Time-resolved tuned diode laser absorption spectroscopy of pulsed plasma

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Abstract A novel method for time-resolved tuned diode laser absorption spectroscopy has been developed. In this paper, we describe in detail developed electronic module that controls time-resolution of laser absorption spectroscopy system. The TTL signal triggering plasma pulse is used for generation of two signals: the first one triggers the fine tuning of laser wavelength and second one controls time-defined signal sampling from absorption detector. The described method and electronic system enable us to investigate temporal evolution of sputtered particles in technological low-temperature plasma systems. The pulsed DC planar magnetron sputtering system has been used to verify this method. The 2" in diameter titanium target was sputtered in pure argon atmosphere. The working pressure was held at 2 Pa. All the experiments were carried out for pulse ON time fixed at 100 μs. When changing OFF time the discharge has operated between High Power Impulse Magnetron Sputtering regime and pulsed DC magnetron regime. The effect of duty cycle variation results in decrease of titanium atom density during ON time while length of OFF time elongates. We believe that observed effect is connected with higher degree of ionization of sputtered particles. As previously reported by Bohlmark et al., the measured optical emission spectra in HiPIMS systems were dominated by emission from titanium ions [1].

1 Introduction

The laser absorption spectroscopy (TDLAS) is a versatile method for analysis of particle density in liquids, gases or low-temperature plasma [2-5]. Especially in sputtering systems, the estimation of sputtered particles density in chamber volume is the crucial parameter for optimisation of deposition process. The LAS method is typically used for determination of ground state concentration of sputtered particles and their kinetic temperature from Doppler broadening of absorption line [6]. The laser beam traversing the magnetron plasma volume is absorbed by e.g. metallic atoms in the ground state at relevant resonant frequency. Consequently, the excitation of these atoms cause a drop in intensity of the passing laser beam. The sweeping of laser frequency enables to observe a absorption curve and as a result to calculate the particle density and temperature of studied atoms along the laser path in the plasma volume [6].

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During the last two decades, the pulsed DC magnetron sputtering has been becoming a very promising method for deposition of thin films with improved structure, stress, composition and adhesion. For better understanding how the plasma parameters influence the thin film growth, the extensive studies employing various time-resolved plasma diagnostics have been carried out [7]. The TDLAS method has been used for time-averaged measurements of Al sputtered particles density in pulsed DC magnetron discharge operates in non-reactive mode and in reactive mode[6]. Results revealed that the measured time-averaged Al density in pulsed DC magnetron discharge is almost by one order of magnitude higher than in DC magnetron for the same mean power delivered into the plasma.

The aim of the research presented in this paper is to develop a technique which will be able to provide the time-resolved measurement of the sputtered particles density and their temperature with help of TDLAS method.

2 Experimental set-up

A schematic configuration of low-pressure planar magnetron sputtering system used for our experimental work can be seen in figure 1. The UHV rig was continuously pumped by a turbo-molecular pump backed by combination of Roots and rotary vane pumps. After achieving a base pressure of $< 10^{-5}$ Pa, the chamber was backfilled with argon to an operating pressure of 2 Pa. The gas flow rate was set on 30 sccm of argon. The Lesker Torus planar balanced magnetron was equipped with 2" in diameter titanium target with typical purity 99.7%. The pulsed DC magnetron discharge was driven using a pulsed DC power supply. The pulsed DC supply comprises an Advanced Energy MDX-1K (U_{max}= 1.5 kV and I_{max}= 1 A) and an in-house made power switch unit based on charging of big capacitors during the idle part of the pulses. The power switch unit was capable of drawing up to

