MONTE CARLO STUDIES ON EVOLUTION OF PRODUCTS FROM LASER-INDUCED CARBON, ALUMINA, AND BARIUM TITANATE PLASMAS

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ABSTRACT: Evolution of pulsed laser ablation products from carbon, alumina, and barium titanate targets is studied using a Monte-Carlo method and assuming elastic collisions amongst the ablated species. In the calculated time of arrival profiles of the ablated species the peak times of arrival closely follow square root mass dependence as observed in mass spectrometric experiments.

I. INTRODUCTION

Pulsed laser deposition is now a frontier technique for making thin films. However, despite its increasing applications, a satisfactory theoretical understanding of the evolution process of the species is far behind the technology because of the complexities of the processes involved. A full theoretical description needs to include the energy transfer to the target material by the laser, composition and thermodynamic state of the plume generated, and collisional processes the constituent particles undergo before they reach the substrate surface where the deposition takes place. The potential returns from a successful model are attractive; it would predict the nature of deposits for a wide variety of laser ablation conditions including cases where non-inert gases are present in the ablation chamber.

We report here the results of a simulation of the evolution process of the species by a Monte Carlo method starting with a known surface temperature and initial plume pressure, and considering only elastic collisions. In an earlier report [1] we presented results on alumina and carbon (graphite). Here we report certain augmentations in the algorithm and additionally present results on barium titanate. We compare the results of the calculation with the mass spectrometric results obtained in some pulsed laser ablation experiments [2-4] on carbon, barium titanate, and alumina targets.

II. METHOD OF CALCULATION

The surface temperature of the substrate and the initial pressure of the plume near the substrate are arrived at from mass spectrometric data of relative concentration of species and relevant calibration factors. From these temperature and pressure the thermal equilibrium temperature and pressure are obtained

using standard gasdynamic relations applicable to expansion through an orifice [3] with the condition of Mach number $M = 1$. The particles are assigned their initial velocities according to Maxwell-Boltzmann's distribution at the the thermal equilibrium temperature. The angular distribution of the velocities is obtained by modelling emission of the particles as a desorption process, so that θ, the polar angle of the velocity with respect to the flow direction, has the following distribution [5]:

$$f(\theta) \propto \cos(\theta) \tag{1}$$

Their initial positions are assigned uniformly randomly inside a small volume which we call the source region.

In the model, an array of multiply structured elements stores the position, velocity, and species type information of all the particles. Each element of the array contains all the relevant information corresponding to a particular particle, updated at every time step. The ablation chamber is partitioned by a 3-dimensional grid into small parallelopipeds in which interparticle collisions are assumed to occur.

The source generates particles of various species, at regular intervals for a duration of 10–30 ns. Parallelly, particles are allowed to collide elastically and evolve. For generation of a particle a species type is selected randomly depending upon the initial relative concentrations. The number of particles for which the calculation is performed is very small compared to the actual number of particles. The number-dependent quantities such as pressure, bonding probabilities etc. are multiplied by the factor by which the actual number of particles exceeds the assumed number of particles.

For simulating the collisions the small boxes of the partitioned chamber are scanned. If there are two or more than two particles in a box then two of them are selected in such a manner that they are most likely to be the high mass species. Three (or more) particle collisions are ignored.

If particles reach within a certain preset proximity of the walls, they are allowed to collide and form chemical bond, stick with the wall, or reflect away from the wall depending upon their relative probabilities.

The particles are counted in a counting region at the height of 0.479 m from the base of the chamber (the height at which the entrance of the mass spectrometer is situated in the experimental setup). With the aid of microscopic collisions the ablated plume particles reach this region and from here onwards they are assumed to escape away. From the plot of the counted number of particles vs time the peak times of arrival (ToA) are obtained.

III. RESULTS

A typical calculated ToA plot of species evolving from alumina target is shown in Fig. 1. Results on the dependence of peak time of arrival on the square root of mass of the species ablated from barium titanate target, both from experiment and calculations, are shown in Fig. 2. A computed spatial mapping of the plume from the barium titanate target is shown in Fig. 3. The proxim-

ity of the calculated peak ToAs with the mass spectrometric results indicates validity of the model.

