17, 101].

Once exceeding the critical coupling parameter larger dust clouds form crystalline fcc and bcc structures [16, 102] like solid-state bodies while finite Yukawa-balls exhibit a shell structure [97] in analogy to atomic clusters. However, a perfectly spherical shaped Yukawa-ball can only be observed at rather large gas pressures. At lower pressures, streaming ions [55] tend to deform the cluster into a more cylindrical shape [56, 58].

In order to investigate the crystallization and melting behavior one needs to change the systems temperature. This is possible by altering the plasma properties [14, 103] i.e. neutral gas pressure and rf-power, changing the number of dust particles [104] or transfer kinetic energy into the system via lasers [52, 105, 106]. As the latter is the tool of choice for this thesis, more detail on it will be given in Ch. 3. Insights into the dynamical behavior of clusters upon melting can be gained by either mapping the collective particle motions onto normal modes [44, 94, 107, 108] or by seeing the cluster as a uniformly charged droplet that is deformed by means of fluid modes [47, 47]. Both approaches yield mode spectra that allow to analyze the phase transition qualitatively [109]. Besides the cluster’s dynamics one can look into statistical information gained from its structure as well. Correlation functions in particular are very well suited for this task and will be presented in Ch. 4 as part of my work.

2.4.3. Open questions

Although the melting behavior of finite Yukawa-balls has already been investigated thoroughly, it is still difficult to determine the point of phase transition. Furthermore, the number of particles in a finite system should have an effect on this point as well, as is known for ion clusters [110]. In general the transition between smaller to larger finite systems and the impact on structure and dynamic is an interesting object of investigation. It was my aim to increase our knowledge of this transition zone as Ch. 4 will reveal.
3. Experimental Setup and Diagnostics

In the preceding chapter the general requirements and characteristics of finite Yukawa-balls were discussed. This chapter will now describe the experimental setup used for the research presented in this thesis including confinement, diagnostic and manipulation tools.

3.1. General setup

The plasma used for the experiments is provided by a capacitively coupled low temperature radio frequency (rf) argon discharge. It is usually operated at neutral gas pressures between 5 and 50 Pa and rf-powers below 2 W. The chamber shown in Fig. 3.1 is a well proven design that was used for various experiments before (see e.g. [38, 44, 52]). It has an inner volume of 4.7 l (without the electrode) with four large observation windows evenly distributed around the horizontal perimeter, two additional smaller manipulation windows along one horizontal diagonal and one circular window on top.

Figure 3.1: a) Diagonal cut through the rf-chamber: 1 - top view window, 2 - side view windows, 3 - diagonal manipulation windows, 4 - reflection reducing blackened aluminum electrode part, 5 - brass electrode part with cavities enabling water heating for thermophoresis, 6 - electrode shield, 7 - rf-Electrode, signal can be applied at the bottom outside the chamber (not shown). b) Beam path of the expanded illumination lasers (red) and focused manipulation lasers (green) converging below the confinement ring inside the chamber.

Inside the chamber the rf-signal is applied to a brass electrode with a diameter of 100 mm that can be heated via hot water for thermophoresis as described in Sec. 2.3.2.
this electrode lies a thinner black anodized aluminum plate that reduces laser reflection in order to raise the contrast of the camera images. Since the discharge is capacitively coupled, the additional plates’ influence can be compensated by a radio frequency matching box. The rest of the chamber is grounded to act as the counter-electrode forming an asymmetric plasma discharge.

The standard dust used for measurements is made of melamine-formaldehyde (MF). Recent findings that are part of this thesis (please refer to Article [A5]) showed that MF particles lose weight and shrink in the plasma environment. However, tests with other substances have been unsatisfactory as well: While polystyrene and polymethyl methacrylate dissolve even faster in the plasma environment, silica particles tend to agglomerate to larger clumps and have an inferior monodispersity. Therefore, MF is still a good choice to study Yukawa-balls when it comes to highly monodisperse particles and long-time observations. Typical particle diameters that have been used are 4.86\( \mu \text{m} \) or 4.7\( \mu \text{m} \) as they are known to produce spherical Yukawa clusters for our experimental conditions. They are injected into the plasma by a dust dropper which is basically a movable reservoir with a small hole in the bottom allowing to shake out the particles mechanically. The exemplary \( N = 39 \) particles cluster shown in Fig. 3.3 features a diameter of about 1.5 mm which is a typical measure for clusters in the \( N \approx 50 \) size-range. Larger cluster consisting of 1000 particles on the other hand can exhibit diameters of up to 10 mm.

3.2. Stereoscopy and manipulation

With a dust plasma frequency (Eq. 2.3) below 100 Hz and the overall cluster being optically thin due to the large inter-particle distance to particle diameter ratio, video microscopy is the preferred diagnostic for finite Yukawa-balls consisting of micron sized particles. With a sufficient optical magnification and a fast camera framerate, it is possible to investigate clusters on the individual particle level. Fig. 3.2(a) depicts the optical setup consisting of three CMOS cameras equipped with 200 mm macro lenses and additional extension tubes resulting in a resolution of about 100 pixels per millimeter in the focus area. Two cameras are mounted in the horizontal plane with an angle of 90\( ^\circ \) towards each other. The third camera is viewing from the top but is tilted by 22\( ^\circ \) with respect to the vertical Z-axis. Tilting the camera decreases the amount of overlapping particles seen from above that are generally vertically aligned due to the ion focus. Such camera arrangement is beneficial for the 3D reconstruction.

The cameras are able to record up to 10,000 frames into the RAM of the respective recording computer with a maximum framerate of 506 frames per second at 1280\( \times \)1024 pixels per image. 2D particle positions in the individual camera images are obtained using an intensity moment method [111]. The reconstruction of 3D particle positions from these 2D images is part of this thesis and a detailed description can be found in Article [A6] and in the work of Himpel et al. [112]. In a nutshell, the procedure is composed of three steps. First, a correspondence between the cameras has to be found, meaning their relative orientation is processed into a virtual camera system. Then, identical particles are searched along common sight lines in the different camera images. Finally, the obtained 3D positions
are compared over time to gain complete trajectories. For the camera correspondence a calibration target as shown in Fig. 3.3(a) is viewed by all cameras. The target is placed at the position of the Yukawa-ball in the plasma experiments. The target has to be rotated in the field of view so that different views of the target can be recorded. Using Matlab toolboxes [113–115] projection matrices are calculated from the calibration target views. From that, a virtual camera system can be generated that gives common sight lines in the different cameras. Then, one looks along those so called epipolar lines [116] to identify individual particles in all cameras. Finally, the particles’ 3D positions are obtained via triangulation. A nearest neighbor algorithm comparing adjacent frames yields trajectories [see Fig. 3.3(b)] and velocities.

Figure 3.2: a) Optical setup: 1 - CMOS camera with attached 200 mm macro lens and additional extension tubes, 2 - galvanometric scanners fed by two green DPSS lasers, 3 - rf-chamber (details in Fig. 3.1), 4 - fiber coupled red DPSS lasers for illumination.

To illuminate the particles, two expanded fiber coupled diode pumped solid state (DPSS) lasers are used that operate with a laser output power of up to 1 W at a wavelength of 660 nm. The lasers are mounted opposite to the two horizontal cameras. The reason for this arrangement is that the forward scattered light has the highest intensity for micron sized particles and the CMOS cameras are most sensitive in the red spectrum. To prevent damage from the camera sensors the lasers are aligned so that they miss the camera lenses while giving as much forward scattered light as possible.

An important tool to heat finite Yukawa-balls are manipulation lasers [52, 105, 106, 117, 118]. The basic idea is that a laser can exert a radiation force on a dust particle as
described in Sec. 2.3.2. Initially, particles hit by the laser beam gain kinetic energy along the beam and travel in its direction. Due to the Yukawa interaction other particles will experience a force exerted by Coulomb collisions in various directions. Thus, although laser energy is fed from only one direction the particles’ kinetic energy dissipates in the cluster in all directions. To mimic a heating process the laser is swept stochastically over the whole cross section of the cluster with a random pattern applied via galvanometric mirrors. Now, adjusting the laser power allows one to tune the cluster’s “temperature” externally while leaving the plasma parameters undisturbed. In the evaluation process the mean squared velocity is usually taken as a measure for the kinetic temperature. Strictly speaking this assumption is only valid for a Maxwellian velocity distribution. While this was found to be accurate in 2D systems [105, 118, 119], studies on 3D Yukawa-balls revealed an only near-Maxwellian distribution [52]. However, the mean squared velocity is still a good approximation and has proven to be the best measure for the temperature of finite Yukawa-balls.

In the experiment the heating is done by two additional DPSS lasers mounted on two galvanometric scanners. They are placed along the diagonal of the two horizontal viewing axes to avoid having the axis of particle movement parallel to the horizontal sight lines. The highly focused manipulation lasers can each apply radiation powers of up to 600 mW at a wavelength of 532 nm. The different wavelength compared to the illumination lasers allows to block the light from the observation setup via longpass filters. All manipulation and illumination lasers converge in the focus area of the cameras under the confinement ring where the Yukawa-ball is trapped as can be seen in Fig. 3.1(b).

Figure 3.3: a) Unprocessed camera images from the setup in Fig. 3.2 and additional view on the calibration target. b) Retrieved trajectories of 39 MF particles moving for 10 s at 14.5 Pa argon pressure and 0.8 W rf-power.
3.3. Confining the dust particles

As stated in Sec. 2.4, Yukawa-balls were first generated in a setup using a glass cuvette that provided an inwards facing radial electric field. Although this approach is robust and rather easy to implement since the cuvette can be placed directly onto the electrode of an existing rf-chamber, it has its shortcomings: The usage of glass through which illumination lasers and scattered light have to travel necessarily leads to unwanted reflections, speckles, ghost images and artifacts from pollutants attached to the glass surface. The latter is especially problematic since dust has to be injected into the cuvette in order to form Yukawa-balls. Thus, the cuvette will eventually become contaminated by the injected particles. Also, the glass will heat up locally due to the laser exposure producing neutral gas flows that can disturb the dust particles. Finally, the confinement potential of a glass cuvette can only be modified by altering the overall plasma parameters. If that does not lead to a desired harmonic confinement or the confinement is not harmonic at a given set of plasma parameters, another cuvette with different dimensions has to be used.

To overcome these limitations I experimented with cylindrical brass cylinders as shown in Fig. 3.4(a) that provide a similar confinement as the glass cuvette but leave the optical path free. Further investigation revealed that just a single brass ring is sufficient to create an excellent trap for Yukawa-balls underneath. The final version shown in Fig. 3.4(b) is attached to an arm that is mounted on a linear vacuum feedthrough at the chamber’s top allowing to alter the position of the ring while operating the experiment. Arm and ring are electrically isolated by a polyether ether ketone (PEEK) connector leaving the latter at floating potential. To account for the asymmetric field produced by the arm three additional brass rods were installed around the ring’s perimeter.

![Figure 3.4](image)

Figure 3.4: a) Prototype of a new confinement cuvette made of brass, shown above the thermophoresis electrode. b) Final design of the new confinement ring mounted at the chamber’s top.

The optical benefits of the new confinement setup can be reviewed in Fig. 3.5 where unprocessed camera images of two example clusters trapped by a glass cuvette (a) and by the brass ring (b) are shown. Obviously, the free optical path in the brass ring setup results in a much cleaner image compared to the previous design using glass. The other not so
obvious benefit is the adaptability of the confinement potential. By raising or lowering the ring the radial confinement at the point where the electrical sheath force counterbalances gravitation changes drastically. This allows to find harmonic trapping potentials for a larger set of plasma parameters without the need to change the cuvette. Additionally, the usage of conducting brass as a material makes it also possible to apply an external DC potential which further broadens the applicability of this new confinement setup although it was not used in this thesis.

Figure 3.5: Unprocessed camera images of Yukawa-balls confined by a glass cuvette (a) and a brass ring (b).

3.4. Movable rf-chamber

Hitherto existing setups rely on movable cameras and laser beams in order to focus particles trapped in a plasma chamber. Unfortunately, the position of clusters in the chamber is not fixed due to changing plasma parameters or shifting of the confinement ring or glass cuvette after opening and closing the chamber. Therefore all optical elements have to be aligned again for a new setup which is rather time consuming considering up to four cameras, two illumination lasers and two manipulation lasers. To account for this, more advanced systems connect all optical parts mechanically and mount the overall system on rails in order to shift the common focus (see e.g. [32, 93, 112, 122]). But again, this becomes more and more challenging engineering-wise as one adds more optical components and wants to distribute them all around the chamber.

Therefore I decided to develop a setup where the chamber is mounted on moving stages while keeping the optical components fixed. The result is shown in Fig. 3.6 where one can see the self-constructed X-Y-Z-stage with the rf-chamber on top. The horizontal X-Y-assembly consists of two aluminum plates with central cutouts that allow to connect rf-voltage, neutral gas inlet and vacuum outlet to the chamber from below the setup. The upper plate has four sliders attached below that run on two profiled linear rails. The rails are mounted on the lower plate which features a similar but perpendicular slider-rail setup below. Its rails, in turn, are attached to the main experiment table. With this setup one can already move heavy loads in all horizontal directions. Together, the two stages
build the foundation for the housing of the vertical Z-stage mounted on top. Therein, the rf-chamber is attached via a connector plate to four sliders that each run on individual vertical rails allowing a displacement in the Z-direction. The individual stages are driven by electric actuators giving the advantage of remote controlling the focus point without needing to intrude the laser safety environment.

Figure 3.6: Movable X-Y-Z stage: 1 - rf chamber, 2 - housing for vertical Z-stage, 3 - horizontal Y-stage, 4 - slider mounted on profiled linear rail, 5 - horizontal X-stage, 6 - electric actuator.

The overall movement range is 50 mm in each horizontal direction and 100 mm in vertical direction. Considering typical cluster diameters between 1 mm for 15 particles and 9 mm for 1000 particles this range is more than suitable for experiments on finite Yukawa-balls. The structure is able to lift about 30 kg with the vertical actuator being the limiting factor which is enough to support and move our plasma chamber securely. The top-mounted plasma chamber dives through the opening of the fixed optical table as seen in Fig. 3.2(a). After all cameras and lasers are aligned onto a common viewing volume one can fix their position. Thus, a movement of the chamber results in an effective shift of this viewing volume in reference to the confinement zone.

3.5. Benefits

Summing up, the tools described in this chapter are well suited for the investigation of Yukawa-balls. Using well proven parts like the plasma chamber as well as the illumination and manipulation systems while adding own components like a more accurate stereoscopic imaging system, a translation stage and an improved particle confinement generated a
perfect setup for observing and manipulating finite Yukawa-balls.

On the one hand the more accurate way of calibrating the camera setup, the better image quality from the use of a brass ring instead of a glass cuvette as well as fixing all focused optical components while moving the rf-chamber yield more reliable reconstructed 3D particle positions. Observations of up to 1000 particles are now possible using a volumetric illumination compared to viewing only laser slices. Thus, 3D trajectories of particles in very large clusters are now obtainable making it possible to investigate the transition zone between very small to larger systems. On the other hand, the possibility to observe clusters at different temperatures via laser heating over long timescales improves the ability to evaluate statistical properties, e.g. correlation functions.

The use of the described setup for investigation of practical physical problems will be presented in the following chapter.
4. Results

This chapter is dedicated to the results of my experimental investigations. First, some basic research on finite Yukawa-balls will be presented. This is followed by a detailed description of Articles [A7] and [A8] where the phase transition of Yukawa-balls and collective vortex motions were investigated respectively. Finally an insight into my yet unpublished research concerning cluster expansion dynamics will be given.


In the beginning of my research I focused on the dynamics of heated and unheated finite Yukawa-balls resulting in my share of work for Articles [A1] and [A2]. There, 2D and 3D clusters were melted via laser heating. In the evaluation process, different analysis techniques were applied to investigate the dynamic behavior of the clusters. Of importance for my later research was the usage of configurational entropies in Article [A1]. The entropies were determined from analyzing shell occupation changes over long time scales to get probabilities for different configurations. Here, different configurations mean different particle numbers on the concentric shells of the Yukawa-ball. From that, Shannon entropies were calculated that showed a clear dependence of the clusters’ particle numbers. This pioneered Article [A7] where configurational entropies are calculated from correlation functions as Sec. 4.2 will present in more detail. Article [A2] on the other hand focused on investigating fluid and normal modes that were already introduced in Sec. 2.4.2. It was shown that both approaches give insight into the particle interaction and the isotropy of the confinement making them perfect tools for the investigation of cluster dynamics. Notable findings were the different properties of spherical and elongated cylindrical clusters formed due to the ion focus. It was pointed out that the ion focus has a non-negligible impact on the dynamical behavior of the cluster and has to be considered upon analysis. This knowledge was picked up in Article [A7] as well, where new correlation functions were derived from spherical models to account for vertically aligned structures.

Further effects of the cluster-elongating ion focus were studied in Article [A3]. Here we analyzed instantaneous normal modes, a modified version of the normal mode analysis, where not the averaged equilibrium particle positions but each configuration observed in the individual time frames are used. This approach yields a density of states that can be divided into a stable and an unstable part allowing a dedicated configuration stability analysis. It was found that few-particle systems under the influence of ion focus can be classified into a stability spectrum ranging from unstable over oscillatory to stable. These regimes are determined by the strength of the ion focus where a strong ion focus accounts for stable systems while a weak focus leads to unstable configurations. Applying the
approach to clusters with higher particle numbers showed that the same effects also have an influence on the stability of larger systems. Understanding the impact of a present ion wind was of particular interest for the subsequent Article [A8] where the cause of collective vortex motions of larger clusters are attributed to the same effect.

After investigating the heating dynamics of finite Yukawa clusters, the question arose how a cluster relaxes into equilibrium after an external heating mechanism is switched off. This can be accomplished by turning the lasers off and observing the recrystallization process as described in Article [A4]. A time-resolved analysis of the Coulomb coupling parameter (see Eq. 2.13) revealed an initial cooling phase in which the coupling parameter rises exponentially followed by a phase of oscillatory convergence towards the equilibrium coupling parameter. By comparing the results with a time-resolved pair correlation function it became obvious that the correlation build-up is slower than the cooling indicated by the coupling parameter. This was accounted to a fast initial shell-formation that affects correlation and coupling followed by individual intra-shell particle ordering that keeps the correlation value down while the more global coupling parameter is already converging towards equilibrium. The basic idea of switching off external influences on the cluster and observing the systems response was picked up again later for investigating the expansion behavior of Yukawa-balls that will be presented in Sec. 4.4.

Besides this fundamental research I participated in the construction of more applied experimental methods. A phase-resolved-measurement setup based on the work of Carstensen et al. [123] was used for Article [A5] where the size distribution of dust particles was measured over time. The result was already mentioned in Sec. 3.1. Particles made of melamine-formaldehyde tend to decrease in mass and size over time in the plasma environment. As a possible cause chemical etching of the particles by oxygen radicals was identified. The overall stereoscopic setup described in Sec. 3.2 and the knowledge gained from it construction and operation was incorporated in Article [A6]. The outstanding performance of the calibration and particle reconstruction approaches described there enabled us to investigate larger dust clouds beyond 100 particles while using volumetric illumination and lead to the findings in Article [A8].
4.2. Phase transitions and configurational entropies - Article [A7]

The melting dynamics of finite Yukawa-balls have already been investigated in-depth as Sec. 2.4.2 made evident. However, there, phase transitions have been analyzed in a phenomenological way [52, 103, 124, 125] while a general quantitative melting criterion for finite Yukawa-balls lacked. As is known from thermodynamics, phase transitions feature a distinct jump or a steep increase in the entropy, depending on the order of the transition, that could be used to determine the melting point. Following this fundamental idea, Thomsen and Bonitz [126] derived configurational entropies from particle correlation functions to provide a more quantitative approach. By analyzing simulated 3D Yukawa-balls, Thomsen and Bonitz concluded that the phase transition of such systems indeed exhibit a distinct entropy jump which can be used as a general melting criterion. The first objective of Article [A7] was to test these findings experimentally and evaluate the configurational entropy approach for laboratory 3D Yukawa-balls.

In a system of particles featuring Coulomb or Yukawa interaction, the coupling parameter (see Eq. 2.13) is the appropriate measure for the phase state as already discussed in Sec. 2.4.2. Being basically an inverse temperature the method proposed by Thomsen and Bonitz allows to identify a critical coupling parameter at which the cluster changes its phase state. Another interesting prediction concerning the point of phase transition of finite ion clusters has been provided by Schiffer [110]. His simulations of clusters of ions under Coulomb interaction showed a dependency between cluster size and melting point. Evaluating whether this size dependency can also be found for particle systems under Yukawa interaction in the laboratory was the second objective of this Article.

A detailed description of the techniques and assumptions needed to accomplish the two presented objectives will be given in the following summary of Article [A7].

Figure 4.1: Sketch of a two-shelled Yukawa-ball illustrating the coordinates used for the C2P and TCF in a) spherical symmetry and b) cylindrical symmetry.
4. Results

The correlation functions used by Thomsen and Bonitz, namely the center-two-particle correlation function (C2P) and the triple correlation function (TCF) were derived for spherical Yukawa-balls. While the C2P takes all possible particle pairs into account, the TCF scans over all particle triples on the same spherical shell [see Fig. 4.1(a)]. The spherical symmetry of the C2P \( r_I, r_{II}, \theta \) and TCF \( \theta_I, \theta_{II}, \phi \) as used in [126] is fundamental to their approach. Analyzing simulated Yukawa-balls, Thomsen and Bonitz found that the corresponding correlation maps give insight into the periodical structure of the clusters. In particular, the C2P mainly represents the intershell arrangement while the TCF preferably visualizes the intrashell structure. From these correlation functions Thomsen and Bonitz derived Shannon configurational entropies

\[
S^{(2)} = -\langle \ln \text{C2P}(r_I, r_{II}, \theta) \rangle \quad \text{and} \quad S^{(3)} = -\langle \ln \text{TCF}(\theta_I, \theta_{II}, \phi) \rangle
\]  

(4.1)

by averaging over their logarithmic value. Evaluating these entropies over a broad temperature range of the same cluster yielded the already mentioned melting criterion: A distinct jump in \( S^{(n)}(\Gamma) \) indicates the phase transition. Compared to the approach in Article [A1], where the shell occupation is used to derive entropies, here additionally the particle arrangements on and between the shells are taken into account leading to a finer identification on melting points.

As already discussed in Sec. 2.4.1, streaming ions cause particles to arrange in more elongated structures featuring a cylindrical shell configuration. A projection of these elongated experimental cluster structures onto the spherical geometry produces an artificial fine-structure in the correlation maps. Upon heating, this fine-structure is highly fluctuating and produces artifacts in the computed entropies. Therefore, we have adapted the C2P and TCF to a cylindrical surface as shown in Fig. 4.1(b). The new C2P uses cylindrical coordinate radii \( R_I \) and \( R_{II} \) seen from the cluster’s center column and the azimuthal angle \( \Omega \) between the corresponding vectors. The adapted TCF on the other hand utilizes the distances \( L_I, L_{II} \) between particle 2 of the triplet 1-2-3 and the other two particles (as indicated in the sketch) and the corresponding bond angle \( \Psi \) on the curved shell surface for the TCF. With these coordinates one scans over all possible particle pairs and triples in a cluster, respectively, to gain a correlation map for a specific time frame. This is repeated for many time steps in order to capture as many metastable and ground state configurations as possible resulting in a good statistical base. Finally, Shannon configurational entropies can be derived from the adapted correlation function in the same way as Eq. 4.1 indicated. To measure the critical coupling parameter \( \Gamma_C \) at the phase transition, the same cluster is heated to different temperatures and the resulting entropies \( S^{(n)} \) are determined for the corresponding coupling strengths.

The experimental setup used for these investigations was already presented in Ch. 3. In short, three cameras are used to observe a cluster heated by two manipulation lasers. By tuning the laser power it was possible to change the cluster’s temperature driving the cluster from a solid-like to a liquid-like state. For each laser-controlled temperature the 3D particle positions were determined over 5000 frames. From the reconstructed particle positions correlation functions as shown in Fig. 4.2 were calculated. With the correlation functions being 3D, integrating over one variable was necessary in order to visualize the
correlation map. For the C2P that scans over all particle-pairs, it is beneficial to integrate over the radial distance of the first particles \( R_I \) in the borders of each cylindrical shell resulting in one hemispheric plot per occupied shell. Thus, for the two-shelled 39-particle cluster shown in Fig. 4.2 two different correlation plots can be generated using the integration borders indicated by the black arrows. Considering the cylindrical symmetry such a C2P map basically gives a top view on the particle correlations “seen” from the respective shell over which has been integrated. More specific, the left hemispheric plots in Fig. 4.2 show the probability to find a second particle at position \((R_{II}, \Omega)\) with respect to an arbitrary first particle on the inner shell. In the solid-like case (a), distinct correlation islands with respective angles of 90° can be identified on the first shell reflecting the 4-fold cylindrical symmetry of the cluster’s inner shell. Looking at the second shell for the same cluster seen in the right hemisphere, a 9-fold symmetry can be found following the same deduction. Since 9- and 4-fold symmetries share no common divisor the respective shell over which was not integrated appears as a continuous band instead of individual islands as it is equally probable to find a second particle at all \( \Omega \) angles. Looking at the heated cluster in (b), the loss of distinct correlation islands becomes evident. Because the particles have more kinetic energy, they are more probable to appear between previous lattice positions and shells resulting in the smeared-out correlation maps. A similar structural analysis can be conducted for the TCF that gives an insight into the correlation on the unraveled cylinder surface. Since the findings concerning the structural changes upon
heating as well as the temperature-dependent trend of the derived entropy is similar, it will not be discussed in detail here, but can be reviewed in Article [A7].

The described correlation functions have been measured for different clusters with particle numbers ranging between $N = 17$ and $N = 72$. In succession, entropies have been calculated from the correlation functions. A comparison between the C2P entropies $S^{(2)}$ of clusters with $N = 17$ and $N = 39$ particles is shown in Fig. 4.3(a). For both clusters a steeper $S^{(2)}(\Gamma)$ region near $\Gamma \approx 350$ is found. We interpret this steeper rise as the point of phase transition. It is also seen that the critical coupling parameter at phase transition, indicated with dashed grey lines, is not the same for the two clusters. To evaluate the findings, the same procedure was applied to two more clusters with $N = 18$ and $N = 72$ particles. It is then seen that the critical $\Gamma$ decreases with particle size. For ion clusters (with Coulomb interaction) Schiffer [110] observed the same behavior. Moreover, he found that the inverse critical coupling parameter, i.e. the transition temperature, scales linearly with the fraction of particles on the cluster surface (which increases for smaller particles). Hence, all our measured critical coupling parameters were inverted to indicate the transition temperature and were plotted against the fraction of particles on the clusters’ surfaces resulting in Fig. 4.3(b). Comparing the experimental results with the theoretical predictions by Schiffer (indicated by the dashed line) shows that the general trend is reproduced rather well, but with a significant offset. Possible reasons we considered for this deviation are summarized in the following list:

- $\Gamma$ depends on the particle charge which we calculated via normal mode analysis (see Sec. 2.4.2 and Article [A2]). The consequential error-offset in $\Gamma$ has been estimated to be 30%, which copes rather well with the distance between Schiffer’s prediction and our results.
- Schiffer investigated simulated ion clusters above 100 particles while our clusters feature particle numbers below 100. It is a reasonable scenario that the linear trend
• In our size-regime “magical numbers” have a huge influence on the stability of Yukawa-balls. Such “magical” configurations feature a much higher transition temperature compared to non-magical clusters [42].

• Streaming ions cause wake-instabilities that have a huge impact on the cluster’s phase-state as discussed in Article [A3]. As this is a plasma-effect, it was not considered in Schiffer’s simulations.

Aside from the discussed offset, we have observed a clear size dependence of the point of phase transition with smaller clusters having a lower transition temperature than larger clusters which is in agreement with the ion-cluster simulations.

Concluding, in Article [A7] we have shown that it is possible to determine phase transitions by evaluating entropies generated from particle correlation functions. We have modified the C2P and TCF proposed by Thomsen and Bonitz [126] to account for spherical clusters formed due to an apparent ion focus. Finally we have shown that the transition temperature of finite Yukawa-balls depends on their respective particle numbers. This is especially interesting since this behavior is a link between dusty plasmas and solid state physics. The transition zone between very small atomic or ionic clusters to large solid state bodies features a size-dependent transition temperature as well. Thus we have been able to reproduce the behavior of such very small systems with our macroscopic and optically observable Yukawa-balls.
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4.3. Collective vortex motions - Article [A8]

Besides the changed interaction in clusters, the ion wind also exerts a shear force on the cluster. For particle numbers higher than 100 the induced forces are strong enough to overpower the solid-like cluster structure resulting in a collective vortex motion. The drive and the influence of the cluster size on the vortex motion as well as general vortex parameters have been studied in this article.

Figure 4.4: 3D Trajectories (left half) and color coded number density profiles (right half) of clusters performing a vortex motion with a,b) \(N = 151\) and c,d) \(N = 1100\) particles.

The setup used for investigations is a modified version of the one in Fig. 3.2 featuring no manipulation lasers but an additional fourth camera. It is mounted next to the top camera but its tilting axis is rotated about 90° degrees around the z-axis. The extra camera helps improving the reconstruction of 3D particle positions described in Sec. 3.2. Even though a three-camera setup is able to identify particles via the epipolar line approach, a high number density eventually deteriorates the algorithm’s performance. Here the fourth camera helps to identify shadowed or ghost particles permitting a more reliable reconstruction. With that setup we were able to reconstruct the 3D positions of clusters with up to \(N = 1100\) particles.

For the characterization of the vortex motions seven different clusters with particle numbers ranging from \(N = 47\) to \(N = 1100\) have been recorded for 20s (2000 frames).
4.3. Collective vortex motions - Article [A8]

A collective vortex motion was only visible after reaching a cluster size of $N = 97$ giving a first hint on the size-dependent properties of the motion. 3D particle trajectories for two exemplary clusters with $N = 151$ and $N = 1100$ can be seen in Fig. 4.4(a,c). For clarity only 5\% (a) and 2\% (c) of the trajectories are shown respectively. In both cases the vortex motion is visible rather clearly. In the center of the cluster the particles stream downwards and after reaching the bottom of the cluster they stream upwards in the outer radial perimeter. However, a distinct difference in the motion of these two clusters becomes apparent: In the smaller cluster the particles are not moving steadily but rather jump between metastable lattice points, a behavior that cannot be found in the larger cluster. This observation is backed by the number density profiles shown in Fig. 4.4(b,d) where the densities were integrated over the azimuthal angle. While the larger cluster (d) shows a smooth density profile, the smaller cluster exhibits clear areas of higher density corresponding to places where particles linger. Since both clusters are confined in the same plasma environment this effect can only be attributed to the number of particles in the clusters. As already presumed in the beginning, the interplay between cluster structure and shear instabilities shifts towards instability with enlarging clusters.

![Figure 4.5: Color coded velocity components $v_x$ (a) and $v_z$ (b) calculated in different horizontal slices of a $N = 528$ cluster.](image)

To further analyze the flow field of the dust components we divided the clusters into horizontal slices and calculated corresponding velocity components in these layers. The horizontal ($v_x$) and vertical ($v_z$) components in a $N = 528$ particles cluster are shown in Fig. 4.5 and confirm the vortex motion’s dynamics that has already been discussed on the trajectories’ basis. A particle at the central bottom of the cluster will first flow radially outwards then bend its trajectory upwards followed by another inwards bend at the top to finally flow vertically downwards, back to its starting point. The fact that the velocity in Fig. 4.5(b) is symmetric around the z-axis suggests that the particle motion essentially takes place in the $\rho - z$ plane (considering cylindrical coordinates). An analysis of the vorticity $\vec{\omega} = \nabla \times \vec{v}$ revealed that only the $\omega_\phi$ component shows a significant rotational field. Thus, the motion is mainly poloidal and a further look into $\omega_\phi$, shown in Fig. 4.6(a), is appropriate. There, vertical slices through the vorticity’s $\phi$-component are shown in 3D with the strength of the rotation colorcoded. Aside from a central column, the vorticity is rather homogeneous and negative. This is in agreement with the already
4. Results

described particle motions and indicates a rigid-body rotation. To investigate this further, we averaged $\omega_\phi$ over $\phi$ and $z$ in order to get a radial vorticity profile. Repeating this for various cluster sizes between $N = 93$ and $N = 1100$ revealed the size dependency of the clusters’ vorticity shown in Fig. 4.6(b). While larger clusters with $N > 528$ perform a rigid body rotation indicated by rather constant vorticity values smaller clusters show a more ragged and unsteady vorticity profile. Here again the influence of metastable lattice positions resulting in a solid-like behavior come into play.

Figure 4.6: a) Vorticity component $\omega_\phi$ in different horizontal slices of a $N = 528$ cluster. b) Cluster size dependency of the averaged vorticity $\bar{\omega}_\phi$. The curves of the individual clusters have a relative offset of $10 \text{s}^{-1}$.

