

Energy and Temperature Relaxation described by Nonequilibrium Green's Functions

J. Vorberger¹, D.O. Gericke¹, Th. Bornath², M. Schlanges³

¹ Centre for Fusion, Space & Astrophysics, University of Warwick, Coventry, UK

² Institut für Physik, Universität Rostock, Rostock, Germany

³ Institut für Physik, E.-M.-Arndt Universität Greifswald, Greifswald, Germany

E-mail: j.vorberger@warwick.ac.uk

Abstract. A quantum kinetic approach is presented to investigate the energy relaxation of dense strongly coupled two-temperature plasmas. We derive a balance equation for the mean total energy of a plasma species including a quite general expression for the transfer rate. An approximation scheme is used leading to an expression of the transfer rates for systems with coupled modes relevant for the warm dense matter regime. The theory is then applied to dense beryllium plasmas under conditions such as realized in recent experiments. Special attention is paid to the influence of correlation and quantum effects on the relaxation process.

1. Introduction

The recent developments of new methods for the creation and probing of warm dense matter and strongly coupled plasmas have brought an impressive progress in the experimental characterization of high energy density matter. For instance, x-ray scattering allows not only the measurement of equation of state (EOS) data, but also to obtain structural, dynamic, and collective properties of matter [1, 2, 3, 4, 5]. For the creation of such high energy density states in the laboratory, one relies on dynamic experiments using intense particle or laser beams to compress and heat the sample. In any case, this leads to the production of highly nonequilibrium states first. Although in most experiments not of primary interest, such nonequilibrium states deserve attention for several reasons. The first one is a practical one for EOS measurements and the like, as the time delay between pump and probe pulses needs to be greater than the relaxation time. The second reason is given by the interest to study the processes that lead from a highly nonequilibrium state towards equilibrium. During the first and shortest stage of a few femtoseconds, equilibrium distributions are established within the electron and ion subsystems. Ionisation and recombination is finished shortly thereafter. On a picosecond timescale, the process of temperature relaxation equals electron and ion temperatures. Apart from the following hydrodynamic motion of the substance, temperature relaxation takes the longest time of all relaxation processes and is also experimentally accessible.

An intense discussion about its order of magnitude started when Dharma-wardana & Perrot [6] found considerably smaller energy transfer rates as predicted by classical Landau-Spitzer (LS) theory [7, 8] and early computer simulations [9] if coupled electron-ion modes were included. Indeed, experiments investigating dense plasmas, found strong indications of longer relaxation times, thus smaller energy transfer rates [11, 12]. One of the shortcomings of the LS approach was

addressed applying a quantum approach for binary collisions which yields however even larger rates [13, 14, 15]. Furthermore, considering independent collective modes in the subsystems within the Fermi-Golden-Rule approach, rates close to the LS results were obtained [16]. Therefore, collective modes seem to be the only candidate to explain the lower electron-ion energy transfer in dense strongly coupled plasmas.

However, a rigorous description of temperature relaxation requires not only appropriate transfer rates but has also to involve the interplay of all contributions to the internal energy. In addition to the kinetic parts, correlations and exchange energies [17, 18, 19], and the ionisation kinetics including excitations [20, 21, 22] can be important.

Here, we assume ionisation, recombination, and charge transfer processes to be completed and concentrate on the electron-ion energy relaxation. We use a quantum kinetic description which enables us to rigorously derive the balance equations and expressions for the transfer rates within an appropriate approximation scheme. The influence of coupled mode effects on temperature relaxation are still under discussion (see Refs. [6, 23, 24, 25, 26, 27]). We therefore re-derive the coupled mode energy transfer rate based on the density operator formalism. Together with adequate expressions for the internal energies/heat capacities, this enables us to determine the time evolution of electronic and ionic temperature in a nonideal system with ionic and electronic correlations and exchange. We demonstrate the effect of the nonideality contributions and the transfer rates on the evolution of the temperatures, on the relaxation time, and on the final temperature.

