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Picosecond pulsed laser ablation of silicon: a molecular-dynamics study

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Abstract

We report here the continuation of our earlier work on the molecular-dynamics simulations of laser ablation of silicon with picosecond laser pulses. A more realistic phenomenon of ablation is observed as, along with a significant expansion of the time scale, carrier diffusion is explicitly taken into account. The motion of approximately 32,000 atoms, contained in a 5 nm × 5 nm × 27 nm surface rectangular box irradiated by a single 308 nm, 10 ps, Gaussian laser pulse, is followed for typically 100 ps of simulation time. Because melting and possibly ablation or desorption of the target following absorption of the laser pulse are described within the thermal annealing model, care is taken not to exceed carrier densities of $\sim 10^{22}$ cm^{-3}. More precisely, the interaction of photons with the target is thought to cause the generation of a dense gas of hot electrons and holes which primarily relaxes through carrier-phonon scattering. Above a characteristic threshold energy of ~ 0.30 J/cm^2, ejection from the target of big chunks of molten material occurs and the latter are expelled with axial velocities of ~ 1000 m/s.

Keywords: Laser ablation; Thermal annealing model; Molecular dynamics; Plume; Ablation threshold; Silicon

1. Introduction

The use of light as a source of energy to cause matter disruption from the surface of a given material has found several important applications in the recent years. More specifically, the development of lasers has led to technological processes involving the controlled removal of matter, a phenomenon known as *laser ablation* [1,2]. However, a detailed comprehension of the various mechanisms underlying laser ablation is still lacking. One computational approach to investigate the properties of various systems on a microscopic scale is molecular dynamics (MD), which allow one to follow, in real time, the motion of atoms when the interatomic forces are known. Previous attempts to model laser ablation through MD simulations are scarce [3–5]. Among them, Zhigilei et al. have successfully applied their breathing-sphere model [3] to the study of laser ablation of organic solids.

In this paper, we present a continuation of our earlier work [5] on the MD simulations of laser ablation of silicon with picosecond laser pulses. More precisely, the recent progress achieved in expanding the time scale allows us to account for the various phenomena in a more realistic manner as ablation is

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explicitly observed with carrier diffusion embedded into the model.

2. The model

We present, in this section, an overview of the model for which a detailed description can be found elsewhere [5]. The simulation box, containing a total of 32,400 atoms, forms a (1 0 0) surface slab with dimensions of 5 nm × 5 nm × 27 nm, even though smaller systems with approximately 20,000 atoms and dimensions of 3 nm × 3 nm × 54 nm have been used. Periodic boundary conditions are imposed in the x and y directions, that is to say parallel to the upper surface. Along the z-axis, however, thermalization is ensured by coupling the system to a heat reservoir.

A 10 ps, 308 nm, laser pulse Gaussian in time, but uniform in space, is simulated by a succession of planes spanning the entire top surface of the supercell, each carrying a well-defined number of photons proportional to the instantaneous irradiance, and separated in time by an interval ranging from Δt to typically $10 \times \Delta t$, where $\Delta t = 0.5$ fs is the value of the MD timestep. At $\lambda = 308$ nm and 300 K, the one-photon interband transition is dominant and other absorption mechanisms can therefore be ignored [5]. The absorption of light is described by the Beer–Lambert law. Following the absorption of a photon, an electron-hole pair is created and is assigned an initial position to allow for subsequent diffusion into the bulk to occur as a result of the carrier density gradient at the surface. The initial kinetic energy of each carrier is then determined according to an instantaneous Maxwell–Boltzmann (MB) distribution at a temperature T_e.

A critical value $n_c \sim 10^{22}$ cm^{-3} for the carrier density n is believed [6] to be separating two distinct regimes and for lower carrier concentrations, *thermal processes only* are operative and described by the thermal annealing model (TAM) [7]. Because the interatomic potential used, the Stillinger–Weber potential [8], is believed to be only suitable for thermal processes, pulses have a minimal duration of 10 ps [7] and fluences are chosen so as not to lead to carrier densities exceeding 10^{22} cm^{-3}. The main relaxation mechanism taken into account in the present model is that of carrier-phonon scattering. The emission of a phonon is carried out by distributing instantaneously an energy $\hbar\omega \sim 62$ eV in a radius of 5 Å of the carrier according to a spatial Gaussian distribution. The other important relaxation mechanisms, namely impact ionization and Auger recombination, can be safely ignored [5]. Finally, carrier diffusion is assumed to occur along the z-axis, that is in the direction of the carrier density gradient, the carrier ambipolar diffusion coefficient being computed from the equations suggested by Berz et al. [9].