Besides investigating the size dependency of the vortex motion we also wanted to determine its drive. A strongly coupled dusty plasma system like the one we investigated can be considered incompressible. Using the vorticity $\vec{\omega}$, the dynamical flow field for dust particles with mass $m$ and charge $Z$ can be described by the Navier-Stokes-equation \[127, 128\]

$$m \beta \vec{\omega} = e \nabla Z \times \vec{E} + \nabla f(E) \times \vec{E}. \tag{4.2}$$

Here, $f(E)$ is the ion “mobility” function \[129\] which connects the ion drag force $\vec{F}_i = f(E) \vec{E}$ with the electric field. This description shows that a vortex is driven by non-vanishing curls in the electric field force (first term) and in the ion drag force (second term). Additionally, gradients in the charge $\nabla Z$ or in the ion mobility function $\nabla f(E)$ respectively are required for the two possible drives. Since the observed motion is poloidal, the corresponding gradients have to be radial. As shown by Bockwoldt et al. \[127\], a radial charge gradient is generated by changes in the shielding length. However, for our rather small clusters, compared to the large clouds investigated by Bockwoldt et al., we expect the shielding to be constant over the radial extent. Thus, we presumed that charge gradients can be excluded as the dominating drive of our observed vortices. Radial gradients in the ion mobility function on the other hand are very possible due to our horizontal confinement utilizing electric sheath forces. Considering only a poloidal rotation driven by radial gradients of the ion mobility function, the Navier-Stokes-equation becomes

$$m \beta \omega_\phi = E \partial_\rho f(E) \tag{4.3}.$$
From this, we were able to calculate a gradient scale length $f(E)/(\partial \rho f(E))$ which turned out to be of the same order as the size of the confinement ring. This supports the assumption that the confinement leads to a radial gradient in the ion wind that drives the vortex motion.

Summarizing, in the article we have observed vortex motions of up to 1100 particles. Compared to previous investigations [127, 128], where dust tori with a dust free region in the center were observed via 2D slices, we have been able to observe the axial symmetry of the vortex motions in a compact Yukawa-ball in full 3D. By applying different analysis techniques we have been able to characterize the collective motion on the one hand and find size-dependent properties in the clusters vorticity on the other hand. By analyzing the underlying mechanics, we have successfully attributed the motion drive to the ion drag force.
4. Results

4.4. Expansion dynamics

While the previously presented investigations focused on the size-dependent properties of very small \( N \leq 72 \), Article \[ A7 \] and medium sized clusters \( 93 \leq N \leq 1100 \), Article \[ A8 \], the following yet unpublished research concentrated on a phenomenon found in very large clusters with particle numbers \( N \gg 1000 \). Here, the shielding effect of the Yukawa interaction becomes dominant for the collective dynamics of the cluster since the cluster size exceeds the screening length. In particular, the expansion behavior of such large systems is significantly different compared to systems without shielding, i.e. Coulomb clusters. A foregoing investigation by Piel and Goree \[ 130 \] based on simulations showed that a cluster with Coulomb interaction explodes self-similar while a Yukawa cluster rather will expand layer-wise. This means that in the Coulomb case all particles start to fly radially outward simultaneously while the declining density stays constant over the whole cluster. The shielding in the Yukawa case however prevents the inner particles from experiencing the lost confinement. Thus, while the outer particle layers will start to move radially away from the cluster, the inner layers stay in place resulting in a faster declining density in the outer layers compared to the inner layers. The aim of my work was to investigate this behavior experimentally in order to test the finding of the simulations.

**Figure 4.7:** Sketch of the modified confinement ring used to switch off the horizontal confinement by moving away the two separable ring halves that are pivot-mounted on the chamber top. The particles are illuminated by an expanded horizontal laser sheet allowing to observe a central cluster slice from the top.

For this, the particle confinement setup [see Figs. 3.1(b) and 3.4(b)] was modified in such a way that the new ring consists of two separable halves that are individually attached to the chamber top as drafted in Fig. 4.7. Now, by turning both mounting poles that are accessible outside the vacuum via linear feedthroughs, the halves can be turned away from the trap center while effectively eliminating the horizontal confinement. Since the characteristic timescale for dynamic changes in the confinement that the particles can follow is about 10 ms, turning the ring-halves in our experiment away faster than that should appear for the particles as an instantaneous loss of horizontal confinement. The expansion is observed with one top camera that sees a horizontal slice through the middle of the cluster. Compared to Article \[ A8 \] a 3D investigation of the cluster was not possible due to the limited depth of field in the observation volume.
4.4. Expansion dynamics

In the experiment only the horizontal confining forces could be switched off by removing the confinement rings, whereas in the simulations of Piel and Goree a loss of the complete 3D confinement was studied. Hence, in the experiment the particles do not expand uniformly in all directions but eventually collapse into a 2D layer while expanding horizontally. Therefore, alongside with the experiment, I have performed simulations similar to Piel and Goree’s using a Langevin molecular dynamics code. In my simulations an initial cluster in its ground state experiences the elimination of horizontal confinement only under the effect of Coulomb or Yukawa interaction. The results for the spatio-temporal evolution of the particle number density in the central horizontal sheath are shown in Fig. 4.8. Fortunately, the clusters show an expansion behavior similar to the 3D case: The ground-state clusters in the harmonic trap show a uniform density distribution for the Coulomb case and a radial density gradient under Yukawa interaction. After switching off the horizontal confinement the Coulomb cluster shows a vertical expansion front in Fig. 4.8(a) (indicated by dotted black lines) meaning that the density declines uniformly over time in the whole cluster. The Yukawa-ball however features a temporally delayed expansion front visible in Fig. 4.8(b) due to the shielding effect already described. Hence, the different expansion dynamics could be tested in our experiments.

The observed cluster consisted of several thousand particles arranged in a rather oblate structure, which was beneficial for our observation of a more horizontal expansion. An overlay of several particle positions seen from the top camera with color-coded timing is depicted in Fig. 4.9(a). The typical behavior of a Yukawa expansion is already visible: While the inner particles only move about 100 pixels in the shown 100 frames, the outer particles are able to travel several hundreds of pixels in the same time, partly out of the observation volume. By analyzing the spatio-temporal density evolution in Fig. 4.9(b) one finds an inclined expansion front similar to the simulated Yukawa case shown in Fig. 4.8(b). This can be explained by the shielding effect that delays the inwards propagation of the

Figure 4.8: Simulated spatio-temporal density evolution after switching off horizontal confinement for Coulomb (a) and Yukawa (b) particle interaction. The confinement was switched off at $t = 100$ as indicated by the black arrows. The following expansion fronts are highlighted by dotted black lines.
density wave generated by the confinement loss. It can be concluded that the experimental observations back the findings of Piel and Goree as well as our own simulations. The expansion dynamics of particles under the influence of shielding is distinctly different compared to a non-shielded Coulomb cluster.

**Figure 4.9:** a) Particle positions at different color-coded times seen from above. b) Experimental spatio-temporal density evolution after switching off the horizontal confinement. The densities have been smoothed by a Savitzky-Golay filter along the temporal axis. The expansion front is again indicated by a dotted black line.

In addition to the shielding, collision between the dust particles and neutral gas atoms have an influence on the expansion dynamics as well. Piel and Goree \[130\] found in their simulations, that collisional expansions of Yukawa-balls exhibit diffusion process characterized by slower dynamics compared to the collisionless case. The model they derived to describe an expansion against friction yields three predictions for the spatio-temporal density evolution: (1) the spatial profiles are inverted parabolas, (2) the maxima of the profiles decay proportional to \( t^{-3/5} \) and (3) the outer radius of the cloud expands with \( t^{1/5} \). While the last statement cannot be tested in the presented experiment due to the limited observation volume that is already completely filled with particles at the start of the expansion, the other two are accessible for investigation. In Fig. 4.10(a) density profiles of the experimentally observed cluster are shown at different time steps. Although the density is cut off at the observation boundary, the visible parts of the profiles do indeed resemble parabolas, which is in agreement with prediction (1). To test prediction (2), the density maxima are plotted as a function of the time after release of confinement in a log-log scaled plot in Fig. 4.10(b). The predicted power law \( \propto t^{-3/5} \) is indicated by a dashed orange line. One sees, that for the first phase of the expansion (\( t = 30 \) to 300 ms) the densities indeed seem to be proportional \( t^{-3/5} \) followed by another phase with a different temporal dependence. An explanation for the deviation at larger times could be the fact that in the experiments the expansion is only quasi-3D due to the still present vertical confinement: After an initial phase of 3D expansion, the cluster collapses to a horizontal particle monolayer that continues to expand in 2D. The observed 2-staged expansion appears to be an expression of this process. However, the parabolic density profiles and the density evolution in the initial expansion phase are in qualitative agreement with the
4.4. Expansion dynamics

collisional model proposed by Piel and Goree. Concluding, there is evidence that the observed expansion is influenced by collision with neutral gas atoms.

In an attempt to switch off both the horizontal and the vertical confinement additional zero-gravity (zero-g) experiments have been conducted at the ZARM drop tower in Bremen. By mounting the confinement ring halves onto metallic anchors that can be detracted horizontally by an external solenoid it was possible to switch off the confinement remotely once zero-g was accomplished. Unfortunately the zero-g experiments have been inconclusive until the present date. Nevertheless, the quasi-3D experiments conducted in the laboratory give very good insights into the expansion behavior of finite Yukawa-balls. By comparing experimental observations with simulated predictions it has been possible to confirm the predictions of Piel and Goree. We have been able to attribute the rather layerwise expansion to the apparent shielding effect while the density profiles have been found to exhibit indications of collisions between neutral gas atoms and dust particles.

Figure 4.10: a) Experimental particle density profiles over radius with color-coded time. b) Temporal progression of the maximum particle densities (blue crosses) compared to the power law \( \propto t^{-3/5} \) (dashed orange line).
5. Summary and Outlook

In the thesis at hand I have investigated properties of finite Yukawa-balls and how they depend on the number of particles in the system. In primary studies the foundation for my later work has been laid: Laser heating of Yukawa-balls has been established, dynamics have been described in terms of normal mode analysis for very small systems, streaming ions have been identified as a cluster stability criterion, instantaneous recrystallization properties have been found and the deteriorating effect of the plasma on melamine-formaldehyde particles has been determined. In order to observe and reconstruct clusters with high particle numbers and large-scale dynamics, I have improved the diagnostic capabilities of the setup by clearing the optical pathways and making the observation volume easily adjustable. With this setup I have been able to explore diverse size-dependent phenomena over the whole particle number spectrum of finite Yukawa-balls.

For smaller clusters with particle numbers below 100 the melting behavior has been investigated. I have shown that spherical correlation functions can be adapted to the cylindrical symmetries found in elongated clusters under the influence of streaming ions. From the modified correlation functions I have calculated configurational entropies that allow to determine the point of phase-transition. Using this general melting criterion, I have been able to find a clear dependency between the size of the cluster and its transition temperature in qualitative agreement with predictions for ion clusters.

In the intermediate size-range between 50 and 1100 particles, the balance between structural forces and instabilities due to streaming ions shifts towards an onset of collective vortex motions. I have been able to observe the size-dependent behavior of such particle flows in full 3D. From the measured vorticity of the poloidal particle flow field the vortex has been found to be driven by radial gradients of the ion drag force: The strong drag in the center forces the dust particles to flow downwards in the central parts of the cloud whereas the weaker drag further outwards allows the particles to move upwards in the outer regions of the cloud.

Finally, I have been able to investigate very large 3D plasma crystals with more than 10,000 particles. Here, the cluster size becomes larger than the shielding length and has an increased impact on the clusters’ structure and dynamics. Especially in the expansion behavior of particle clusters a distinct difference between unshielded Coulomb and shielded Yukawa interaction was expected from simulations. By physically removing the horizontal confinement I have been able to study the expansion dynamics: The exterior particles immediately start to flow outwards while the inner particles stay confined due to their shielding neighbors. From the analysis of the spatio-temporal density evolution and by the comparison with simulations I have been able to identify an inclined expansion front that is a sign of the expansion under shielded particle interaction. An additional comparison
5. Summary and Outlook

between the observed densities and predictions of a model that takes collisions between gas atoms and dust particles into account has revealed indicators of a collision influenced expansion. By testing the experimental setup under zero gravity conditions I have made a first attempt to induce a full 3D expansion in which the vertical confinement is removed as well.

Concluding, finite Yukawa-balls provide an interesting link between single particles and extended dust clouds in which the interplay between collective particle interactions and confining plasma forces results in fascinating size-dependent phenomena. I conclude my thesis with an encouraging list of research topics that can and hopefully will be further investigated based on my findings.

- Observing more clusters with smaller size differences would help to understand the influence of “magical” particle numbers on the size-dependent transition temperature. Furthermore, it would be interesting to see whether the transition temperature continues its linear trend shown in Fig. 4.3.
- As the observed vortices have been attributed to gradients in the ion drag force, the next step could be to manipulate or counter-balance this cause of instability. Doing so would allow to observe very large, stable clusters in 3D as the transition region between medium sized systems and bulk-like clouds. In particular, the changeover from shell-like pentagonal and hexagonal structures to crystalline bcc and fcc lattices deserves deeper investigation given the experimental insights already gained.
- The mentioned attempt to observe full 3D expansions of finite Yukawa-balls under zero gravity conditions was inconclusive. Thus, a continuation of these studies seems very promising.
- Finally, the overall confinement setup using a brass ring allows to alter the properties of the harmonic trap via external voltages. New ways of manipulating the phase state or the isotropy of finite Yukawa-balls may emerge from this technique.
Bibliography


Bibliography


Bibliography


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A. Thesis Articles

Author Contribution

The following list contains the individual contributions to the thesis articles listed in chronological order. The list is succeeded by reprints of all listed publications.

Here, the individual contributions to the thesis articles are listed in chronological order. Below, reprints of the original articles are implemented.

A1: “From transport to disorder: Thermodynamic properties of finite dust clouds”
AS, MM and JS conducted the measurements. AS evaluated the data. The manuscript was written by AS. It was edited by all authors. (The experiments and data evaluation have been performed alongside MM’s work for his Master thesis but were not part of it.)

A2: “Crystal and fluid modes in three-dimensional finite dust clouds”
AS and MM conducted the measurements. AS evaluated the data by means of NMA and fluid modes. The manuscript was written by AS. It was edited by all authors. (The experiments and data evaluation have been performed alongside MM’s work for his Master thesis but were not part of it.)

A3: “Nonequilibrium finite dust clusters: Connecting normal modes and wakefields”
A. Melzer, A. Schella and M. Mulsow, Phys Rev. E 89, 013109 (2014)
MM conducted part of the experiments. Another part of the experiments has been conducted by AS. AM analyzed the data and performed the calculations. The manuscript was written by AM. The manuscript was co-edited by MM and AS.
A. Thesis Articles

A4: “Correlation buildup during recrystallization in three-dimensional dusty plasma clusters”
A. Schella, M. Mulsow and A. Melzer, Phys. Plasmas 21, 050701 (2014)
MM developed the camera-laser-triggering setup for the experiment. AS conducted the measurements and analyzed the data. The manuscript was written by AS and co-edited by MM and AM.

A5: “Spatio-temporal evolution of the dust particle size distribution in dusty argon rf plasmas”
CK conducted the experiments, analyzed the results and wrote the manuscript. MM provided the experimental setup and guidance for the phase-resolved resonance experiments. Data interpretation was performed by CK and AM. The manuscript was co-edited by all authors.

A6: “Stereoscopic imaging of dusty plasmas”
MM, MH and CK provided the knowledge of the experimental approach of stereoscopic imaging. AM wrote the manuscript which was co-edited by all authors.

A7: “Experimental determination of phase transitions by means of configurational entropies in finite Yukawa balls”
MM developed the experimental setup, conducted the experiments, performed the data analysis and wrote the manuscript. The interpretation of the results has been done by MM and AM. The latter also co-edited the manuscript.

A8: “Analysis of 3D vortex motion in a dusty plasma”
MM developed and built the experimental setup. MM and MH conducted the experiments. AM and MM interpreted the data and AM wrote the manuscript. MM and MH co-edited the manuscript.

Confirmed:

(Matthias Mulsow) (Prof. Dr. André Melzer)
A1

From transport to disorder: Thermodynamic properties of finite dust clouds

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From transport to disorder: Thermodynamic properties of finite dust clouds

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The quantities entropy and diffusion are measured for two- and three-dimensional (3D) dust clusters in the fluid state. Entropy and diffusion are predicted to be closely linked via unstable modes. The method of instantaneous normal modes is applied for various laser-heated clusters to determine these unstable modes and the corresponding diffusive properties. The configurational entropy is measured for 2D and 3D clusters from structural rearrangements. The entropy shows a threshold behavior at a critical temperature for the 2D clusters, allowing us to estimate a configurational melting temperature. Further, the entropic disorder increases for larger clusters. Finally, the predicted relation between entropy and unstable modes has been confirmed from our experiments for 2D systems, whereas 3D systems do not show such a clear correlation.

I. INTRODUCTION

The statistical properties of fluid (liquid) states of an ensemble are often associated with quantities such as entropy or diffusion [1–14]. Diffusive transport itself is closely related to the onset of unstable modes [15–22]. Based on the work of La Nave et al. [23], Keyes predicted a close relationship between configurational entropy $S_c$ and unstable modes [24]. The configurational entropy $S_c$ is a measure of structural rearrangement. Unstable modes describe crossing events over potential barriers. In the framework of a random energy model Keyes found a rather robust relation of the form

$$S_c = a + b \ln(f_b),$$

(1)

with $a$ being a free parameter. The parameter $b$ was only specified to be in the range of $1 \leq b \leq 2$. The fraction of unstable modes $f_b$ describes the relative amount of unstable modes in the density of states of the system. The density of states can be retrieved by instantaneous normal mode (INM) analysis [19–21]. At first glance, it seems counterintuitive to speak in terms of normal modes in the context of liquids. However, below the Maxwell relaxation time $\tau_M$, liquids show many aspects of solidlike behavior [15]. In typical dusty plasma experiments, $\tau_M$ is of the order of $\approx 0.1$ s [25]. Hence fast video imaging is needed to account for these short-time dynamics. In contrast, structural rearrangement can be found at time scales up to a few seconds.

Dusty plasmas are ideally suited to study these fluid state properties [4,26,27]. They are formed by trapping dust particles, e.g., melamine formaldehyde particles of micron size, in a gaseous plasma. The particles become highly negatively charged due to the fluxes from the plasma species. Moreover, the large mass of the microspheres slows down their dynamics to time scales in the millisecond range. This allows for video imaging of the dust component on the kinetic level. Fluid two- and three-dimensional (3D) dust clusters can be realized by laser heating [25,29–31].

Now we study the dynamics of finite 2D and 3D dusty plasmas. These are generated by trapping a small number of dust particles in a harmonic confinement [32–35]. Finite ensembles offer the possibility to study the complex interplay between the particle interaction and the system boundary on the particle dynamics [12,36]. The pairwise interaction between the $N$ dust particles is generally assumed to be of Yukawa type, where shielding by the ambient plasma is considered. With the additional isotropic confinement, the Hamiltonian of the finite system can thus be written as [32]

$$E = \sum_{i=1}^{N} r_i^2 + \sum_{i<j}^{N} \exp(-\kappa r_{ij})/r_{ij}. \quad (2)$$

Here dimensionless units are used by introducing the normalized energy $E_0 = (m_0Q^2/32\pi\varepsilon_0^2)^{1/3}$ and the normalized length $r_0 = (Q^2/2\pi\varepsilon_0\Omega^2)^{1/3}$ with $\Omega_0$ the trap frequency, $Q$ and $m_0$ the dust charge and mass, respectively, and $\kappa = r_0/\lambda_D$ the screening strength (where $\lambda_D$ denotes the Debye shielding length). The distance to the trap center is $r_i = (x_i^2 + y_i^2)^{1/2}$ in two dimensions and $r_i = (x_i^2 + y_i^2 + z_i^2)^{1/2}$ in three, with the indices $i$ and $j$ denoting the particles.

In this paper the INM approach is used to investigate the transport properties of 3D dust clusters, so-called Yukawa balls, in the fluid phase, whereas the fluid quantities, such as diffusion coefficients, are mainly deduced from the unstable INM modes [16,17]. Additionally, the INM technique allows us to determine the fraction of unstable modes $f_u$. The INM approach for 2D systems has been demonstrated previously in Refs. [21,22].

Moreover, the configurational entropy is measured from long video sequences of fluid particle arrangements in two and three dimensions. To calculate the configurational entropy, a certain number of structural transitions has to be observed. Thus the INM approach and entropy as statistical methods require long-time series on the one hand recorded at high frame rates on the other.

II. EXPERIMENT

We report on laser-heating experiments of finite dust clusters in two and three dimensions. The 2D setup was extensively described in Refs. [21,31], whereas the 3D setup has been

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reported in Ref. [30]. Both experiments were conducted in asymmetrically driven rf argon discharges at 13.56 MHz where the lower electrode was powered and the discharge chamber was grounded. A brief repetition of the devices will be given below for clarity.

A. Experiments on 3D dust clouds

The rf discharge was driven at powers between 1.2 and 1.5 W ($U_{pp} = 70–95$ V at the driven electrode) and at neutral gas pressures between 4.8 and 7.8 Pa. For the 3D experiments [see Fig. 1(a)], isotropic confinement is achieved inside a cubic glass box placed on top of the lower electrode. The particles are trapped by a superposition of the electric field force from the dielectric walls and a thermophoretic force from the heated lower electrode (55 $^\circ$C), which, together with the sheath electric field force, compensates for gravity and levitates the particle cloud. Melamine formaldehyde particles of 4.86 μm diameter have been used here. Yukawa clusters with total particle number in the range $5 < N < 70$ were trapped in that way [see Figs. 1(b)–1(e)].

Two laser beams at 532 nm were used to illuminate the particles. The scattered laser light is observed via three orthogonal complementary metal-oxide semiconductor cameras to retrieve the 3D particle positions using well-established techniques [37,38]. The camera frame rate was 100 frames per second in the experiments described here. For most of the experiments, long video sequences (runs) with approximately 30,000 frames per camera were evaluated. The stereoscopic setup thus allows us to follow both the short-time kinetics due to the high frame rate and the long-time dynamics.

To heat the 3D dust ensemble, two diode lasers with two opposing laser beams at 660 nm wavelength with 1 W maximum output power were used. To mimic a heating process, the two beams were swept independently in a random way over the cluster’s cross section by galvanometer scanners with a dwell time of $\tau = 0.1$ s at each position [30].

B. Experiments on 2D dust clouds

Here also an asymmetric rf discharge was used. The rf power was set to 3 W and the neutral gas pressure was held constant at 7 Pa. Melamine formaldehyde particles of 12.26 μm diameter were used for the experiments on laser-heated 2D dust clusters. The 2D setup is illustrated in Fig. 2(a).

When immersed into the discharge, the particles sediment into a position where the sheath electric field from the lower electrode compensates for gravity. An additional radial confinement is achieved by a shallow spherical depression. Since no thermophoretic levitation is used in this case, the dust particles are trapped as a finite single-layer system in the sheath [see Figs. 2(b) and 2(c)]. A top-view CCD camera observed the particle positions at a frame rate of 60 frames per second with a sequence length of 15,000 frames per experiment.

A four-axis laser-heating system was used to heat the 2D clusters with a sophisticated random frequency method [31]. This technique allows us to mimic a true heat bath for the dusty...
plasma subsystem \[31,39\]. Here two green diode-pumped solid state lasers with total output powers of 5 and 6 W were used.

### III. THEORETICAL CONCEPTS

At this point, only a brief description of the INM technique is given; details can be found in Refs. \[15–20\]. Our starting point is the Hessian matrix, which is given by the second derivative of the energy of the system in Eq. (2) as

\[
H(t) = \frac{\partial^2 E(\vec{r}, t)}{\partial r_{\alpha,i} \partial r_{\beta,j}} |_{\vec{r}(t)}.
\]  

(3)

Contrary to the normal mode analysis (NMA) \[33,40–42\], the eigenvalue problem of Eq. (3) is solved for each instant of time. The 2N and 3N eigenvectors (in two and three dimensions, respectively) obtained at each time step describe the momentary mode oscillation pattern and the 2N and 3N eigenvalues \(\omega_l^2\) the mode frequency. The eigenfrequencies are averaged over the time series resulting in a dimensionless density of states

\[
\rho_\omega = \left\langle \delta(\omega - \omega) \right\rangle.
\]

(4)

where we use the normalization \(\int d\omega \rho_\omega d\omega = 1\).

Since the ordinary NMA uses the equilibrium positions of the particles as input, the NMA eigenvalues are always positive definite, i.e., their eigenfrequencies are always real \[40\]. In the INM approach the eigenvalues \(\omega_l^2\) can be either positive or negative, hence, depending on the sign of the eigenvalues, the INM eigenfrequencies \(\omega_l\) can be either purely real or purely imaginary. Thus, by convention, the INM density of states is split into a stable \(\rho_s(\omega)\) and an unstable part \(\rho_u(\omega)\) with real and imaginary \(\omega\), respectively. Integrating over all imaginary eigenfrequencies defines the fraction of unstable modes as

\[
f_u = \int_0^{\infty} \rho_u(\omega) d\omega.
\]

(5)

Real eigenfrequencies describe the oscillation of a particle in the local potential cage of the neighboring particles; they describe the solid properties of the system. Imaginary eigenfrequencies describe the crossing over potential hills. Consequently, the liquid properties can be deduced mainly from the unstable part \(\rho_u(\omega)\) \[15–20\].

A measure of crossing over potential hills is the so-called hopping rate \(t_{5,1}^{-1}\). In general, \(t_{5,1}^{-1}\) is a rather complicated function due to the multidimensional potential energy landscape of the involved particles \[19,20\]. In the INM approach used here, we follow Vijayadamodar and Nitzan \[19\], resulting in

\[
t_{5,1}^{-1} = c \int d\omega \rho_u(\omega) \frac{\omega^3}{2\pi} A \exp \left( -\frac{B \omega^2}{k_BT} \right).
\]

(5)

Here \(c \approx 3\) is used to describe the possible escape routes \[21,22\]. The free parameters \(A\) and \(B\) are obtained from the fit \(\rho_u(\omega)/\rho_s(\omega)\) to the function \(A \exp(-B\omega^2/k_BT)\). The exponent in the exponential is mainly related to the energy of vibration (proportional to \(\omega^2\)), i.e., reflecting the barrier height over which to cross and the thermal energy (proportional to \(k_BT\)).

Finally, the diffusion coefficient \(D\) can be derived from the hopping rate by

\[
D = \frac{k_BT}{m} \int d\omega \rho(\omega) \frac{\tau_3}{1 + \tau_3^2 \omega^2}.
\]

(6)

This equation allows us to measure the diffusion coefficient as a function of the INM density of states.

As already mentioned, diffusion is closely connected to configurational changes, or rearrangement. A classical quantity to describe the disorder or, equivalently, the probability for configurational changes in a system, is the configurational entropy. The classical textbook definition for the entropy (see, for instance, Ref. \[3\]) follows from the probability of finding a state \(\rho_s\) by

\[
S_c = -\sum_{\omega} p_\omega \ln p_\omega.
\]

(7)

Radzvilavičius and Anisimovas investigated the entropy of charged particle clusters by means of Monte Carlo simulations \[5\]. They found that the entropy as a function of the particle number can be described by two terms. The first term comes from a combinatorial contribution to the entropy, thus giving

\[
S_c = \gamma \ln(N!) + S_{\omega},
\]

(8)

where \(\gamma\) is a factor depending on the dimensionality of the system and the screening strength. This equation can be interpreted as a general increase of \(S_c\) with the cluster size. The second term \(S_{\omega}\) describes fluctuations of the configurational entropy as a consequence of the exact configuration of the finite ensemble and the specific interaction between the particles due to shielding. The simulation data of Ref. \[5\] will be used for comparison in the following.

### IV. RESULTS

Laser-heating and long-run experiments enable us to investigate finite dust clusters with statistical methods over a wide thermodynamic range and thus to retrieve both entropy and unstable modes.

#### A. The INM analysis for 3D dust clusters

In the first part, the INM analysis is performed to evaluate unstable modes and connected thermodynamic properties of 3D dust clusters consisting of \(N = 33, 48, 49,\) and 60 particles (see Fig 1). All clusters were laser heated as described above. As a measure of heating, the kinetic temperature \(T_{\text{kin}} = m(v^2)/3k_B\) is determined from the particle velocities \[30\].

As a representative to illustrate the INM technique in more detail, the cluster consisting of \(N = 60\) particles [shown in Fig. 1(e)] is chosen. The Yukawa ball has a well-established two-shell structure with 16 particles on the inner shell and 44 particles on the outer.

Figure 3 shows the INM spectra as a function of the normalized frequency in units of \(\omega_0/\omega_h\) and the corresponding trajectories over a short period of time of about 2 s at different cluster temperatures. By convention, the unstable branch of the INM spectra is plotted on the negative frequency axis.

Finally, the diffusion coefficient \(D\) can be derived from the hopping rate by

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where \(\gamma\) is a factor depending on the dimensionality of the system and the screening strength. This equation can be interpreted as a general increase of \(S_c\) with the cluster size. The second term \(S_{\omega}\) describes fluctuations of the configurational entropy as a consequence of the exact configuration of the finite ensemble and the specific interaction between the particles due to shielding. The simulation data of Ref. \[5\] will be used for comparison in the following.

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Figure 3 shows the INM spectra as a function of the normalized frequency in units of \(\omega_0/\omega_h\) and the corresponding trajectories over a short period of time of about 2 s at different cluster temperatures. By convention, the unstable branch of the INM spectra is plotted on the negative frequency axis.
FIG. 3. (Color online) (a) The INM spectra for the $N = 60$ particle cluster for three different temperatures. By convention, the unstable density of states $\rho_u(\omega)$ resulting from imaginary eigenfrequencies is plotted as $\rho_u(\omega)$ on the negative frequency axis ($\omega \rightarrow -|\omega|$). The INM spectra shown here are plotted with a constant offset. (b) The corresponding trajectories of the cluster particles are shown over a time span of 2 s.

For the lowest temperature $T = 2930$ K, several peaks occur in the stable branch: The sharp peak at $\omega/\omega_0 = 1$ belongs to the sloshing mode. The sloshing oscillation is an exact solution of the Hessian with threefold degeneracy in three dimensions [41,43]. The broader peak at $\omega/\omega_0 = 1.2$ belongs to a large-scale vortex-antivortex mode [42]. The peak at approximately $\omega/\omega_0 = 1.72$ comprises the breathinglike oscillations.

By heating the cluster to $T = 2930$ and 27 810 K the high-frequency features near $\omega/\omega_0 = 2$ vanish. Only the sloshing oscillation and the large-scale oscillation remain. The INM spectra are generally less structured than in the corresponding 2D cases (see Refs. [21,22]).

The unstable branch shows a continuum curve with its maximum position at about $|\omega_u,\max| = 0.75\omega_0$. The fraction of unstable modes $f_u$ increases from 16% for $T = 2930$ K to 23% for $T = 27810$ K. These values agree well with the unstable mode fraction of 3D Lennard-Jones fluids [16]. In contrast, the unstable mode fractions derived here are decisively larger than for 2D dust clusters, which were in the range of up to only 8% [21,22]. The reason for the higher fraction in three dimensions can be understood since in three dimensions a particle has more low-energy pathways to cross the energy barrier formed by the surrounding particles than in a flat 2D cluster. Additionally, the larger reconstruction error in three dimensions might contribute to a broadening of the spectra since they lead to a larger variation of the eigenvalues [37]. However, the simulations, which do not suffer from reconstruction errors, show exactly the same behavior.

From fitting the ratio $\rho_u(\omega)/\rho_s(\omega)$ of each INM spectrum, it is straightforward to calculate the hopping frequency according to Eq. (5). The total error in fitting the curves did not exceed 3%. The so-determined hopping frequency $\tau^{-1}_h$ is shown as a function of the cluster temperature $T$ in Fig. 4 for all dust clusters shown in Figs. 1(b)–1(e).

Most hopping rates are in the range of 0.05 s$^{-1} < \tau^{-1}_h < 0.17$ s$^{-1}$. These rates correspond to 15–50 hopping events per video sequence, which corresponds well with the visual impression. The hopping rate generally increases as the temperature of the cluster particles increases to higher values. It seems that $\tau^{-1}_h$ is only weakly dependent on the particle number here.

The temperature dependence of the hopping frequency is consistent with the point of view that the thermal energy mainly drives configuration changes in the system. These lead to a hopping from a particle into another local equilibrium position formed by the potential cage of neighboring particles [44]. To consider the vertical elongation of some of the clusters shown in Fig. 1, we also tested the influence of an anisotropic trap by assuming weaker confinement in the vertical direction in the INM analysis, but the results do not differ significantly.

FIG. 4. (Color online) Hopping frequency $\tau^{-1}_h$ as a function of the temperature $T$ for various dust clusters. The hopping frequency increases slowly as the dust cluster temperature increases.
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A. Thesis Articles

B. Comparison to simulations

Ohta and Hamaguchi investigated the diffusion of a frictionless Yukawa one-component plasma (YOCP) over wide parameter regimes of $\Gamma$ and $\kappa$ by means of simulations [7]. The authors revealed that the self-diffusion coefficient obeys a universal scaling near the melting point of the form $D^* = \alpha (T^* - 1)^\beta + \gamma$, with $T^* = T/T_M$ the temperature normalized to the melting temperature $T_M$, and $\alpha$, $\beta$, and $\gamma$ fit parameters depending on $\kappa$. The formula is valid as long as the temperatures are not too high ($1 < T^* < 10$). There the diffusion constant is normalized to $D^* = D/\omega_E b^2 k_0$, with $\omega_E$ the Einstein frequency of the lattice and $b$ the Wigner-Seitz radius as a measure of the interparticle spacing.

Moreover, to account for the finite system size and friction, we performed Langevin molecular dynamics simulations with 60 particles in a harmonic confinement interacting via isotropic Yukawa pair potentials with moderate screening ($\kappa = 1$) according to Eq. (2). Friction was set to be at the order of the confinement frequency $\gamma/\omega_E = 1$, which is valid for our experiment. To model the heat bath at a certain temperature $T$, the sympletic low-order scheme of Ref. [49] was used. The time step was chosen to be $\Delta t = 0.2/\omega_E$, with $\omega_E$ the dust plasma frequency. After an equilibration phase, the particle positions in the simulated clusters were simulated for 18,000 time steps further and then analyzed with the INM technique in the same manner as the experimental ones.