2. The energy balance equation

We consider a spatially homogeneous nonequilibrium plasma. To investigate the relaxation of the plasma towards thermodynamic equilibrium, balance equations can be used. With the single particle Hamiltonian H_a and the two-body potential V_{ab} , the mean kinetic and the mean potential energy of species "a" can be defined by

$$\langle K_a(t) \rangle = \text{Tr}_1 \{ H_a \rho_a(t) \}, \quad \langle V_a(t) \rangle = \frac{1}{2} \sum_b \text{Tr}_{1,2} \{ V_{ab} \rho_{ab}(t) \}, \quad (1)$$

where ρ_a and ρ_{ab} are the one- and two-particle density operators, respectively. Note that definition (1) implies the symmetric distribution of electron-ion contributions to the internal energy into the electron and ion subsystems. The reduced density operators are determined by the following equations of motion which are the first two equations of the quantum version of the BBGKY hierarchy (time variables dropped) [28]

$$i\hbar \frac{\partial}{\partial t} \rho_a = [H_a, \rho_a] + \sum_b \text{Tr}_2 [V_{ab}, \rho_{ab}], \quad (2)$$

$$i\hbar \frac{\partial}{\partial t} \rho_{ab} = [H_{ab}, \rho_{ab}] + \sum_c \text{Tr}_3 [(V_{ac} + V_{bc}), \rho_{abc}]. \quad (3)$$

The square brackets denote the commutator $[A, B] = AB - BA$.

Using the first equation, one can find the following expression for the change of the mean kinetic energy over time [19]

$$\begin{aligned} \frac{\partial}{\partial t} \langle K_a \rangle &= \frac{1}{2} \sum_b \text{Tr}_{1,2} \frac{1}{i\hbar} \{ (H_a + H_b) [V_{ab}, \rho_{ab}] \} \\ &\quad + \frac{1}{2} \sum_b \text{Tr}_{1,2} \frac{1}{i\hbar} \{ (H_a - H_b) [V_{ab}, \rho_{ab}] \}. \end{aligned} \quad (4)$$

The first term on the r.h.s. of Eq. (4) can be transformed using Eq. (3). Then one can show easily that it is equal to the time derivative of the mean potential energy of species a , i.e. equation (4) can be cast into [19]

$$\frac{\partial}{\partial t} \langle K_a \rangle + \frac{\partial}{\partial t} \langle V_a \rangle = \sum_b Z_{ab}, \quad (5)$$

where Z_{ab} defines the energy transfer rate between species a and b

$$Z_{ab} = \frac{1}{2} \text{Tr}_{1,2} \frac{1}{i\hbar} \{ (H_a - H_b) [V_{ab}, \rho_{ab}] \}. \quad (6)$$

Note that only terms with $b \neq a$ give non-zero contributions to the total rate and that $Z_{ab} = -Z_{ba}$. Summation over species a in Eq. (5) then gives

$$\sum_a \frac{d}{dt} \langle K_a \rangle + \sum_a \frac{d}{dt} \langle V_a \rangle = \sum_{a,b} Z_{ab} = 0, \quad (7)$$

which expresses the conservation of the total energy.

3. Energy transfer rate

We write the transfer rate in the form

$$Z_{ab} = \frac{1}{2} \text{Tr}_{1,2} \frac{1}{i\hbar} \{ (H_a - H_b) [V_{ab}, \rho_a \rho_b + \rho_{ab}^{\text{corr}}] \}, \quad (8)$$

where ρ_{ab}^{corr} is the correlation part of the two-particle density operator ρ_{ab} . Our goal is to derive an expression for the energy transfer rate for dense nonequilibrium plasmas where collective phenomena can play an important role. For this purpose, it is useful to apply the following relation ($b \neq a$)

$$\langle 12 | \rho_{ab}^{\text{corr}}(t) | 2'1' \rangle = i\hbar L_{ab}^<(11't, 22't')|_{t=t'}. \quad (9)$$