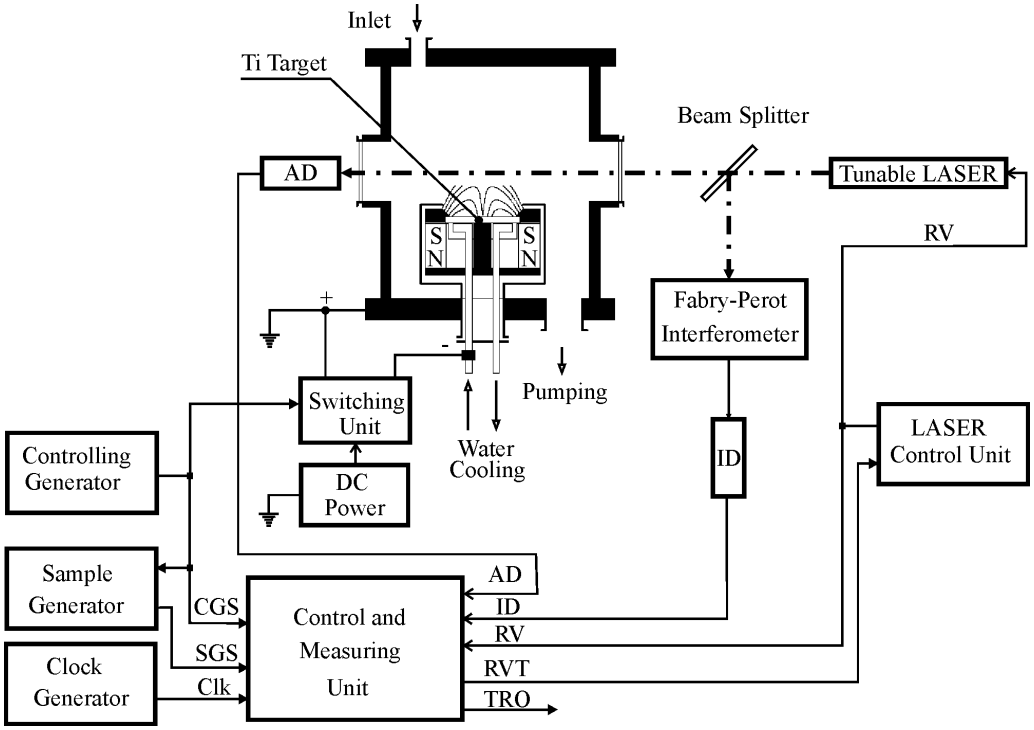

Figure 1. Experimental arrangement.

200 Amps. A ballast resistor of 5.5 Ω incorporated into the power switch unit stabilised the discharge current. Repetition frequency of pulsed discharge was held at 250 Hz (duty cycle 50%) and 500 Hz with the length of the active plasma pulse time 100 μs (duty cycle is then 5%). The mean current was set on I_D = 250 mA.

The TDLAS system used in our experiments is commercially available and consists of: Toptica™ DL100 single mode extended cavity diode laser with optical feedback into the laser diode from the first order of spectrally selective grating, Fabry-Perot etalon with free spectral range 1 GHz for frequency calibration of absorption signal, the laser control unit allowing to adjust the laser wavelength coarsely by temperature changing and finely by changing of current injected into laser diode. The sweeping of the laser wavelength was implemented by means of a piezoelectric element moving with the optical grating in the laser set-up to scan laser frequency over 10 GHz without mode hoping. In this paper, we have studied argon metastable atoms with absorption wavelength 811.53 nm and titanium atoms with absorption wavelength 398.29 nm both excited from ground state. The laser beam was traversed the processing chamber on path length 20 cm and at distance of 70 mm below the target face.

2.1 Control and measuring unit

In figure 1, the block diagram of prototype of control and measuring unit (CMU) is depicted. The CMU is the essential part of the diagnostics system and performs the time-resolved measurements.