Figure 5(b) now shows the normalized self-diffusion coefficients $D^*$ of the measured $N = 60$ cluster as a function of the temperature together with our Langevin simulations and the YOCP diffusion constants [7]. For normalization of the experimental values, the Wigner-Seitz radius was calculated from the cluster volume as $b_{WS} = 560 \mu m$. The charge number of the particles is $Z = 6900 \pm 1100$ [30]. Then the Einstein frequency in the experiment follows as $\omega_E = (32.06 \pm 2.56) \pi^{-1}$.

The YOCP results of Ohta and Hamaguchi [7] are systematically larger in the observed temperature range. In contrast, the diffusion coefficients obtained from the simulated finite cluster are close to the experimental values. There are two reasons for this. On the one hand, friction, which hinders the movement of the particles, was neglected in Ref. [7]. On the other hand, our observed 3D system is finite. Particles sense the system boundary so that, in general, the diffusion processes in particle traps should be smaller than for extended matter [9].

C. Configurational entropy

To measure the configurational entropy, different dust cluster configurations have to be identified. To separate different configurations, boundaries between the different shells are defined from the equilibrium positions of the clusters. By counting the particles within each shell, the cluster configuration is determined at each time step. Then the occurrence of different configurations, i.e., the shell occupation numbers, along the time series is counted to calculate the probabilities.

An illustrative example is depicted in Fig. 6. In Figs. 6(b)–6(d) the corresponding observed different states together with the shell occupation number and their probabilities in the experiment are visualized.

The most probable configuration at $T = 25730 K$ is the $(1,7,11)$ configuration with $p_1 = 87.37\%$ followed by the $(1,6,12)$ configuration with $p_2 = 12.47\%$ and the $(1,8,10)$ configuration with a rather low probability $p_3 = 0.06\%$. Note that the most probable configuration is a metastable state [33,50] and not the ground state.
FIG. 6. (Color online) (a) Trajectories of the particles in an $N = 19$, 2D particle cluster over a time span of 250 s. (b)–(d) Observed cluster configurations together with the shell occupation numbers and their probabilities. Dashed lines indicate borders between different shells.

Consequently, the configurational entropy simply follows from the probabilities as $S_c = 0.38$ according to Eq. (7).

This procedure was repeated for all measured 2D clusters at different temperatures along the time series of 15,000 frames and all 3D clusters of Figs. 1(b)–1(e) along the time series of 30,000 frames.

Figure 7 shows the configurational entropy as a function of the dust particle temperature for selected 2D and 3D clusters.

For the 2D clusters, the configurational entropy always starts at $S_c = 0$ (i.e., a single stationary configuration) for very low temperatures. Above a certain critical temperature, the entropy grows fast and reaches higher values. This critical temperature differs for the different clusters. Higher values for the configurational entropy are reached for larger clusters in the limit of high temperatures. Due to the limited number of experimental runs, a clear saturation as in the simulations of Ref. [5] is not found.

According to Radzvilavičius and Anisimovas [5], the sudden increase of the configurational entropy with temperature can be attributed to a configurational phase transition. This implies that one can estimate a further melting temperature $T_M$ from the curve $S_c(T)$. For the 2D clusters shown in Fig. 7(a), configurational melting was estimated at the critical temperature where the entropy starts to soar. The so-determined melting temperatures are summarized in Table I. For comparison, the INM melting temperatures of Ref. [21] are added for exactly the same 2D clusters, where the melting temperature has been derived from the zero crossing of the diffusion constant, as discussed in Sec. IV A. The values of the two approaches differ, since the method of configurational entropy is only sensitive to radial transitions, whereas the melting temperatures of Ref. [21] seem to indicate angular melting. The relatively large error in $T_M$ from the $S_c(T)$ curve results from a sensitive dependence of $S_c$ on the number of different cluster configurations detected during the experimental runs, especially around the transition from $S_c = 0$ to finite values.

The highest melting temperature is obtained for the $N = 19$ dust cluster. The $N = 20$ dust cluster has a much lower melting temperature. This is consistent with the INM results of Ref. [21] since the difference of the configurational melting temperature well reflects the geometrical stability of the 2D clusters. The 19-particle cluster has a high stability due to the commensurable number of particles in the inner and outer shells. The 20-particle cluster with incommensurable numbers has a much lower stability [32].

The configurational entropies as a function of the dust particle temperature for the 3D clusters, except for the lowest temperature of the $N = 48$ cluster, are more or less constant over temperature [see Fig. 7(b)]. As a general trend the entropy increases with cluster size.

The findings agree with the results from the INM analysis. The Schweigert instability leads to elevated kinetic temperatures for the 3D clusters [30]. As a result of the high kinetic energies, more different states are energetically accessible. Hence the low-temperature branch with a single stationary configuration (i.e., $S_c = 0$) was not monitored. However, even

<table>
<thead>
<tr>
<th>Number of particles</th>
<th>$T_M$ (K) from $D$ (Ref. [21])</th>
<th>$T_M$ (K) from $S_c$</th>
</tr>
</thead>
<tbody>
<tr>
<td>19</td>
<td>6700 ± 2200</td>
<td>14520 ± 1890</td>
</tr>
<tr>
<td>20</td>
<td>1650 ± 550</td>
<td>1550 ± 200</td>
</tr>
<tr>
<td>27</td>
<td>1800 ± 600</td>
<td>3640 ± 1780</td>
</tr>
<tr>
<td>34</td>
<td>2190 ± 730</td>
<td>820 ± 400</td>
</tr>
</tbody>
</table>
for clusters observed at high pressures and low temperature, spontaneous configurational changes are not so seldom [38].

Now the question arises as to how the configurational entropy behaves with increasing size of the dust cloud. Figure 8 shows the configurational entropy $S_c$ of the 2D and 3D dust clusters as a function of $\ln(N!)$. For comparison, the results from the simulations of Radzvilavičius and Anisimovas [5] for different screening strength are also given. For the 2D clusters, only the configurational entropies obtained from the experiments were used.

In general for 2D systems, the entropy rises with increasing particle number, ranging from $S_c = 0.58 \pm 0.15$ for the $N = 19$ cluster to $S_c = 1.45 \pm 0.63$ for the $N = 50$ cluster. The overall agreement with the simulations of Ref. [5] is good. Exact quantitative agreement cannot be expected since, on the one hand, the finite sampling time in the experiment allows only for a limited number of different determined configurations and, on the other hand, even the simulation results massively depend on the exact value of $\kappa$.

FIG. 8. (Color online) Configurational entropy $S_c$ as a function of the particle number $N$ for finite dust clusters. The symbols represent experimental values for (a) 2D and (b) 3D dust clusters and the lines are results from simulations of Ref. [5] for different values of $\kappa$. In (b), the (blue) circles represent unheated clusters and (red) squares mark laser-heated clusters.

D. Unstable modes and entropy

The main aim of this paper is to address the relationship between the unstable modes from the INM and the concept of the configurational entropy, as given by Eq. (1). For that purpose, the unstable mode fraction $f_u$ derived from the INM is shown in Fig. 9 as a function of the configurational entropy $S_c$ obtained from the cluster states. These thermodynamic quantities were derived from the experiments using independent methods. It should be noted that both the unstable mode fraction and the configurational entropy are depicted for diverse 2D and 3D dust clusters at all realized temperatures [23,24]. As illustrative examples, the 2D clusters shown in Fig. 2 with $N = 20$ and 27 particles are highlighted. The configurational entropy and the fraction of unstable modes for the simulated clusters were also added.

For the 2D clusters, the expected linear relation between $\ln(f_u)$ and $S_c$ can be assigned. A linear relationship between $S_c$ and $\ln(f_u)$ implies that a lower number of unstable modes coincides with a lower number of realized configurations [23,24]. The unstable modes connect different cluster states via diffusion. Thus a higher fraction of unstable modes leads to a larger configurational entropy. The parameter $b$ in

FIG. 9. (Color online) Logarithm of the unstable mode fraction $f_u$ as a function of the configurational entropy $S_c$ for 2D dust clusters (closed circles) and 3D clusters (open triangles). The solid line represents a linear fit to the 2D data. As representatives, the 2D clusters $N = 20$ (closed squares) and $N = 27$ (open squares) are highlighted. The data of the simulated $N = 60$ cluster are added as closed upward triangles.

Figure 8(b) shows the configurational entropies for the 3D dust clusters. The circles represent unheated clusters. Here, only the number of particles was varied. Due to the Schweigert instability, these clusters are nevertheless at elevated temperatures. The error in estimating the configurational entropy then is roughly represented by the symbol size. The squares in Fig. 8(b) denote the configurational entropy of the laser-heated clusters and the error bars result from averaging $S_c$ over different cluster temperatures. The general trend given by the simulations [5] is well reproduced for the 3D clusters, with a slight underestimation for larger clusters.
Eq. (1) can be estimated as $b \approx 1.7$ for the 2D dust clusters. This is fully consistent with the parameter range predicted by Keyes, where it was argued that $1 \lesssim b \lesssim 2$ [24].

For the 3D clusters, the fraction of unstable modes is generally higher than in two dimensions. Moreover, the unstable mode fraction seems to be nearly constant. For the simulated $N = 60$ cluster, the values of $\ln(f_u)$ and $S_c$ above the melting point lie in the same range as the experimental Yukawa balls. The overall higher fraction was already attributed to the higher dimensionality of the 3D clusters. No clear trend of increasing entropy with increasing $f_u$ is observed. A possible reason can be that wake field effects by the streaming ions lead to a more complex interplay between the diffusive properties and the configurational entropy of the 3D dust clouds. A modification of Eq. (2) that considers also the ion focus could lead to an improvement and leaves space for future investigations.

V. CONCLUSION

We investigated statistical properties of finite fluidlike Yukawa systems. The major question was how unstable modes, which are related to the diffusion of the particles, are connected to the configurational entropy. Finite 2D and 3D dust clusters serve as ideal model systems since the particles in the charged particle cluster can be traced individually and the ensemble can be heated by lasers to fluidlike regimes.

The INM analysis was applied to laser-heated 3D finite dust clouds. We found that the fraction of unstable modes is relatively large compared to 2D systems even for the unheated clusters. The hopping rates seem to be nearly independent of the size of the cluster, but increase with the temperature of the cluster particles. The diffusion coefficients were derived for various clusters using the unstable modes of the INM. As an example, the melting transition was investigated by extrapolating the $D(T)$ curve to the freezing point for an $N = 60$ cluster. A comparison between measured and simulated clusters showed that the diffusive behavior of the cluster particles is influenced by boundary effects and friction.

Further, the configurational entropies of 2D and 3D clusters were evaluated using long-time series. In two dimensions the configurational entropy shows a threshold behavior at a certain temperature. Melting temperatures were derived for the 2D clusters from the $S_c(T)$ curve and compared to the literature. For the 3D Yukawa balls, no such threshold behavior for the configurational entropy was observed since even the unheated clusters exhibit large kinetic temperatures. The influence of the cluster size on the configurational entropy was compared with simulations of Ref. [5], showing good agreement in two dimensions as well as in three dimensions. In general, the entropy increases for bigger clusters because of the larger number of possible states.

Finally, the relation between unstable modes and configurational entropy as predicted by Ref. [24] was tested. For 2D dust clusters, a clear relation was observed, whereas for the 3D Yukawa balls, the fraction of unstable modes was nearly constant for all measured configurational entropies.

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Crystal and fluid modes in three-dimensional finite dust clouds

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Crystal and fluid modes in three-dimensional finite dust clouds

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Abstract. The spectral properties of three-dimensional dust clusters confined in gaseous discharges are investigated using both a fluid mode description and the normal mode analysis (NMA). The modes are analysed for crystalline clusters as well as for laser-heated fluid-like clusters. It is shown that even for clusters with low particle numbers and under presence of damping fluid modes can be identified. Laser-heating leads to the excitation of several, mainly transverse, modes. The mode frequencies are found to be nearly independent of the coupling parameter and support the predictions of the underlying theory. The NMA and the fluid mode spectra demonstrate that the wakefield attraction is present for the experimentally observed Yukawa balls at low pressure. Both methods complement each other, since NMA is more suitable for crystalline clusters, whereas the fluid modes allow to explore even fluid-like dust clouds.

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1. Introduction

One access to study the dynamics of finite systems are dusty plasmas (see [1–3] for an overview). These are plasmas with additionally embedded, typically micron-sized, particles. Once immersed in the discharge, the particles attain a high negative charge through the ion and electron fluxes to the particles’ surface. Due to the high charge and a thermal energy near room temperature the dust system generally is strongly coupled. In dusty plasmas, the particle dynamics can be studied on the kinetic level of individual particles.

When the system consists of less than a thousand dust particles, it is often considered to be finite. Finite systems are known to differ from extended bulk matter in many ways. Theoretical concepts and quantities had to be developed to account for boundary-related effects dominating over volume effects [4–8].

In dusty plasmas, finite dust clusters can be formed by trapping the particles in a cubic glass box inside a discharge plasma [9–17]. There, the dust particles arrange under the influence of their mutual Yukawa interaction and a harmonic confinement into nested spherical shells, the so-called Yukawa balls [9, 10].

While the shell structure and energy states of finite three-dimensional (3D) dust clouds are well understood [18–26], the dynamical properties of finite 3D systems are less explored [14, 27–34]. They offer the possibility to study the interplay between the external confinement and the particle–particle interaction.

Two alternative approaches to study the mode dynamics of finite dust clouds have been put forward: the normal mode analysis (NMA) together with its offspring, the instantaneous NMA and, more recently, the fluid mode description. In the NMA approach, the particle dynamics is described as a harmonic oscillation pattern around the particles’ equilibrium positions, the so-called eigenmodes of the system. The NMA has been applied to two-dimensional (2D) dust clusters [4, 35, 36]. So far, 3D systems have been investigated by NMA only at high gas pressure, where the modes are heavily damped [27].
A different, recently developed approach has been proposed by Kählert and Bonitz [8, 37]. They have treated the Yukawa ball similar to a fluid droplet, deriving the fluid modes of a 3D dust cluster from a cold fluid plasma description.

In the following, we adopt both approaches to describe the dynamical properties of experimentally observed Yukawa balls. The scope is to compare fluid modes and NMA both for crystalline and liquid-like, laser heated clusters. The dust clouds under investigation are heated systematically and identically by means of two laser beams. Laser-heating enables to drive the clusters from a crystalline to a fluid-like state, and hence to study the mode dynamics over a wide range of the strong-coupling regime [14, 38–45].

The dust clouds are investigated at different gas pressures to study the influence of ion streaming motion on the mode properties. At low gas pressure the ion streaming motion leads to the onset of attractive forces between the dust particles [46–51].

Finally, NMA and fluid mode approach are compared to each other. Also, the limitations of the approaches are discussed in section 3.

2. Experiment

The experimental setup to trap 3D dust clusters has been described extensively in [13, 14, 23, 27] (see figure 1) and will only be briefly reviewed here. The experiments have been carried out in an asymmetric capacitively coupled rf discharge in argon. The rf power was varied between 1.2 and 1.5 W (corresponding to $U_{pp} = 70$ and 95 V at the driven electrode), the neutral gas pressure was set to rather low values ranging between 4.8 to 7.8 Pa.

Inside a cubic glass box of 2.5 cm wall-length placed on a heated electrode, a small number of particles (melamine–formaldehyde particles of 4.86 µm in diameter) is trapped in a harmonic 3D confinement [10]. The electric field from the dielectric glass walls confines the particles horizontally. A temperature gradient in the neutral gas due to the heated lower electrode ($55 \, ^\circ\text{C}$) leads to an upward thermophoretic force that, together with the electric field force, levitates the particle cloud against gravity.

Figure 1. Scheme of the experimental setup. The particles confined in the glass box are illuminated by two Nd:YAG lasers from two sides and manipulated by two diode lasers from opposing directions. The three orthogonal high-speed cameras allow for a full 3D dynamic tracking of particle trajectories.
Figure 2. Trajectories over a time span of about 10 s of the N = 60 particle cluster for (a) 0 mW laser power, (b) 250 mW laser power and (c) equilibrium particle positions in cylindrical coordinates $\rho = \sqrt{x^2 + y^2}$ and $z$ for case (a). (d)–(f) The same for a N = 49 particle cluster which has a structure influenced by the ion focus. A single particle chain is highlighted in (d) and (f), e.g. particles 1–4. See text for details.

In that manner, up to hundred dust grains can be confined in an isotropic potential well where they form ordered 3D structures, the so-called Yukawa balls, see figure 2 and [9, 10]. To follow the kinetics of each individual particle in the cluster, we use the stereoscopic setup described in detail in [23, 27]. The particles are illuminated via two expanded Nd:YAG laser beams at 532 nm with 600 mW power each. Three C-MOS cameras observe the scattered light from the dust from orthogonal directions. Here, they were operated at a frame rate of 100 frames per second. In the experiments here, long runs with up to 30 000 frames per camera were recorded.

Two focused laser beams from diode lasers at 660 nm with maximum output power of 1 W are used to laser-heat the Yukawa balls from two opposing sides. The beams are swept randomly over the cross section of the cluster similar to the procedure of [14, 32], leading to heating primarily in the horizontal direction. In the experiments shown here, the two lasers are adjusted to the same output power with a maximum power of $P_L = 400$ mW at the cloud position. With this setup we are able to heat Yukawa balls with particle numbers $N < 100$ to the liquid regime [14].

We conducted several experiments upon laser heated finite 3D dust clouds by varying the particle number ($5 < N < 100$), the rf power of the discharge and the neutral gas pressure.

It is well known from many dusty plasma experiments that the ion focusing effect becomes important below a certain neutral gas pressure. The ion focus is due to ions flowing through a dust cloud. By the electric field of the dust the ions are focused below the highly charged particles [46]. There, the ions create a local positive space charge, which on the one hand leads to a flow-alignment of particles [47–49, 52–60]. On the other hand, the flow-aligned state can exhibit unstable oscillations, so-called Schweigert instabilities [46]. The trend to form aligned particle chains was reported in various experiments with finite 3D dust clouds confined at low pressure under the influence of an ion focus [12–14, 16, 50, 51, 61].
To study the mode dynamics of Yukawa balls with and without the trend to form aligned particle chains, we choose two representative clusters. Firstly, we have confined and analysed a cluster above the critical neutral gas pressure for the onset of Schweigert oscillations (6.4 Pa and 1.3 W rf power). The Yukawa ball with \( N = 60 \) particles is spherical in shape and consists of two shells, see figures 2(a)–(c). Secondly, we selected a cluster that was confined at 4.8 Pa and 1.5 W, just below the threshold neutral gas pressure. The cluster with \( N = 49 \) particles is slightly elongated along the ion streaming direction (aspect ratio about 1.7:1) and the particles tend to align below each other, see figures 2(d)–(f). The four highlighted particles shown in figure 2 build a single vertical chain emphasizing the trend to chain formation.

3. Mode properties

The fluid mode as well as the normal mode technique to analyse the mode properties of finite dust clusters are briefly described. In dimensionless units, the Hamiltonian for the ground state of the \( N \) particle ensemble can be written as

\[
E = \sum_{i=1}^{N} r_i^2 + \sum_{i<j}^{N} \exp(-\kappa r_{ij}) \frac{1}{r_{ij}},
\]

where coordinates and energies are dimensionless with the units \( r_0 = \left( \frac{Q^2}{2\pi \varepsilon_0 m \omega_0^2} \right)^{1/3} \) and \( E_0 = \left( \frac{m \omega_0^2 Q^4}{32 \pi^2 \varepsilon_0^2} \right)^{1/3} \). Furthermore, \( \omega_0 \) is the trap frequency, \( Q \) and \( m \) the dust charge and mass, \( r_i = \sqrt{x_i^2 + y_i^2 + z_i^2} \) is the distance of particle \( i \) to the trap centre. The indices \( i \) and \( j \) denote the particles and \( \kappa = r_0 / \lambda_D \) the screening strength, which is inversely proportional to the Debye length \( \lambda_D \). The first term in equation (1) describes the isotropic harmonic 3D confinement and the second the pairwise interaction between the charged particles. The interaction is of Yukawa type, since shielding by the ambient plasma should be taken into account.

Although the \( N = 49 \) cluster confined at low pressure lacks a perfect isotropic shape and even though wake field effects become important for this cluster [50–52], we can test the validity of state of the art theories [4, 8] in the limit of realistic experimental situations.

3.1. Fluid modes analysis

When the number of dust particles becomes sufficiently large, it is convenient to treat the system as a continuum. In addition, when the cluster is not in a solid state, a fluid description might be more adequate.

Kähler and Bonitz [8, 37] treated the Yukawa cluster as a fluid droplet and derived the modes by solving the plasma fluid equations. In their approach, the total potential of the dust cloud with time-dependent density \( n_d(\vec{r}, t) \) is expanded into a set of radial and angular eigenfunctions. Multipole moments of the density can be defined in terms of these eigenfunctions as

\[
q_{lm}(t) = Q \sqrt{\frac{4\pi}{2l+1}} \int n_d(\vec{r}, t) \hat{i}_l(kr) Y^*_{lm}(\theta, \phi) d\vec{r},
\]

with \( l \) and \( m \) being the angular mode numbers, \( \hat{i}_l(kr) \) the modified spherical Bessel function, and \( Y^*_{lm}(\theta, \phi) \) the spherical harmonics, respectively. In our analysis, weak screening was...
assumed by fixing the screening strength to $\kappa = 0.6$ but other values of $\kappa$ did not reveal any qualitative differences [23, 24].

In order to derive the experimental fluid modes, the particle positions at each time step are taken to calculate the multipole moments $q_{lm}$, where the density $n_d$ is treated as a series of $\delta$-functions according to $n_d(\vec{r}, t) = \sum_i \delta(\vec{r} - \vec{r}_i(t))$. For all fluid modes shown here, we calculated the power spectral density from 30,000 frames. The power spectral density $q_{lm}(\omega)$ of each fluid mode is taken as the Fourier transform of $q_{lm}(t)$ via

$$q_{lm}(\omega) = \frac{2}{TN} \left| \int_0^T q_{lm}(t) \exp(-i\omega t) \, dt \right|^2$$

over a time interval $T$.

### 3.2. Normal mode analysis (NMA)

For the sake of a broader picture, the dynamics has also been analysed by normal modes [4, 27, 35, 62, 63]. Thereby, it is assumed that the particles only perform small oscillations around their local equilibrium positions. Thus, using the dynamical matrix, i.e. the second derivative of the total energy given in equation (1),

$$H = \frac{\partial^2 E}{\partial r_\alpha, \partial r_\beta, i, \partial r_j},$$

the $3 \times N$ eigenvalues and eigenvectors are calculated with $\alpha$ and $\beta$ being the Cartesian coordinates $x, y, z$ and $i, j$ denoting the particle number. The eigenvalues define the frequencies of the different modes of the Yukawa cluster. The resulting 3D eigenvectors $\vec{e}_{i,l}$ describe the oscillation pattern of particle $i$ in each normal mode $l$.

From the measured time series, the velocities $\vec{v}_i(t)$ of each particle are mapped onto the eigenmode pattern according to

$$f_l(t) = \sum_{i=1}^N \vec{v}_i(t) \vec{e}_{i,l}.$$  

From this, the NMA spectral power density $S_l(\omega)$, i.e. the energy per frequency interval of each individual crystal mode $l$, follows from the Fourier transform of equation (5) similar to equation (3) above [27]. Contrary to the fluid mode technique, where the momentary particle positions inside the dust cloud are required, the NMA needs the full trajectories of each individual dust grain. For the NMA performed here, the full 3D trajectories of more than 1600 frames per experiment were used.

While both approaches are well applicable to dust clouds at high pressure, the analysis faces problems at low pressure since both methods are based on the isotropic Yukawa potential. In the latter case, the streaming ions make the potential anisotropic and lead to non-reciprocal inter-particle forces [52]. In principle, the ion focus can be taken into account by introducing an additional positive charge below each dust grain [46, 58, 64, 65] or by using the linear response formalism to calculate an improved interaction potential [50, 52]. However, the anisotropy of the clusters in the present experiment is not very pronounced, which is why we neglect these effects in our analysis. The comparison with the measurements is thus expected to yield useful information on the importance of wake effects in the theoretical description.
4. Results

For the analysis of fluid modes and NMA of Yukawa balls two clusters are considered. The first cluster is of spherical shape. The pressure (6.4 Pa) was high enough to suppress the onset of Schweigert oscillations. The second dust cloud under investigation has aligned particle chains. Here, the cluster was trapped at 4.8 Pa where an influence of the ion streaming motion onto the dust dynamics is expected. Both clusters were laser-heated in the same manner.

4.1. Fluid modes of the spherical dust cluster $N = 60$

We have observed a spherical dust cluster and recorded the particle motion over 30,000 frames. From that, the particle positions have been determined. Then, the spectra of the fluid modes were calculated for all angular mode numbers up to the order $l = m = 5$ according to equations (2) and (3). Here, the $q_{00}$, $q_{10}$ and $q_{11}$ fluid modes are presented as representatives for the monopole and dipole modes (with $q_{00}$ being breathing mode like $[8, 37]$, $q_{10}$ containing the sloshing mode in vertical direction and $q_{11}$ containing the sloshing mode in horizontal direction). As examples for fluid modes with a more complex structure, the spectra of the $q_{2m}$, $m = 0, 1, 2$ modes are depicted, corresponding to quadrupole modes.

Figure 3(a) shows the spectra of the fluid modes $q_{00}$, $q_{10}$ and $q_{11}$ and figure 3(b) the spectra of the fluid modes $q_{20}$, $q_{21}$ and $q_{22}$ for the unheated $N = 60$ cluster. Figures 3(c) and (d) show the same modes for the same cluster in the liquid regime at a laser heating power of $P_{L} = 250$ mW. In the absence of any external laser heating, figures 3(a) and (b), no fluid modes are detectable.

For the spectra of the heated cluster shown in figures 3(c) and (d), mainly two fluid modes are excited. This can be seen from the elevated signal of the $q_{11}$ fluid mode at 2.5 Hz and the $q_{22}$ fluid mode at 3.0 Hz. The value of the peak positions for all fluid modes shown here and in the following were determined by fitting a Gaussian distribution to the peak. The peak width was typically found from the fit as $\sigma = 1.4$ Hz. The reason for the excitation of the horizontal $q_{11}$ and $q_{22}$ modes can be understood by the fact that the beams are pointing in the horizontal direction to the dust cloud. Therefore, the lasers transfer momentum to the cluster particles mainly in this direction.

Interestingly, the ratio of the two mode frequencies $\omega_{22}/\omega_{11} = f_{22}/f_{11} = 3.0/2.5$ Hz = 1.2, is in good agreement with the fluid theory which predicts two limiting cases: (a) for $\kappa = 0$ one gets $\omega_{01} = \sqrt{3/2 + 1}$, $\omega_{22}/\omega_{11} = \sqrt{3 \cdot 2/(2 \cdot 2 + 1)} = 1.095$ and for (b) $\kappa \to \infty$ one finds $\omega_{01} = \sqrt{1}$, $\omega_{22}/\omega_{11} = \sqrt{2} \approx 1.41$ [8]. In particular, equating the theoretical ratio to 1.2 we obtain $\kappa = 0.57 \approx 0.6$, which is in good agreement with earlier findings (due to the weak dependence of the frequency on the screening parameter (see figure 2 in [8]), the error is relatively large).

A possible cause not to find any peak in the $q_{00}$ spectra (which describes a monopole breathing oscillation) is the presence of neutral drag. As pointed out by Kähler and Bonitz low damping rates are necessary to detect fluid modes [37]. In our experiment, the neutral gas pressure was held constant at 6.4 Pa. From this, the Epstein friction coefficient can be calculated as $\nu = 8 \text{s}^{-1}$ [66]. When we now assume that the peak of the $q_{11}$ mode is a sloshing oscillation at the trap frequency $f_{11} = 2.5$ Hz $\approx \omega_{01}/(2\pi)$ the normalized friction is $\nu/\omega_{01} \approx 0.5$. Even though the gas pressure is relatively low compared to most other Yukawa ball experiments [22, 23, 27], only a few broad peaks can be seen in the interesting frequency domain.

Figure 3. Spectra of fluid mode analysis of a spherical \( N = 60 \) particle cluster. Spectra of fluid modes \( q_{00}, q_{10} \) and \( q_{11} \) in (a) and \( q_{20} \) to \( q_{22} \) in (b) for the unheated cluster. Spectra of fluid modes \( q_{00}, q_{10} \) and \( q_{11} \) in (c) and \( q_{20} \) to \( q_{22} \) in (d) for the cluster in the liquid regime \((P_L = 250 \text{ mW})\). The unheated cluster does not show any characteristics at all. The laser heated cluster has a transverse sloshing mode \( q_{11} \) at 2.5 Hz and a transverse quadrupole mode \( q_{22} \) at 3.0 Hz.

4.2. Fluid modes of the wake-affected dust cluster \( N = 49 \)

It is tempting to reduce the neutral gas pressure in order to decrease friction of the particles. This should lead to richer fluid mode spectra [37]. As discussed in section 2, lowering the neutral gas pressure leads to an increasing influence of the ion focus on the particles [36, 49]. Thus, we can study the influence of damping and the influence of anisotropic particle–particle interactions to the fluid modes at once. The spectra of the same fluid modes as above are shown in figure 4 for a cluster with \( N = 49 \) particles to compare directly with the results of the isotropic cluster.

In the absence of any external laser heating, figures 4(a) and (b), indeed a few collective excitations appear in the elongated cluster. No peak occurs in spectra of the \( q_{00} \) and \( q_{10} \) fluid mode. An elevated signal can be found in the \( q_{11} \) mode at about 3.5 Hz. The \( q_{22} \) fluid mode, which represents a transverse quadrupole oscillation, has also a broad peak at the frequency domain of 4 Hz. Moreover, a sharp peak occurs in all quadrupole spectra and in nearly all spectra of the higher order modes at 7.8 Hz for this cluster indicating that this peak is not a pure fluid eigenmode.
Figure 4. Spectra of fluid mode analysis of an elongated $N = 49$ particle cluster as in figure 3. Laser heating destroys the ion focus related unstable oscillation at 7.8 Hz and mainly excites a sloshing movement at 3.2 Hz.

Even though the friction is rather low no $q_{00}$ mode is detectable (for 4.8 Pa and assuming the trap frequency to be $\omega_0/(2\pi) \approx 3.5$ Hz one finds $\nu/\omega_0 \approx 0.27$). The reason is in the elongation of the cluster along the $z$-axis and the particle-interaction, which is influenced by the ion focus. Such a cluster does not possess a uniform monopole mode.

High mode numbers generally correspond to more localized oscillations in the cluster. The observed sharp peak at 7.8 Hz in the spectra of the higher-order fluid modes for the $N = 49$ cluster thus belongs to an oscillation that does not involve large-scale motion. Moreover, the fact that quadrupole oscillations transverse to the ion flow $q_{22}$ are favoured over the longitudinal quadrupole oscillations seem to confirm that restoring forces are mainly in the direction perpendicular to the ion streaming direction even without laser excitation [48, 49, 60].

The origin of the transverse restoring forces can be attributed to the ion focus [46, 48, 49, 60]. The alignment can result in unstable oscillations—the Schweigert instability—since there are repulsive forces between the dust particles and attractive forces between ion cloud and the dust particles [47, 48, 52–59].

Additional evidence for the Schweigert instability in our experiment is given by the fact that the dust system is ‘heated’ by the instability [14, 31, 36, 46, 58, 64, 67–69]. For the dust cluster under investigation, the temperature (mean kinetic energy) of the cluster particles is found at $T_{\text{kin}} = m \langle v^2 \rangle / 3k_B \approx 7370$ K, i.e. far above room temperature even in the unheated case [14, 67]. For comparison, the cluster in section 4.1 has $T = 2930$ K.

Figure 5. (a) Transverse movement of selected particles in the cluster over a time span of 2 s. All particles belong to one particle chain where the particles are numbered from top to bottom. The correlated oscillatory movement of particles 2–4 is clearly visible. (b) The corresponding power spectra from the full trajectories (300 s), showing a peak in the spectra at 7.8 Hz.

To underline the above argument, the motion of individual particles was analysed. Therefore, the four representative particles of the particle chain were chosen (marked in figure 2).

In figure 5(a) the transverse (to the ion stream) movement $\rho(t)$ of the four particles within the cluster is shown over a time span of 2 s. The corresponding power spectra from the full trajectories (300 s) are depicted in figure 5(b).

For the uppermost particle 1, the trajectory does not show any particular oscillations, consequently the power spectrum is nearly flat. In contrast, the lower particles 2–4 respond to the ion wakefield from particles placed above it. This can be seen from the trajectories correlated oscillatory motion and the corresponding peak in the spectra of the particles, respectively. Clearly, the frequency of the peak in the spectrum of the individual particle motion $\rho(t)$ coincides with that of the $q_{2m}$ fluid modes. Furthermore, the amplitude of the oscillatory motion increases with particle positions further down the ion stream. This supports that these oscillations are due to the Schweigert instability. A second peak in the spectrum for particles 3 and 4 at the first harmonic ($\approx 16$ Hz) hints at the nonlinear character of these oscillations.

The ion wakes induce local small-scale perturbations of the cloud potential, leading to signals in the fluid spectra for higher mode numbers, see figure 4. Even when the fluid mode technique starts from the assumption of an isotropic Yukawa model, equation (1), it allows for identification of such instabilities.