Here, $L_{ab}^<$ is the correlation function of density fluctuations given by

$$i\hbar L_{ab}^<(11't, 22't') = \langle \psi_b^\dagger(2', t') \psi_b(2, t') \psi_a^\dagger(1', t) \psi_a(1, t) \rangle - \langle \psi_b^\dagger(2', t') \psi_b(2, t') \rangle \langle \psi_a^\dagger(1', t) \psi_a(1, t) \rangle, \quad (10)$$

with ψ_a^\dagger and ψ_a being the creation and annihilation operators of species a , respectively. Spin variables will be suppressed for simplicity. With Eq. (9) and using the equations of motion for the function $L_{ab}^<(t, t')$, one can derive the following expression for the energy transfer rate for spatially homogeneous systems [29],

$$Z_{ab}(t) = -2\mathcal{V} \text{Im} \int \frac{d^3q}{(2\pi\hbar)^3} \int_0^\infty \frac{d\omega}{2\pi} \omega V_{ab}(q) i\hbar L_{ab}^<(\mathbf{q}; \omega, t), \quad (11)$$

where \mathcal{V} denotes the volume, and $L_{ab}^<(\mathbf{q}; \omega, t)$ is the Fourier transform of $L_{ab}^<(\mathbf{r}_1 t_1, \mathbf{r}_2 t_2)$ with respect to the difference variables $\tau = t_1 - t_2$ and $\mathbf{r} = \mathbf{r}_1 - \mathbf{r}_2$.

Expression (11) allows for the determination of the energy transfer rate applying appropriate approximation schemes for the correlation function of density fluctuations. For this purpose, we start from the equation for the function L_{ab} defined on the Keldysh time contour [30, 31]

$$L_{ab}(t, t') = \Pi_{ab}(t, t') + \sum_{c,d} \int_C d\bar{t} \Pi_{ac}(t, \bar{t}) V_{cd} L_{db}(\bar{t}, t'). \quad (12)$$

Here, Π_{ab} is the polarization function, V_{ab} is the Coulomb potential, and the symbol \int_C stands for the integration along the contour. For simplicity, only the time variables were given explicitly. Applying the well-known techniques originally introduced by Keldysh, the equation for the correlation function $L_{ab}^<$ can be obtained from Eq. (12) [30, 31]. Furthermore, the equations for the retarded and advanced functions follow easily.

Now, we will specify our considerations on a two-component plasma consisting of electrons with density n_e and ions of density n_i . As a first approximation we assume $\Pi_{ab} = \delta_{ab}\Pi_a$ where Π_a is considered to be the polarization function of the subsystem of species a , i.e. contributions from the other species are neglected. Then a system of equations follows for the functions L_{ee} , L_{ei} , L_{ie} , and L_{ii} defined on the contour.

For further considerations, it is useful to introduce the auxiliary functions \mathcal{L}_e and \mathcal{L}_i , defined by

$$\mathcal{L}_a = \Pi_a + \Pi_a V_{aa} \mathcal{L}_a, \quad (a = e, i), \quad (13)$$

which describe the pure electron and ion subsystems, respectively. Then, the equation for L_{ei} takes the form

$$\begin{aligned} L_{ei} &= \mathcal{L}_e V_{ei} \mathcal{L}_i + \mathcal{L}_e V_{ei} \mathcal{L}_i V_{ie} L_{ei} \\ &= \mathcal{P}_{ei} + \mathcal{P}_{ei} V_{ei} L_{ei}, \end{aligned} \quad (14)$$

with $\mathcal{P}_{ei} = \mathcal{L}_e V_{ei} \mathcal{L}_i$. Here, the equations are written in a compact form where the variables are suppressed.

The equation for the function $L_{ei}^<$ for physical times can be obtained using the Langreth-Wilkins rules [32]. In this equation, we introduce difference and sum variables, and we account for only the terms of lowest order in the gradient expansion [31]. Finally, after Fourier transformation with respect to the difference variables we get the following expression for the electron-ion correlation function of density fluctuations [29]

$$L_{ei}^<(\mathbf{q}; \omega, t) = \frac{\mathcal{P}_{ei}^<(\mathbf{q}; \omega, t)}{|1 - \mathcal{L}_e^R(\mathbf{q}; \omega, t) V_{ei}(q) \mathcal{L}_i^R(\mathbf{q}; \omega, t) V_{ie}(q)|^2}, \quad (15)$$

where $\mathcal{P}_{ei}^< = \mathcal{L}_e^< V_{ei} \mathcal{L}_i^A + \mathcal{L}_e^R V_{ei} \mathcal{L}_i^<$ with $\mathcal{L}_a^{R/A}$ being the retarded and advanced response functions of the subsystems ($a = e, i$).

