

Modelling of surface evaporation by laser ablation

R. Mendes Ribeiro ^{a,*}, Marta M.D. Ramos ^a, A.M. Stoneham ^b, J.M. Correia Pires ^a

^a *Departamento Física, Universidade do Minho, Largo do Pazo, 4709 Braga Codex, Portugal*
^b *Physics Department, University College London, Gower Street, London WC1E 6BT, UK*

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Abstract

This paper presents some preliminary results on simulations of the absorption of high fluence laser radiation by a target surface. We show the role of absorbing defects and their concentration as well as the influence on target roughness of places that absorb radiation more strongly than others. We then develop a mesoscopic model, and find that the grain boundary width (including any atmosphere of defects or space charge region) is an important parameter.

Keywords: Laser ablation; Surface; Simulation

1. Introduction

Pulsed laser ablation has proved to be a reliable technique for thin film deposition [1]. It has become popular because it makes possible the congruent evaporation of very complex materials, like multi-component oxides. Among other advantages, it is easy and cheap to implement laser ablation experimentally [2]. Nevertheless, the effect of a high laser fluence (2–10 J/cm², for 5 to 30 ns) on a material is not easily predictable, and in fact, the experimentalist has to make hundreds of experiments in order to achieve a good result. This is time and money consuming. It would be very good to save most of this work by a proper modelling of the process, which could reduce significantly the range of experimental parameters to vary.

Ideally, this model should be such that one would

only have to give some parameters of the material (preferentially those easy to obtain experimentally or tabulated; at least ones that you could make a reasonable estimation), and run a program for a range of laser parameters such as the fluence, the wavelength and the duration of the laser beam.

The main material properties that influence the interaction of a laser beam with a material, in a simplistic view, are the absorption coefficient and the target roughness. Of course, the problem is that both vary as the laser impinges on the material. This implies that the model has to include this change in time. If the changes cannot be given as a parameter, which is usually the case, the model has to calculate them as a function of time. Let us consider the optical absorption. There are three main types of absorption: band transition, defect excitation and nearly free carrier absorption [3]. It is important to know the role of each of these processes in a particular material. This is measured by the cross section and the density of absorption centres. So the model should keep track of these. The mechanisms for the

* Corresponding author. Tel.: +351-53-604335; Telex: 32135 RUTMIN P; fax: +351-53-678981; e-mail: ribeiro@ci.uminho.pt.

generation and annihilation or evaporation of these absorption centres are the ones that actually define the model and its simplicity or complexity.

Band to band absorption is only possible when considering photon energies greater than the band gap, unless you take account of the two or more photon absorption. Two photon absorption is a second order mechanism which has to be taken into account if the other absorption processes have low probability to happen. Of course, this can be a plausible mechanism because of the high fluences involved.

Defect excitation seems to be very important for laser ablation of sub-band gap materials. Actually, it is observed that there is an emission of particles under the laser ablation threshold fluence for dielectric breakdown of sub-band gap materials [4]. Defects can be of various types and can have several possible charge states, like interstitials, vacancies, grain boundaries and steps at the surface. They may absorb light and be generated by the heat and tensions produced during the laser pulse. They also may cause carrier capture or generation.

The free carriers can absorb light very strongly if their density becomes greater than the plasma frequency. If the material is metallic, this is the initial situation. If it is an insulator, carriers have to be generated in a sufficient amount in order to start absorption. This introduces a delay in carrier absorption which may not happen at all if the generation rate is not sufficiently high.

The roughness of the target seems to be a puzzling problem: in fact, sometimes laser ablation is used to smooth a surface [5], and other times it generates a very inconvenient rough surface, with cone formation, that makes the target unusable for thin film deposition [2,6].

The models we are developing try to be a tool in the study of laser ablation phenomena, in order to help the user to rapidly select control parameters which are reasonable, and which allow direct further experimental optimisation.

2. The model: a mesoscopic model with point defects

The model we present in this paper is for a material with a band gap much greater than the

photon energy, and so we assume that there is no band transition. The absorption is made by the defects in the crystal which are characterised by an ionisation energy and a cross section. The initial density of nearly free electrons (electrons in the conduction band) is zero, and these carriers are generated by the ionisation of defects. Defects are assumed to be static, leaving a positive charged background as they ionise. A work function is assumed for the evaporation of the electrons, and an evaporation energy is also assumed for the crystal atoms (defects evaporate at the same rate as the atoms).

The processes in this model occur as follows: a defect is ionised by the absorption of a photon becoming positively charged and giving an electron to the conduction band. This electron may recombine with another ionised defect, giving in this process all its energy (both potential and kinetic energy) to the lattice. This is the only way the lattice increases its energy in this model. The electrons in the conduction band may also absorb the incident radiation, if they have enough density, and drift through the sample, recombining in a place different from the one where they were created. The atoms and electrons may evaporate due to the energy increase of the lattice and of the electrons, respectively, and in an independent way.

We present the results of the model assuming a matrix of $5 \times 5 \times 3$ parallelograms (Fig. 1), with $50 \text{ nm} \times 50 \text{ nm} \times 50 \text{ nm}$ except for the first layer which is much thinner than the others (it only has 0.5 nm height). This is because we are simulating a surface, and so we want to have a greater concentration of

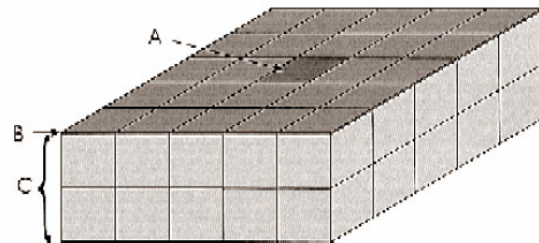


Fig. 1. The elements used in the model: (A) this element and the ones below it belong to a grain boundary; (B) these elements belong to the surface of the sample and they are much thinner than the others; (C) these are the elements that represent the bulk of the material.

