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Energy distribution of ions produced by laser ablation of silver in vacuum

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ABSTRACT

The ion energy in a silver ablation plume for fluence in the range of 0.6–2.4 J cm^{-2}, typical for a pulsed laser deposition (PLD) experiment has been investigated. In this fluence range the ion fraction of the ablated particles becomes gradually dominant and can be utilized to characterize the ablation process. A silver target in vacuum was irradiated with a Nd:YAG laser at a wavelength of 355 nm and detailed measurements of the time-resolved angular distribution of plume ions were made. In contrast to earlier work, the beam spot was circular such that any flip-over effect of the plume is avoided. The angular energy distribution of ions in forward direction exceeds values of 500 eV, while at large angles the ion energy tail is below 100 eV. The maximum for the time-of-flight distributions agrees consistently with the prediction of Anisimov's model in the low fluence range, in which hydrodynamic motion prevails.

1. Introduction

Laser ablation is a versatile technique which can be used as an analytical tool for atomic emission (e.g. LIBS [1,2]), direct ion generation (e.g. LA-GDMS [3]) as well as a thin film deposition tool. Thin-film deposition using laser ablation has been widely used due to its versatility. The technique allows production of films of high crystalline quality, good uniformity and complex stoichiometry. The high-energy particles generated in the ablation process contribute to an excellent microstructure of the deposited films, but can also induce re-sputtering of the growing layers [4–6] and mixing at the interface. The positive ions are the most energetic particles in the plume [7] and thus they are most likely responsible for re-sputtering or mixing at the layer/layer interface or film/substrate interface during deposition. The ablated ions are much easier to detect than the neutrals and are thus important for studies of the plume dynamics. The ion yield dependence on fluence and the energy distribution of ions as a function of fluence are key-quantities for a better understanding of the plume expansion. We have chosen an elemental target, silver, to avoid stoichiometric effects in the plume or at the impact on the film, and all quantities were also measured in vacuum in order to avoid the complexity introduced by the interaction of the ablated particles with the ambient gas.

The current work is a continuation and refinement of our previous studies on the ablation plume from silver. In Hansen et al. [8] we studied the energy distribution of ions emitted from laser ablation of silver in vacuum at a wavelength of 355 nm. A similar investigation at 248 nm was performed by Doggett and Lunney on silver as well [9]. However, the beam spot on target was not rotationally symmetric. In contrast to [8] we have used a circular beam spot, since any deviation from a circular beam spot on target changes the lateral, angular distribution of the ablated particles, [10–12]. The time-of-flight spectra were reported earlier by Hansen et al. [13] and Toftmann and Schou [14] in which a circular beam spot was used as well. It will be shown that the peak of the TOF-distributions is at considerably lower energy for the measurements with a circular beam spot. In the present investigation we have used Langmuir probes, which are particularly well suited for angular resolved measurements. Also fast image analysis has occasionally been used for angular resolved plume studies [15].

We have investigated the angular distribution of silver ions from a low fluence (0.68 J cm^{-2}) to an intermediate fluence (2.4 J cm^{-2}). In the same fluence range the fraction of ions rises from the threshold at 0.6 J cm^{-2} up to about 0.5 at 2.4 J cm^{-2} [16]. At the lowest fluence we may consider the plume as a neutral gas with a dilute component of ions and the ions actually behave as markers of the neutral ablation flow. This case turns out to agree consistently with Anisimov's isentropic model of hydrodynamic plume expansion.

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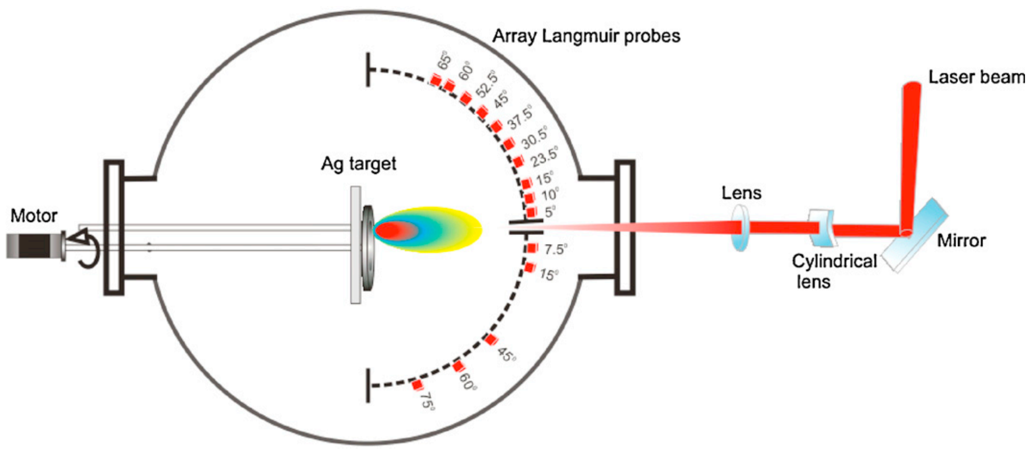

Fig. 1. Schematic of PLD ablation geometry showing a circular array of ion probes at a distance of 80 mm. Each cube on the hemisphere represents an ion probe.