In the following, a nonequilibrium plasma state is described corresponding to a situation where the electron and ion subsystems can be considered to be in local equilibrium with the electron temperature T_e and the ion temperature T_i . The correlation functions of the subsystems, $\mathcal{L}_a^<$, can then be expressed in terms of spectral functions \mathcal{A}_a and the Bose function as follows

$$i\mathcal{L}_a^<(\mathbf{q}; \omega, t) = \mathcal{A}_a(\mathbf{q}; \omega, T_a) n_B(\omega/T_a), \quad \mathcal{A}_a(\mathbf{q}; \omega, T_e) = -2\text{Im}\mathcal{L}_a^R(\mathbf{q}; \omega, T_a), \quad (16)$$

and $n_B(\omega/T_a) = 1/[\exp(\hbar\omega/k_B T_a) - 1]$. Inserting the expression (15) with (16) into the general expression (11) for the energy transfer rate, we arrive at

$$\begin{aligned} Z_{ei}(t) &= -\mathcal{V} \int \frac{d^3q}{(2\pi\hbar)^3} \int_0^\infty \frac{d\omega}{2\pi} \hbar\omega [V_{ei}(q)]^2 \\ &\times \frac{\mathcal{A}_e(\mathbf{q}; \omega, t) \mathcal{A}_i(\mathbf{q}; \omega, t) [n_B(\omega/T_e) - n_B(\omega/T_i)]}{|1 - \mathcal{L}_e^R(\mathbf{q}; \omega, t) V_{ei}(q) \mathcal{L}_i^R(\mathbf{q}; \omega, t) V_{ie}(q)|^2}. \end{aligned} \quad (17)$$

Expression (17) has the same structure as that given by Dharma-wardana and Perrot [6] for systems with coupled modes. If the denominator is set equal to unity, which corresponds to the

approximation $L_{ei}^< = \mathcal{P}_{ei}^<$, the expression reduces to the Fermi-Golden-Rule formula valid for the regime of weak coupling between electrons and ions [29, 6]. A detailed discussion of coupled mode effects compared to the Fermi-Golden-Rule approach is given in [27].

The key quantities in the transfer rate (17) are the density response functions of the subsystems. Strong correlations in the subsystems can be accounted for by local field corrections (LFC). Using static local field corrections, the retarded density response function can be written as

$$\mathcal{L}_a^R(\mathbf{q}; \omega, t) = \frac{\Pi_a^0(\mathbf{q}; \omega, t)}{1 - V_{aa}(q) [1 - G_a(\mathbf{q}, t)] \Pi_a^0(\mathbf{q}; \omega, t)}. \quad (18)$$

Π_a^0 is the retarded polarization function in random phase approximation (RPA)

$$\Pi_a^0(\mathbf{q}; \omega, t) = \int \frac{d\mathbf{p}}{(2\pi\hbar)^3} \frac{f_a(\mathbf{p}, t) - f_a(\mathbf{p} + \mathbf{q}, t)}{\hbar\omega + \epsilon_a(\mathbf{p}) - \epsilon_a(\mathbf{p} + \mathbf{q}) + i\varepsilon}, \quad (19)$$

where f_a is the Fermi function for the subsystem of species a being in local equilibrium with temperature T_a . The local field correction factor G_a is given by [33]

$$G_a(\mathbf{q}, t) = 1 - \frac{k_B T_a}{n_a V_{aa}(q)} \left(\frac{1}{S_{aa}(\mathbf{q}, t)} - 1 \right), \quad (20)$$

with $S_{aa}(\mathbf{q}, t)$ ($a = e, i$) being the static structure factors of the electron and ion subsystems. For numerical calculations, it is sometimes convenient to use the coupled mode formula (17) in another form given in terms of the dielectric functions. For the derivation of this formula, we refer to Refs. [27, 29].