3. Results and discussion

All simulations were run for a single 10 ps, 308 nm, laser pulse interacting with a (1 0 0) silicon surface. Fig. 1 shows the value of the surface temperature as a function of time (in ps), obtained for various fluences, below and at the predicted threshold of 0.30 J/cm^2 for ablation. The laser pulse starts at $t = 0$ ps and the evolution of the system is followed for typically 100 ps of simulation time. If the melting fluence, F_m, is defined as the energy required to bring the surface temperature to $T_m = 1685$ K, the predicted value for the melting fluence can, upon inspection of Fig. 1, be safely estimated to lie between 0.10 and 0.15 J/cm^2, in a relative good agreement with experiment [10,11]. Additionally, it can be seen that, as the fluence is increased, the predicted temperature at surface tends toward an asymptotic value of ~2000 K, a value consistent with typical experimental data [10,11].

The minimum fluence for which ablation occurs, that is the ablation threshold energy F_{th}, is 0.30 J/cm^2, a value typically obtained in experiment [10]. In our previous work [5], it had been mentioned that carrier diffusion could be suppressed in order to recreate artificial conditions where ablation occurred within about 5 ps. The observed temperatures were then very high, that is between 20,000 to 40,000 K, resulting in a plume mainly composed of single atoms. However, a recent expansion of the time scale has allowed us to observe ablation in normal conditions, where the carriers were given the possibility to diffuse into the bulk. When doing so, ablation occurs after ~30 ps. However, the composition of the plume is totally different: instead of single atoms, matter is expelled through the ejection of big chunks of molten

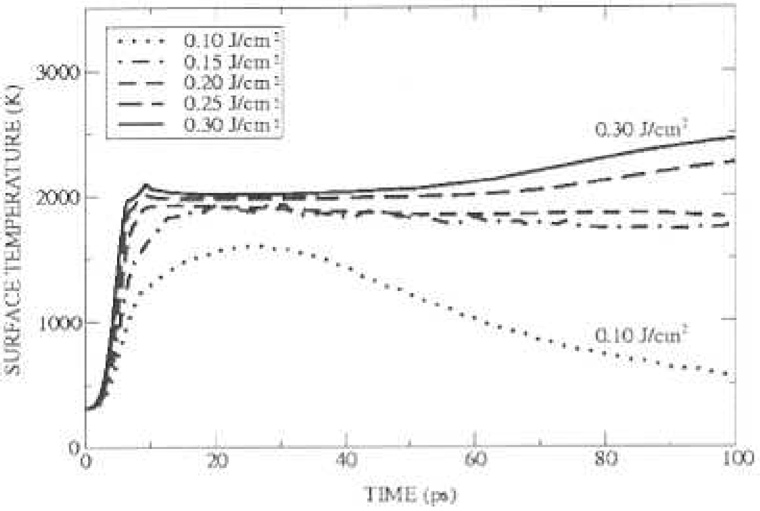

Fig. 1. Surface temperature as a function of time for various laser fluences.

material. This observation can be explained by the fact that the temperature corresponds to only about one tenth of the cohesive energy of silicon (~4.65 eV) and is therefore insufficient, on average, to break all the bonds of the four-coordinated silicon atoms.

Finally, Fig. 2 gives the average axial (perpendicular to the surface) and radial (within the plane of the surface) components of the velocities of the ejected atoms as a function of laser fluence.

One can see that increasing the laser fluence results essentially in an increase of the *axial* component of the velocity, that is, raising the fluence causes the additional energy to be mainly converted into translational kinetic energy in the direction *perpendicular* to the surface. The predicted velocities are observed to have a typical value of ~1000 m/s.

Full details of the calculations and complete results will be presented elsewhere [12].

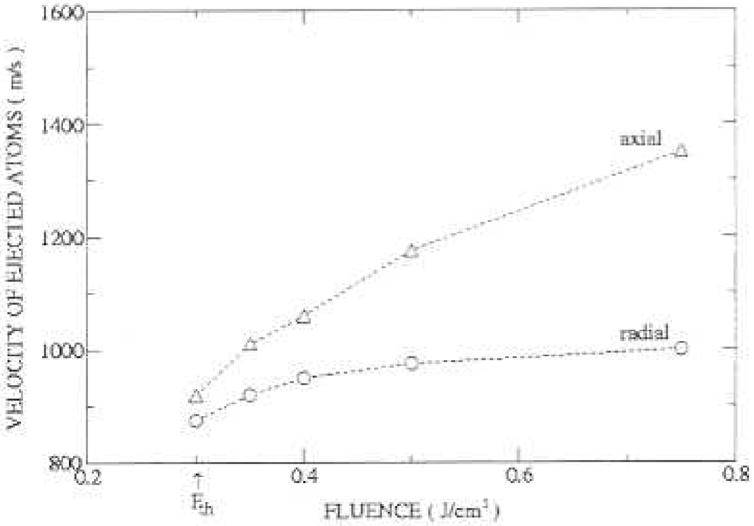

Fig. 2. Average velocity of the ejected atoms as a function of laser fluence.

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