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Direction-Selective Free Expansion of Laser-Plasmas From Planar Targets

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Direction-selective expansion of laser-plasmas from planar, slab targets of Al, Ni, Mo and Ta are reported. Angular distributions of the particles emitted from the targets, produced by a 130mJ-5nsec, Nd:YAG laser, were obtained by means of a retarding potential analyzer and a quartz crystal. It was observed that the angular distribution of the particles was preferentially focussed towards the target normal with the increase in the focal spot size, atomic mass of the target material and the ionization state of the expanding particles, for a given value of the laser energy. Our experimental results confirm, for the first time, the Monte Carlo simulation results of the earlier workers, taking into account collision and recombination processes.

I. Introduction

In the field of material preparation such as fabrication of thin films of high T_c superconductors, oxides, semiconductors and diamond-like carbon laser pulsed-deposition technique (LPD) has been found to be interesting and useful [1-3]. The partition of energy as well as the angular distribution of the emitted materials have a great influence on the quality of the deposited layers [4]. Experimental investigations of Mueller and Rohr [5] demonstrated that the free-expansion of laser-produced plasmas from planar targets is directed essentially towards the normal to the target surface. The causes for this direction-selective behaviour of the freely expanding plasma are not well known. Itina et al [6] observed that the non-equilibrium chemical reactions in the gas phase during ablation affected the angular distributions in the way opposite to elastic collisions. Itina et al [7], in their subsequent works, presented the results of a Monte Carlo simulation of the laser desorption-process. They concluded that the chemical process, the recombination-dissociation one, gave rise to the broadening of the angular distribution and the collisions were found to be responsible for the focussing effect.

In the present work we have investigated the direction-selective free-expansion of laser-produced plasmas from the slab targets of Al, Ni, Mo and Ta using a 130mJ-5nsec, Nd:YAG laser. We report the experimental observation of the following results. For a given laser energy and the target material the angular distribution showed more preferential focusing towards the target normal as the values of the focal spot size B increased. For the given laser-energy and the given focal spot diameter, the focusing toward the target normal was more pronounced as the value of the atomic mass number increased. For a given laser-energy, a given focal spot diameter and a given target element, the angular distribution showed more preferential focusing as the ionization state of the emitted particles increased. The results are well explained on the basis of the estimates of recombination and collision rates.

II. Experiment

The plasma is created by an Nd:YAG laser ($E_L = 130\text{mJ}$, $\tau = 5\text{nsec}$ and $\lambda = 1.06\mu\text{m}$) incident on planar, slab targets at a fixed angle of -45° with respect to the target normal [5]. Target materials consisted of Al, Ni, Mo and Ta. The particles of the freely expanding plasma were detected in an angular range relative to the target normal between 50° to -10° for ions and 80° to -15° for the total number of particles by moving the analyzers within the plane of incidence, at a distance of 35° from the target. Positive sign before the angle signifies the analyzer being away from the direction of incident laser with reference to target normal. The ion-spectra were fully resolved by the time-of-flight/retarding-potential method, which made it possible to obtain the absolute number of each species. The second detector consisted of an rf-excited quartz crystal. From the frequency change of the crystal after the plasma has been deposited, one obtains the total mass or, in the case of monoatomic beams, the total number of particles. The number of the neutral particles could be deduced from the difference between the total particle signal and the summed spectra of the different ion-species. Full experimental details are given in the works of Mueller and Rohr [5].

III. Results

Fig.1 typically displays the integrally normalized angular distributions of the total number of particles of Ni as a function of the angle θ for three sizes of the spot size B , where θ is the angle between the direction of observation and that of the target normal. We note that although all the distributions are focussed towards the direction of the target normal, the focussing effect increases as the focal spot size increases. The combined result from Al, Ni, Mo and Ta were observed and found that the focussing significantly increases with increase in atomic mass number. For two focal sizes $B = 8.34\text{mm}^2$ and 0.61m^2 ,

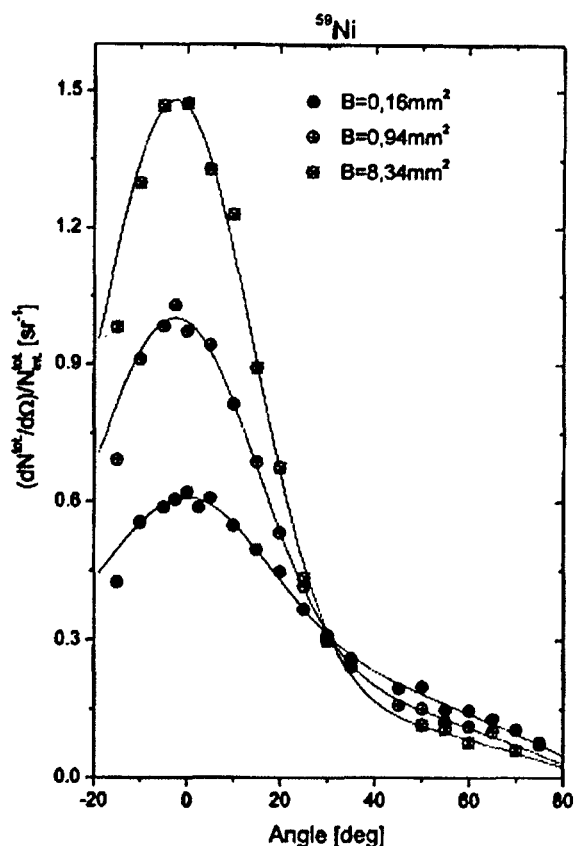


Fig.1 A typical distribution of normalized integrated emissions from Ni for three focal spot sizes

the experiments were repeated on the four materials and the angular distributions of neutral as well as upto three times ionized particles were obtained. The results showed that the particles with higher ionization states were much more focussed towards the target normal. Approximate calculations of the recombination rates and the particle collision frequencies were performed. It was seen that when recombination rate dominated, the focussing was less pronounced and when particle collision frequencies dominated, the focussing effect was sharper, in agreement with the results of Monte Carlo simulations of Itina et al [6,7] and Sibold and Urbassek [2].

Experimentally obtained maximum ionization states for Al, Ni, Mo and Ta were observed to be approximately as 2.4, 1.9, 1.5 and 1.3 respectively. It should be noted that these values represent the average values of the ionization states after the recombination, in the core of the plasma, has taken place. The estimated values of the average ionization states from Mosher's calculations [8] are approximately 11, 16, 17 and 22. This means that a strong recombination has taken place during the time-interval of the laser pulse duration. But this is not enough. One has to know which process,

collisional or recombination is dominating, which seems, at this stage, a difficult question. When the focal spot size varies from $B=0.16\text{mm}^2$ to $B=8.34\text{mm}^2$, Te decreases from 85 to 75 eV i.e. by a factor of 1.2 and the average decrease in the average ionization states, for all the four elements is also around a factor of 1.17. That is to say, the fractional decrease in Te and Z is nearly the same. From a simple calculation one can see that, because of this change in the plasma parameters three-body recombination rate decreases by a factor of approximately 1.3. The electron-ion collision frequency ν_{ei} also increases by a factor of about 1.1. Hence, these two processes nearly neutralize each other. But when we consider the radiative recombination rate and the electron-electron collision frequency ν_{ee} we find encouraging results. For the same amount of variation in the plasma parameters the radiative recombination rate decreases by a factor of 1.31 and, at the same time, the electron-electron collision frequency also increases by a factor of 1.31. As a result, for larger values of the focal spot size, the focusing toward the target normal increases. These experimental results support the simulation results of Itina et al [6,7].

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