4. Energy relaxation, temperature relaxation, and quasi equation of state

In order to study temperature relaxation, a connection between temperature and internal energy needs to be introduced into Eq. (5). This can formally be done via heat capacities and leads for a two component electron ion system to two coupled differential equation for the temporal evolution of the temperatures of the species

$$\frac{\partial T_e}{\partial t} = \frac{Z_{ei}(T_e, T_i)}{\Delta} \left(\frac{\partial U_e}{\partial T_i} + \frac{\partial U_i}{\partial T_i} \right), \quad (21)$$

$$\frac{\partial T_i}{\partial t} = -\frac{Z_{ei}(T_e, T_i)}{\Delta} \left(\frac{\partial U_e}{\partial T_e} + \frac{\partial U_i}{\partial T_e} \right), \quad (22)$$

$$\Delta = \frac{\partial U_e}{\partial T_e} \frac{\partial U_i}{\partial T_i} - \frac{\partial U_e}{\partial T_i} \frac{\partial U_i}{\partial T_e}. \quad (23)$$

The internal energies of the subsystems are defined via $U_a = \langle K_a \rangle + \langle V_{aa} \rangle + 1/2 \langle V_{ab} \rangle$ with $\{a, b\} = \{e, i\}$ ($a \neq b$, $\langle V_{ab} \rangle = \langle V_{ba} \rangle$).

The internal energies for the electron and ion subsystems have to be calculated taking into account different electron and ion temperatures. This presents some challenges in the general case (see e.g. discussions in Refs. [10, 6]) and can only fully be overcome by taking one step back describing the whole process on the basis of kinetic equations.

Here, we need a theory that can handle electron degeneracy and strong ion coupling. So far, no unified approach that includes electrons and ions on the same basis is able to achieve this. For our purpose, we therefore use a perturbation expansion in the electron coupling to approximate the electron-electron internal energy contributions. This part is

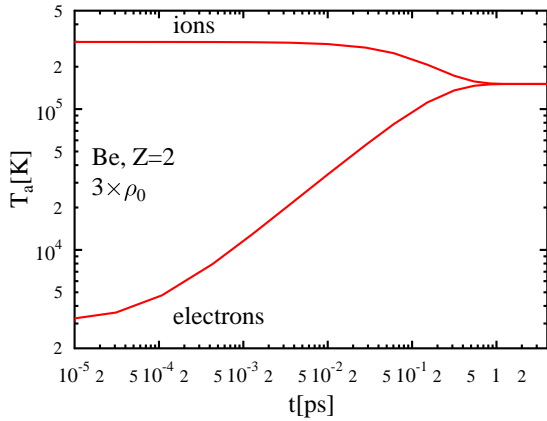


Figure 1. Nonideal temperature relaxation in a shock produced beryllium plasma at threefold solid density $\rho_0 = 1.848g/cm^3$. The ion charge state is $Z = 2$. The conditions are the same as in a recent experiment [36]. The energy transfer rate is in coupled mode approximation including local field effects for the ions. Heat capacities as described in Eqs. (4) & (25).

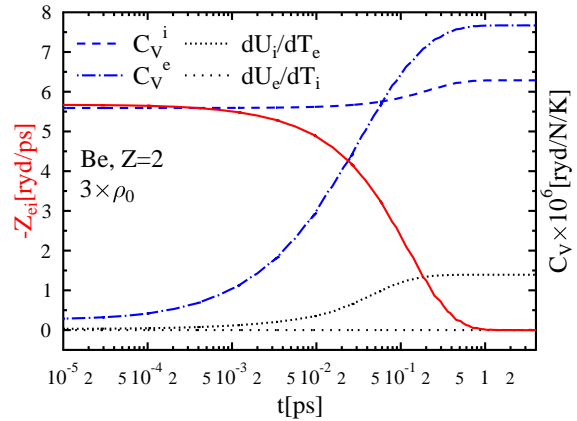


Figure 2. Energy transfer rate and heat capacities for the temperature relaxation presented in Fig. 1. The red solid line with the left ordinate gives the energy transfer rate. The heat capacities are plotted in blue (normal terms) and black (cross terms). The heat capacities include the factor of 2 from the densities: $n_e = 2 \cdot n_i$.

