Formation of nanostructures at metal surface exposed to femtosecond laser pulses

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Abstract

Ultrafast laser irradiation of free-standing gold film was simulated using molecular dynamics. The spatially non-uniform deposition of laser energy was modeled by a two-dimensional temperature profile applied during time of electron-ion energy exchange. Our simulations show that the ultrafast two-dimensional heating results in the melting and pressurization of a thin surface layer. Due to a non-uniform stress distribution, this molten layer expands to form a jet-like protrusion at the laser pulse’s focal point. Above some critical stress, many voids start to nucleate forming a foam-like material covered by a thin liquid shell/cupula. The still expanding cupula may rupture forming a rim around the newly-developed crater. All these processes lead to the experimentally observed complicated surface morphology, which becomes frozen at the nanosecond time scale. Geometrical characteristics of simulated surface profiles, including crater depth and size of frozen bubbles, agree well with experiment. Our simulations help to provide better insight into the atomistic mechanisms of nanostructure formation.

Experimental Background

- Femtosecond laser pulses for forming nanostructures
- Ultrashort heating of electrons (10¹⁵ K/s)
- Heating depth very small (~100 nm)
- Response of metal to ultrashort heating
- Build-up of high pressure in surface layer
- Formulation & propagation of ultrashort pressure waves
- Strong tensile wave results in cavitation
- Ablation & surface morphology
- Ultrafast cooling (10¹² K/s)

Typical experimental set-up:
- Ti-Sapphire laser system
  - λ = 800 nm
  - θ_sample = 45°
  - τ_pulse ~ 100 fs
  - F_instant < 1 J/cm²

Ablation threshold:
- F_absorbed = 100 - 200 mJ/cm²
- Crater depth at threshold:
  - d_crate = 110 - 130 nm

Fig. 1: Diagram of experimental setup. Experiments were done by S. Ashitkov.

Computational Method

Past molecular dynamics (MD) simulations of ablation were done in 1-D geometry, which didn’t allow for the generation of 2-D or 3-D nanostructures. Ours simulates a 2-D energy distribution on the surface, which allowed for the development of complex surface morphology. Due to ultrafast heating of the sample, there are 2 phases to our model. The first is a two-temperature hydrodynamic (2T-HD) model, which simulates electron-ion thermalization. From 2T-HD simulations, we obtain a temperature distribution T(x, y) of the ions, which is input for a Langevin thermostat in MD simulations.

Fig. 2: Experimental results from ultrashort laser irradiation of Al film a) image of crater and rim formation, b) preparation of platinum covering for ion sputtering, c) cross sectional view of sample with visible rim and cavitation underneath. Images done by Y. Emrion at Nanotechnology Research & Education Center, USF

Results

Our purpose was to investigate the processes leading to the formation of the surface morphology shown in experiment. We determined and investigated three processes that were important to these structures. We simulated the regime of simple melting, jetting of molten gold, and cavitation under a thin molten gold surface (or cupola).

Fig. 3: The basic setup of the MD simulation with the temperature profile obtained from the hydrodynamic model. The two-dimensional profile of the target temperature is given by

\[ T(x, y) = T_0 \exp\left(-\frac{x^2}{d^2}\right) \cos\left(\frac{\pi y}{2L}\right) \]

Our MD simulations use periodic boundary conditions on the y,z-axes and free boundary conditions on the x-axis. The interatomic forces are modeled by a new Embedded Atom Model (EAM) potential designed for wide ranges of temperatures and pressures.

Fig. 4: Sketch of the cross section of sample from experiment showing rim formation with cavitation beneath the surface.

The combination of these processes followed by re-crystallization results in the final morphology, which includes the crater and frozen bubbles.

Fig. 5: Results from MD simulations a) simple melting, b) jetting of molten material, c) cavitation under cupola.

Fig. 6: Simulation results of an Au sample with original crystal size of 160 x 600 x 10 [nm].
   a) density of the atoms at an early time of the simulation (~150 ps)
   b) velocity at later times (after 1.1ns), green is in the positive x-direction and red is in the negative x-direction. This shows the cupula formation and eventual destruction along with the cavitation and bubbles under the molten surface on the edges.

Funding provided by NSF Grant DMR-0755256