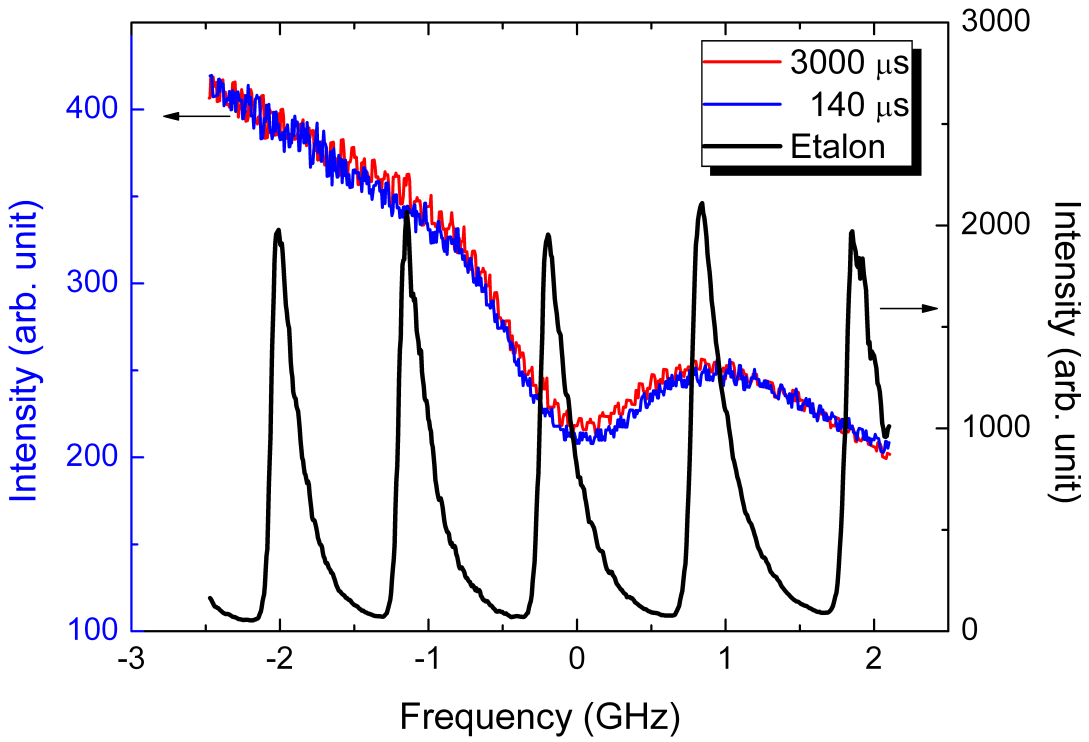

Figure 2. Typical measured time-resolved absorption peaks of Ar* with Fabry-Perot signal. Processing gas pressure 2 Pa, pulsing frequency 250 Hz, duty cycle 50%

Hardware of the control and measuring unit is driven by Clk signal of clock generator. A triggering of the commercial laser control unit Toptica® DL100 generates the time defined ramp up and ramp down of controlling voltage for tunable diode laser. The triggering signal RVT for laser control unit is derived from the signal CGS of controlling generator and signal SGS of sample generator. A special software running at the Personal Computer that support fast hardware of the unit has been developed. The CMU enables by means of controlling computer to set mode of the fast hardware control circuit, starting and stopping of measuring process and data acquisition from the A/D converters digitising signals from the absorption detector AD, Fabry-Perot interferometer ID and ramp voltage RV. Measured data may be averaged for selected number of repeated measurements and then saved into the text files at the hard disc.

The measured data transferred into the PC are evaluated by special programs for further analyzation and presentation. The detected absorption peak by detector AD, the wavelength calibrating signal from the Fabry-Perot interferometer ID and the sweeping ramp voltage RV of the laser control unit are displayed on-line during a experiment. The on-line displaying of all the signals manages the visual checking of the correct function of the apparatus. The on-line monitoring of AD, ID and RV signals are following: checking of the laser frequency stability and observation of intensity of noise of the absorption detector signal. Laser frequency hop is visualized by distorted interference peaks of calibrating signal from the Fabry-Perot interferometer detector. The intensity of the absorption detector noise is possible to suppress by averaging of several repeated measurements. The number of repeated measurements is also adjustable by controlling software. The signal TRO is optional for triggering of measurement with other processes.

3 Results and discussion

Typical measured time-resolved absorption spectra of Ar metastable atoms for for time 140 μs and 3000 μs, i.e. within the active plasma pulse and during the plasma off-time are depicted in figure 2.