independent of the ion temperature and given in terms of Feynman diagrams to be [34, 35, 31]

$$U_{ee} = U_{ee}^{id} + \text{[diagrams]} \quad (24)$$

The terms considered are from left to right the Hartree, Hartree-Fock, Montroll-Ward, and normal and anomalous e^4 exchanges terms. This approximation is sufficient for most laser produced plasmas with hot electrons and also for shock produced plasmas with degenerate electrons. In both cases, the electron heat capacities resulting are very small compared to the ion heat capacities. In the first case this is due to the high coupling of the ions, in the second case due to the degeneracy of the electrons. The pure ion and electron-ion contributions can be evaluated using the effective one component plasma model [37]. The ions interact via an electronically linearly screened potential. Using the hypernetted chain approximation the internal energy can be written as

$$U_{ii}^{corr} + U_{ei} = \frac{n_i}{2} \int \frac{d\mathbf{k}}{(2\pi\hbar)^3} [\phi_{ii}^{eff}(k) - \phi_{ii}(k)] + \frac{n_i^2}{2} \int d\mathbf{r} [g_{ii}(r, T_i, T_e, n_e) - 1] \phi_{ii}^{eff}(r, T_e, n_e). \quad (25)$$

The pure Coulomb potential is denoted by ϕ , whereas ϕ^{eff} is the effective interaction. The cross term can be approximately evaluated by subtracting the contribution that arises in this equation when using a pure Coulomb potential. It turns out that writing the internal energy as in Eq. (25) means to give the electron-ion contribution the electron temperature. There exist further arguments that Eq. (25) misses terms with temperature derivatives [38]. However, for the physical systems under consideration here, we estimate that those terms are of minor importance.

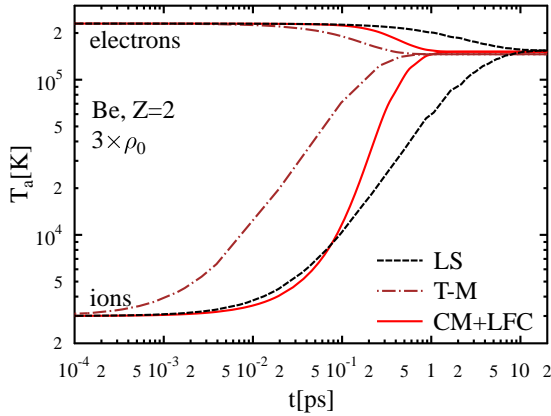


Figure 3. Nonideal temperature relaxation in a laser-heated beryllium with threefold solid density $\rho_0 = 1.848 \text{ g/cm}^3$. The ion charge state is $Z = 2$. The curve labelled LS displays a classical, ideal relaxation. Its final temperature is 154000 K. T-M uses ideal quantum relaxation and a T-Matrix energy transfer rate yielding $T(\infty) = 146000$ K. The red curve includes a coupled mode energy transfer rate and nonideal contributions to the heat capacity; the final temperature is here 151000 K.

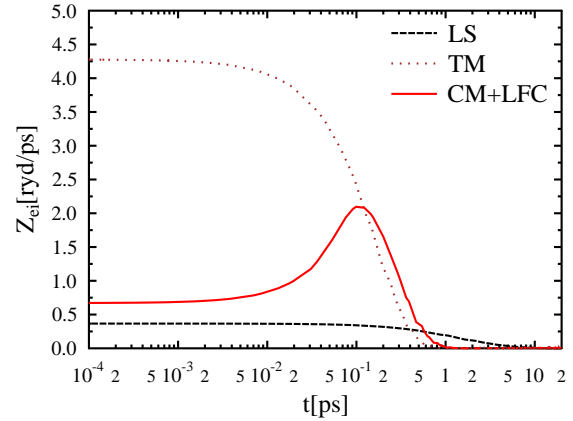


Figure 4. Energy transfer rates for the temperature relaxation presented in Fig. 3. TM and CM rates merge when the temperature difference is small so that the condition for ion acoustic modes is not fulfilled anymore ($T_i < 0.27 T_e$) [27]. For small times, CM effects lower the rate. The LS rate (with the Coulomb logarithm chosen here) is so low because the approximations made break down [27].