To display the influence of heating, the fluid mode spectra for the laser heated $N = 49$ cluster are shown in figures 4(c) and (d). Heating was applied in the same manner as for the spherical cluster. Here, the peak at roughly 3.2 Hz is even more pronounced in the $q_{11}$ mode. A slightly higher signal is also found in the spectra of the $q_{10}$ mode at 5 Hz.

Contrary to the unheated case, no sharp peaks occur at 7.8 Hz for higher angular mode numbers $l = 2$. Instead, several other modes are excited at 5.6 and 11.4 Hz in the $q_{20}$ mode, at 2.6 and 6 Hz in the $q_{21}$ mode and at roughly 3.6 Hz in the $q_{22}$ fluid mode.
The ratio of the two mode frequencies $\omega_{22}/\omega_{11} = 4/3.5 \text{ Hz} = 1.14$ for the unheated, and $3.6/3.2 \text{ Hz} = 1.13$ for the heated case, respectively, agree well with the ratio for the spherical cluster and literature [8]. However, one must take into account that the assumptions of the theory, i.e., isotropic confinement and interaction, are not fully fulfilled here.

According to the fluid theory of [8], the frequency for excitations in the $q_{lm}$ fluid modes for the same $l$ but different $m$ should be independent of mode number $m$, since the underlying theory assumes isotropic confinement and isotropic particle–particle interaction. The fact that the frequencies of the $q_{1m}$ and $q_{2m}$ fluid modes change as $m$ changes could either indicate that the confinement in the experiment is not perfectly isotropic or that the symmetry in the particle–particle interaction is broken due to the presence of an ion focus.

### 4.3. Evolution of fluid modes during laser heating

To analyse the fluid behaviour of finite dust clouds more closely, the evolution of the $q_{11}$ mode and the $q_{22}$ mode with temperature is investigated. Here, we restrict to the $N = 49$ cluster, since the fluid modes are more pronounced than for the spherical cluster, but the general results are qualitatively the same. The mode evolution is shown in figure 6 as a function of laser heating power. The power of the two beams was varied between 0 and 400 mW in steps of 50 mW thus covering a wide range of fluidity.

For comparison, the kinetic temperature of the cluster particles as function of the applied laser power is shown in figure 6(c) together with the corresponding Coulomb coupling parameter. The Wigner–Seitz radius for the $N = 49$ cluster was estimated as $b_{WS} = (3/4\pi n)^{1/3} = 340 \mu \text{m}$ from the overall density. For the dust charge, the value of $Q = 6900e$ was used. The solid curve represents a parabolic fit of the temperature versus laser heating power. This parabolic dependence corresponds to a laser particle interaction via radiation pressure [38, 70]. It was shown previously [14] that the velocity distribution is not purely Maxwellian for the dust particles, but sufficient to assign a temperature to the 3D dust clusters.

In each spectrum of the $q_{11}$ fluid mode shown in figure 6(a), a peak is found at a frequency range of $(3.3 \pm 0.2) \text{ Hz}$. The position of the peak maximum does not change significantly with laser heating. This transverse sloshing mode was already discussed in the previous section. The peak height slightly increases as the laser power increases, and the peak gets broader. This is exactly what is expected for the fluid modes [37].

Figure 6(b) shows the evolution of the $q_{22}$ fluid mode. Here, a peak is found for all modes at a frequency interval of about $(3.7 \pm 0.2) \text{ Hz}$. As in figure 6(a), the peak height increases and broadens continuously. Similar behaviour was found for all fluid modes. The oscillation associated with the Schweigert instability at 7.8 Hz is seen for the unheated cluster at 0 mW. At higher heating this peak is destroyed, probably due to the frequent particle exchanges in the liquid regime.

### 4.4. NMA of finite dust clusters

One goal of this paper is to compare the fluid and crystal modes of finite dust clouds. Contrary to the fluid mode description, where the Yukawa ball is treated like a fluid droplet, the NMA describes the harmonic movement of individual dust grains in terms of crystal modes [4, 71]. To compare the crystal modes with the fluid mode results, a NMA is performed from the trajectories of the spherical and the aligned cluster for the unheated crystalline case ($P_L = 0 \text{ mW}$) and
Figure 6. Evolution of the $q_{11}$ fluid mode (a) and the $q_{22}$ fluid mode (b) during laser heating. In (c) the kinetic temperature of the cluster particles and the Coulomb coupling strength for the applied laser power are shown. The solid curve corresponds to a parabolic fit.

The reason for choosing the NMA instead of the recently established instantaneous normal mode (INM) technique is that the NMA allows to investigate the mode resolved spectral properties, whereas the INM only enables to reveal information about the total distribution of the eigenfrequencies [32, 72].
Figure 7. Normal mode spectra for the spherical $N = 60$ particle cluster ($p = 6.4$ Pa); (a) power spectral density per mode and (b) corresponding PSD for the unheated cluster, (c) power spectral density per mode and (d) corresponding PSD for a cluster in the liquid regime ($P_L = 250$ mW). In the mode resolved spectra, darker colours correspond to a higher energy per frequency interval. The circles correspond to calculated mode frequencies. Without external heating, the energy per mode is concentrated in a narrow frequency band.

Figure 7 shows the mode resolved NMA spectral power density for the unheated cluster and the heated cluster together with the total power density $S(\omega)$ (PSD), which follows from the summing up $S_l(\omega)$ over each individual mode $S(\omega) = \sum S_l(\omega)$.

For the crystalline cluster, figure 7(a), regions with higher power density can be found for each mode. These regions shift slightly to higher frequencies as the mode number increases. This is the expected trend and has been previously demonstrated for 2D and 3D clusters [27, 36]. The spectral width is much less than that of [27] due to the much lower pressure used in the present experiment. The corresponding PSD, figure 7(b), that is obtained from the integration over all modes, shows evenly distributed energy at each frequency in this representation. However, the interpretation of individual normal modes is not as straightforward as in the fluid mode description. For the heated cluster, see figure 7(c), the energy in the modes is not located in a narrow frequency band as for the solid cluster. Higher energy can be found in a much broader region up to 15 Hz for all modes. In the PSD, figure 7(d), most of the energy in the system is seen to be located in a broad frequency range with its maximum at about 2.6 Hz. Compared to the PSD for the solid cluster, figure 7(b), laser heating increases the values of the PSD about one order of magnitude since kinetic energy is deposited to the cluster particles. Frequent particle exchanges occurring in this liquid regime prevent the establishment of normal mode oscillations. Thus, the spectrum is much broader as for the cluster in the solid phase, so as the NMA is mainly suited for crystalline states.
The $N = 49$ cluster with aligned particle chains was investigated by means of NMA in the same manner as described above. For the solid cluster, figure 8(a), the features of the mode resolved NMA spectra of a solid weakly damped Yukawa cluster are recovered, too. Moreover, in this unheated case, a significant amount of energy is stored in a narrow frequency band around 7.8 Hz in most of all modes. Consequently, this feature is no eigenmode of the system [36]. The peak at 7.8 Hz becomes more distinct in the corresponding PSD, see figure 8(b). The contribution at 7.8 Hz is by far the most dominant input in the PSD (note that energy density in this narrow frequency band is about two magnitudes larger than for the unheated spherical cluster, figure 7). Thus, the Schweigert instability at 7.8 Hz can be identified within both, the crystal and the fluid mode approach.

The mode resolved NMA spectra of the heated chain-like cluster, see figure 8(c), looks very similar to that of the spherical cluster with laser heating. In the PSD, figure 8(d), the maximum of the energy is now found at 3.3 Hz due to the different discharge parameters.

It is a remarkable result that even when both approaches assume isotropy and use different data as an input (fluid modes require the 3D density, i.e. the particle positions, and NMA uses the particles velocities), the dominant spectral properties of the system are well recovered:

In the fluid regime, the peak of the $q_{11}$ fluid mode coincides with the maximum of the PSD for both clusters. Moreover, in the unheated case of the elongated cluster, the sharp peak at 7.8 Hz due to the ion focus is covered by the NMA and the fluid mode approach as well. A difference in both methods lies however in the allocation of the total energy of the system onto the individual crystal or fluid modes.
5. Summary

A comparison between two different approaches, the NMA and the fluid mode technique, describing the mode structure of finite 3D Yukawa type systems in solid and liquid phases was made. For that purpose, a cluster consisting of \( N = 60 \) particles representing a spherical Yukawa ball and a cluster formed from \( N = 49 \) particles, which was elongated along the ion streaming direction, were investigated. Both clusters, which were solid in the unheated case, were gradually heated by means of two opposing laser beams to a liquid state.

The fluid mode technique was applied to the experimental data. Here, the dust cluster is treated as a continuous fluid, similar to a fluid droplet. Diverse fluid modes could be identified in the motion of the finite dust clouds. Furthermore, for the crystalline but elongated cluster, the Schweigert instability at 7.8 Hz was seen as a sharp peak in high order modes, that corresponds to more local cloud oscillations.

Transverse laser heating mainly excites fluid modes that correspond to a transverse particle movement. The evolution of fluid modes was followed during laser heating. The mode frequencies were found to be nearly independent of heating power, but the peaks themselves were getting broader, in agreement with theoretical calculations.

A NMA was performed to compare the fluid modes with crystal eigenmodes. A clear mode spectrum has been derived for the unheated, crystal-like 3D clusters. Again, it has been revealed that the ion focus leads to an unstable oscillation of the cluster particles due to the Schweigert instability for the cluster which leads to particle alignment. With laser heating, i.e. when the clusters are in a liquid-like regime, the NMA spectra becomes less structured due to frequent particle exchanges.

Both methods yield comparable results concerning the spectral features of the system, even for the low particle numbers of the investigated cluster and under the influence of the ion focus. Also, both techniques hint at the non-isotropy of the confinement and particle interaction.

The results are a helpful input and feedback for theory and simulations. Future extensions of the theory, for instance for systems in a flowing environment, are desirable, as they are important for a quantitative comparison. This will allow to widen up the fluid mode description to a broader area of physical systems, even beyond dusty plasmas.

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A3

Nonequilibrium finite dust clusters: Connecting normal modes and wakefields

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Nonequilibrium finite dust clusters: Connecting normal modes and wakefields

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The dynamics of systems in a nonequilibrium environment is of enormous interest in many situations. Specifically, dusty plasmas allow a fundamental insight into the microphysical behavior of such systems since the individual particles can be traced on the kinetic level. In dusty plasma systems the particles are typically trapped in a (plasma-mediated) confining potential and interact via a shielded Coulomb interaction. The nonequilibrium environment stems, among others, from the ions streaming through the dust system, e.g., in the space-charge sheath where the dust particles generally are confined [1].

The particles in a dusty plasma usually are highly charged microspheres and, hence, the dust-dust interaction is strongly coupled [1–3]. The spatial and temporal scales of the dust motion allow for a detailed observation by video microscopy [4] where the particle motion can be resolved on the microscopic dynamic level. Here, we will focus on three-dimensional (3D) dust clusters of a small number of particles ($N < 50$, say) in a spherical confinement (so-called Yukawa balls [5–7]).

Under typical particle trapping conditions the dust particles are found in an environment with streaming ions. The ion streaming motion then leads to the formation of an ion wake, or ion focus, which is a region of positive space charge downstream from the dust particle [8–21]. This ion focus leads to a nonreciprocal interaction between the dust particles [10,12,22–25] where a dust particle sitting further downstream in the ion flow feels an attraction from the ion focus of the upstream particle, whereas the upstream particle is repelled from the downstream particle. The influence of the ion focus on the structure formation has been studied (see, e.g., [23,25–27]). Further, this nonequilibrium ion focus leads to the onset of instabilities (so-called “Schweigert” oscillations) and to the heating of the dust system and, subsequently, to solid-liquid phase transitions [22,28–30]. The occurrence of ion-focus-driven oscillatory instabilities in extended and finite systems has been demonstrated [15,18,31–33].

To analyze the dynamical properties of finite (dust) systems the normal mode analysis (NMA) is a well suited technique [34–37]. In finite systems, the normal mode spectra assume the role of wave spectra [16,38–40] of extended systems. A related technique, the instantaneous normal mode (INM) analysis [41–46] has also been successfully applied to dusty plasmas [47–49]. These techniques, however, have always relied on the equilibrium energy to derive the mode patterns and oscillations.

It is now tempting to study how the techniques of NMA and INM, which are well established for equilibrium situations, can be extended to account for this nonreciprocal ion-focus interaction and the corresponding instabilities. We will demonstrate how this nonreciprocity can be taken into account in the NMA and INM analysis of a 3D finite system. From that, an analytical model for a few-particle system is derived. The NMA and INM technique extended by the ion focus will then be applied to dust systems in the experiment and compared to the model. The differences arising between NMA and INM lead to interesting consequences in the stability analysis of these finite systems.

I. INTRODUCTION

The dynamics of systems in a nonequilibrium environment is of enormous interest in many situations. Specifically, dusty plasmas allow a fundamental insight into the microphysical behavior of such systems since the individual particles can be traced on the kinetic level. In dusty plasma systems the particles are typically trapped in a (plasma-mediated) confining potential and interact via a shielded Coulomb interaction. The nonequilibrium environment stems, among others, from the ions streaming through the dust system, e.g., in the space-charge sheath where the dust particles generally are confined [1].

The particles in a dusty plasma usually are highly charged microspheres and, hence, the dust-dust interaction is strongly coupled [1–3]. The spatial and temporal scales of the dust motion allow for a detailed observation by video microscopy [4] where the particle motion can be resolved on the microscopic dynamic level. Here, we will focus on three-dimensional (3D) dust clusters of a small number of particles ($N < 50$, say) in a spherical confinement (so-called Yukawa balls [5–7]).

Under typical particle trapping conditions the dust particles are found in an environment with streaming ions. The ion streaming motion then leads to the formation of an ion wake, or ion focus, which is a region of positive space charge downstream from the dust particle [8–21]. This ion focus leads to a nonreciprocal interaction between the dust particles [10,12,22–25] where a dust particle sitting further downstream in the ion flow feels an attraction from the ion focus of the upstream particle, whereas the upstream particle is repelled from the downstream particle. The influence of the ion focus on the structure formation has been studied (see, e.g., [23,25–27]). Further, this nonequilibrium ion focus leads to the onset of instabilities (so-called “Schweigert” oscillations) and to the heating of the dust system and, subsequently, to solid-liquid phase transitions [22,28–30]. The occurrence of ion-focus-driven oscillatory instabilities in extended and finite systems has been demonstrated [15,18,31–33].

To analyze the dynamical properties of finite (dust) systems the normal mode analysis (NMA) is a well suited technique [34–37]. In finite systems, the normal mode spectra assume the role of wave spectra [16,38–40] of extended systems. A related technique, the instantaneous normal mode (INM) analysis [41–46] has also been successfully applied to dusty plasmas [47–49]. These techniques, however, have always relied on the equilibrium energy to derive the mode patterns and oscillations.

II. THEORETICAL TREATMENT AND MODEL

The normal modes of a finite system under equilibrium conditions are derived by starting from the ground-state energy of the system which is given by

$$E = \frac{1}{2} m_0 \sum_{i=1}^{N} \sum_{j=1}^{N} \frac{Z^2 e^2}{4 \pi \varepsilon_0} \sum_{i<j} \exp(-r_{ij}/\lambda_0) \cdot \mathbf{r}_{ij}.$$  (1)

where $r_{ij}$ is the distance of particle $i$ from the trap center and $r_{ij}$ is the distance between particles $i$ and $j$. The dust mass is denoted by $m$, and the dust charge number by $Z$. Further, $e$ is the elementary charge and $\varepsilon_0$ the vacuum permittivity. The first term describes the potential energy in the (assumed harmonic) confining potential of strength $\lambda_0$ and the second term is the Coulomb repulsion between the particles including shielding by the ambient plasma with the Debye shielding length $\lambda_D$.

A. NMA and INM

In the NMA the modes are derived from a harmonic approximation to the potential energy surface around the equilibrium configuration. They are computed as the eigenvalues and eigenvectors of the dynamical matrix (Hessian) of the energy,
Real values of $\omega_\ell$ correspond to potential wells in the momentary energy landscape of the system, in which the particles can oscillate around their current equilibrium in the cage of the nearest neighbors. Imaginary values of $\omega_\ell$ correspond to the potential hills that separate the minima. It is now argued [41–45] that especially the unstable part can be related to the liquid properties, such as the diffusion constant $D$, since the thermal energy drives configurational transitions to take place over these potential hills associated with $\rho_v(\omega)$. Using this approach, the diffusion and liquid-solid phase transitions have recently been studied in finite dust clusters [47–49].

B. Accounting for the ion focus

The presence of an ion focus (or wakefield) is a well-known phenomenon in dusty plasmas with streaming ions (such as in the space charge sheath) [8–21,51,52]. The streaming ions are deflected in the electric field of the negatively charged particle and are scattered into a region downstream from the dust particle. There, they form a region of enhanced positive charge, the ion focus. It has earlier been noted [10,22] that the interaction between two dust particles at different heights in the sheath is not reciprocal, but that rather the downstream particle feels an attraction due to the ion focus of the upstream, but the upstream particle is repelled from the downstream (i.e., Newton’s third law of action and reaction is seemingly violated). This behavior has been demonstrated experimentally [12,23–25]. Ion-neutral collisions somewhat affect the strength of the ion focus, but its nonreciprocal attraction is severely diminished only at much higher gas pressures than used here [23,25,51,53].

Often (see, e.g., [10,14,15,19,21,22,51,52]), the ion focus has been modeled as a positive point charge $Z_+$ rigidly attached to the dust particle at a distance $d_\ast$ below the dust [see Fig. 1(a)]. Following this assumption together with the nonreciprocity of the attraction, we have added the interaction of a dust particle with the ion focus of another particle by an

![FIG. 1. (Color online) (a) Model of the dust-dust interaction. The ion focus is modeled as a point charge below the dust. Here the situation is illustrated for three vertically interacting particles. (b) and (c) Horizontal and vertical modes of the three-particle system. See text for details.](image-url)
additional term in the energy equation as
\[ E_W = \frac{Z^2 Z}{4\pi \epsilon_0} \sum_{j=1}^{N} \sum_{i=1}^{N} \exp\left(-r_{ij}/\xi_0\right) r_{ij}, \] (6)
where \( r_{ij} = |\vec{r}_i - (\vec{F}_i - d_i \hat{z})| \) is the distance between particle \( i \) and the ion focus of particle \( j \), and \( \hat{z} \) is the unit vector in vertical direction. Note that \( E_W \) is attractive. As indicated by the prime at the sum symbol, the nonreciprocity of the interaction is accounted for by adding \( E_W \) only for particles \( i \) that are below the ion focus of particle \( j \), i.e., \( z_i < z_j - d_i \). Further, this interaction is taken into account only in the horizontal motion in the \( x \) and \( y \) directions without any coupling to the vertical component, i.e., only contributions to \( \partial^2 E/\partial r_{i\alpha} \partial r_{j\beta} \) with \( \alpha, \beta = \{x, y\} \) are considered. This kind of interaction is proposed, e.g., by Refs. \([22, 29, 51]\). Hence, the pairwise interaction force is pure Coulomb (\( \alpha = \beta \)).

These unstable types of oscillations (so-called Schweigert oscillations) have been observed, e.g., also in the phase transition of multilayer dust systems \([11, 29, 32]\). The situations where these oscillations with complex eigenvalues occur will be termed Schweigert unstable in the following.

It should be noted that our model does not account for any mode coupling between horizontal and vertical oscillations (see discussion in Sec. V).

C. Three-particle model

We like to illustrate the above model for a three-particle system that can be treated analytically (a system of only two particles cannot become Schweigert unstable within the framework of this model). We consider a situation with three particles confined in a potential well of strength \( \omega_0 \) where the particles are vertically aligned due to the action of the ion-focus attraction, as shown in Fig. 1. This model will later be compared to the experimental situation.

The equation of motion for the three particles in the horizontal (\( x \)) direction can be written as, with \( F = -\nabla E \) with the energies \( E \) given by Eqs. (1) and (6),
\[ \ddot{x}_1 + \beta \dot{x}_1 + \omega_0^2 x_1 = -\omega_0^2 (x_2 - x_1), \]
\[ \ddot{x}_2 + \beta \dot{x}_2 + \omega_0^2 x_2 = (\xi - 1) \omega_0^2 (x_1 - x_2) - \omega_0^2 (x_3 - x_2), \] (7)
\[ \ddot{x}_3 + \beta \dot{x}_3 + \omega_0^2 x_3 = (\xi - 1) \omega_0^2 (x_2 - x_3), \]
where \( x_i \) is the horizontal excursion of particle \( i \) from the aligned situation and \( \beta \) is the Epstein friction coefficient due to the neutral gas damping. Here, we have boiled down the situation to the minimum complexity assuming that the particle interaction is pure Coulomb (\( \lambda_0 \rightarrow \infty \)). The horizontal interaction strength is then \( \omega_0^2 = Z^2 e^2/(4\pi \epsilon_0 m d^3) \) and the ion-focus strength \( \xi = (Z/\lambda_0)(d/d - d_i)^3 \) accounts for the ion-focus attraction \([23]\). The ion focus of particle 1 is acting on particle 2 (but not vice versa, since particle 2 is found below particle 1). Equivalently, the ion focus of particle 2 influences particle 3. Hence, in this model only nearest-neighbor interaction is considered. In principle, one can account for the interaction of all particles (repulsion of particle 1 by particle 3 and attraction of particle 3 by ion focus of particle 1). However, in the above approximations, the interaction strength between particle 1 and 3 is only 1/8 of the interaction of particle 1 and 2 (due to the \( d \rightarrow \infty \) scaling of \( \omega_0^2 \) leading to only minor corrections and, more importantly, one would lose the simple analytical expression for the mode frequencies (see below).

From this model, the dynamical matrix is asymmetric as mentioned above and found as
\[ A_{xx} = \begin{pmatrix} \omega_0^2 - \omega_0^2 & 0 & -\omega_0^2 \\ -\omega_0^2 & \omega_0^2 + (\xi - 2) \omega_0^2 & \omega_0^2 \\ 0 & -\omega_0^2 & \omega_0^2 + (\xi - 1) \omega_0^2 \end{pmatrix}, \] (8)
where only the \( x \times x \) component is given which is sufficient for the following analysis. It is straightforward to verify that the eigenvalues \( \omega_0^2 \) of \( A_{xx} \) are
\[ \omega_0^2 = \omega_0^2, \] (9)
\[ \omega_0^2 = \omega_0^2 + \omega_0^2 (\xi - 2 - \sqrt{1 - \xi}), \] (10)
\[ \omega_0^2 = \omega_0^2 + \omega_0^2 (\xi - 2 + \sqrt{1 - \xi}) \] (11)
which for the case of no ion focus (\( \xi = 0 \)) reduce to \( \omega_0^2 = \omega_0^2 - 3\omega_0^2 \) and \( \omega_0^2 = \omega_0^2 - 3\omega_0^2 \). The corresponding mode oscillation patterns are given in Fig. 1(b). The first mode is just the “slashing” oscillation of all particles in the confining potential, the second mode is the zigzag transition mode \([54, 55]\), and the third is a “weaker” variant of the zigzag mode. In the absence of the ion focus, the relative magnitude of \( \omega_0^2 \) and \( \omega_0^2 \) determine whether the aligned situation is the equilibrium situation. For \( \omega_0^2 > 3\omega_0^2 \) the vertical chain is stable, otherwise it is absolutely unstable, i.e., \( \omega_0^2 = \sqrt{\omega_0^2 - 3\omega_0^2} \) is purely imaginary.

For \( \xi > 1 \) the eigenvalues \( \omega_0^2 \) become complex (complex conjugate pairs, to be precise). This indicates the onset of the “Schweigert” oscillations with growing amplitudes. No mode coupling to the vertical oscillations is required, here. Obviously the complex eigenvalues go along with complex eigenvectors. The resulting mode oscillation pattern is a combination of modes 2 and 3 in Fig. 1(b), but with phase lags between the individual particle oscillations (reflecting the real and imaginary parts of the eigenvector).

Similarly, the vertical excursions \( z_i \) of the three-particle chain are described as
\[ \ddot{z}_1 + \beta \dot{z}_1 + \omega_0^2 z_1 = \omega_0^2 (z_2 - z_1), \]
\[ \ddot{z}_2 + \beta \dot{z}_2 + \omega_0^2 z_2 = \omega_0^2 (z_3 + z_2 - 2z_1), \] (12)
\[ \ddot{z}_3 + \beta \dot{z}_3 + \omega_0^2 z_3 = \omega_0^2 (z_3 - z_2). \]
The vertical interaction is not influenced by the ion focus and, hence, the interaction is pairwise symmetric and repulsive. The vertical interaction strength is described by \( \omega_0^2 = 2Z^2 e^2/(4\pi \epsilon_0 m d^3) = 2\omega_0^2 \) and the vertical interaction
part of the dynamical matrix then is symmetric and
\[ A_{zz} = \begin{pmatrix}
\omega_z^2 + \omega_x^2 & -\omega_z^2 & 0 \\
-\omega_z^2 & \omega_x^2 + 2\omega_y^2 & -\omega_x^2 \\
0 & -\omega_z^2 & \omega_y^2 + \omega_z^2
\end{pmatrix}, \]
(13)
with the eigenvalues
\[ \begin{align*}
\omega_1^2 &= \omega_x^2 \\
\omega_2^2 &= \omega_y^2 \\
\omega_3^2 &= \omega_z^2 + 3\omega_y^2.
\end{align*} \]
(14)
Again, the first mode corresponds to the vertical sloshing motion, and the other two to relative vertical motions as indicated in Fig. 1(c). As stated above the ion focus is considered not to influence the vertical interaction (and also no coupling of the vertical motion to the horizontal is taken into account here).

Concluding, from this model the nine modes of the three-particle system are then the three vertical modes at \( \omega_{1,2,3} \) and the 2 \times 3 horizontal modes at \( \omega_{01,2,3} \) which are twofold degenerate in \( x \) and \( y \) due to the symmetry in the horizontal plane.

\section*{III. RESULTS: THREE-PARTICLE EXPERIMENT}

The experiments have been performed in a capacitively coupled radio-frequency (\( f_{rf} = 13.56 \text{ MHz} \)) discharge in argon described in detail elsewhere \cite{5,56,57}. Melamine-formaldehyde microspheres of 4.86 \( \mu \text{m} \) diameter are trapped inside a cubic glass box that provides a harmonic 3D confinement for the particles \cite{58}. The particle motion is followed in three dimensions using a stereoscopic camera setup \cite{56} and time series of 30,000 frames at a frame rate of 100 Hz have been recorded.

Figure 2 shows the full particle trajectories for a three-particle system at a gas pressure of 6 Pa at different discharge powers. At the lower powers the three particles are vertically aligned. At 0.8 W [Fig. 2(a)] the particles form a triangle, where, however, two of the three particles still seem to be vertically aligned. The dynamics of this system will be analyzed in the following and compared to the analytical calculations.

The INM spectrum is obtained from the particle configurations in each frame as described in Sec. II A. The mode-resolved NMA spectrum \cite{34} is determined experimentally via
\[ S_\ell(\omega) = \frac{2}{T} \left| \int_{-T/2}^{T/2} f_\ell(t) \exp(-i\omega t) dt \right|^2, \]
(17)
where \( f_\ell(t) = \tilde{V}(t) \cdot \tilde{e}_\ell \) is the projection of the particle velocities \( \tilde{V}(t) = \Delta \tilde{X}(t)/\Delta t \) (\( \Delta t = 0.01 \text{ s} \) according to the camera frame rate) onto the mode pattern \( \tilde{e}_\ell \) of mode number \( \ell \). The spectral density \( S_\ell(\omega) \) of mode \( \ell \) is then just the Fourier transform of this projection. The total power spectral density (PSD) \( S(\omega) \) is the summation over the mode-resolved spectral density \( S_\ell(\omega) = \sum_\ell S_\ell(\omega) \).

\subsection*{A. Analysis ignoring the ion focus}

We start analyzing the three-particle system at 0.8 W shown in Fig. 2(a) with the standard approach that neglects the ion-focus effect. The mode-resolved spectrum is shown in Fig. 3 in false colors together with the total power spectral density and the INM spectrum. Here, we have taken the limit of infinite...
screening length $\lambda_D \to \infty$ to allow for comparison with the simple analytical model.

The mode numbers in Fig. 3 correspond to those in Figs. 1(b) and 1(c), i.e., the modes 5 and 6 are the two vertical modes and the modes 2x (2y) and 3x (3y) are the horizontal modes, where the modes 2x and 2y as well as 3x and 3y are identical, but oriented along the x and y directions, respectively. The center-of-mass modes at $\omega = \omega_0$ are omitted for clarity, here. The normal modes and the PSD reveal that most energy is stored at three distinct frequencies, namely, $f_{2,3} = 4.3$ Hz, $f_5 = 7.0$ Hz, and $f_6 = 9.8$ Hz. It is interesting to note here, that the mode-resolved NMA spectrum yields nearly the same mode frequency of $f_{2,3} = 4.3$ Hz for the modes 2 and 3, although these modes describe different oscillation patterns.

The INM spectrum shown in the upper panel shows three distinct frequencies with $f > 0$, namely, at $f = 10.2$ Hz ($\approx f_6$), at $f = 6.1$ Hz ($\approx f_5$), and at $f = 1.5$ Hz, and a fourth absolutely unstable frequency of $f = 15.4$ Hz (which absolute value is plotted, by convention, on the negative frequency axis as $-5.4$ Hz). These last two frequencies do not really match the mode frequencies of either of the first four modes. This means that the vertical modes 5 and 6 are adequately modeled using the standard approach of NMA, but the horizontal modes 2x (2y) and 3x (3y) are not well described.

The INM mode frequencies are well reproduced from the analytical model where the confinement frequency is obtained as $\omega_0/(2\pi) = 3.8$ Hz, the vertical frequency is $\omega_0/(2\pi) = 5.6$ Hz, and $\omega_0/(2\pi) = 3.5$ Hz $\approx (1/\sqrt{2})\omega_0/(2\pi)$. From that, the dust charge number follows to be $Z = 8100$. This would result in the following mode frequencies (compare Sec. II C):

\[
\begin{align*}
    f_2 &= \sqrt{f_0^2 - 3f_0^2} = 14.7 \text{ Hz}, \\
    f_3 &= \sqrt{f_0^2 - f_0^2} = 1.5 \text{ Hz}, \\
    f_5 &= \sqrt{f_0^2 + f_0^2} = 6.7 \text{ Hz}, \\
    f_6 &= \sqrt{f_0^2 + 3f_0^2} = 10.4 \text{ Hz}.
\end{align*}
\]

The slight numerical differences between the analytical model and the INM values (derived from the experimental configuration) stem only from the fact that, in the experiment, the vertical distances between the three particles are not identical as assumed in the analytical model. Hence, the analytical model allows one to generally nicely retrieve the mode frequencies in this three-particle system.

To conclude, the standard approach of NMA reveals oscillatory motions for modes 2 and 3 at a distinct frequency. The INM (and the analytical model) fail to explain these horizontal mode frequencies when the effect of an ion focus is neglected. Especially the fact that these two modes show the same oscillation frequency cannot be recovered (not even an anharmonic confinement can resolve this issue). Instead both INM and the analytical model would characterize this vertically aligned arrangement as absolutely unstable.

We now analyze the same data, but including the ion-focus effect according to Eq. (6) using $Z_{\parallel} = 0.7Z$ and $d_{\parallel} = 0.33d$ (other combinations of $Z_{\parallel}$ and $d_{\parallel}$ that lead to the same ion-focus parameter $\xi = 2.3$ show the same outcome). The results are shown in Fig. 4.

First of all, it is seen that the INM spectrum shows complex eigenvalues. In the INM spectrum, we find real, positive eigenvalues and complex eigenvalues where the real part is positive, and the imaginary part shows that we have complex conjugate pairs. The following mode frequencies $f_{2,3} = 4.5 \pm i0.8$ Hz, $f_5 = 6.6$ Hz, and $f_6 = 10.8$ Hz are obtained. Hence, these eigenvalues indicate only stable modes (modes 5 and 6 with real, positive eigenvalues) or Schweigert unstable modes (modes 2 and 3; complex eigenvalues with positive real part). We do not find any eigenvalues which are purely imaginary which would indicate absolutely unstable configurations. We can thus conclude that the observed configuration is adequately...
FIG. 5. (Color online) Variation of the ion-focus strength $\xi$ for the three configurations shown in Fig. 2. The lower panels show the normal mode spectra of modes numbers 2 and 6. The upper panel shows the eigenvalues of the dynamical matrix $f_2$ and $f_6$ for these two modes as a function of the ion-focus strength $\xi$. For the horizontal modes real and imaginary parts of $f_2$ are indicated.

C. Strength of the ion focus

Now the role of the ion-focus strength $\xi$ will be investigated in more detail. Figure 5 shows the behavior of the horizontal mode number 2 and the vertical mode number 6 as a function of frequency for the three-particle configurations of Fig. 2. The corresponding mode frequencies as obtained from INM as a function of the ion-focus strength are also indicated. For the horizontal mode number 2 both real and imaginary parts are given. The real part corresponds to the observable oscillation frequency [59].

Starting with the situation for 0.8 W as (described in the previous sections) the INM frequencies $f_2$ of mode number 2 in the upper panel show purely imaginary eigenvalues for ion-focus strengths below $\xi < 0.8$. There the real part is zero and the imaginary part is finite. This corresponds to the absolutely unstable situation described in Sec. III A where this vertically aligned situation of the three particles is always unstable. Obviously, absolutely unstable regimes cannot be realized in long-run experiments.