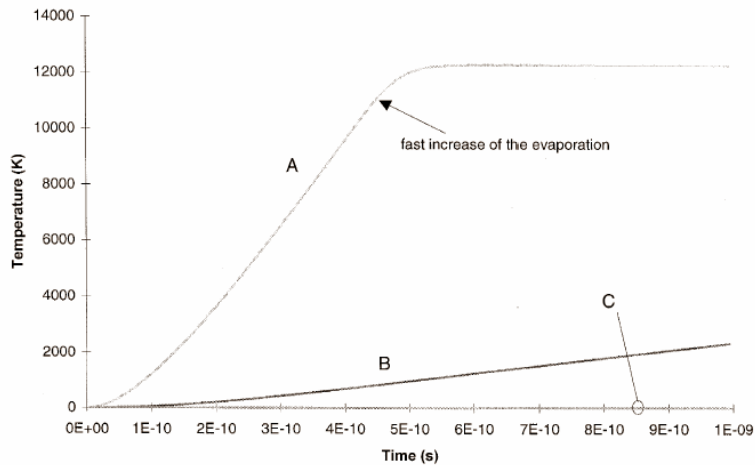


Fig. 2. The variation in time of the temperature of the elements. The letters correspond to different positions of the material as shown in Fig. 1.

defects there. At the surface, it is assumed that all the atoms can be ionised (all of them are considered to be a defect because of its lower coordination). The parallelograms in the middle of the matrix are assumed to belong to a grain boundary and so the density of defects there is 1% the density of atoms. In the bulk, the density of absorbing defects is considered as 0.01% the density of atoms.

The energy assumed for an atom to evaporate is 19.1×10^{-19} J, the photon energy is 5 eV and the laser fluence is 2 J/cm^2 . The absorption cross-section of the defects is 10^{-21} m^2 and the density of atoms is $5.35 \times 10^{28} \text{ m}^{-3}$. Other parameters are taken from values close to the ones of MgO.

Fig. 2 shows the evolution of the temperature of the elements as a function of time, for the first nanosecond. The temperature of the first layer of the grain boundary increases very fast and starts evaporation before any other part of the surface. After

about 0.45 ns the evaporation of this element increases significantly and its temperature stops increasing, since it loses energy as the atoms are evaporated. The rest of the surface also heats faster than the bulk. The grain boundary in the bulk does not heat so fast, although its temperature is always higher than the rest of the material.

So the results from the model above show that evaporation starts in places of high concentration of defects, like grain boundaries. Using this assumption, we developed a mesoscopic model to see what happens to the surface of a target with grain boundaries, assuming a faster evaporation of these parts of the target.

In this other model, the laser is incident from a direction that makes an angle of 45° with the vertical and the evaporation is assumed to be proportional, on each point, to the energy deposited per unit area.

At the beginning, the target surface is plane, but

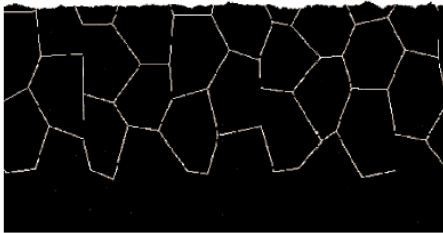


Fig. 3. Surface of the target: thin grain boundaries.

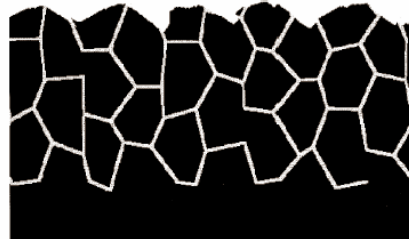


Fig. 4. Surface of the target: intermediate grain boundaries.

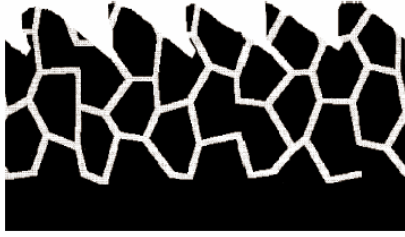


Fig. 5. Surface of the target: thick grain boundaries.

as it evaporates some roughness shows up as a consequence of the faster evaporation at the grain boundaries. This roughness continues increasing until a steady state is reached, in which the surface of the target has cones pointing in the direction of the incident laser beam. This is observed in laser ablation experiments [2,5].

Figs. 3–5 show the effects of the laser on three simulated targets with different grain boundary widths, after the same evaporation time. By “grain boundary width” we mean the width including any space charge region or any atmosphere of point defects associated with the grain boundary. The target with the wider boundaries develops cones much faster than the others. The target with the thinner boundaries will never develop cones, but will keep the little roughness it shows. A simulation with a target without grain boundaries but with an initial surface roughness of the size of the grains shown was performed. The result was a smoothening of the surface to about half the initial roughness and without any preferred orientation.

3. Conclusion

A proper modelling of the laser ablation process is useful for making decisions on the nature of the target used for evaporation. The role of defects in the absorption of sub-band gap radiation was studied. Their high concentration in places like grain boundaries provokes a preferred evaporation in these places of the target. These preferred evaporation seems to be the origin of the observed cone formation at the surface of the targets after several laser pulses during laser ablation. Thinner grain boundaries seem to delay or even inhibit the cone formation.

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