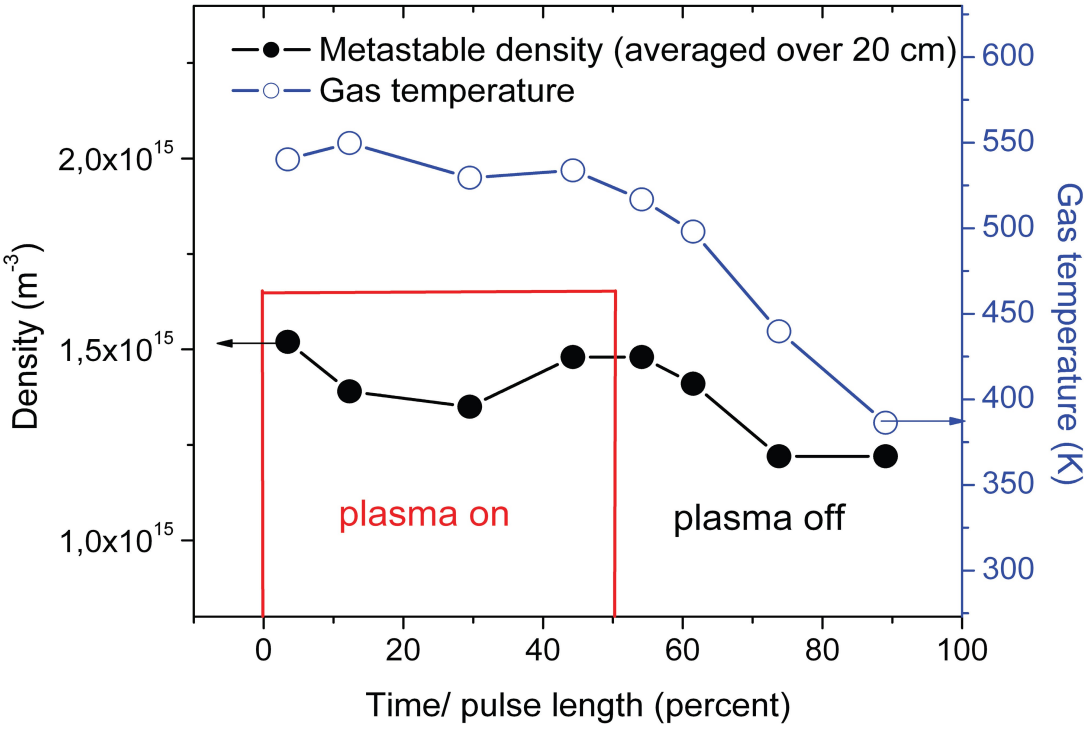

Figure 3. Evaluated time-resolved density and temperature of Ar^* under the same plasma condition as in figure 2.

The magnetron plasma was operated at pulsing frequency 250 Hz and duty cycle was set on 50%. Data demonstrate different absorption peak minimum indicating a change in the Ar^* density. The absorption spectra have been evaluated according to Lambert-Beer law of absorption and atom density and temperature have been numerically calculated according to the method published elsewhere – see [6]. The measured absorption profiles clearly demonstrate the ability of the CMU to acquire time-resolved data of the absorption signal correlated with sweeping frequency of the controlling laser unit.

Evaluated Ar metastable density during the plasma period is plot on a graph in figure 3. It can be seen that Ar^* is roughly 1.5 10^{15} m^{-3} during the active plasma pulse. The time evolution of Ar metastable density demonstrates the decrease in density during the first half of the active plasma pulse followed by the gradual increase to the initial values as at the plasma pulse beginning. During the off-

time we can observe a almost linear decrease in the Ar^* density followed by the constant value up to the end of the plasma period. We can assume that at the beginning of the off-time the number of Ar metastable is depleted by collisions in relatively dense plasma and as the plasma density decreases the long-life Ar metastable remain in the gas.