For $\xi > 1.7$ the imaginary part is zero and the real part is finite indicating a situation where this aligned situation is stable without any self-excited Schweigert oscillations. In the medium range $0.8 < \xi < 1.7$ the frequency $f_2$ is complex with finite real and imaginary parts indicating the possibility of Schweigert oscillations. The best agreement between the NMA spectra of the two modes and the INM frequencies is obtained for ion-focus strengths of $\xi \approx 1.5$. This is just in the range of Schweigert oscillations, but very close to the stable situation. This is also seen directly in the trajectories that show only relatively small excursions around the equilibrium configuration [60].

For the second configuration at 1.2 W, the Schweigert oscillation range is much broader and agreement between INM values and the NMA spectra is seen for $\xi \approx 3$. Hence, here the effect of the ion focus is much stronger than for 0.8 W. This can also be seen from the trajectories that show much stronger oscillatory motion. Consequently, the NMA spectra have a

which very well reflect the measured mode frequencies (again slight numerical deviations arise from the slight difference in the vertical separation between the three particles).
much higher power density (note the different ordinate scales). The instability in this case is so strong that the horizontal mode number 2, whose power density peaks around 4 Hz couples to the vertical oscillation that, while having its main peak around 9.5 Hz, shows a second peak at the horizontal mode frequency.

Finally, for the triangular configuration at 1.6 W the situation is slightly different. There we do not find a regime of ion-focus strengths where the observed configuration can be identified as stable. In contrast, at low focus and high focus strengths the situation is absolutely unstable: At low strength the vertically aligned pair of particles at $y \approx -0.1$ mm in Fig. 2(c) tends to a not-aligned situation, whereas at high strength the configuration tends to become a three-particle vertically aligned chain similar to that in Fig. 2(a). For medium values of $\xi$ the configuration is oscillatory unstable, and a reasonable agreement between measured NMA frequencies and INM frequencies is found for $\xi \approx 5$. Also here, a coupling of the unstable horizontal oscillations to the vertical mode is seen. Even though this triangular configuration looks similar to the expected equilibrium configuration without ion focus the present analysis suggests a strong influence of the ion focus.

From the above investigations it is seen that the ion-focus strength increases with higher discharge power. How the discharge power affects the formation of the ion focus is not obvious. In the experiment it is seen that, with increased power, the levitation height of the dust cloud above the lower electrode decreases. Also the particle separation decreases (see Fig. 2). This might hint at a decreased sheath width with stronger electric fields that might result in higher ion streaming velocity and thus increased ion-focus strength.

### IV. RESULTS FOR LARGER PARTICLE NUMBER

The above NMA-INM analysis can, of course, also be used to describe larger systems. First, we show an example of a five-particle system consisting of a three-particle and a two-particle vertically aligned string. The gas pressure in this experiment was 7.2 Pa and the discharge power 1.3 W. The corresponding mode spectrum consisting of the 12 eigenmodes excluding the three sloshing modes is shown in Fig. 6 together with the INM eigenvalues. An ion-focus strength of about $\xi \approx 2.5$ has been obtained as the best fit using the method described above. For comparison, when an ion focus is neglected ($\xi = 0$), four absolutely unstable modes are obtained. The number of absolutely unstable decreases with increasing $\xi$ and for an ion-focus strength of about $\xi \approx 2.5$ no absolutely unstable mode is found. For this value, there are three pairs of Schweigert unstable modes which comprise the highest power spectral density. This again substantiates that these modes are indeed best described using the ion-focus model.

In a second example, a 30-particle cluster has been analyzed that shows a strongly prolate shape (gas pressure 7.8 Pa, discharge power 1.2 W) (see Fig. 7). The ion-focus strength used for the NMA/INM analysis is about $\xi \approx 6$, here. As in the previous cases, the adoption of an ion focus strongly reduces the number of absolutely unstable modes. Nevertheless, possibly due to the strongly prolate shape, we were not able to completely get rid of all absolutely unstable modes in this case.
cluster in relation to the vertical. The second is the vertical order parameter VOP, that describes the number of vertical bonds related to the total number of bonds. For this cluster, we obtain $a = 0.35$ and VOP = 0.1 which corresponds to $Z_+/Z$ of 0.45 giving an ion-focus strength of $\xi \approx 7$ (an ion-focus distance of $d_+ / d_+ \approx 0.4$ has been used there) [compare Fig. 9(a) of Ref. [61]]. This correlates very well with our dynamic results.

V. DISCUSSION OF THE MODEL

Finally, we like to add a few notes on the ion-focus model that has been used here.

A. Comparison to similar models

Our approach is on the one hand somewhat comparable to but on the other hand principally different from that of others, e.g., Refs. [14,15,18,21,52]. These authors aim at the description of a mode-coupling instability between horizontal and vertical modes (compare also Ref. [62]). Therefore, they analyze a two-dimensional system of dust particles with an ion focus as a point charge below the negative dust particle, whereas NMA uses the particle velocities and their ion foci are essentially in the same layer, and only small horizontal and vertical excursions are treated. The wave propagation in the horizontal direction couples to the vertical motion, which is essential for the instability in their model.

In contrast to our approach, the interaction between dust and ion focus in Refs. [14,15,21,52] is pairwise symmetric resulting in a dynamical matrix that also is symmetric. However, the mode coupling of horizontal and vertical motion leads to complex interaction terms in the $x$-$z$ or $y$-$z$ components. These complex entries then lead to complex eigenvalues, describing unstable hybrid (mode-coupled) oscillations.

In our situation the downstream particles are located beneath the ion focus of the upstream particles. Hence, our investigations essentially refer to vertically elongated systems. The interaction is nonreciprocal resulting from the non-Newtonian interaction between upstream and downstream particles, as explained above, and this asymmetry specifically manifests only in the horizontal modes $x$ or $y$. The nonreciprocal interaction directly yields an asymmetric matrix, but with real elements.

As a consequence, the mode-coupling instability [14,15,18,21,52] is a quite subtle instability that requires relatively low neutral-gas damping and horizontally extended systems, whereas our model describes a comparatively robust instability of vertically extended and aligned clusters.

B. Dynamical information from INM and NMA

The dynamical analysis without accounting for an ion focus has led to differences between INM and NMA in the prediction of oscillation frequencies (see, e.g., Fig. 3). One might now suspect that using such an inappropriate model to describe the dynamics naturally leads to such discrepancies. However, both INM and NMA make use of the same energy equation, Eq. (1), and of the same data: The INM use directly the particle positions, whereas NMA uses the particle velocities that are derived from the particle positions in different frames.

So, it is not obvious that the two methods should give different results. Or oppositely, what can be learned when NMA and INM agree with each other (using, e.g., the proposed ion-focus model)? To check this, we will investigate here how an oscillating particle configuration is treated in NMA and INM, respectively.

To start with, we consider a particle configuration that oscillates at a certain frequency $\omega^*$ with amplitude $\vec{X}_0$ around a particular configuration $\vec{X}_0$ (that is not necessarily an equilibrium configuration). Hence, the motion is given as

$$\vec{X}(t) = \vec{X}_0 + \vec{X}_* \sin(\omega^* t). \quad (18)$$

Here, as above, $\vec{X}$ is the state vector of all particle positions. In the NMA, now the mode projection becomes

$$f_\ell(t) = \frac{d\vec{X}(t)}{d^\iota} \cdot \hat{e}_\ell = \omega^* \cos(\omega^* t) \vec{X}_0 \cdot \hat{e}_\ell$$

and the mode-resolved spectra are found as [see Eq. (17)]

$$S\ell(\omega) = \frac{2}{T} \left| \int_{-T/2}^{T/2} \omega^* \cos(\omega^* t) \exp(-i\omega t) \vec{X}_0 \cdot \hat{e}_\ell dt \right|^2$$

$$= \frac{2\omega^* \sin(\omega^* T)}{T} (\vec{X}_0 \cdot \hat{e}_\ell)^2 \delta(\omega - \omega^*). \quad (19)$$

This means, in the NMA a sharp peak is obtained at a frequency $\omega = \omega^*$ when the oscillation vector has a nonzero contribution along the eigenmode pattern, i.e., $\vec{X}_0 \cdot \hat{e}_\ell \neq 0$. If the assumed oscillation is purely along the eigenmode pattern $\hat{e}_\ell$, we only find a contribution at this particular eigenmode due to the orthogonality of the eigenmodes. Otherwise, we find a contribution at $\omega = \omega^*$ for more modes. Most important, the mean configuration $\vec{X}_0$ does not contribute to the NMA spectral density directly.

In contrast, for the INM the mode frequencies are derived as the eigenvalues of the dynamical matrix that in turn depends on the instantaneous particle positions. Hence, the eigenvalues can be developed in first order as

$$\omega_\ell(t) = \omega_\ell(\vec{X}_0) + \vec{V}_{\ell\ell} \cdot \vec{X}_0 \sin(\omega^* t). \quad (20)$$

As a result, the density of states $\rho(\omega)$ is centered around $\omega_\ell(\vec{X}_0)$ with a width that depends on the oscillatory part. So, the oscillatory motion only enters in the width of the distribution of mode frequencies, but the central value is determined by the configuration $\vec{X}_0$. Since in our assumptions the configuration $\vec{X}_0$ is not necessarily the equilibrium configuration, also the mode frequencies $\omega_\ell(\vec{X}_0)$ in general differ from that of the equilibrium. Obviously, NMA and INM “see” different aspects of the particle dynamics. NMA is sensitive to the oscillatory part and INM to the configurational part.

But now, and this is the important point here, when the cluster oscillation is around the equilibrium configuration $\vec{X}_0$ with the corresponding mode frequency $\omega_\ell$, then both methods, INM and NMA, give the same frequency $\omega_\ell$. In NMA the mode-resolved spectrum is peaked at $\omega_\ell$ and in INM, the central frequency is $\omega_\ell \equiv \omega_\ell(\vec{X}_0)$ with a width related to the oscillation amplitude. Conversely, when one finds the same frequencies of cluster oscillations in INM and
NMA one can conclude that the observed configuration is an equilibrium configuration under the assumed interaction model.

Hence, the fact that we find the differences in frequencies between INM and NMA in Sec. III A where we have ignored ion-focus effects states that this is not the right model to account for the ion-focus cluster configuration. In contrast, in Secs. III B and IV where the INM frequencies agree with the NMA spectrum our ion-focus model suggests that the observed cluster configurations are indeed equilibrium configurations in the presence of an ion focus.

VI. SUMMARY

We have studied how the dynamic properties of finite 3D dust clusters can be described under the influence of an ion focus. The normal mode analysis has been extended to account for the ion-focus using the point-charge model for the horizontal interaction of the dust particles.

In a three-particle analytical model and corresponding experiments the role of the ion focus has been described quantitatively. There we have combined analytical descriptions and dynamical analysis both from NMA and INM. Three distinct regimes have been identified, namely, absolutely unstable configurations preferably at low ion-focus strengths, a regime of fully stable configurations at very high focus strengths, and the Schweigert oscillatory regime at medium strengths. In the experiments, we find configurations close to stability, and also fully in the Schweigert regime. Absolutely unstable regimes are naturally not realized in these long-run experiments.

We then have used the normal mode analysis (NMA and INM) for larger systems and have shown that these clusters are equally well described by a mode analysis including the ion focus. For the prolate cluster, the dynamical derivation of the ion-focus strength has been found to agree with previous investigations using structural information. Since NMA and INM are sensitive to different dynamic aspects of the particle motion equilibrium configurations in this nonequilibrium ion-focus environment can be deduced.

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[59] In the calculation of the mode frequencies, here we used a finite screening length of $\lambda_D \approx 400 \mu m$ to account for a more adequate consideration of the plasma.
[60] The value of $\xi$ differs here from value in the previous section since we have taken shielding into account here.
Correlation buildup during recrystallization in three-dimensional dusty plasma clusters

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Correlation buildup during recrystallization in three-dimensional dusty plasma clusters

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The recrystallization process of finite three-dimensional dust clouds after laser heating is studied experimentally. The time-dependent Coulomb coupling parameter is presented, showing that the recrystallization starts with an exponential cooling phase where cooling is slower than damping by the neutral gas friction. At later times, the coupling parameter oscillates into equilibrium. It is found that a large fraction of cluster states after recrystallization experiments is in metastable states. The temporal evolution of the correlation buildup shows that correlation occurs on even slower time scale than cooling. © 2014 AIP Publishing LLC [http://dx.doi.org/10.1063/1.4875750]

One access to study crystallization processes on the kinetic level are dusty plasmas, see Refs. 1 and 2 and references therein. These are discharge plasmas that contain additionally embedded, negatively charged dust grains of micron size. Due to their convenient charge to mass ratio the particles can be traced individually by means of video cameras. Recrystallization processes in dusty plasmas were investigated experimentally by quenching and relaxation of metastable states. The temporal evolution of the correlation buildup shows that correlation occurs on even slower time scale than cooling.

In this letter, we study the recrystallization process of a 3D dust cloud consisting of a small number of dust particles in a harmonic confinement. Under typical conditions, the particles are strongly coupled and arrange into spherical, nested shells. Due to the pairwise, shielded interaction between the dust grains these system are often termed Yukawa clouds. The energy of such a dust cloud is given as

\[ E = \frac{1}{2} m_0 e^2 \sum_{i,j=1}^{N} \frac{Q_i Q_j}{4 \pi \epsilon_0 r_{ij}} \exp\left(-r_{ij}/\lambda_D\right) \]  

with \( i \) and \( j \) denoting the particle number, \( r_{ij} \) is the distance to the trap center and \( r_{ij} = |r_i - r_j| \) is the distance between particles \( i \) and \( j \). Dust mass and charge are denoted by \( m \) and \( Q \) and \( \epsilon_0 \) is the frequency of the confinement potential. The first term describes the potential energy \( E_{\text{pot}} \) in the confining potential, the second term the screened Coulomb interaction energy \( E_{\text{cycl}} \). The electron Debye length can be expressed by the relation \( \lambda_D = b_{\text{WS}}/\kappa \) using the Wigner-Seitz distance \( b_{\text{WS}} \) as a measure for the particle density and the screening strength \( \kappa \), which characterizes the effect of shielding by the ambient plasma. Together with the Coulomb coupling parameter

\[ \Gamma(t) = \frac{Q^2}{4 \pi \epsilon_0 b_{\text{WS}} k_B T(t)} \]  

which describes the ratio of interaction energy to thermal energy \( k_B T(t) \) in the ensemble, the phase behavior of Yukawa-type systems is completely described.

The goal of this letter is to compare both, the correlation buildup and the evolution of coupling of a finite 3D system during recrystallization. We perform 3D dynamic measurements of individual particle motion that allows us to study the kinetics of the recrystallization process in detail. From that, we will compute the time-dependent Coulomb coupling parameter during the transition from a laser heated, fluid state to the solid, ordered state of the dust cluster. The initial phase of recrystallization will be used to calculate the cooling rate of the dust system. The emergence of correlations in our dust clusters will be estimated by means of the temporal evolution of the pair correlation function.

Strongly coupled finite 3D systems of charged particles are formed in a discharge plasma, see e.g., Refs. 10, 11, and 15. Here, a capacitively coupled rf discharge was operated in argon at 13.56 MHz at about 4 W with a neutral gas pressure of 8 Pa. A 3D harmonic trap for the dust particles can be created inside a cubic glass box of 2.5 cm wall length that is placed onto the electrode. The cloud is confined sideways due to the electric field from the dielectric glass walls. To balance gravity, the heated lower electrode (\( T = 75 \, ^\circ C \)) provides a thermophoretic force that, assisted by the sheath electric field force, levitates the particle cloud. In that manner, up to hundred particles can be trapped in a 3D harmonic confinement. Melamine formaldehyde microspheres of 4.04 \( \mu \)m size (\( m = 5.23 \times 10^{-14} \) kg) were used for the presented experiments and clusters with different particle numbers were investigated. Here, we study clusters with \( N = 19 \) and \( N = 36 \) particles in detail. The dust charge can be estimated by means of a normal mode analysis (NMA) as \( Q \approx 2900 e \) for the \( N = 36 \) cluster confined at 3.8 W rf power and \( Q \approx 3400 e \) for the \( N = 19 \) cluster trapped at 4.1 W rf power (\( e \) is the elementary charge). The error in charge measurement is about 20% (Ref. 18) and the charge difference can be explained by the slightly different rf power applied to confine both clusters.

A common way to observe the particles is by means of high speed cameras. We use two Nd:YAG lasers (600 mW...
per laser) for illumination of the dust clouds and three orthogonal C-MOS cameras to image the particles. To follow the fast kinetics during the recrystallization experiments, the frame rate of the cameras was set to 0.5–1.0 kHz. With the stereoscopic setup, the full 3D phase space information of the system is retrieved, see e.g. the trajectories in Fig. 1.

Driving this strongly coupled system into a fluid state requires heating of the particles. 19–21 Feeding the clusters with kinetic energy is achieved by directly “kicking” the particles via random motion of two opposing additional diode laser beams (maximum output power 1 W).17,22 Here, the heating lasers and cameras are synchronized to get a fixed starting point for the heating and recrystallization events in our experiment. In that manner, all clusters were heated for approximately 1 s. The particle motion was tracked during heating and subsequent recrystallization for a total of about 4 s for the \( N = 36 \) cluster or 2 s for the \( N = 19 \) cluster. To increase statistics, the procedure was repeated ten times for the \( N = 36 \) cluster and eight times for the \( N = 19 \) cluster. The trajectories of the \( N = 36 \) cluster during laser heating (over a time span \( \Delta t = 0.9 \) s, heating power \( P_L = 400 \) mW) are shown in Fig. 1(a). The particles along the laser beam are displaced from their equilibrium positions.5 The cluster is visibly molten. The mean particle velocities are of the order of 2 to 8 mm/s. In contrast, when the heating is switched off, see Fig. 1(b), the particles lose their kinetic energy very quickly and sediment into an equilibrium position where they form a solid dust cluster.

To assess the recrystallization dynamics, we first determine the time-dependent Coulomb coupling parameter, see Eq. (2). The dust charge is considered unaffected by our laser experiments2 and the Wigner-Seitz distance is calculated as \( b_{WS} = 250 \) μm for \( N = 36 \). Apparently, the coupling parameter is determined by the kinetic energy \( k_B T(t) \) of the particles. An instantaneous temperature is derived from the kinetic energy using the relation \( (3/2) k_B T(t) = (1/2) m v(t)^2 \), with \( v(t) \) being the absolute value of their 3D velocity. To improve statistics the velocities \( v(t) \) are averaged over ten consecutive frames as well as over the dust ensemble. An analysis of the velocities in different spatial directions reveals that the kinetic energy in laser heating direction is larger by about 60% than perpendicular to it.

The temporal evolution of the Coulomb coupling parameter for both clusters is depicted in Fig. 2(a). The recrystallization process is timed to start at \( t = 0 \) s, i.e., the heating lasers were “on” between \( t = -1 \) s and \( t = 0 \) s. After switching on the lasers, the coupling parameter drops to a low, fluctuating level of \( \Gamma_{fluid} \approx 40 \pm 6 \) for \( N = 36 \) (in the range of \(-0.75 \) s < \( t < 0 \) s). At these values of \( \Gamma \), finite dust clusters are in the fluid state.22

When the lasers are switched off, \( \Gamma \) quickly rises in the initial phase of recrystallization. We found that the slope of \( \Gamma(t) \) in this phase was almost equal for all individual recrystallization experiments. Thus, the initial phase of recrystallization must obey a generic mechanism. An inflection point is reached at approximately \( t = 0.3 \) s, afterwards \( \Gamma \) rises further monotonically. For \( t > 1 \) s, \( \Gamma \) seems to oscillate into equilibrium, where \( \Gamma_{cryst} \approx 440 \pm 37 \), see inset in Fig. 2. A profound oscillatory behavior was found for some, but not all of our recrystallization experiments in this later stage of recrystallization. This can be due to the fact that the observed system is rather small and individual particles might have an influence onto the cluster dynamics.24,25 At the high camera frame rates, the main error in the velocity estimation arises...
from pixel noise, which might affect the measurement of the small velocities only in the crystalline state.

To evaluate the initial stage of recrystallization, we fitted an exponential to \( \Gamma(t) \) in the range from \( t = 0 \) s to 0.3 s, where we observed the exponential increase of \( \Gamma(t) \). The time constants \( \tau_x \) are evaluated individually for each experimental run from fits to our data sets. The mean and rms values are found in Table I. The cooling rate is almost equal for both clusters and is systematically lower than the Epstein friction coefficient \( \nu = 21 s^{-1} \) for our recrystallization experiments. Simulations of the crystallization process reveal that the cooling rate depends on the normalized damping rate \( \nu/\omega_0 \). The trap frequency \( \omega_0 \) in our experiments is derived from the NMA as \( \omega_0 \approx 18.5 s^{-1} \) (\( N = 36 \)) and \( \omega_0 \approx 24.1 s^{-1} \) (\( N = 19 \)), hence \( \nu/\omega_0 \approx 0.9 - 1.1 \) in our experiments. In this moderate damping regime, simulations for 3D crystals predict a cooling rate \( \tau_x \leq \tau \), in agreement with our observations.

Figures 2(b) and 2(c) show the temporal evolution of both the potential energy in the trap \( \Delta E_{\text{pot}} \) and the interaction energy \( \Delta E_{\text{int}} \) relative to the equilibrium value, which was determined from the behavior at \( t > 1 \) s. The potential energy is found higher when the system is laser heated \( (t < 0 s) \) and decreases during recrystallization \( t > 0 s \). We find large fluctuations for the estimated energies, especially for \( \Delta E_{\text{int}} \). The interaction energy fluctuates strongly when the system is fluid, which is due to collision events between the particles. Both energy terms oscillate towards their equilibrium value, which is reached roughly at \( t > 1 \) s. The first minimum (maximum) of the oscillation of \( \Delta E_{\text{int}} \) (\( \Delta E_{\text{pot}} \)) after the lasers are switched off is found at \( t = 0.3 s \). This is exactly when \( \Gamma \) changes from an exponential growth into a more moderate slope, see Fig. 2(a). Switching off the external laser heating at \( t = 0 s \) leads to an instantaneous contraction of the cluster and therefore to the excitation of a damped monopole oscillation, which is seen in \( \Delta E_{\text{pot}} \) and \( \Delta E_{\text{int}} \).

Since the observed dust clouds are finite, particles within the cluster can—statistically—follow different individual paths towards an equilibrium state after the system has cooled down. This equilibrium state must not necessary be the ground state of the system. The ground state for a \( N = 36 \) dust cluster is found as (6,30) with \((N_1, N_2)\) being the number of inner and outer shell particles (for \( N = 19 \) the ground state is (1,8)). Observed metastable states were \((5,31)\) and \((7,29)\) for \( N = 36 \) and \((2,17)\) for the \( N = 19 \) cluster.

We observed settling into ground states only in 70% (50%) of the recrystallization experiments for the \( N = 36 \) (\( N = 19 \)) cluster. The probability of the observed metastable states corresponds very well to a screening strength of \( \kappa = 1 \). The energy that is needed to achieve metastable states is usually of the order of about 50 meV per particle. As can be seen from Figs. 2(b) and 2(c), laser heating can provide sufficient energy to reach a metastable state. Due to friction the dust clusters remain in these metastable state instead of sedimenting into the ground state again.

It is our major goal to compare the temporal evolution of the coupling parameter with the emergence of correlations during the recrystallization process. To measure these, the time-dependent pair correlation function was computed using \( g(r,t) = \langle \delta(r - r(t)) \rangle \). Again, we average over ten consecutive frames and normalize \( g(r,t) \) to unity, \( \int g(r)dr = 1 \). The temporal evolution of the pair correlation function for the \( N = 36 \) cluster is shown in Fig. 3(a).

### Table I: Cooling rate \( \tau_x \), normalized cooling rate \( \tau_x/\nu \) and correlation rates for the correlation buildup of ground \( \langle \tau_x \rangle \) and metastable state clusters \( \langle \tau_x \rangle \) for \( N = 36 \) and \( N = 19 \).

<table>
<thead>
<tr>
<th>( N )</th>
<th>( \tau_x ) (s)</th>
<th>( \tau_x/\nu )</th>
<th>( \tau_x/\nu )</th>
<th>( \tau_x/\nu )</th>
</tr>
</thead>
<tbody>
<tr>
<td>36</td>
<td>5.16 ± 1.28</td>
<td>0.25 ± 0.06</td>
<td>4.2 ± 2.78</td>
<td>3.61 ± 2.64</td>
</tr>
<tr>
<td>19</td>
<td>5.19 ± 2.30</td>
<td>0.25 ± 0.11</td>
<td>2.53 ± 0.54</td>
<td>3.63 ± 0.54</td>
</tr>
</tbody>
</table>

![FIG. 3.](image-url)
two regions corresponds to the nearest neighbor and next-nearest neighbor distance, respectively.

To find a measure for the temporal behavior of the pair correlation function, we approximated the nearest neighbor peak, i.e., the maximum of $g_1(r < 0.7 \text{ mm})$ by inverted parabolas. The maximum of these parabolas then correspond to the height of the peak of $g_1$, the curvature of the fitted parabola is a measure of the peak width and thus of the localization of the particles. The time-dependent peak height is depicted in Fig. 3(b) for both clusters (Due to the normalization of $g_1$, the peak height seems to be overall larger for the $N = 19$ cluster). The inset in Fig. 3(b) shows the curvature of $g_1$ for the $N = 36$ cluster.

During the heating period, the peak height of $g_1$ is low and the curvature is large. Thus, in the time period $t < 0 \text{s}$, the particles are less correlated and less localized, as one would expect for fluid systems. When the system cools down, the peak heights increase and reaches equilibrium values approximately 1 s after the laser heating was switched off. The same trend was found for the region of enhanced correlation at distances of $r = 1 \text{ mm}$ for both clusters. To compare the correlation buildup with the coupling parameter at the initial phase of recrystallization, the curves in Fig. 3(b) are also fitted to exponentials. To address the role of individual cooling paths, we distinguished between clusters which sediment into ground and into metastable states after recrystallization. The time scales are then defined as $\tau_{g,ss}$ for ground and $\tau_{m,ss}$ for metastable cluster states, respectively. Interestingly for the correlations, an exponential behavior was found for times up to $t = 1 \text{s}$. Here, the time dependent coupling parameter $\Gamma(t)$ already reaches its equilibrium value. The obtained values for the correlation rates $\tau_{g,ss}$ and $\tau_{m,ss}$ are given in Table I. Within the uncertainties, for both ground and metastable states, correlation occurs with the same rate. As found for the coupling parameter, the correlation buildup of individual measurements follows the general slope of the averaged results shown in Fig. 3(b). For both investigated clusters, the correlation buildup seem to be slower than the cooling rates estimated from $\Gamma(t)$. Hence, the clusters are faster cooled down than correlated. The initial emergence of the shells occurs fast, but the individual ordering of particles within the shells, which mainly affect the correlations and leave the coupling parameter nearly unaffected, takes place on a slower time scale. Minor adjustments on the shells do not significantly affect $\Gamma$, but the correlation function $g$.

The recrystallization dynamics for finite 3D dust clouds were studied kinetically. Two dust clouds were heated by means of lasers into fluid states. A cooling rate for the initial phase of recrystallization was estimated from the temporal behavior of the Coulomb coupling parameter and compared to recent experiments and simulations. After laser heating, dust clusters sediment either in ground or in metastable states. The correlation buildup was studied during the recrystallization experiments. Measurements indicate that the correlation rate obtained from the pair correlation function is lower than the cooling rate computed from the coupling parameter.

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A5

Spatio-temporal evolution of the dust particle size distribution in dusty argon rf plasmas

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Abstract
An imaging Mie scattering technique has been developed to measure the spatially resolved size distribution of dust particles in extended dust clouds. For large dust clouds of micrometre-sized plastic particles confined in a radio frequency (rf) discharge, a segmentation of the dust cloud into populations of different sizes is observed, even though the size differences are very small. The dust size dispersion inside a population is much smaller than the difference between the populations. Furthermore, the dust size is found to be constantly decreasing over time while the particles are confined in an inert argon plasma. The processes responsible for the shrinking of the dust in the plasma have been addressed by mass spectrometry, ex situ microscopy of the dust size, dust resonance measurements, in situ determination of the dust surface temperature and Fourier transform infrared absorption (FT-IR). It is concluded that both a reduction of dust size and its mass density due to outgassing of water and other volatile constituents as well as chemical etching by oxygen impurities are responsible for the observations.

Keywords: dusty plasma, Mie scattering, plasma diagnostics

1. Introduction
In all dusty plasmas, the dust particle size is a crucial parameter, since all forces acting on a dust particle depend on its size (here denoted by the radius $a$). The gravitational force is proportional to $a^3$ and therefore only significant for particles with a size of at least a few 100 nm. The electric field force is proportional to the dust charge, which is generally assumed to scale linearly with the radius $a$. Drag forces like the neutral drag, ion drag, radiation pressure or the thermophoretic force are proportional to $a^2$ [1].

Low temperature plasmas containing dust particles are widely studied for fundamental research as well as technological applications [1, 2]. In many technological processing plasmas, dust particles grow from sputtered material or due to chemically reactive plasma constituents [3]. The formation of dust in these plasmas can severely influence the plasma conditions and therefore disturbs the plasma processing [4, 5]. In other situations, dust growth can be useful and is employed e.g. to improve the efficiency of polymorphous silicon solar cells [6].

In early experiments on the dust formation in plasmas, the particles were detected and studied in situ by laser light scattering, but a reliable conclusion about the particle size was only possible by ex situ scanning electron microscopy (SEM) [4] or transmission electron microscopy (TEM) [7] measurements. Soon afterwards, Mie scattering ellipsometry was used to measure the size of injected particles [8] as well as growing particles in situ [9, 10]. Recently, Greiner et al introduced an imaging Mie ellipsometry technique, which is able to monitor the spatio-temporally resolved evolution of the particle growth in argon–acetylene plasmas [11].

Apart from the applications in industrial processing plasmas, dusty plasmas are also studied with respect to fundamental science. In this field of research, dust particles (usually melamine formaldehyde (MF) or other plastic particles of a well-defined size in the µm-range) are injected into chemically inert plasmas, where they acquire a highly
negative electric charge of $10^3$ e to $10^4$ e. With an appropriate confinement, strongly coupled Coulomb systems can be formed, ranging from 2D layers [12, 13] over finite 3D clusters [14] to spatially extended 3D clouds [15, 16]. There, one exploits the advantage that the dust particles can be observed individually and, that due to their slow timescales, can be tracked in 3D throughout time, providing the full phase space information.

The MF particles generally used in fundamental studies offer a very narrow size dispersion and a highly spherical shape. Only few works exist where the size of MF particles has been investigated, since it is considered to be well-defined by the manufacturer. However, Liu et al found the MF particles to be slightly smaller than specified by the manufacturer using TEM [17]. Swinkels et al employed Mie ellipsometry to monitor the etching of MF particle clouds in oxygen plasmas [18] and Stoffels et al investigated the etching of a single MF particle in an oxygen plasma by angular resolved Mie scattering [19]. Since most experiments on dusty plasmas use argon or other noble gases, chemical etching of the dust is not considered relevant. However, the outgassing of water, leading to a reduction of the MF mass density, has been observed by Carstensen et al by a precise measurement of the dust resonance frequency over a long time [20]. There, the authors conjectured that the outgassing might be caused by an increased surface temperature of the dust particles in the plasma (which was observed by other groups [21, 22]). The assumption of a temperature-dependent outgassing is endorsed by thermogravimetry [20] and quadrupole mass filters measurements [23]. The dust size was, however, assumed to be constant in all of these experiments.

In this work, we developed a spatially resolving Mie imaging technique, which is based on the angle-resolved measurement of the Mie scattering intensities. This method allows a precise determination of the dust size distribution in spatially extended dust clouds. In experiments featuring large, thermophoretically levitated and seemingly homogeneous dust clouds consisting of monodisperse MF particles, a somewhat segmented structure of the dust cloud is visible (see e.g. our previous work [24] and, presumably, experiments from other groups with similar conditions [25]). Since this segmentation might be caused by differently sized particles, this issue will be addressed by Mie imaging in this work.

Our imaging Mie diagnostic is furthermore used to monitor the time evolution of the dust size distribution. The variation of the dust size with time is confirmed by ex situ microscopy of plasma-exposed dust. Further indirect evidence is found by measuring the dust plasma frequency over time and the response of the dust to different (thermophoretic) force fields.

In order to gain insight into the physical processes leading to the changes in dust size and mass, mass spectrometry has been used to identify possible signatures of dust outgassing or etching. Furthermore, the surface temperature of the dust particles has been measured using fluorescent dust particles and the chemical structure of MF dust was evaluated using Fourier transform infrared (FT-IR) absorption.

2. Experimental setup

Our main experiments have been performed in a capacitively coupled radio frequency (ccrf) plasma that has a 360° optical access. Some supporting experiments have been carried out in a typical rf discharge chamber. In both cases, argon plasmas are ignited at 13.56 MHz with a gas pressure on the order of 10 Pa (measured by a capacitance vacuum gauge) and low plasma powers in the range of 1–10 W. The dust particles are stored in dispensers under vacuum conditions (the base pressure of the plasma chamber is approximately 0.1 Pa) prior to the experiments.

2.1. Main experimental setup

A symmetric parallel plate setup with temperature-controlled electrodes is used to confine spatially extended dust clouds (details in [24]). The circular electrodes with a diameter of 8 cm are separated by a 3 cm discharge gap and are operated in push–pull mode. A volume-filling dust cloud is produced by using the thermophoretic force to compensate for gravity. Therefore, a temperature gradient is created in the neutral gas by heating the lower and cooling the upper electrode, resulting in an upwards directed thermophoretic force.