ACKNOWLEDGEMENT

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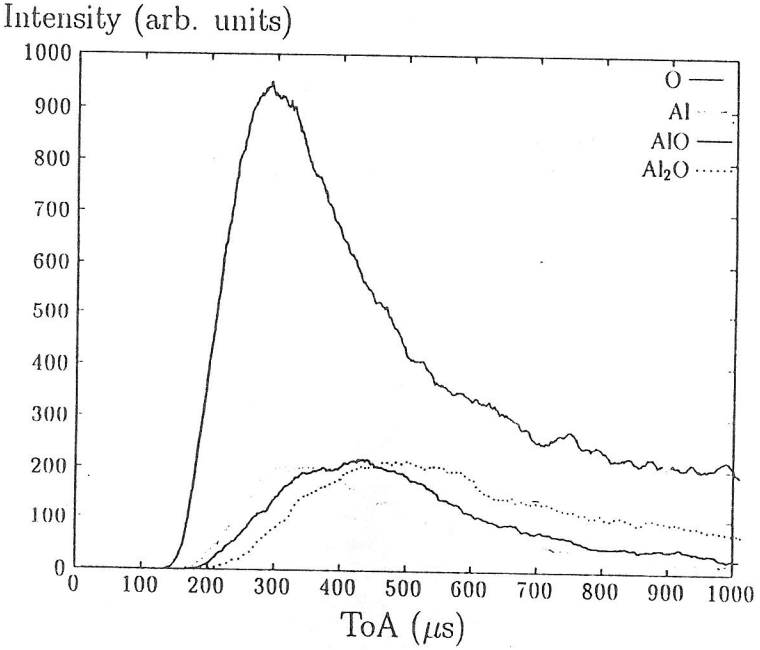

Figure 1: Time of arrival plots of O, Al, AlO, and Al_2O from alumina target.

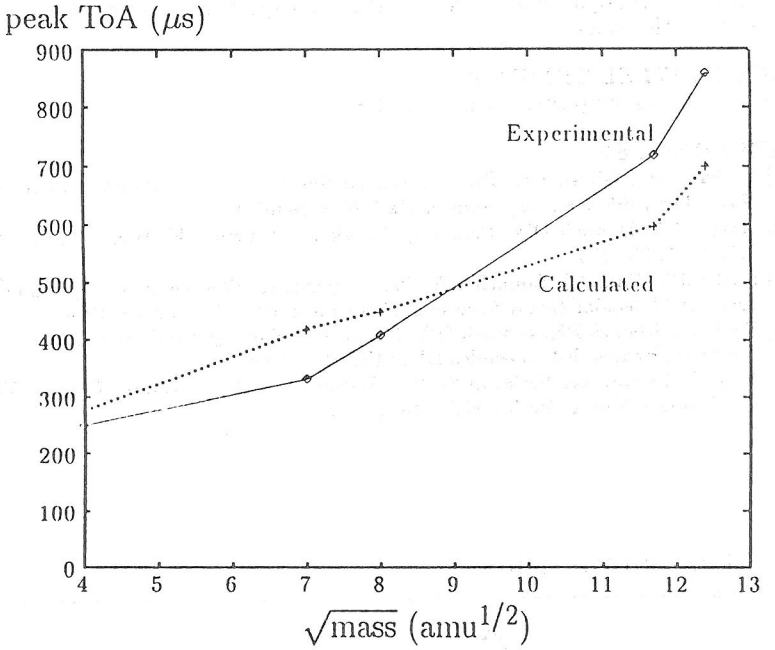

Figure 2. Plot of peak time of arrival vs $\sqrt{\text{mass}}$ of species ablated from barium titanate target (O, Ti, TiO, Ba, and BaO).

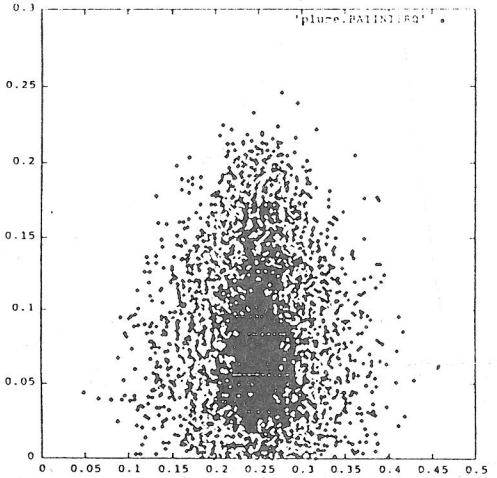

Figure 3. Computed spatial distribution of the plume from barium titanate target, after a time of 80 μs.