5. Temperature relaxation in compressed Beryllium

Beryllium is of particular interest due to its use in inertial fusion and related experiments [39, 3, 36]. We therefore study here two examples for temperature relaxation in dense compressed beryllium. Both cases are chosen with conditions as in the experiment - threefold compression, final temperature about 13 eV, charge state $Z = 2$ for the ions. Figure 1 shows the resulting equilibration from a shock produced state as in Ref. [36]. The initial temperatures are results of the nonideal relaxation under the boundary condition that the final temperature is 13 eV. This choice of initial temperatures would have resulted in a final temperature of 8.7 eV in case of an ideal classical relaxation and in a final temperature of 12.7 eV for an ideal quantum relaxation. Quantum effects for the electrons are very important as they decrease the electron heat capacity to almost zero for small times (see Fig. 2). The ideal relaxation using the quantum heat capacity for the electrons is already very close to the full result, because the correlation contributions to the heat capacities of electrons and ions partially cancel. The pure electron-electron heat capacity is always very small as electron degeneracy is very high. However, as temperature differences between ions and electrons become smaller, the electron-ion contributions grow faster than the single species terms. Since the electron-ion internal energy gets distributed evenly between electrons and ions, the electron subsystem receives considerable heat capacity to balance the massive ion-ion correlation heat capacity. Contributions to the heat capacity from $\partial U_a / \partial T_b$ ($a \neq b$) terms are small compared to the full heat capacities. The final temperature is reached after 1.7 ps. This value is sensitive to the choice of energy transfer rate. In general, coupled mode energy transfer rates predict slower relaxation than T-Matrix or Fermi Golden Rule transfer rates. Not much can be said about Landau-Spitzer energy transfer rates as their behavior strongly depends on the choice of Coulomb logarithm. For an extensive discussion see Refs. [27, 29].

An overview of the relaxation using different energy transfer rates is given in Fig. 3. In this case, we study the opposite case to the shock produced plasma of Fig. 1. The hypothetical case of Fig. 3 assumes a laser produced and compressed state of beryllium. The initial temperatures are still the same but since the electrons are hot and the ions cold on the outset, the final temperature is different (13.3 eV). The electronic heat capacity is still dominated by quantum effects, although the higher temperature increases the ideal electron heat capacity to 88% of the classical value. In Fig. 3, the T-Matrix relaxation is fastest and the Landau-Spitzer relaxation slowest. The influence of the heat capacities (correlation contributions) on the relaxation time is small compared to the influence of the energy transfer rates. In general, correlations in the heat capacities slow relaxation down and quantum effects (which lower heat capacities) speed relaxation up. The coupled mode energy transfer rates are the smallest rates up to date which can be justified quantum statistically. It was therefore predicted that temperature relaxation using coupled mode rates gives relaxation times much longer than previously obtained. However, as Figs. 3 & 4 show, the influence of the coupled mode rate is not so much about the relaxation time as about the time evolution of the relaxation. There are two important factors: Relaxation is driven by temperature difference - the bigger the temperature difference the bigger the energy transfer. In Fig. 3 for instance, one has to observe that the temperature difference in the coupled mode case after about 0.1 ps is an order of magnitude larger than in the T-Matrix case. This drives a faster relaxation. The second point to make is that the coupled mode effect ceases to exist for small temperature differences which gives additional increase to the energy transfer. The relaxation time obtained are 1 ps for T-Matrix rates, 1.7 ps for coupled mode rates, and 16 ps for the Landau-Spitzer calculation. The significance of the Landau-Spitzer result can be doubted as the classical approximation scheme breaks.

6. Summary & Outlook

Relaxation times in the order of hundreds of picoseconds as seen in experiment [11, 12] cannot be found with the theoretical methods presented in this paper. Neither the coupled mode energy transfer rate nor the inclusion of correlation and quantum effects in the heat capacities slow down the relaxation process on a necessary scale. An improvement of our theory is possible by inclusion of bound states in the energy transfer rates. It will furthermore be necessary to put the quasi equation of state theory on a firm theoretical foundation.

Acknowledgements

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