In order to perform angle-resolved measurements of the Mie scattering intensity, the plasma chamber is completely transparent by using a glass cylinder as the outer wall. A CCD camera is placed on a rotating stage (details in [26]), allowing to observe the dust cloud from any angle with respect to a stationary laser. The Mie scattering experiments described in section 3, the thermophoretic force field measurements (section 4.2) and the mass spectrometry (section 4.5) have been carried out in this device. A sketch of the plasma chamber and the rotating stage is shown in figure 1.

2.2. Supporting experiments

This setup consists of an asymmetric discharge with a powered lower electrode (diameter of 170 mm), where the chamber walls act as the grounded electrode [14, 27]. This plasma chamber is used for the measurement of the resonance frequency of small dust clusters over time using the phase resolved resonance method of Carstensen et al [20], which is described in section 4.3. Furthermore, the dust surface temperature measurements have been carried out in this setup. Also, particles have been collected from this device for ex situ microscopy (section 4.4).

3. Angular resolved Mie scattering

For dust particles with a size of a few micrometres, the Mie scattering signal has a characteristic angular dependence [28]. With our diagnostic we aim to combine imaging techniques with the angle-resolved measurement of Mie scattering signals. The angular Mie scattering signals from a single particle have been used by Stoffels et al to measure the particle size evolution of this single MF particle being etched in an oxygen plasma [19]. In our setup with a completely transparent plasma chamber, we are able to observe the dust cloud from a wide
angular space, as sketched in figure 1(b). To avoid too heavy optical distortion for the imaging, however, our measurements are limited to an angular range of 90°, stretching roughly from 45° to 135° with respect to the incident laser beam. For our Mie scattering experiments, we use highly spherical MF particles with a nominal radius of 1.775 µm and a refractive index of n = 1.68 (real part), as given by the manufacturer (Microparticles GmbH).

A thin slice of the dust cloud is illuminated by a vertical laser sheet (about 1 mm thick). The laser light has a wavelength of 532 nm and is linearly polarized in the horizontal plane. Hence, only the parallel component of the Mie scattering signal is measured. The scattered laser light is observed by a CCD camera, which is equipped with a telecentric lens and an optical filter to suppress the plasma light emission.

Two representative images of a typical dust cloud, taken from two slightly different angles, are shown in figure 2 in inverted greyscale. Besides the presence of the central void, the dust cloud appears to be segmented into different parts, which are separated by sharp intensity edges. Some segments of the dust cloud are particularly bright at a scattering angle of 84.7° (figure 2(a)), while others show a higher intensity at 90.0° (figure 2(b)). Observing the dust cloud from many different angles, we find that each segment of the dust cloud features a distinct dependence of the scattered intensity from the observation angle. To illustrate this fact, four representative parts of the dust cloud are further examined in figure 3. Each of the four arbitrarily chosen points (whose locations are indicated by small circles in figure 2(b)) resembles an area averaged over 5 × 5 pixels in the original images in order to enhance the data quality. In figure 3(a), the scattering intensity for each point is shown as a function of the observation angle. It is clearly visible that each graph has a distinct behaviour.

With all required parameters for Mie scattering at hand (laser wavelength and polarization, dust refractive index), we can deduce the particle size from the scattering signals in figure 3(a). Since the dust particle radius is now the only free parameter for the Mie scattering problem, it can be determined by least-squares fitting the Mie theory signals to the experimental results.

We found that fitting the intensity oscillations together with the angular positions of the intensity peaks is much more reliable than using the actual scattering amplitudes, which are influenced by experimental uncertainties (e.g. small fluctuations in the dust cloud, laser beam quality, CCD camera sensitivity). Hence, a moving average filter is applied to the angle-resolved scattering signals for each position, resulting in intensity oscillations with a zero mean value. Now, the radius is determined by fitting the Mie theory scattering signals (also with moving average subtracted).

The results are shown in figures 3(b)–(e) for each of the four selected positions separately. The Mie theory fits agree very well with the course of the individual experimental graphs. Surprisingly, the fitted radii differ by about 6% with respect to each other, which is about three times larger than the standard deviation of the particle size given by the manufacturer. Furthermore, the particles are specified to have a
mean radius of 1.775 µm and therefore seem to be considerably smaller in our measurement.

Monitoring the dust size distribution over the course of a few hours, a drastic further reduction of the particle size is observed. In figure 4, the size distribution in the dust cloud from figure 2 is shown for different times after the dust injection at \( t_0 \) (with figure 4(a) corresponding to the situation in figures 2 and 3). Besides the obvious reduction of the particle size, the overall shape of the dust cloud is also changing. This effect can be explained by the constant size (and mass) loss of the dust particles. As a result of such a process, the force balance between gravity (scaling with \( a^2 \)) and the thermophoretic force (scaling with \( a^3 \)) is disturbed. For shrinking dust particles, the thermophoretic force wins over gravity, leading to the accumulation of the dust in the upper part of the cloud.

Comparing the dust size in a specific area of the cloud over time, one can try to calculate a size loss rate. This value will not describe the actual loss rate of a single particle, but rather the size evolution of the dust particles in that area (which will not necessarily be the same particles over time, e.g. due to transport). The dust size at four arbitrarily chosen points (indicated in figure 4(c)) as a function of time is shown in figure 4(d). The dust size decreases with a roughly constant rate at all four locations. From 10 measurements over the course of about 2 h (after more than 2 h, all the dust is compressed in a flat layer at the upper sheath), we find an average size loss rate of 0.04 nm s\(^{-1}\). This rate is almost as large as the etching rate for MF particles in oxygen plasmas, which was found to be in the range of 0.06–0.13 nm s\(^{-1}\) for plasma powers of a few watts [18, 19].
oven-baked particles were exposed to a temperature of 120 °C (typically 10–20) in the respective situation. The reference plasma-exposed dust under a microscope are shown for (a) fresh dust and (c) plasma-exposed dust.

4. Additional diagnostics

The Mie imaging results in the previous section strongly indicate that the dust particle size heavily decreases with time. Possible reasons may include chemical etching, sputtering and outgassing. In order to evaluate the importance of each of these processes, additional experimental techniques have been employed. As a general approach, we assume that the dust radius \(a\) and the mass density \(\rho\) may both change over time.

For some experiments, the dust particles were baked in an oven at 120–130 °C prior to the experiments. This procedure has been proposed due to evidence of the outgassing of water from the MF particles in the plasma [20, 23]. With the baking process, most of the water bound in the MF is supposed to evaporate.

4.1. Ex situ microscopy

In order to verify the shrinking of dust particles in the plasma with a very reliable technique, dust particles have been examined with an optical video microscope before and after plasma exposure. Furthermore, the influence of oven-baking on the particle size is investigated. A typical image using a magnification of 500 is shown in figures 5(b) and (c), where fresh and plasma-exposed particles are shown. The dust size is evaluated by fitting circles to the edge of the particles.

The resulting dust sizes are shown in figure 5(a), where each data point represents an average of all dust particles (typically 10–20) in the respective situation. The reference at \(t = 0\) corresponds to unused, fresh dust particles. The oven-baked particles were exposed to a temperature of 120 °C and are marked in red. These particles already show a slightly decreased size after baking, which accumulates to a shrinking of almost 6% after 4 h baking (with the size ratio of exposed dust (radius \(a\)) and fresh dust (radius \(a_0\)) determined as \(a/a_0 = 0.945\). This leads to the conjecture that the dust particles contract due to the outgassing of volatile components such as water and formaldehyde.

For the plasma-exposed dust, a more drastic change of the dust size is observed, with ratios of \(a/a_0 = 0.82\) after 4 h and \(a/a_0 = 0.80\) after 20 h. In these experiments, MF particles with a specified radius of 5.1 µm have been confined in the sheath of the asymmetric discharge (see section 2.2) at an argon pressure of 20 Pa and a plasma power of 4 W. Surprisingly, the difference between 4 and 20 h of plasma operation is quite small, indicating a saturation of the process that leads to the dust size reduction. However, the ratio \(a/a_0\) agrees well with the values obtained from the Mie imaging technique in the symmetric discharge.

4.2. Thermophoretic force fields

The dust size maps in figure 4 do not only reveal a decreasing dust size but also a shifting overall shape of the dust cloud. For a confined particle, the force balance results in a zero net force. For dust particles in the plasma volume (where the electric field is small), the vertical confinement is mainly influenced by the interplay between the gravitational force (pointing downwards) and the thermophoretic force (pointing upwards). Since both forces scale differently (\(F_T \sim a^3\rho\) and \(F_{th} \sim a^3\)), a reduction of particle size or mass density will lead to the thermophoretic force gaining the upper hand over gravity. Hence, the dust will move upwards until another force compensates the excess of thermophoresis over gravity. This other force may be due to the electrostatic pressure of the dust cloud or, finally, the sheath electric field close to the upper electrode.

Therefore, the dust cloud shifts upwards as a whole (figure 4), and after a few hours of plasma exposure, the entire dust cloud is compressed into a thin layer at the upper sheath due to the drastic mass or size loss of the particles. Now, if the plasma is turned off, the sheath electric field instantly vanishes and the dust particles will be accelerated upwards due to the thermophoretic force being stronger than gravity. Because of friction of the dust with the neutral gas, the dust reaches a terminal velocity almost instantly. The force balance for the dust particles at the terminal velocity can be written as

\[
F_T(a, \rho) + F_{th}(a) + F_i(a, v) = 0, \quad (1)
\]

where \(F_T\) is the neutral gas friction, which depends on \(F_{th} \sim a^3\) (with the dust velocity \(v\)). For this experiment, the temperature gradient in the neutral gas is set to a value where freshly injected dust particles with the radius \(a_0\) and mass density \(\rho_0\) are almost ideally well levitated after the plasma is switched off, due to \(F_t(a_0, \rho_0) = -F_{th}(a_0)\). Combining the force balances for the plasma-exposed dust and the fresh dust, we find

\[
\frac{a_0 \rho}{a \rho_0} = 1 + \frac{C_1}{C_2} v, \quad (2)
\]

where \(C_1\) and \(C_2\) correspond to the coefficients for the neutral drag force and thermophoretic [29] force, respectively:

\[
C_1 = \frac{32}{3} \frac{p}{v_i} \frac{\delta}{\nu} \quad \text{and} \quad C_2 = -3.33 \frac{k_B}{\sigma} \frac{\nabla T}{\sigma}, \quad (3)
\]

Here, \(p\), \(v_i\) and \(\nabla T\) are the pressure, thermal velocity and temperature gradient of the neutral gas; \(1 \leq \delta \leq 1.44\) is a geometric scattering coefficient and \(\sigma\) is the gas kinetic collision cross section.
In experiments similar to the situations in section 3, dust particles which have been exposed to the plasma for 2h, attain terminal velocities of about 25 mm s\(^{-1}\) after the plasma is switched off. Estimating \(C_1/C_2 \approx 10 \text{ s mm}^{-1}\) for our conditions, this velocity corresponds to a ratio of \((a\rho)/(a_0\rho_0) \approx 0.75\). Even if a mass density loss due to outgassing is accounted for \((\rho/\rho_0 \approx 0.9)\), this technique reveals a heavily decreasing dust size on the order of \(a/a_0 \approx 0.8\).

In a different experiment involving 5 h of plasma exposure, an even more drastic effect was found. There, the particle attained velocities of about 50 mm s\(^{-1}\), resulting in a ratio of \((a\rho)/(a_0\rho_0) \approx 0.5\).

### 4.3. Phase resolved resonance

To illustrate this further, we investigated the force balance for a small ensemble of dust particles located in the plasma sheath of the supporting discharge. There, the particles are vertically confined by the force balance between gravity and electric field force due to the sheath electric fields (thermophoresis is not used in this situation). If the electrode voltage is sinusoidally modulated with a low frequency, the dust particles behave as damped harmonic oscillators with an eigenfrequency \(\omega_0\), which is proportional to the dust’s charge-to-mass ratio \(\sqrt{q/\rho}\) [30].

Recently, Carstensen et al. established a very precise technique for the determination of relative changes in the dust particles’ plasma frequency, called the phase resolved resonance method [20]. There, the authors found that dust plasma frequency of an MF particle confined in the sheath of an argon rf plasma increased by 5–6% over the course of a few hours. Since the dust plasma frequency scales with \(\sqrt{q/\rho}\), they concluded that during the measurement time the dust mass was reduced by 10–12%, assuming a constant size and therefore constant charge \(q_0\). The most probable cause for this behaviour was reasoned to be the outgassing of water from the MF material. This assumption is supported by the temporal evolution of the dust plasma frequency, featuring a steep increase at the beginning, later turning into a slow saturation, and ultimately resulting in a constant eigenfrequency after 10 h.

We adapted the phase resolved resonance method from [20] and found an even stronger effect in our experiments, which were performed in the asymmetric plasma chamber described in section 2.2. The experimentally obtained dust plasma frequencies are presented in figure 6 and reveal a linear increase with time. After 5h, a total increase of 37% is observed, corresponding to a relative change in the charge-to-mass ratio of

\[
\frac{\omega^2}{\omega_0^2} = \frac{q}{m} \frac{q_0}{m_0} = \frac{a}{a_0} \frac{\rho}{\rho_0} = 1.87.
\]

Here, the relations \(q \sim a\) and \(m \sim a^2\) have been used. This drastic result is probably not purely caused by a mass loss due to outgassing, as it was proposed in [20].

### 4.4. Evidence of outgassing or chemical processes

The process of water gassing out from the MF material strongly depends on the MF temperature [20, 23]. Often, the temperature of the dust particles confined in the plasma is assumed to be equal to the gas temperature, which is roughly at room temperature. At this temperature, no significant outgassing is expected. There are however a few works, where the dust material temperature in the plasma has been measured using fluorescent dye [21, 31] or phosphor particles [22].

Here, we employed the method described by Swinkels et al. [21], which uses MF dust with incorporated fluorescent dye (Rhodamine B), whose emission spectrum is temperature-dependent. First, two calibration measurements have been performed, where the fluorescent dust particles were suspended in water and glycerol, respectively. The jars containing the suspensions were placed on a heatable magnetic stirrer. The fluorescence has been excited by a green laser (532 nm) and was observed with a compact spectrometer. An optical bandpass filter suppressed the green laser light from the spectrometer.

The emission curves have been measured and the full widths at half maximum (FWHM) have been determined as a function of the temperature, see figure 7. The FWHM is a reliable quantity for the temperature measurement from the emission spectra, since with increasing temperature the spectra become flatter and broader, while the overall shape of the emission curve does not change much (one sample spectrum is shown in the inset of figure 7). From the calibration curves of FWHM versus temperature, a linear dependence is found for both solvents. The different slopes of the obtained calibration curves can probably be attributed to absorption by the solvents.

Now, the emission curve of fluorescent dust particles confined in the plasma sheath has been measured (using the asymmetric chamber described in section 2.2). The same laser is used for excitation and a background spectrum of the plasma emission (without laser and fluorescence) is recorded. From the emission of the particles in the plasma, again the
FWHM is determined and found as 40.5 ± 0.15 nm. Using the linear fits obtained from the calibration curves in figure 7, a dust temperature of \( T = 119.8 \pm 4.4 \) °C (calibration in water) and \( T = 144.5 \pm 9.7 \) °C (calibration in glycerol) is found. These values agree very well with results reported in [21, 31]. Reasons and possible mechanisms for the high dust temperature (compared to the neutral gas and the heated electrode, which are both below 40 °C) are discussed in [21]. In our situation, the energy influx due to the recombination of argon ions on the dust surface is thought to be the most important factor.

This elevated dust temperature will favor the outgassing of water and possibly formaldehyde (as stated by the manufacturer [32]). To verify the outgassing process, FT-IR absorption measurements have been performed with untampered dust as well as pre-baked dust, which has been in an oven at elevated temperatures for 2 h. The absorption spectra in figure 8(a) reveal only a minor change when the dust was heated to 75 °C, but a stronger effect for the dust baked at 120 °C. To emphasize the effect of baking, the difference of the transmission spectra is shown in figure 8(b). The dip between 3000 and 3500 cm\(^{-1}\) corresponds to the typical FT-IR absorption wavenumbers of water, while the changes at lower wavenumbers represent the chemical constituents of the MF material [33]. The decrease of the water absorption at higher temperature indicates that water has outgassed from the dust due to the baking. Furthermore, the chemical structure seems to be significantly altered by heating the dust to 120 °C.

### 4.5. Plasma mass spectrometry

The most probable causes for the decrease of dust size and mass in the plasma are chemical etching and the outgassing of volatile contents from the dust material. These processes naturally evoke a transport of water or organic chemical compounds from the dust material into the plasma. To analyse the composition of the gas in our plasma chamber, a quadrupole mass spectrometer for residual gas analysis has been used. Besides the argon feed gas, some residual molecules and atoms such as C, N, O, HO, H\(_2\)O, N\(_2\), CO, O\(_2\), CO\(_2\) and others are found in the (dusty) plasma.

For the evaluation of the influence of the dust presence on the gas composition, the symmetric chamber (see section 2.1) has been evacuated to the base pressure of about 0.1 Pa (using a rotary pump). After reaching base pressure, the argon flow is started and the plasma is switched on. Performing a time-resolved measurement of these characteristic masses, already a significant influence of the plasma operation on the gas contents is found. Igniting a plasma (at otherwise constant parameters) leads to a heavy increase of the H\(_2\) signal and, to a smaller extent, to an increase of C and CO/N\(_2\) (both \( m = 28 \)), while the O\(_2\) signal decreases (due to dissociation in the plasma), as is depicted in figure 9(a). Switching the plasma off or reducing the plasma power leads to an opposite behaviour of these mass signals (increase of O\(_2\), decrease of H\(_2\), C and CO/N\(_2\)). This is also the explanation for the behaviour of the mass signals prior to plasma ignition in figure 9(a): the plasma was switched off earlier, some minutes before \( t = 0 \).

Now, a large amount of MF dust particles is injected into the plasma with an electromagnetic dust dispenser at \( t = 30 \) min and \( t = 60 \) min. The injection results in an...
almost instantaneous increase of the signals for \( \text{H}_2, \text{C}_x, \text{H}_2\text{O} \) and \( \text{CO}/\text{N}_2 \), which is shown in figure 9(b). To emphasize the relative changes, the time derivative of the mass signals is plotted here. The narrow peaks of the time derivatives upon dust injection indicate a jump of the overall concentration of the respective mass numbers. This result further confirms that outgassing and/or chemical reactions take place.

5. Discussion

The Mie imaging technique presented in this work together with the additional diagnostics gives insight into the dust size distribution and its temporal evolution. In this last section, we will discuss these two main aspects.

5.1. Spatial dust size distribution

The de-mixing of differently sized particles in dusty plasmas is known to take place in situations involving binary mixtures with large size differences between the two species [34-36]. Now, our Mie imaging method provides an excellent resolution, which allows to discriminate size differences of a few nm for micrometre-sized particles. The resulting dust size maps in figure 4 reveal a de-mixing of slightly differently sized particles, which results in a segmentation of the dust cloud. It is interesting to note that the dust size maps indicate a rather discrete dust size distribution instead of a smooth distribution, which is naturally expected [17]. However, since our Mie diagnostic reveals the dust sizes only in a thin 2D slice of the entire cloud, other dust sizes might be found in other parts of the cloud.

A general limitation concerning the spatial resolution of the dust size maps is given by the fact that we do not observe single dust particles but rather an ensemble of a few particles corresponding to a pixel in the camera images. Therefore, our Mie scattering technique is limited by the requirement for homogeneous dust populations in the area of investigation (typically \( 5 \times 5 \) pixels, but the same applies for smaller averaging areas). Hence, the angle-resolved scattering signals from a heterogeneous part of the dust cloud, which contains particles of different size, cannot be interpreted with regard to Mie theory. Such parts of the dust cloud are shown in grey colour in figure 4.

This limitation can however be used to estimate the timescale required for the de-mixing, which was found to be on the order of a few minutes for our supposedly monodisperse particles featuring only small size differences (\( \Delta a/a \leq 10\% \)). This can be inferred from a dust cloud, that has been confined in the plasma for at least 30 min (similar to the ones in figure 4), and subsequent injection fresh dust particles. Directly after injection, the observed Mie signal cannot be interpreted by dust particles of a unique size, indicating mixed populations. In some situations, it took up to 30 min to observe clear, unambiguous scattering signals. Hence, de-mixing might take a long time compared to most other dust processes. It should also be kept in mind that de-mixing in the plane perpendicular to the laser illumination cannot be observed by our diagnostic.

<table>
<thead>
<tr>
<th>Method</th>
<th>Exposure</th>
<th>Measured ratio</th>
<th>Result</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mie scattering</td>
<td>1 h plasma</td>
<td>( a/a_0 )</td>
<td>0.90</td>
</tr>
<tr>
<td>Ex situ</td>
<td>2 h plasma</td>
<td>( a/a_0 )</td>
<td>0.81</td>
</tr>
<tr>
<td>Microscopy</td>
<td>4 h plasma</td>
<td>( a/a_0 )</td>
<td>0.82</td>
</tr>
<tr>
<td>1 h oven</td>
<td>20 h plasma</td>
<td>( a/a_0 )</td>
<td>0.80</td>
</tr>
<tr>
<td>4 h oven</td>
<td>1 h oven</td>
<td>( a/a_0 )</td>
<td>0.97</td>
</tr>
<tr>
<td>Phase resolved</td>
<td>2 h plasma</td>
<td>( (a^2)<em>{\rho}/(a</em>{\rho}^2)_{\text{pl}} )</td>
<td>0.75</td>
</tr>
<tr>
<td>Resonance</td>
<td>4 h plasma</td>
<td>( (a^2)<em>{\rho}/(a</em>{\rho}^2)_{\text{pl}} )</td>
<td>0.59</td>
</tr>
<tr>
<td>Thermophoretic</td>
<td>2 h plasma</td>
<td>( (ap)/(a_{\text{th}}p_{\text{th}}) )</td>
<td>0.75-0.80</td>
</tr>
<tr>
<td>Force field</td>
<td>5 h plasma</td>
<td>( (ap)/(a_{\text{th}}p_{\text{th}}) )</td>
<td>0.50-0.60</td>
</tr>
</tbody>
</table>

Finally, the validity of the absolute values for the dust size determined by Mie imaging depends on the refractive index of the dust. We have used \( n = 1.68 \), as specified by the manufacturer [32]. However, the absolute size determined from Mie scattering does not match the size specified by the manufacturer, but is somewhat smaller (assuming a different value for \( n \) naturally affects the determined particle size). Nonetheless, the relative size differences shown in figure 4, and therefore the de-mixing phenomenon, are independent of the choice of the refractive index.

5.2. Temporal changes of dust size and mass

Besides the Mie imaging technique, various additional experimental methods have been employed in this work to investigate the temporal changes of dust particle sizes. To draw a complete picture, the results of all experimental techniques are listed in table 1. For comparison, the kind (plasma or oven) and duration of the particle exposure is given in each situation.

The most unambiguous evidence of the particle size loss in the plasma is given by the Mie scattering and the ex situ microscopy, since these techniques directly measure the (relative) particle size. Comparing the results from these two techniques indicates that the size loss is mainly occurring in the first hours of plasma exposure, since the final ratio of \( a/a_0 = 0.8 \) found by microscopy after 20 h is observed by Mie imaging already after 2 h. The size reduction is therefore supposedly a saturating process. Unfortunately, this cannot be further investigated by the Mie imaging technique, since after more than 2 h, the entire dust population is compressed in a thin layer below the upper electrode (see section 4.2). In this situation, no clear signal can be derived from Mie scattering theory, probably due to the mixing of different dust sizes. Furthermore, after a few hours in the symmetric discharge, the increasingly smaller and lighter dust cannot confined any more, and ultimately no dust is left in the plasma.

It might also be possible that the different experimental conditions for Mie imaging and microscopy have influenced the size loss rate: for the microscopy, MF particles with a radius of \( a_0 = 5.1 \mu m \) were confined in the sheath of an asymmetric discharge, while the Mie scattering experiments were performed with smaller \( (a_0 = 1.775 \mu m) \), thermophoretically levitated MF particles confined in the plasma volume of a symmetric discharge. Finally, it could be...
conjectured that the refractive index of the MF particles is not constant during plasma exposure. A change of the refractive index due to chemical reactions might lead to inaccurately determined dust sizes using Mie imaging.

While the dust size changes determined by Mie imaging and microscopy are very reliable, the last two techniques in Table 1 provide more indirect, but still very profitable evidence. Assuming a reduction of the mass density of \( \rho/\rho_0 = 0.9 \) \cite{20}, the phase resolved resonance experiments result in \( a/a_0 \approx 0.9 \) after 2 h and \( a/a_0 \approx 0.8 \) after 4 h. The results of the force field experiments, again assuming \( \rho/\rho_0 = 0.9 \), also yield \( a/a_0 \approx 0.85 \) after 2 h, and a more drastic value of \( a/a_0 \approx 0.6 \) after 5 h of plasma exposure.

Comparing the different experimental approaches, the investigations in the symmetric plasma chamber with the volume-filling, thermophoretically levitated dust clouds tend to result in a higher size loss rate than the experiments on small dust ensembles in the asymmetric discharge. It could be conjectured that this might be a self-enhancing effect. More dust particles release more volatile gases by outgassing, which can dissociate into oxygen radicals. The oxygen radicals can then, in turn, chemically etch the dust and therefore further reduce its size and mass.

This assumption on the mechanism behind the changing dust size and mass is supported by the mass spectrometry results (see section 4.5), where an instant increase of characteristic mass signals is observed upon dust injection. The initial outgassing, which is known to be temperature-dependent \cite{20, 23, 32}, is to be expected from the fluorescence-based temperature measurements, which revealed a dust temperature of about 125 °C (see section 4.4). Furthermore, baking of unused MF particles was found to lead to the outgassing of water and other organic compounds. Plasma-related mechanisms such as sputtering by argon ions can probably be neglected in our experiments, since the dust potential is not larger than 10 V (with respect to the plasma potential), leading to a low energy of impinging argon ions. A basic calculation for the sputter rate in a comparable argon plasma is presented in \cite{20}, where the authors conclude that the argon sputter rate is orders of magnitude too low to account for the size and mass reduction in the data analysis.

6. Summary

In this work, we have presented a Mie imaging technique which provides the spatially resolved size distribution of dust particles confined in an inert argon plasma. The high resolution of the dust size determination is achieved by measuring the Mie scattering signal angle-resolved over a wide angular range. Our measurements of spatially extended dust systems reveal a segmented structure of the dust clouds, where each segment contains a homogeneous population of dust particles with a distinct size. Since seemingly monodisperse particles are injected into the plasma, a self-excited de-mixing phenomenon is observed even if the size differences between the particles are very small (\( \Delta a/a \leq 10\% \)).

Observing the particle size distribution over time reveals a constantly decreasing dust size. This effect has been confirmed by \textit{ex situ} microscopy of dust particles collected after plasma exposure. Further evidence of changes in the dust size and mass has been obtained from thermophoretic force field measurements and investigations of the dust particles’ resonance frequency.

The physical processes behind this phenomenon have been evaluated by different approaches: mass spectrometry of the plasma indicated that the injection and presence of dust particles in the plasma increases the concentration of impurities in the argon plasma (notably H₂, C, H₂O, CO/N₂). Furthermore, fluorescence-based temperature measurements of the dust particles’ surface temperature in the plasma revealed elevated values of more than 100 °C. Such a high temperature was found to result in the outgassing of water and possibly organic constituents of the dust material by FT-IR absorption spectroscopy.

We therefore conclude that a combined action of outgassing of volatile constituents from the dust material and chemical etching due to impurities (possibly oxygen radicals) reduces the dust size and mass during confinement in the plasma. This result is of high importance for many experiments with dusty plasmas, which often use particles made of melamine formaldehyde or other plastics. We recommend either to regularly replace the dust in experiments or to account for the size and mass reduction in the data analysis.

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Stereoscopic imaging of dusty plasmas

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Stereoscopic imaging of dusty plasmas

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The fundamentals of stereoscopy and their application to dusty plasmas are described. It is shown that stereoscopic methods allow us to measure the three-dimensional particle positions and trajectories with high spatial and temporal resolution. The underlying technical implications are presented and requirements and limitations are discussed. The stereoscopic method is demonstrated for dust particles in dust-density waves under microgravity conditions.

1. Introduction

Laboratory experiments on dusty plasmas usually make use of solid particles in the size range between 100 nm and 20 µm diameter immersed in a gaseous plasma environment. In the plasma, the particles typically attain high negative charges due to the collection of plasma charge carriers (electrons and ions). This makes the dust particles a unique plasma species that is susceptible to various forces that are otherwise unimportant in plasmas. Moreover, the dust system usually is strongly coupled and this acts on very different time scales than electrons and ions. Indeed, the dust size, the interparticle distance and the time scales associated with the particle motion are ideally suited to study the dust by video microscopy (Bouchoule 1999; Shukla & Mamun 2002; Melzer & Goree 2008; Bonitz, Horing & Ludwig 2010; Piel 2010; Ivlev et al. 2012; Bonitz et al. 2014). A wide variety of experiments have been performed on the structure and dynamics of these dust systems, see e.g. Shukla (2001), Fortov, Vaulina & Petrov (2005), Bonitz et al. (2008), Piel et al. (2008), Shukla & Eliasson (2009), Melzer et al. (2010), Merlino (2014).

It is easy to see that it is necessary to measure the (three-dimensional) particle positions $r_i$ of (each) particle $i$ to reveal the structure of a given particle arrangement. In the same way, one needs to know the velocities $\dot{r}_i = v_i = \Delta r_i / \Delta t$ and accelerations $\ddot{r}_i = a_i = \Delta v_i / \Delta t$ to determine the particle dynamics. Hence, the equation of motion

$$m\ddot{r}_i + m\beta \dot{r}_i = F_i(r, v)$$ (1.1)

becomes accessible to extract the relevant forces $F_i$ on particle $i$. Here, $m$ is the particle mass (which might or might not be known) and $\beta$ is the corresponding (Epstein) friction coefficient (Epstein 1924; Liu et al. 2003).

Here, video stereoscopy provides a very versatile and reliable technique to measure the individual three-dimensional (3-D) particle positions (and subsequently the particle

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velocities and accelerations in (1.1), where the time step $\Delta t$ is just given by the frame rate of the cameras). In general, this technique requires multiple cameras (at least two) that observe the same volume under different angles. From the appearance of the particles under the different camera viewing angles, the three-dimensional particle position can then be reconstructed. In this article we will focus on stereoscopy. Other techniques that are able to retrieve 3-D particle positions include inline holography (Kroll, Block & Piel 2008), colour-gradient methods (Annaratone et al. 2004), scanning video microscopy (Pieper, Goree & Quinn 1996; Arp et al. 2004; Samsonov et al. 2008), light field imaging with plenoptic cameras (Hartmann, Donko & Donko 2013) or tomographic-PIV (Williams 2011) which, however, will not be reviewed here.

2. Diagnostics

The principal idea of stereoscopy is that a common observation volume is imaged by multiple cameras from different viewing angles. As an example, our stereoscopic camera set-up used for parabolic flight experiments (Buttenschön, Himpel & Melzer 2011; Himpel et al. 2012, 2014) is shown in figure 1. For the parabolic flight experiments, three CCD cameras with $640 \times 480$ pixels at a frame rate of approximately 200 frames per second (f.p.s.) are used. The cameras are fixed with respect to each other, but the entire system can be moved on two axes to freely position the observation volume within the discharge chamber. In our laboratory experiments, a stereoscopic system with three orthogonal cameras at megapixel resolution ($1280 \times 1024$) at 500 f.p.s. is installed (Käding et al. 2008). In both cases,
the cameras have to be synchronized to ensure that the frames of all cameras are recorded at the same instant.

To retrieve the 3-D particle positions, first of all the viewing geometry of the individual cameras has to be determined. The best results for our dusty plasma experiments with intense dust dynamics have been achieved when, second, the (two-dimensional) positions of the dust particles are identified and tracked through the image sequence in each camera individually. The next and most difficult step then is to identify corresponding particles in the different cameras. The problem here mainly lies in the fact that the particles in the camera images are indistinguishable. When this step has been accomplished one has the full 3-D particle positions at hand for the physical interpretation of structure and dynamics of the dust ensemble.

The individual steps together with the requirements and limitations will be discussed in the following.

2.1. Camera calibration

In a simple pinhole camera model (Hartley & Zisserman 2004) the viewing geometry of the camera can be described as follows (see Salvi, Armangué & Batlle (2002) for camera calibration accuracy). A point in the world coordinate system \( M = (X, Y, Z)^T \) is projected by the lens system onto a point \((x_p, y_p)^T\) on the camera image plane by

\[
\begin{bmatrix}
    x'_p \\
    y'_p \\
    z'_p
\end{bmatrix}
= P \cdot \begin{bmatrix}
    X \\
    Y \\
    Z
\end{bmatrix}
\quad \text{and} \quad
\begin{bmatrix}
    x_p \\
    y_p
\end{bmatrix}
= \begin{bmatrix}
    x'_p/z'_p \\
    y'_p/z'_p
\end{bmatrix},
\]

(2.1a,b)

where \( P \) is the \( 3 \times 4 \) projection matrix. This projection matrix

\[
P = K \cdot [R \mid t]
\]

(2.2)

contains the intrinsic \( 3 \times 3 \) camera matrix \( K \) as well as the \( 3 \times 3 \) rotation matrix \( R \) and the translation vector \( t \) that describe the camera orientation and position with respect to the origin of the world coordinate system. The intrinsic camera matrix contains the camera-specific properties such as focal length \( f \), principal point (image centre) \( p \) and the skew \( \alpha \) (which generally is zero for modern cameras) and is then given by

\[
K = \begin{bmatrix}
    f_x & \alpha & p_x \\
    0 & f_y & p_y \\
    0 & 0 & 1
\end{bmatrix}.
\]

(2.3)

From this it can be determined how a point in the real world is imaged onto the plane of the camera. In a stereoscopic camera system, this projection matrix \( P \) has to be determined for each camera individually.