2. Experimental

The experimental setup consists of a vacuum chamber with a base pressure of 10^{-5} Pa and a Nd:YAG laser ($\lambda = 355$ nm, $\tau = 6$ ns) operated at a frequency at 1 Hz, with a laser fluence varied from 0.68 J cm^{-2} to 2.4 J cm^{-2} (Fig. 1) [16,17]. The laser beam was partly focused onto a silver target using a spherical and a cylindrical lens to a circular beam spot of an area of 0.04 cm^2. The angular resolved time-of-flight (TOF) spectra of the ablated ions were measured with an array of 13 ion probes mounted at a distance of 80 mm from the ablation point on the target and at angles varying from 5° to 75° with respect to the normal (Fig. 1). Each ion probe consisted of a tip of a tin-soldered copper wire with a projected area of 0.033 cm^2 at the end of an insulated cable. The active probe was biased at a saturation value of – 40 V in order to attract ions and repel electrons, while all other probes were grounded during the signal collection. The normal incidence of the laser beam on the target provides a rotational symmetry and enables us to perform measurements of the signal ions over the full hemispherical array of ion probes. In this geometry the total number of silver ions at each fluence can be determined [14,16].

3. Results and discussion

As already reported in Ref. [14] the ion signal increases strongly with fluence and the angular distribution of the plume becomes increasingly narrow with increasing fluence. The peak of the time-of-flight (TOF) distributions, corresponding to the arrival of the maximum ion flux to the probe at 5°, was calculated from the ion probe current at different fluences and it is shown in Fig. 2a. The peak of the ion TOF-distribution moves toward shorter time of flight with increasing fluence, while the corresponding kinetic energy increases with fluence (Fig. 2b). The kinetic energy of ions close to normal (probe at 5°) increases from 74 eV at a fluence of 0.68 J cm^{-2} to 150 eV at a fluence of 2.4 J cm^{-2}.

The energy distribution of silver ions, $N(E)$, for the free expansion in vacuum was calculated from the TOF-signals using the relation $N(E) = (I(t) \times t^3)/(Ze \times m \times d^2)$, where $I(t)$ is the current of ions with charge Ze, m is the ionic mass and d is the distance to the probe. The ion energy spectra for two distinct angles of 5° and 45° and for a wide range of laser fluences are shown in Fig. 3a and b, respectively. (The underlying TOF-distributions are shown in Ref. [14].) The decrease of the number of particles with increasing angle is evident. The characteristics of the ion energy distribution can be described as follows: (i) the energy tail of ions close to the normal extends up to 500 eV and beyond and (ii) at large angles the ion energy does not exceed values of 100 eV (Fig. 3b). Another remarkable feature is that the ion spectra at 5° exhibit a strong fall-off which can be approximated by a tail proportional to $E^{-7.5}$ for the highest energies for all values of the fluence studied here. At 45°

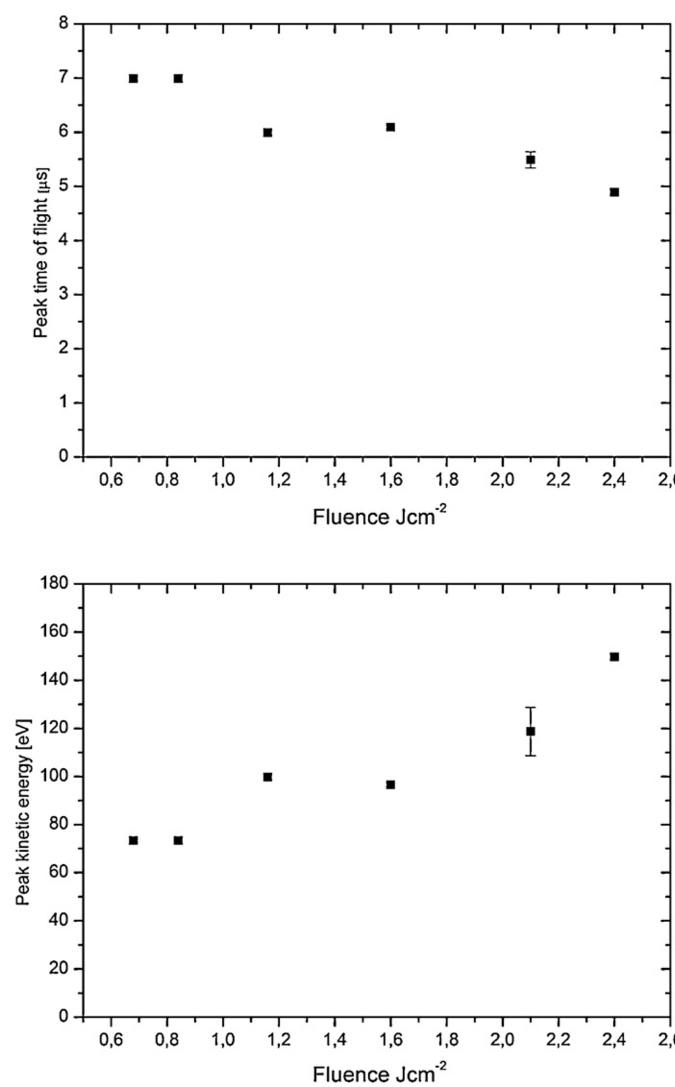

Fig. 2. (a) The "peak" of time-of-flight (TOF) distributions (b) and kinetic energy of the ions at the peak time close to normal (probe at 5°) as a function of laser fluence. Typical error bars have been indicated.