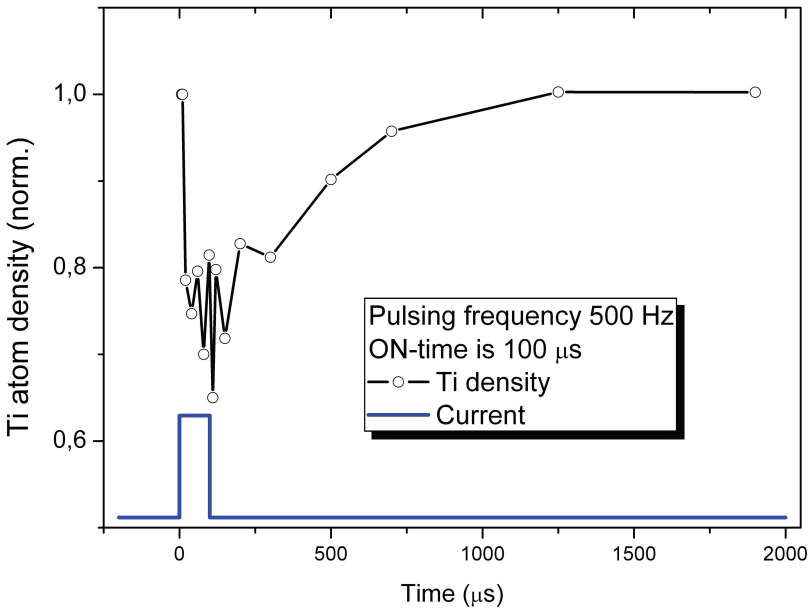

Figure 4. The time-resolved relative Ti atom density for working gas pressure 2 Pa, pulsing frequency 500 Hz and duty cycle 5%.

The Ar metastable temperature reaches 550 K during the active plasma pulse and remains practically constant during the on-time. When plasma decays, we observed the gradual decrease in the temperature almost up to room temperature. The measured time-resolved data indicate enhancement of Ar^* density and temperature when magnetron discharge operates in pulsed dc mode. It can results in higher plasma activity to produce more complex thin film.

The normalised time-resolved Ti atom density is depicted in figure 4. Because of a high level of noise in the absorption detector signal the evaluation of absorption lines was not completely available. For that reason, we plotted only the relative absorption peak height into the figure 4. When pulsing frequency decreased the signal to noise ratio was more and more smaller (not show in this paper) as the Ti atom density during the active plasma pulse decreased due to Ti atom ionisation by very dense HiPIMS plasma [1]. Nevertheless, time-resolved results clearly demonstrate a rapid drop in the Ti atom density as the plasma pulse is developing. While the data measured during the off-time indicate gradual increase in the Ti atom density up to values measured during the early stage of the active plasma pulse. We assume during the plasma afterglow the Ti ions recombine and by diffusion process travel to the chamber walls where they are deposited. We believe that our time-resolved measurements prove the massive ionisation of sputtered metallic particles when high power impulse magnetron sputtering system is used. Furthermore, the Ti ions can dwell in the chamber volume for the relatively long time after the plasma pulse was switched off and can still bombard the substrate. The relative Ti atom density enable us to estimate ion to neutral density ratio as far as we assume the sputtered Ti atom density is measured at the end of plasma period. Then up to 30% of the sputtered Ti atoms is ionised at the distance 70 mm from the target face during the plasma pulse. The previous study demonstrated by means of OES number of ionised sputtered particles up to 90% in vicinity of the target [1]. Therefore our measurements seems to be realistic.

4 Conclusions

Herein presented time-resolved measurement of Ar metastable density, temperature and Ti atom density in pulsed dc magnetron sputtering system confirmed a possibility of using the developed measuring system for time-resolved laser absorption spectroscopy. This method is applicable for diagnostics of discharges working in the pulsed dc mode with ability to observe time evolution of concentration and temperature of neutrals and many other kinds of particles. Actual disadvantage lies in limited frequency of laser sweeping.

Acknowledgements

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