An efficient way to determine the projection matrix for the cameras is to observe a calibration target with known properties simultaneously with all cameras. Wengert et al. (2006) and Bouguet (2008) have developed Matlab toolboxes that allow us to reconstruct the projection matrices from the observation of a calibration target. Alternatively, ‘self-calibration’ routines can be applied (Svoboda, Martinec & Pajdla 2005) which yield camera projection models from observation of a small, easily detectable, bright spot. However, for measurements in calibrated real world units (e.g. mm) a calibration target with a known reference geometry is required.
For our dusty plasma experiments, a calibration target with a pattern of dots of 0.5 mm diameter and a centre-to-centre distance of the dots of 1 mm has been found to be useful (see figure 2). In addition, according to Wengert et al. (2006), the target has two perpendicular bars in the centre for unique identification of target orientation in the cameras. For further information on the target see Himpel, Buttenschön & Melzer (2011), a different target and camera calibration concept can be found in Li et al. (2013).

This calibration target is now moved and rotated in the field of view of all cameras and simultaneous images for the different target positions or orientations have to be captured. Approximately 30–40 different target images per camera are sufficient to reliably determine the projection matrices \( P \) for each camera. For each image, the calibration dots are extracted using standard routines (Gaussian bandwidth filtering and successive position determination using the intensity moment method, see e.g. Feng, Goree & Liu (2007), Ivanov & Melzer (2007)), the central bars are identified from their ratio of major to minor axis as well as their absolute area (Wengert et al. 2006). The calibration dots are indexed by their position relative to the central bars. These coordinates are unique for every dot and are the same in all cameras and for all the different target orientations. This then allows to retrieve the line of sights...
of all cameras individually and the relative camera orientations, in short, the camera projection matrices (Bouguet 2008).

The calibration target with the perpendicular orientation bars together with the Matlab toolboxes (Wengert et al. 2006; Bouguet 2008) allow an automated analysis of the projection properties (Himpel et al. 2011). The camera calibration has to be performed in the actual experiment configuration (including mirrors etc.) and has to be updated each time the camera set-up is changed (adjustment of focus, adjustment of mirrors etc.). In a parabolic flight campaign, the calibration is performed before and after each flight day. The projection properties will be required to identify corresponding particles as described in §2.3. An accurate calibration will return more reliable correspondences.

2.2. Particle position determination

In actual experiments, the particles are viewed with the different cameras in the stereoscopic set-up and the images are recorded on a computer. The first step in data analysis then is to identify the particles and their (2-D) positions in the camera images. This can be done using standard algorithms known from colloidal suspensions or from dusty plasmas, see e.g. Crocker & Grier (1996), Feng et al. (2007), Ivanov & Melzer (2007). Depending on image quality, first Sobel filtering or Gaussian bandwidth filtering of the image might be applied. Position determination can be done using intensity moment method or least-square Gaussian kernal fitting (Crocker & Grier 1996; Feng et al. 2007; Ivanov & Melzer 2007). Such methods are usually able to determine particle positions with subpixel accuracy in the 2-D images when 5–10 pixels constitute a particle in the image. Problems occur when two or more particle projections overlap or are close to each other.

From our experience, we find it useful, as the next step, to track the particles from frame to frame in each camera separately. So, 2-D trajectories \((x_p(t), y_p(t))^T\) of particles in each camera are obtained. This is done by following each particle through the subsequent frames. Thereby, all particles that lie within a certain search radius in the subsequent frame are linked to the particle in the actual frame. Hence, a particle in the starting frame may constitute more than only a single possible trajectory through the next frames. Then, a cost function for all possible tracks of the start particle is computed. The cost of a trajectory increases with acceleration (both in change of speed and change of direction). The trajectory with lowest cost over the following 10 frames is chosen (Dalziel 1992; Sbalzarini & Koumoutsakos 2005; Ouellette, Xu & Bodenschatz 2006). This way, smoother trajectories are favoured.

This 2-D particle tracking usually reliably works for up to 200 particles per frame (for a megapixel camera). When more particles are visible in an individual frame, a unique identification of 2-D particle trajectories becomes increasingly difficult. In general, this also limits the number of particles that should be visible in the 3-D observation volume to approximately 200 (see also §3).

2.3. Identifying corresponding particles

To retrieve the 3-D particle positions it is necessary to identify corresponding particles in the different cameras (‘stereo matching’), i.e. we have to determine which particle in camera \(C'\) is the same particle that we have observed in camera \(C\). As mentioned above, this task is quite difficult since the particles are indistinguishable. In standard techniques of computer vision one can exploit a number of image properties to identify correspondence, such as colour information, detection of edges or patterns etc.
These do not work here since all the particles usually appear as very similar small patches of bright pixels against a dark background. This problem can be solved in two ways. When three (or more) cameras with good data quality are available, corresponding particles can be identified from the set of camera images taken at each instant by exploiting the epipolar line approach. When only two cameras can be used, then in the first step, we identify all possible corresponding particles in camera C′ for a given particle in camera C using epipolar lines. Then, in the second step, we exploit the information from the 2-D trajectories \((x_p(t), y_p(t))^T\) to pin down the single real corresponding particle.

2.3.1. Correspondence analysis using epipolar lines

The possible corresponding particles are found from the epipolar line approach (Zhang 1998). In short, the epipolar line \(l'\) is the line of sight of the image point \(m\) in the image plane of camera C of a 3-D point \(M\) as seen in the image plane of camera C′, as shown in figure 3. Here, \(m\) is the 2-D position of a dust particle in the image of camera C and the point \(M\) corresponds to the real world 3-D coordinate of the particle, which is to be determined. Now, the corresponding projection \(m'\) of this point \(M\) in the image plane of the second camera C′ has to lie on the epipolar line \(l'\).

This condition can be formulated as

\[
\tilde{m}'^T \cdot F \cdot \tilde{m} = 0,
\]

where \(\tilde{m} = (m_x, m_y, 1)^T\) and \(F\) is the so called fundamental matrix

\[
F = K'^{-T}[t]_xRK^{-1},
\]

which can be constructed from the projection matrices of camera C and C′, see (2.2). Here, \([t]_x\) is the antisymmetric matrix such that \([t]_x x = tx\).

Now, for a point \(m\) in camera C, all possible candidates \(m'\) lying within a narrow stripe around the current epipolar line \(l'\) are taken as the possible corresponding particles to the particle with image point \(m\). Empirically, the stripe width around the epipolar line is chosen to be approximately 1 pixel in our case, see figure 4.

The figure shows a snapshot of a dust particle cloud from the three cameras of our stereoscopic set-up. In the left camera, a particle \(a\) is chosen and the corresponding epipolar line \(l'_a\) in the right camera is calculated. In this case, there are 4 particles \(a'\) to \(d'\) in the right camera that have a small distance to the epipolar line and, thus,
are possible corresponding particles to particle $a$. When a third camera is present (top camera) the epipolar line $l''_a$ of particle $a$ in the top camera can be calculated as well as the epipolar lines of the particles $a'$ to $d'$ of the right camera. Now, it is checked whether there is a particle in the top camera that is close to the epipolar line $l''_a$ of particle $a$ from the left camera and either of the epipolar lines of the possible candidates $a'$ to $d'$ from the right camera. Here, in the top camera, there is only a single particle that is close to both the epipolar line $l''_a$ of particle $a$ and, in this case, the epipolar line of particle $a'$ (the epipolar lines $l''_a$ of particle $a$ and $l''_{a'}$ of particle $a'$ are shown). Hence, here, a unique correspondence is found between particle $a$ in the left camera, $a'$ in the right camera and $a''$ in the top camera from a single snapshot.

2.3.2. Correspondence analysis using trajectories

When a third camera is not available or the epipolar line criterion does not uniquely define particle correspondences, the dynamic information of the particle motion in camera $C$ and $C'$ can be exploited. From the tracking we have the 2-D trajectory of the chosen particle $a$ in the left camera $C$ ($x_C^a(t)$, $y_C^a(t))^T$ as well as the trajectories of the possible correspondences in camera $C'$ ($x_{C'}^{a'}(t)$, $y_{C'}^{a'}(t))^T$. It is then checked how far each possible correspondence in camera $C'$ moves away over time from the (time dependent) epipolar line of particle $a$. For a real corresponding particle, the deviation of its 2-D particle position from the epipolar line is found to be less than approximately 1 pixel throughout the entire trajectory, see figure 4(d). It can

\begin{figure}
\centering
\includegraphics[width=\textwidth]{figure4.png}
\caption{(a–c) Snapshot of a dust particle cloud from the three cameras of our stereoscopic set-up together with epipolar lines to check for particle correspondences (the raw images are inverted: particles appear dark on a light background). See text for details. (d) Deviation of the particles $a'$ to $d'$ from the (time dependent) epipolar line $l''_a$ over several frames. Note the logarithmic axis scaling.}
\end{figure}
be clearly seen that only the corresponding particle $a'$ stays close to the projected epipolar line whereas the others ($b'$ to $d'$) develop much larger excursions.

In our analysis the matching trajectories must have a minimum length of 50–100 frames. The advantage of this procedure is that the identified particle pairs are reliable corresponding particles. The disadvantage is that, due to the restrictions, a number of matching particles are not further considered. This happens mainly when the 2-D trajectories are not correctly determined over their temporal evolution. Nevertheless, in typical parabolic flight experiments we can identify several hundred reliable 3-D trajectories with a minimum length of 100 frames in a sequence of 1500 frames.

2.4. Three-dimensional trajectories, velocities and accelerations

Having identified the particle correspondences, the 3-D position is determined from triangulation. An optimized triangulation procedure according to Hartley & Sturm (1997) is used here. Because the approach is specially designed for triangulation with two cameras, there are three pairwise triangulations possible within a set of three cameras. We take the mean location of the three pairwise triangulations as the final particle position. The variance of each pairwise triangulation is called the triangulation error. The triangulation error results from the error in the projection matrices and camera vibrations as well as the 2-D particle position error. In our set-up, the triangulation error is usually of the order of 10 $\mu$m when a 2-D particle position error of approximately 0.5 pixel is assumed.

Quite often, overlapping particle images will occur. Nevertheless, 3-D trajectories can still be reliably reconstructed for particles that have overlapping images in one camera, but not the other(s). More cameras help in minimizing the problems with overlapping images.

The stereoscopic algorithm hence determines the 3-D particle positions $r_i(t)$ over time. As mentioned above, the particle velocities $v_i = \Delta r_i/\Delta t$ can then directly be determined (with $\Delta t$ being the time between successive video frames, i.e. $\Delta t$ is the inverse frame rate).

However, due to uncertainties in the accuracy of the 3-D particle position, the 3-D trajectory is quite ‘noisy’ which makes it difficult to determine the velocity directly. Therefore, we found it useful first to smooth the original position data by a Savitzky–Golay filter (Savitzky & Golay 1964) (in many cases a second-degree polynomial with a window width of nine frames has been used, see e.g. Buttenschön et al. (2011)). From the smoothed position data, the difference quotient is computed to obtain the particle velocities $v_i = \Delta r_i/\Delta t$. For the particle acceleration, the velocity data are Savitzky–Golay smoothed again and used for computing the second difference quotient $a_i = \Delta v_i/\Delta t$.

2.5. Algorithm

The determination of the 3-D particle positions from the stereoscopic camera images can be summarized in the following ‘algorithm’:

(1) Calibrate all cameras with the calibration target and compute the projection matrix $P$ including the camera matrix $K$, rotation matrix $R$ and translation vector $t$ for all cameras.

(2) Determine the 2-D particle positions and particle trajectories in the recorded images for each camera separately retrieving the 2-D trajectories $(x_{Cp}^i(t), y_{Cp}^i(t))^T$ of the particles in all cameras.

(3) Pick a particle $p$ in camera C.
(4) Determine its epipolar line \( l_p' \) in camera \( C' \) using (2.4).

(5) Determine the possible corresponding particles \( p' \) in \( C' \) within a narrow stripe around the epipolar line \( l_p' \).

(6) (i) When three or more cameras are available: pin down the correspondences \( p \) and \( p' \) by comparing the epipolar line criterion in the other camera(s).

(ii) When only two cameras are available: calculate the distance of the possible correspondences \( p' \) from the (time dependent) epipolar line of particle \( p \). A corresponding particle \( p' \) should not deviate by more than approximately 1 pixel from the epipolar line over 50 or so frames.

(7) Determine the 3-D position/3-D trajectory from the corresponding pair \( p \) and \( p' \) from triangulation.

(8) Return to step 3 for next particle \( p \).

2.6. Requirements, conditions and limitations

From the above described stereoscopic reconstruction techniques the following requirements, conditions and limitations follow for successful application of stereoscopy:

(i) Two or more synchronized cameras with a common observation volume are required.

(ii) Stable, vibration-free mounting of all cameras is necessary.

(iii) Homogeneous illumination of the dust particles in the observation volume increases image quality and hence particle determination.

(iv) Larger angular distance between the cameras (the best is 90°) improves triangulation accuracy.

(v) A successful 3-D reconstruction of particle positions is possible when up to approximately 200 particles are visible in each camera (for a megapixel camera, see § 2.2).

(vi) In dense particle clouds, the visible particle number density can be decreased using fluorescent tracer particles (see § 3).

(vii) Correspondence analysis becomes increasingly difficult the more particles are found close to an epipolar line.

3. Example of measurement

As an example, we now discuss measurements of the microscopic particle motion in a dust-density wave (DDW) under weightlessness. DDWs are compressional and rarefactive waves of the dust component excited by an ion flow through the dust ensemble (Barkan, Merlino & D’Angelo 1995; Prabhakara & Tanna 1996; Khrapak et al. 2003; Schwabe et al. 2007; Hou & Piel 2008; Merlino 2009; Thomas 2009; Arp et al. 2010; Flanagan & Goree 2010; Menzel et al. 2010; Himpel et al. 2014; Williams 2014). These waves have frequencies of the order of 10 Hz and wave speeds of the order of a few centimetres per second. These waves are very prominent features in volume-filling dusty plasmas, see e.g. figure 5, they feature very strong density modulation between the wave crest and the wave trough.

Recently, the microphysics of particle motions in the crests and troughs, as well as the trapping of particles in the wave crest, have been of interest (Hou & Piel 2008; Teng et al. 2009; Chang, Teng & I 2012; Himpel et al. 2014). Since the observation of a 2-D slice of the dust can only give a limited view on the particle dynamics, it is beneficial to measure the 3-D particle motion. This has been done using our stereoscopic set-up shown in figure 1.
The experiments have been performed on a parabolic flight campaign during 2013 in Bordeaux, France. An argon plasma is produced in a capacitively coupled rf parallel-plate discharge. The discharge gap in the plasma chamber (IMPF-K2, (Klindworth, Arp & Piel 2006)) is 3 cm. The discharge was operated with an rf-power of 3 W at a gas pressure of 20 Pa. The electrodes have a diameter of 8 cm. The dust particles in this experiment are melamine–formaldehyde (MF) microspheres with a diameter of $6.8 \mu m$. The ion-excited DDWs then appear above a critical dust density (Menzel, Arp & Piel 2011).

To further reduce the number of visible particles in the observation volume of the stereoscopic cameras (see § 2.2) we have added to the ‘standard’ MF particles a small fraction of approximately 1–2 % of Rhodamine-B-doped fluorescent MF particles of the same mass and size. When illuminated by a Nd : YAG laser at 532 nm these particles show a fluorescence around 590 nm. By using bandpass filters for wavelengths from 549 to 635 nm in front of the camera lenses we only observe the small number of fluorescent particles as tracer particles in our cameras (see Himpel et al. (2012, 2014) for details). Figure 6 shows a snapshot of the fluorescent tracer particles inside a DDW as seen by the three stereoscopic cameras. In each camera approximately 50 particles are visible in the fluorescent light. The field of view was chosen to achieve single-particle resolution on the one hand, and to capture collective wave motion on the other. The density of visible (fluorescent) particles then is approximately 600 cm$^{-3}$, which corresponds to a total particle density of roughly $4 \times 10^4$ cm$^{-3}$ since the fluorescent particles constitute only 1–2 % of all particles.

These images are taken from a single parabola of our parabolic flight campaign. The analysed image sequence had 1500 frames recorded at 180 f.p.s., hence covering 8.3 s of the 22 s of weightlessness during the parabola (at the beginning of the parabola the dust was injected and an equilibrium situation with self-excited DDWs had to develop before the sequence was taken).
Stereoscopic imaging of dusty plasmas

As described above, the 2-D positions of the particles have been identified in the individual cameras, their correspondences among the different cameras have been determined, and the 3-D trajectories of the particles have been derived. In total, 3-D trajectories of 233 particles with a minimum trajectory length of 75 frames have been obtained. Hence, it is observed that quite a large number of particle trajectories can be reconstructed. This even allows the derivation of statistical properties of the dust particle motion: as an example, the velocity distribution functions of the particles along the \( z \)-direction and perpendicular (\( x \) and \( y \)-directions) to the wave propagation has been determined and analysed in Himpel et al. (2014).

Here, figure 7(a) shows the trajectory of a single particle during its oscillatory motion in the DDW. It is seen that the wave motion is mainly in the \( z \)-direction (compare figure 5). For a more quantitative analysis the instantaneous phase of the particle oscillation has been determined from a Hilbert transform (see Menzel et al. (2011), Killer et al. (2014)) and is shown colour coded with the trajectory. As expected, it is found that near oscillation maximum the instantaneous phase is near zero whereas near oscillation minimum it shifts towards \( \pm \pi \).

We now investigate the full 3-D dust trajectories of all particles in a DDW. In figure 7(b) the trajectories, together with their instantaneous phases, are shown in a moving reference frame that accounts for the DDW propagation along the \( z \)-direction, i.e. the \( z \)-axis has been rescaled \( z \to z - ct \) with the measured wave speed \( c = \omega / k \approx 20 \text{ mm s}^{-1} \). Also, a stroboscopic-like approach has been applied: with known wave period, the trajectories are mapped onto a single oscillation period of the DDW. For that purpose, from the times \( t \), entire multiples of the oscillation period \( T = 2\pi / \omega = 0.172 \text{ s} \) have been subtracted so that \( 0 < t < T \) (Himpel et al. 2014).

It is found that there are distinct stripes of fixed phases in this DDW representation indicating that here the DDW exhibits a coherent wave field. The DDW can be seen

**Figure 6.** Raw images of the particles in fluorescent light in the three stereoscopic cameras (here, for better recognition in the publication, a ‘dilate’ filter has been applied to the images using image processing tools). Also, the approximate region illuminated by the laser in the focal range is indicated by the lines. The images are 640 × 480 pixels corresponding to approximately to 8 mm × 6 mm.
FIGURE 7. (a) Trajectory of a single dust particle in the DDW. A clear oscillatory motion is seen. The instantaneous phase of the particle oscillations, as determined from a Hilbert transform, is colour coded. At oscillation maximum the phase angle is near zero (light/yellow colours) and at oscillation minimum it is near $\pm \pi$ (dark/blue colours). (b) 3-D dust particle trajectories forming a DDW. The data are plotted in a moving reference frame accounting for the wave motion along the $z$-direction. Again, the colour indicates the instantaneous phase. (c) As (b), but averaged over the $y$-direction. The arrows indicate the wave crests with phase angle near zero.

to extend approximately 3 wavelengths in the $z$-direction whereas the phase is flat in the $x$ and $y$-directions, see also figure 7(c).

4. Future developments

Recently, other techniques have been developed to extract 3-D information of particle positions or velocities in dusty plasmas, namely light field imaging with plenoptic cameras (Hartmann et al. 2013) or tomographic-PIV (Williams 2011).

A light field image can be recorded by a so-called plenoptic camera using a microlens array directly in front of the camera chip. The lens array produces a large number of similar images of the object on the chip that usually has some to some ten megapixels. These multiple images then allow to compute refocused images representing different depth layers from the light field function and hence to determine the depth of the object, see e.g. (Levoy 2006) for details. Hartmann et al. (2013) have introduced this concept to dusty plasmas. It is very tempting and straightforward to combine stereoscopy with the imaging by plenoptic cameras.

Similarly, with tomographic-PIV a common observation volume is imaged by multiple cameras. After camera calibration, the intensity recorded along a line of sight is distributed over the observation volume which is divided into voxels. Typically,
algebraic reconstruction techniques are employed to derive a ‘density’ distribution and from successive reconstructions a 3-D velocity field can be extracted by standard PIV adapted to three dimensions. Williams (2011) has applied this tomographic-PIV to dusty plasmas, recovering 3-D velocity fields. The general set-up and the camera calibration requirements seem to be quite comparable between tomographic-PIV and stereoscopy. In the case of high dust densities, tomographic-PIV is certainly better suited than stereoscopy since particle positions do not need to be determined. For low densities, stereoscopy allows us to determine individual trajectories. A cross-over from stereoscopy to tomographic-PIV certainly deserves to be studied.

5. Summary
We have presented the basic techniques for the application of stereoscopy to dusty plasmas. Stereoscopic methods allow to retrieve the full 3-D particle positions with high spatial and temporal resolution.

The underlying techniques have been demonstrated together with their requirements and limitations. As an example, stereoscopy has been applied to measure the microphysics of particles in a dust-density wave under the weightlessness conditions of a parabolic flight.

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A7

Experimental determination of phase transitions by means of configurational entropies in finite Yukawa balls

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*(Editors’ Suggestion)*
The phase transition of finite Yukawa balls (ordered systems of microspheres in a gaseous plasma environment) with less than 100 particles is studied experimentally by means of configurational entropies. We have developed cylindrical two- and three-particle-correlation functions to measure these entropies for multiple cluster sizes over a wide temperature range. The cluster temperature is finely tuned using a stochastic laser heating setup. It is shown that the correlation functions give a detailed insight into the structural properties of the cluster. The derived configurational entropies give a clear indication of the transition temperature from a solid-like to a fluid-like state. Comparing the transition temperatures of different sized clusters it is found that the transition temperature increases with cluster size in general agreement with theoretical predictions.

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I. INTRODUCTION

The transition from very small clusters with only few constituents to an extended system is essential for the understanding of how macroscopic effects of matter develop on multiple scales. Well-known objects of investigation for these effects are, e.g., atomic and molecular clusters [1–3] or the many forms of granular matter [4]. Such systems inherit properties that depend on the number of constituents and how they interact on the individual level. The focus of this work will be on the determination of phase transitions of rather small conglomerates and how they are affected by the number of constituents.

The objects of investigation in this paper are Yukawa balls in a dusty plasma environment. Besides the neutral gas component, dusty plasmas consist of ions and electrons with an additional third charged species, the dust (for details, see, e.g., [5–7] and related references therein). These macroscopic dust particles with a size between a few nanometers and several micrometers charge due to the inflow of plasma electrons and ions. Usually, they gain very high negative charges since the electrons are more mobile than the ions. Yukawa balls [8–11] are finite three-dimensional systems made of only a few to thousands of microspheres. From the competition of Coulomb repulsion and confining plasma forces the particles arrange in a spherical structure on nested, concentric shells [12,13]. Being strongly coupled but optically thin makes them a unique and rich tool for investigating size-depending effects of phase transitions on the individual level. In such a finite particle system one can directly measure the velocity $v$ of individual particles with mass $m$. From that one can define the kinetic temperature $T = m v^2/3k_B$ of the system from the particles’ velocity distribution. Increasing the kinetic temperature of the particles one can drive and study phase transitions in these small systems.

In a simplified picture the point of phase transition of a macroscopic solid state body can be determined by feeding heat to the system and monitor the temperature change. First, the temperature will rise as the atoms oscillate stronger around their fixed lattice positions. At a certain point the temperature rise will stop because the additional heat is used to break up the bond structure. This point is the transition temperature in a first-order phase transition. Upon further heating the atoms overcome their lattice positions entering the liquid state where the atoms can move more freely. Accordingly, the system exhibits a jump in the entropy at the transition temperature.

It is a question of high interest how this can be translated into a finite system of less than 100 charged particles that form an ordered structure like a Yukawa ball. A strongly coupled charged-particle system is described by the coupling parameter which is the ratio of thermal and Coulomb energy

$$\Gamma = \frac{Z^2 e^2}{4\pi \epsilon_0 \mathbf{b}_{WS} / \lambda_D} \frac{1}{k_B T}$$

with the electrical charge number $Z$ and the Wigner-Seitz distance $b_{WS} = \sqrt{3/4\pi n}$, which is of the order of the inter-particle distance and is calculated from the particle number density $n$. It has been shown in simulations [14] that an infinite three-dimensional (3D) one-component plasma (a system of a single charged species in a homogeneous neutralizing background) will change its phase from solid to liquid at a critical value of $\Gamma_{\text{crit}} \approx 168$ being solid above it and liquid-like below it. In an experimental dusty plasma the particles interact with a shielded Coulomb potential which affects the crystalization properties. To account for this, Ikezi [15] estimated a modified coupling parameter $\Gamma_{\text{eff, Ikezi}} = \Gamma \exp(-\kappa) \approx 168$ with an added shielding term $\kappa = b_{WS}/\lambda_D$ which gives the Wigner-Seitz radius in units of the Debye length $\lambda_D$. This has been studied further by Vaulina et al. [16] revealing a more complex dependency between coupling parameter and screening. However, in general the transition line for a Yukawa system runs in a $\Gamma \sim \kappa$-plane where the (unmodified) critical coupling parameter increases with the strength of the shielding [17].

Schiffer [18] studied in simulations how the critical coupling parameter depends on the number of particles for finite systems. He evaluated the transition temperature as an inverse coupling parameter for different sized Coulomb clusters. He has shown that the critical coupling parameter increases with the decreasing particle number.

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The aim of this paper is to study the size dependence of the transition temperature for Yukawa balls with less than 100 particles. To reach it we want to induce and investigate phase transitions and identify the critical point by using statistical entropy methods.

II. CONFIGURATIONAL ENTROPIES

Phase transitions in finite systems can be described in a phenomenological way, for example by analyzing fluctuations in the calculated inter-particle distance [19] or by looking at the decay of radial and angular correlation functions [20–22]. Recently, a melting criterion has been derived from a thermodynamics quantity by Thomsen and Bonitz using configurational entropies from different particle correlation functions [23]. This allows identifying phase transitions in finite systems on a quantitative level and it is the aim of this paper to study these entropies from experiments.

Two correlation functions used by Thomsen and Bonitz are the center two-particle correlation function (C2P) and the triple correlation function (TCF) to measure the configurational entropy of a system. These correlations are evaluated from the 3D positions of the particles in a specific set of coordinates exploiting the spherical symmetry of the problem as illustrated in Fig. 1(a). For the C2P the length of the respective position vectors \( r_1 \) and \( r_2 \) relative to the trap center and the solid angle \( \theta \) between them are considered. The TCF uses a particle triplet 1-2-3 on a selected shell evaluating the angular distances \( \theta_1, \theta_2 \) between particles 2-1 and 2-3 and the bond angle \( \varphi \) on the spherical shell surface.

For the C2P, the probability of finding a particle pair at \( r_1, r_2 \) and \( \theta \) is determined by averaging over all particle pairs. Similarly, for the TCF the probability of finding particle triplets with \( \theta_1, \theta_2 \), and \( \varphi \) is determined. Moreover, the averaging extends over a large number of manifestations of the finite system at a fixed temperature. Hence, one needs the 3D positions of all particles at many instants in the described coordinate sets \( Q \). This poses a challenge to the experiments. Finally, the space-resolved histograms \( \tilde{\rho}(Q) \) have to be normalized by geometrical factors \( V_i \) that depend on the chosen coordinates and are given in [23] for spherical symmetry. Hence, the correlation functions are given as

\[
C2P(r_1, r_2, \theta) = \frac{\tilde{\rho}(r_1, r_2, \theta)}{V_i(r_1, r_2, \theta)}
\]

(2)

and

\[
TCF(\theta_1, \theta_2, \varphi) = \frac{\tilde{\rho}(\theta_1, \theta_2, \varphi)}{V_i(\theta_1, \theta_2, \varphi)}
\]

(3)

where \( k \) indicates the number of correlated particles. From those, the reduced Shannon entropies

\[
S^{(2)} \equiv -\langle \ln C2P \rangle \quad \text{and} \quad S^{(3)} \equiv -\langle \ln TCF \rangle
\]

(4)

measure the configurational entropy of the system at the temperature \( T \) (see [23,24]). As usual, it is possible to derive heat capacities as \( C = \delta S/\delta T \) but this paper will focus purely on the configurational entropy. The described correlation functions and configurational entropies have been investigated in simulations by Thomsen and Bonitz using Yukawa balls with perfect spherical symmetry.

III. EXPERIMENTAL SETUP

The experimental setup shown in Fig. 2 is a refined version of the one used by Schella et al. [22,25]. Its centerpiece is a capacitively coupled radio frequency (rf) discharge operated in argon at gas pressures typically between 5 and 50 Pa. The electrode at the bottom of the chamber is driven with rf powers below 2 W while the rest of the chamber is grounded, hence forming an asymmetrical low temperature discharge.

Melamine-formaldehyde (MF) particles with a radius of 4.7 \( \mu \)m are trapped in the sheath region above the rf-electrode. For confinement of the particles we use a brass ring instead of the conventional glass cuvette [10]. The ring floats electrically and is positioned about 20 mm above and parallel to the electrode. The upwards sheath electric field, the radial inwards electric field due to the ring and the downward gravitational field create a nearly harmonic 3D confinement in which the
particles form a Yukawa ball. The experiments on the dust clusters shown here have been performed at a gas pressure of 14.5 Pa and an rf power of 800 mW. In order to observe the particles they are illuminated by two fiber-coupled DPSS lasers at 660 nm with a light output of up to 1000 mW each. The forward scattered light is captured by three CMOS cameras that record images at a resolution of $1280 \times 1024$ pixels at a frame rate of up to 500 fps. Two cameras are viewing from the side under 90° relative to each other, while the third is viewing from top, however slightly tilted by 22° with respect to the vertical direction. Camera calibration and epipolar-line particle reconstruction developed by Himpel et al. [26–28] based on the work of Wenger et al. [29] and Bouguet [30] has been applied to retrieve the 3D particle position from the individual camera images. To investigate the phase transition of the Yukawa ball one needs to observe it at different temperatures. Established experimental methods to induce phase transitions [22,31–33] are based on the neutral gas pressure, plasma power or particle number in dusty plasmas. These methods affect more than a single quantity in the coupling parameter. Furthermore they rely partly on additional instabilities due to ion streaming [34,35] which are not considered in the coupling parameter making it difficult to evaluate their impact on the phase transition. To overcome this we chose a pure laser heating setup to raise the kinetic energy of the particles directly via radiation pressure leaving the confining plasma system undisturbed [22,25,36–38]. The focused light of two green (532 nm) laser modules with up to 600 mW each is distributed over the cluster by two galvanometer scanners using random scan patterns. With the laser power the kinetic temperature of the particles is controlled. Increasing the laser power leads to higher particle temperatures, see [39] for details. In the measurements, Yukawa balls with particle numbers from 17 to 72 are trapped and heated at various laser power settings. For each laser-driven temperature the motion of all particles is recorded over 5000 frames at 100 fps. The 3D particle positions have been reconstructed for all 5000 frames at a fixed temperature. From that the particle correlation functions and the corresponding configurational entropies are calculated as described in the following. IV. RESULTS A. Structure of Yukawa balls

The typical structure of Yukawa balls that we investigate in our experiment can be seen in Fig. 3(a) for an example of a 39-particle cluster, consisting of two shells. It can be seen that especially the particles of the inner shell are located atop of each other. Obviously, in the experimental environment streaming ions [34] lead to chain-like vertical particle alignment. Hence, it seems that the particles arrange in a cluster structure with a more cylindrical symmetry [35,40].

To illustrate this further, the particle density of the cluster is shown in Fig. 3(b). The density has been calculated from the particle positions as a mean over all frames. The density is shown averaged over the azimuthal angle $\Omega$ in cylindrical $\mathbf{R} = (\mathbf{r}, \Omega)$ coordinates where $\mathbf{R} = \sqrt{X^2 + Z^2}$ is the radial coordinate and $Z$ is the vertical direction (along gravity).
we chose to integrate \( R_I \) over one cylindrical shell. This results in as many 2D visualizations as there are shells where the first of the two correlated particles is always chosen from this particular shell.

We start with the discussion of the cylindrical C2P in Fig. 4(b). In the left hemisphere the C2P was integrated over the inner shell (where \( 0.2 \text{ mm} < R_I < 0.55 \text{ mm} \)). This inner shell of the 39-particle cluster features four equidistant vertical particle chains (see Fig. 3), so an arbitrary particle on this shell will see other particles on the same shell at an angle \( \Omega \) of about 0°, 90°, or 180°. Correspondingly the C2P shows distinct islands of high correlation at these angles and a radius around \( R_2 \approx 0.3 \text{ mm} \). The outer shell on the other hand features a nine-fold symmetry seen from above (see Fig. 3) which does not share a common divisor with the four-fold inner shell. This means the shells do not share a common symmetry and a particle on the first shell can see a particle on the second shell at various \( \Omega \) resulting in the smeared out area at \( R_2 \approx 0.8 \text{ mm} \). When we now look into the right hemisphere of Fig. 4(b), where the first particle was chosen to be in the outer shell (0.55 mm < \( R_I < 0.85 \text{ mm} \)) we see high correlation in the outer shell at angles \( \Omega \) of 0°, 40°, 80°, 120° and 160° and at a radius of \( R_2 \approx 0.8 \text{ mm} \) in agreement with the nine-fold rotation symmetry. The inner shell now appears smeared out.

We conclude that the cylindrical C2P represents the structure very well due to the rotational symmetry of the cluster seen from above.

Now, how does this compare to the spherical C2P shown in Fig. 4(a)? In both cases (where integrating over the inner spherical shell in the left hemisphere or the outer spherical shell in the right hemisphere) no clear correlations can be identified. In order to observe distinct correlation islands the cluster would need not only rotational symmetry around one axis but 3D isotropy which it lacks. So if one would try to analyze the evolution of a correlation island over a larger temperature range the finely structured spherical correlations would produce artifacts in the computed entropies. Similar behavior can be observed for the triple correlation function which will not be discussed in detail here. We therefore choose to use the cylindrical C2P and TCF to derive heat capacities and entropies for clusters of various particle numbers.

**B. Correlation functions and heating**

With the manipulation lasers, the cluster was heated in 26 steps from a kinetic temperature of 600 K up to 15000 K. The average particle charge number has been determined to be about \( Z \approx 3400 \) by normal mode analysis [41]. The Wigner-Seitz radius is measured as \( b_{WS} = 0.28 \text{ mm} \) calculated from the static particle positions yielding a coupling parameter range between 1168 and 44. Hence, one should expect a phase transition happening inside that range.

At first the C2P and TCF distributions for a solid and a liquid phase state will be discussed. Starting with the C2P one can directly look into the changes due to the temperature increase since the structural properties that can be deducted for the solid case of \( \Gamma \approx 440 \) [Fig. 5(a)] were already described in Sec. IV A. The C2P for the heated cluster with \( \Gamma \approx 130 \) is shown in Fig. 5(b). Again, the distribution was integrated over \( R_I \) in the range of the first shell (left hemisphere) or second shell (right hemisphere). It is apparent that for \( \Gamma \approx 130 \) the overall distribution is smeared out compared to the solid case (\( \Gamma \approx 440 \)). More precisely on the one hand the space between the two radial shells is now “filled” as well, which means that it is now possible to find a particle between the shells. Nevertheless, the individual shells are easily identified. Moreover, correlation peaks can still be identified as in the solid state, however less pronounced. On the other hand one can see in the right hemisphere of Fig. 5(b) that the long range angular order of the outer shell gets lost for \( \Omega > 90^\circ \). This stepped heating behavior with earlier loss of orientational order followed shortly by the loss of radial order is well known for finite systems [22,42] and confirms the applicability of the C2P.

Moving on to the TCFs shown in Figs. 5(c) and 5(d) it is important to mind the differences to the C2P. While the C2P uses all particle pairs of the whole cluster and the plot hemispheres are generated due to integrating over different shells, the TCF uses particle triples on one specific shell. Therefore the hemispheres in Figs. 5(c) and 5(d) show different correlation functions for the two shells. Nevertheless it is still necessary to integrate over one of the variables to generate a 2D plot. Here we integrate over \( L_I \) with the range chosen so that the first and second particle are nearest neighbors. While the C2P polar plots can be interpreted as correlations from a top view.
on the clusters particle density the TCF show probable particle triplets on the unrolled cylinder side surface. Consequently, it is sufficient to have \( L_3 \) in the range from 0 to half the perimeter of the cylindrical shell and \( \Psi \) in the range of 0 to 180°. Since the perimeter is different for the two shells, we normalize \( L_3 \) to the perimeter length. In view of the nine-fold symmetry of the outer shell one would expect an arbitrary particle of the outer shell to “see” eight other particle columns around the perimeter of the cylinder. And that is exactly what one can find in the right TCF hemisphere of Fig. 5(c), which corresponds to the outer shell of the solid cluster. Since this plot only shows half the perimeter, there are four areas of high correlation at \( L_3 \approx 0.11, 0.22, 0.33, 0.44 \) perimeter fractions (p.f.) due to the nine-fold symmetry. This being visible already in the C2P, the TCF gives additional insight in the intrashell structure of which some exemplary features will be described now. The first one is the correlation gap in all TCFs in the range of the nearest neighbor distance \( d_{\text{NN}} \approx 0.25 \) p.f. for the first shell and \( d_{\text{NN}} \approx 0.1 \) p.f. for the second shell between \( \Psi = 0° \) and \( \Psi = 30° \). The existence of this area is evident because the second particle is chosen by the integration to be a direct neighbor so a third in the same distance at a bond angle around 0° is not possible in a strongly coupled system. Further, in the outer shell of the solid cluster [right hemisphere of Fig. 5(c)] the first high correlation areas appear in the nearest neighbor region at \( \Psi \approx 45° \). This means the vertical particle columns are not interlocked in a way that forms a hexagonal structure on the cylinder surface, which would result in a bond angle of \( \Psi = 60° \), but are rather aligned in a square lattice. This characteristic arises due to the final boundaries of the cluster, the strong vertical confinement and the non-commensurate number of particles to form a hexagonal lattice. In such a square lattice an arbitrary particle with an arbitrary neighbor will eventually “see” a third particle directly above, below or at same height all around the cylinder perimeter. This leads to the elongated islands of high correlation for larger \( L_3 \) which have their center at bond angles of \( \Psi \approx 0°, 90°, 180° \).

Interestingly, the inner shell shown in the left hemisphere of Fig. 5(c) does not show correlation islands at those positions. Here, they start to form at \( \Psi \approx 60°, 120°, 180° \) for second and third particles of the triplet in the nearest neighbor region. Thus the inner shell does in opposition to the outer shell feature a more hexagonal than square lattice. As the cluster heats up, one can again observe the loss of long range order resulting in the blurred correlation functions shown in Fig. 5(d).

In summary, the presented modified distribution functions are well suited to investigate the intershell and intrashell cluster structure.

C. Configurational entropies

The next step is to derive the configurational entropies for each cluster temperature by using Eq. (4) and then look for indicators of a phase transition. Figure 6 shows the thermal progression of the configurational C2P entropies \( S^{(2)} \) and TCF entropies \( S^{(3)} \) of the 39-particle cluster and will now be described in more detail.

To compare the entropies from the different correlation functions, the curves have been normalized to their respective maximum value which is always the one at the lowest \( \Gamma \). The error bars indicate a rough estimation of the data consistency. To generate them, every entropy has been calculated with only half of the data set (frames) and the difference between that and the entropy from the full data set is given as the error.
Starting at low temperatures the entropies calculated from the different correlation functions have their minimum at $\Gamma \approx 1000$. Then, upon heating, the entropies increase slowly. The region between $410 > \Gamma_{\text{cut}} > 320$ features a steeper slope after which a region with an again decreased slope follows. We interpret the range of $\Gamma \approx 370$ as the region of phase transition due to the relatively sharp increase in entropy. The dashed gray line indicate the middle point of that region and can be identified as the critical coupling parameter $\Gamma_{\text{cut}}$. For $\Gamma < \Gamma_{\text{cut}}$ the fluid range is seen, whereas for $\Gamma > \Gamma_{\text{cut}}$ the solid phase is found. Finally, the grey marked area above $\Gamma \approx 1000$ indicates a region where the entropy counterintuitively again increases with $\Gamma$. This happens due to metastable configurations in which the cluster is trapped [43–45]. At these high $\Gamma$ values it is difficult for the systems to reach the ground state. With enough kinetic energy, the cluster is able to “fall down” into its ground state, where the entropy is minimal. This is an effect that we were able to observe multiple times for different clusters.

The overall trend of all three curves in Fig. 6 is generally very similar. Thomsen and Bonitz however, have observed distinct differences between the $S^{(2)}$ and $S^{(3)}$ for clusters well below the melting temperature [23]. Their simulations featured additional changes in the slope of the TCFs $S^{(3)}$ for the different shells of a 80-particle cluster and identified them as the early onset of intrashell disordering. If we were able to heat up the cluster from much lower temperatures resulting in an entropy curve that spans one or two orders more of magnitude on the $\Gamma$-scale it should be possible to see this onset of intrashell-disordering as well. With the experimental setup at hand this is not possible and therefore it is well expected that the $S^{(2)}$ and $S^{(3)}$ curves are very similar. Due to this fact and since its curve features the highest dynamic range we will focus on the C2P entropies in the following analysis of different clusters.

D. Size effect

To discuss the effect of different particle numbers Fig. 7 shows the C2Ps $S^{(2)}$ entropy curves for a 17-particle cluster and the already discussed 39-particle cluster.

At the beginning (highest $\Gamma$, lowest $T_{\text{fin}}$) the smaller cluster is trapped in a metastable configuration as already explained in the last section. It stands out that the error bars in that area are much larger than in other regions. It turns out, that there are some particle rearrangements happening in the observation time, which is typical for a cluster that is not in its ground state. The repetition timescale for those rearrangements is almost as large as the observation time. Thus, by bisecting the frames for the error estimation they happen mainly in one of the data halves resulting in much different derived entropies. This shows that a much larger observation time would be needed for a cold cluster to attenuate the effect of metastable configurations. But since our main focus lies in the identification of the phase transition this is a negligible effect.

Looking into the phase transition it is apparent that the small cluster has a very distinct and sharp increase in entropy, while the larger clusters transition is smoother and seems to feature a two-staged melting indicated by the drop of entropy at $\Gamma \approx 250$. We assume that this is a side-effect of the cylindrical geometry assumed for the distribution. On the one hand, the manipulation lasers are only heating along one axis resulting in an anisotropic force that elongated the cluster horizontally at higher laser powers. On the other hand a hotter cluster tends to assume a more spherical shape because the force of the ion wind forming the chains in the solid state becomes less dominant. Nevertheless, the phase transition is clearly identified for both clusters.

Figure 8 shows the critical temperature derived from the experiments for four clusters with particle numbers of 17, 18, 39, and 72 particles. One can see, as a general trend, that the critical temperature increases with the particle number. This effect has already been seen by Schiffer [18] in his simulations. He proposes a linear dependency between the ratio of particles on the outer shell of a Coulomb cluster and the transition temperature. This linear trend is drawn together with our experimental data. The blue circles indicate the transition temperatures of the four Yukawa balls with 16/17,
FIG. 8. Cluster size effect on the transition temperature of four different clusters ($N = 17, 18, 39, 72$) compared to the predicted curve for Coulomb clusters by Schiffer [18].

6/18, 32/39, and 50/72 particles on their outer shells. Our experiments show a very similar linear trend but the absolute transition temperatures are a factor of 1.5 above Schiffer’s predicted curve. Including the effect of shielding would result in even larger distance between our experimental data and Schiffer’s line.

There are some reasons that we believe lead to the rather large difference between Schiffer’s simulations and our experiments. First, the calculated coupling parameters highly depend on the particle charge [see Eq. (1)]. The particle charge is determined as $\frac{e}{m}$ = 3400 ± 530 from our normal mode analysis. Hence, the $\Gamma_{\text{crit}}$ is uncertain to about 30% which would just come close to Schiffer’s trend line. This error in $\Gamma$ is the absolute error for all four clusters, the relative error between the experiments is much smaller since they have been performed under identical conditions.

Another question is, whether Schiffer’s predicted linear trend holds true towards very low particle numbers. The smallest cluster Schiffer simulated had 100 particles and was already somewhat above his linear approximation with a larger error bar than the bigger clusters. It is possible that the linear trend only holds true for particle numbers above 100. Our clusters are all below 100 particles. For such small clusters effects of “magical numbers” (clusters with certain particle numbers, that are more stable due to a symmetrical particle configuration) come into play as well. Apolinario and Peeters [46] showed that such “magical numbers” do exist for spherical, two-shelled Yukawa balls and that the transition temperature is much higher for those configurations compared to non-magical clusters of similar size. The particle numbers of our investigated cluster are different from Apolinario’s. However due to the different geometry (cylindrical instead of spherical and additional wake-field influence) certain very stable configurations might exist for other particle numbers. To evaluate this a finer variation of particle number would be necessary which is beyond the scope of this article. Our observed dependency between cluster size and transition temperature does not hint toward very stable configurations yet.

Summarizing, we observe a clear size dependence of the phase transition using cylindrical correlation functions to measure a statistical configurational entropy.

V. CONCLUSION

Our experiments showed that the usage of correlation functions as proposed by Thomsen and Bonitz [23] are highly suitable for evaluation of structural properties and determination of phase transitions of finite Yukawa balls. Furthermore, it is possible to adapt the original spherical correlation functions to anisotropic confinement properties leading to symmetries like the cylindrical we presented, which confirms the robustness of the approach. Using multiple cluster sizes we were able to find a clear dependence between cluster size and melting temperature. Larger clusters need more kinetic energy thus a higher temperature to break up the inner bonds and change into another phase state which is in agreement with the findings of Schiffer [18] for Coulomb clusters.

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A. Thesis Articles
Analysis of 3D vortex motion in a dusty plasma

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(Editor’s Pick)
Analysis of 3D vortex motion in a dusty plasma

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Dust clusters of about 50–1000 particles have been confined near the sheath region of a gaseous radio-frequency plasma discharge. These compact clusters exhibit a vortex motion which has been reconstructed in full three dimensions from stereoscopy. Smaller clusters are found to show a competition between solid-like cluster structure and vortex motion, whereas larger clusters feature very pronounced vortices. From the three-dimensional analysis, the dust flow field has been found to be nearly incompressible. The vortices in all observed clusters are essentially poloidal. The dependence of the vorticity on the cluster size is discussed. Finally, the vortex motion has been quantitatively attributed to radial gradients of the ion drag force. Published by AIP Publishing. https://doi.org/10.1063/1.5006841

I. INTRODUCTION

Dusty plasmas allow to study dynamical phenomena on the kinetic level of individual (dust) particles. Typically, monodisperse micrometer-sized spherical particles are immersed and trapped in a gaseous plasma discharge of electrons, ions, and neutrals. In the plasma, the dust particles acquire a highly negative charge and form a strongly coupled electron–ion–neutral system where the temporal and spatial scales are ideally suited to study the motion of the particles by video microscopy.1–3 Larger dust clouds can behave like incompressible fluids and can provide insights into Kelvin-Helmholtz or Rayleigh-Taylor instabilities4–7 on a kinetic, single-particle level. Additionally, macroscopic fluid-like behavior manifests in vortex motion which has attracted great interest, recently, in experiments, theory, and simulations; see, e.g., Refs. 8–19.

In most of the experiments, only two-dimensional cross-sections of the dust clouds were available for the analysis of the dust motion. Further, the vortices were often observed in the outer edges of the dust cloud quite far from the symmetry axis of the dusty plasma discharge. This makes it difficult to gain a complete overview over the vortex torus. Similarly, in theoretical descriptions of the vortex motion, rotational symmetry around the discharge axis was assumed. Hence, a full three-dimensional measurement of the properties of a complete vortex in dusty plasmas is missing.

In this article, we present experiments on the compact dust vortices in dust clusters of various sizes. We present an experimental set-up to confine dust clusters with about 50 up to 1100 particles exhibiting strong vortex motion. By stereoscopic methods,21–28 the full three-dimensional (3D) information of the individual dust particle motion has been measured over a long observation time. Hence, the full 3D dynamics of the dust vortex can be analyzed with high accuracy. We will look into the particle densities, flow fields, and related questions of compressibility and vorticity of the flow and the dependence on the cluster size. Finally, the vortex drive will be analyzed.

II. EXPERIMENTAL SET-UP

The dust clusters used for the vortex experiments have been trapped in a radiofrequency (rf) plasma discharge similar to the one used in previous experiments.27–29 The rf power at 13.56 MHz was chosen as 1.1 W at an argon gas pressure of 20.9 Pa for the experiments presented here.

Melamine-formaldehyde (MF) particles of 2a = 4.7 μm diameter have been dropped into the discharge. The dust is confined in the sheath region below a brass ring placed 20 mm above the lower electrode (see Fig. 1). The brass ring has an inner diameter of 24 mm and is electrically floating. The brass ring provides an electric confining potential that together with the plasma forces acting on the dust (electric field force of the plasma sheath, gravity, and ion drag force) is nearly harmonic in the 3 spatial directions. In the experiments described here, several three-dimensional clusters with varying particle number from N = 47 to 1100 particles have been formed by successively removing more and more particles under the same discharge conditions.

The dust particles are illuminated by two fiber-coupled DPSS lasers at 660 nm with a light output of up to 1000 mW each. The lasers are expanded to illuminate the whole dust cloud. Due to the expansion of the beams, the particles remain undisturbed by the radiation pressure. The scattered light is recorded by a stereoscopic camera system consisting of 4 cameras that observe the particles from various directions, two cameras looking from the side and two cameras looking from top under a small angle relative to the vertical axis (see Fig. 1). The cameras had a resolution of 1280 × 1024 pixels and operated at a frame rate of 100 frames per second. From the 4 cameras, the 3D positions of the particles are recovered using standard techniques.23–26 The clusters have been recorded for 20 s (2000 frames) and the 3D trajectories of the particles have been retrieved.

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III. EXPERIMENTAL RESULTS

We now investigate the three-dimensional dynamical behavior of the particles in the different clusters starting with the trajectories and densities and then look into the velocity and vorticity profiles before the size dependence of the vorticity and the vortex drive are discussed.

A. Trajectories and densities

Figure 2 shows the example trajectories of particles in a small cluster with particle number $N = 151$ and in a large cluster with $N = 1100$. Note that only a small fraction of the available trajectory data is shown for clarity. In both cases, it is clearly seen that the particles follow a vortex-like motion where the particles move downwards in the central part of the cloud and upwards in the outer parts. In general, the trajectories look quite symmetric around the central axis. Naturally, the cluster with the smaller number of particles has a smaller spatial extent compared with the one with larger particle number.

In a first step, the particle density is derived for the two clusters discussed earlier (see Fig. 3). Here, due to the supposed cylindrical symmetry, the particle number density is calculated and shown in a $\rho$–$z$-plot, where $\rho = (x^2 + y^2)^{1/2}$ is the radial position relative to the symmetry axis and $z$ is the vertical position.

It is easily seen that the clusters are not spherical: the smaller cluster features a heart-like shape, whereas the larger cluster is more torus-like. For the smaller cluster, the density distribution is quite non-uniform. It shows density maxima at certain radial and vertical positions. This reflects the higher probability of finding particles at these positions, indicating that the cluster is still in a quite solid-like state where certain structural positions are preferred. One can identify the vertically aligned structure due to the wakefield interaction between the particles. The average number density is about $11 \text{ mm}^{-3}$.

The larger cluster, in contrast, shows a more homogeneous density distribution with a triangular central region of higher density that slowly drops to the edges. The density distribution is much more fluid-like than compared with the smaller cluster. Nevertheless, traces of preferred “shells” of rotation can be identified. The average number density in this case is about $7 \text{ mm}^{-3}$.

We find a strict trend that the clusters with higher particle number have a smaller average density. This is, as seen above, due to the fact that the smaller clusters are much more solid-like than the larger ones. The larger clusters explore a much larger volume during the vortex-like rotations and, hence, their average density is reduced.
In an alternative approach, since we have the full 3D position information, we have performed a 3D Voronoi analysis of the structure [see Fig. 4(a)]. We then have calculated the volume of the Voronoi cells and, correspondingly, the density distribution as the average over the inverse Voronoi cell volume. This is shown in Fig. 4(b) for the large cluster with \(N = 1100\) particles. In general, this Voronoi density distribution is qualitatively and quantitatively very similar to the one shown in Fig. 3(b). Again, the triangular region of high density is quite pronounced. However, the Voronoi density behaves much smoother near the central axis. In comparison to Fig. 3(b) where the particles per volume element are counted, the problem of vanishing radial volume elements near the central axis does not appear for the Voronoi analysis. This is since simply the (average) inverse Voronoi volume is assigned to each \(\rho-z\) coordinate. However, in contrast, the outer regions of the cloud are less well characterized due to difficulty to construct Voronoi cells for particles lying on the outer edge.

### B. Velocities

For a more quantitative, three-dimensional analysis, the volume occupied by the clusters is divided into 30 \(\times 30 \times 30\) volume elements (“voxels”). Now, for each voxel, the velocities in Cartesian coordinates \((v_x, v_y, v_z)\) as well as in cylindrical coordinates \((v_q, v_\phi, v_z)\) are determined as the average of the respective velocity component over all particles crossing the voxel. Figure 5 shows these velocity components as volumetric slices for a medium-sized cluster with \(N = 528\).

The measured velocity distribution is quite what one would expect for a vortex-like motion. For the Cartesian velocity \(v_x\), the uppermost slice shows a positive velocity at negative \(x\)-positions and vice versa, indicating a motion towards the center. In the lowermost slice, the velocities are just opposite indicating a motion away from the center. The maximum velocity is of the order of 5 mm/s. The corresponding behavior is seen for the Cartesian velocity \(v_y\). The vertical velocity component \(v_z\) shows the expected downward motion on the central axis and the upward motion in the outer parts. The vertical velocity is somewhat higher than the \(x\) and \(y\) components.

Similarly, the radial velocity component \(v_q\) shows positive outward motion in the lower part of the cloud and radially inward motion in the upper part. Looking at \(v_q\) and \(v_\phi\), the velocity components seem to be symmetric in azimuthal direction. Indeed, the azimuthal velocities \(v_\phi\) are much smaller than all the other components, but nevertheless a slight asymmetry can be seen within the cloud probably due to slight asymmetries in the confinement.

### C. Stream function

Having verified that the dust particle motion and rotation are essentially only in the \(\rho-z\)-plane, we now take a brief look in to the azimuthally averaged velocity data. Figures 6(a)–6(c) show the averaged radial and vertical velocity components \(v_r\) and \(v_z\) as well as the total velocity \(|v|\). As already shown above, in the lower part of the cloud, we see the radial outflow of particles and at the upper part, the radial inflow. Correspondingly, the particles move upward in the outer part of the cloud and downward in the inner part. The magnitude of the velocity [see Fig. 6(c)] is very small in the center of the vortex and increases nearly linearly with distance from the vortex center. This would correspond to a nearly
rigid-body rotation around the vortex center (see also below). Along the rotation path, the velocity is nearly constant. However, near the central cloud axis, the dust particles seem to get accelerated downwards until they reach a region of near stagnation in the lowest areas of the dust cloud where one sees a relatively sharp bending of the trajectories from the vertical downward motion near the axis to an inclined upward motion afterwards. In the upper part of the cloud, no such sharp changes of direction are seen.

In axisymmetric flows (here, the symmetry axis is the central vertical axis of the dust cloud), a stream function $\psi(\rho, z)$ can be defined to characterize the particle flow where then the flow velocity components are determined as the derivatives of the stream function via

$$
v_x = -\frac{1}{\rho} \frac{\partial \psi}{\partial \rho} \quad \text{and} \quad v_z = \frac{1}{\rho} \frac{\partial \psi}{\partial z},
$$

in cylindrical coordinates. Using the experimental flow data, the stream function is calculated via integration using the algorithm of Ref. 34 as shown in Fig. 6(d). It shows a singular well characterizing a single-mode source vorticity with mode number $l = 1$ as discussed in Ref. 15. For this mode, the first zero of the stream function eigenmode is located on the boundary.

**D. Compressibility**

One assumption often made when describing the flow of dust particles is that the flow is incompressible, i.e., $\text{div} \vec{v} = 0$. Correspondingly, we have calculated that quantity for the measured flow field and the result is shown in Fig. 7.

It is seen that the flow field mainly shows random fluctuations of $\text{div} \vec{v}$ around zero, confirming that the flow is indeed essentially incompressible. However, at the very top of the cluster, there is a small ring-like region that features somewhat stronger (negative) values of $\text{div} \vec{v}$. At that region, the dust particles enter the central region of the cluster and are accelerated downwards [compare Fig. 6(c)].
flow is slightly thinner compared with the remainder of the vortex. In contrast, a compression region at the lower edge of the cluster where the “stagnation” was observed cannot be identified. In summary, the assumption of incompressibility can be justified.

E. Vorticity

To quantify the vortex motion of the cluster, we now have calculated the corresponding vorticity of the threedimensional flow field, \( \omega = \nabla \times \mathbf{v} \). Figure 8 shows the measured vorticity in cylindrical coordinates \( \omega_\rho, \omega_\phi, \omega_z \). It can be seen that \( \omega_\phi \) and \( \omega_z \) exhibit only random fluctuations, whereas only \( \omega_\rho \), as one would expect, carries the vorticity. The vorticity is mostly negative (indicating the above-discussed rotation direction) and has a maximum magnitude of the order of \(-15 \text{ s}^{-1}\).

On the central axis, also positive values of the vorticity are found. This, however, stems from the fact that the particles at the center at the top of cluster move downwards very slowly if at all [see also Fig. 6(c), where in the top part of the cluster near the center, the vertical velocity is nearly zero]. So, when particles that are a little further away from the center move downwards with greater speed, this results in a (seemingly) positive vorticity near the top center.

F. Size dependence

We now have averaged the poloidal vorticity \( \omega_\phi \) over the azimuthal angle and the vertical position \( z \), yielding the radial dependent vorticity \( \bar{\omega}_\phi(\rho) \). This averaged vorticity \( \bar{\omega}_\phi(\rho) \) is now compared among the differently sized clusters in Fig. 9. It is seen that the large clusters have zero vorticity in the center, then the vorticity drops to a value of the order
of $\bar{\omega}_x = -8 \text{ s}^{-1}$ and stays nearly constant throughout the cluster indicating a rigid-body-like vortex motion.

The smaller clusters feature a zero or slightly positive vorticity in the center and a negative vorticity at larger radial distances. Finally, in the very outer parts, they again show a zero or slightly positive vorticity. Due to the much more solid-like behavior of the smaller clusters, sometimes stationary particles are found at the outer rim and on the central axis of the cluster. These particles do not participate in the total rotation and, hence, lead to a zero or slightly positive vorticity in comparison with the particles rotating past them.

The fact that some particles are almost stationary might be attributed to the action of the ion wakefield. For example, on the central axis, there are only small transverse forces which are easily counterbalanced by the ion wakefield. Hence, it is not easily possible to remove a dust particle from the central chain. These are the dust particles usually observed at the top center of a cluster mentioned above.

For the larger clusters, where these solid-like, stationary particles do not play an essential role, one finds a constant, rigid-body rotation of the cluster with an average rotation frequency of $22 \pm 8 \text{ s}^{-1}$.

G. Vortex drive

It has been shown in Refs. 14, 16, and 17 that the vorticity of the dust particle flow field $\vec{\omega}$ can be derived from the dust particle’s force balance and the Navier-Stokes equation as

$$m\vec{\beta} = e\nabla Z \times \vec{E} + \nabla f(E) \times \vec{E}. \quad (2)$$

Here, $m$ is the dust particle mass, $Z$ is its charge, $\beta$ is the Epstein dust-neutral collision frequency, and $E$ is the electric field strength. Thus, a vortex is driven by a non-vanishing curl of the electric field force $\vec{F}_E = eZ\vec{E}$ when a charge gradient in the dust cloud exists or by a non-vanishing curl of the ion drag force $\vec{F}_{\text{ion}} = f(E)\vec{E}$ with a gradient of the “mobility” function $f(E)$. Here, as in Ref. 17, it is assumed that the ion drag force is in the flow direction of the ions where the ion flow velocity $\vec{u}_i = \mu(E)\vec{E}$ depends on the
electric field via the ion mobility \( \mu E \).\(^{35}\) Using the well-established expressions for the ion drag force of Refs. 36–38, the mobility function \( f(E) \) can be calculated as a rather complicated function of the electric field.

In our experiment, we essentially observe only a poloidal rotation of the dust cloud. Such a poloidal rotation is driven by radial gradients of the dust charge density \( \partial_r Z \) or by radial gradients of the ion mobility function \( \partial_r f(E) \). These radial gradients might most probably be due to radial changes of the density of the plasma species (see also Refs. 14, 16–18, and 20). However, a radial gradient of electron and ion density does not cause a dust charge gradient \( \partial_r Z \) as long as the electron temperature and electron-to-ion density ratio are constant over the radius. Hence, we presume that the vortex is not driven by charge gradients.

In contrast, radial density gradients lead to gradients of the ion mobility function \( \partial_r f(E) \) (see also Refs. 16 and 17). Hence, the vortex drive can be expressed as

\[
m \partial_r \phi_0 = E \partial_r f(E),
\]

where \( m \) is the dust mass.

As an estimate, the electric field is derived as \( E = 620 \text{ V/m} \) from the condition of vertical force balance between the electric field force and gravity, i.e., \( mg = Eq \). Here, \( g \) is the gravitational acceleration and the dust potential was assumed as \( \phi = -5 \text{ V} \) (the electron temperature was assumed as \( 2 \text{ eV} \)) corresponding to a dust charge number \( Z \) as long as the electron temperature and electron-to-ion density ratio are constant over the radius. Hence, we presume that the vortex is not driven by charge gradients.

Using the vorticity value of \( \partial_r \phi_0 \approx 8 \text{ s}^{-1} \) found for most of the large clusters, we find the radial derivative of the mobility function from Eq. (3) as

\[
\partial_r f(E) = 4.5 \times 10^{-14} \frac{\text{m}}{\text{Vs}}.
\]

Note that the vertical gradients of the electric field and the mobility function do not contribute to the observed poloidal vortex motion.

Finally, the gradient scale length can estimated as

\[
\frac{f(E)}{\partial_r f(E)} = 30 \text{ mm},
\]

which corresponds quite nicely to the electrode radius of 40 mm. Hence, one could assume that the vortex is indeed driven by the gradient of the plasma density over the radial extent of the electrode.

IV. SUMMARY

Using an electrically floating metal ring above the electrode, we were able to confine dust clusters from about 50 to 1100 particles in an rf discharge. All investigated clusters show a vortex motion. For the smaller clusters, a competition between solid-like cluster structure and vortex motion is observed, whereas larger clusters feature pure vortices. The full three-dimensional motion of all particles has been reconstructed from stereoscopy. Hence, the vortex motion of all particles in a compact cluster is available for the analysis on the individual particle level.
34. K. Pankratov, see http://pordlabs.ucsd.edu/matlab/stream.htm for How to compute a streamfunction?, 1994.
B. Scientific Contributions

Publications in peer-reviewed journals

1. M. Mulsow, M. Himpel and A. Melzer
   “Analysis of 3D vortex motion in a dusty plasma”

2. M. Mulsow and A. Melzer
   “Experimental determination of phase transitions by means of configurational entropies in finite Yukawa balls”

3. A. Melzer, M. Himpel, C. Killer and M. Mulsow
   “Stereoscopic imaging of dusty plasmas”

4. C. Killer, M. Mulsow and A. Melzer
   “Spatio-temporal evolution of the dust particle size distribution in dusty argon rf plasmas”

5. A. Schella, M. Mulsow and A. Melzer
   “Correlation buildup during recrystallization in three-dimensional dusty plasma clusters”

6. A. Melzer, A. Schella and M. Mulsow
   “Nonequilibrium finite dust clusters: Connecting normal modes and wakefields”

7. A. Schella, M. Mulsow, A. Melzer, H. Kähler, D. Block, P. Ludwig and M. Bonitz
   “Crystal and fluid modes in three-dimensional finite dust clouds”

8. A. Schella, M. Mulsow, A. Melzer, J. Schablinski and D. Block
   “From transport to disorder: Thermodynamic properties of finite dust clouds”
B. Scientific Contributions

Oral presentations at Workshops and Conferences

1. M. Mulsow and A. Melzer
   *Experimental determination of phase transitions by means of configurational entropies in finite Yukawa-balls*

2. M. Mulsow and A. Melzer
   *Experimental determination of phase transitions by means of configurational entropies in finite Yukawa-balls*
   8th International Conference on the Physics of Dusty Plasmas, Prague, Czech Republic (2017)

3. M. Mulsow and A. Melzer
   *Investigating structure and dynamics of Yukawa-balls*
   DPG Spring Meeting, Hannover (2016)

4. M. Mulsow and A. Melzer,
   *Yukawa-balls: New experimental methods*
   4th International Workshop on Diagnostics and Simulation of Dusty Plasmas, Kiel (2015)

5. M. Mulsow, A. Schella and A. Melzer,
   *Experimental investigation of specific heat in finite 3D Yukawa-balls*
   5th HEPP-Colloquium at the DPG spring meeting, Bochum (2015)
Poster presentations at Workshops and Conferences

1. M. Mulsow and A. Melzer,  
*Experimental investigation of specific heat in finite 3D Yukawa-balls*  
43rd European Physical Society Conference on Plasma Physics, Leuven, Belgium (2016)

2. M. Mulsow and A. Melzer,  
*Experimentelle Beobachtung expandierender Yukawa-Cluster*  
DPG Spring Meeting, Bochum (2015)

3. M. Mulsow, A. Schella and A. Melzer  
*Fluid mode excitation in finite 3D Yukawa-balls*  
XXII Europhysics Conference on Atomic and Molecular Physics of Ionized Gased, Greifswald (2014)

4. M. Mulsow, A. Schella and A. Melzer  
*Selektive Modenanregung in finiten 3D Yukawa-Clustern*  
DPG Spring Meeting, Berlin (2014)

5. M. Mulsow, A. Schella and A. Melzer  
*Selective fluid mode excitation in finite 3D Yukawa-balls*  
7th International Conference on the Physics of Dusty Plasmas, New Delhi, India (2014)

6. M. Mulsow, A. Schella and A. Melzer  
*Scheranregung in finiten 3D Yukawa-Clustern*  
DPG Spring Meeting, Jena (2013)
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