Petascale atomistic simulations of short pulse laser-induced surface nanostructuring

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Material modification with short (ps and fs) laser pulses

short pulse laser irradiation of a metal target

- ultrafast heating/cooling + laser-induced stresses
- ultrahigh density of crystal defects
- nonequilibrium/metastable phases
- complex nano/micro-scale surface structures

Petascale atomistic simulations are needed!

Savolainen, Christensen, Balling

TTM-MD model for laser interaction with metals

The combined TTM-MD model adds **physics missing in classical MD**

1. \[ C_e(T_e) \frac{\partial T_e}{\partial t} = \nabla \cdot [K_e(T_e, T_l) \nabla T_e] - G(T_e)(T_e - T_l) + S(\vec{r}, t) \]

2. \[ C_l(T_l) \frac{\partial T_l}{\partial t} = \nabla \cdot [K_l(T_l) \nabla T_l] + G(T_e)(T_e - T_l) \]

3. \[ m_i \frac{d^2 \vec{r}_i}{dt^2} = \vec{F}_i + \xi m_i \vec{v}_i^{th}, \quad T_i^{cell} = \sum_{cell} m_i \left( \frac{v_i^{th}}{3k_B N_{cell}} \right)^2 \]

4. pressure-transmitting, heat-conducting boundary conditions

**TTM** (two-temperature model)


**MD**

\[ \xi = \frac{GV_N(T_e - T_l)}{2K^T} \]

- Laser energy absorption by the conduction band electrons
- Electron-phonon equilibration
- Electronic heat conduction

Laser-metal interaction: from **melting** to **spallation** and to **phase explosion**

Al (001) target, 100 fs pulse

large-scale simulation of 100 fs laser pulse irradiation of Ag (001) at 900 J/m²

fast heating → compressive stresses → tensile (unloading) stress wave

**melting** 650 J/m²

**spallation** 900 J/m²

**phase explosion** 2000 J/m²

94 × 94 × 200 nm³, 107M atoms

Ultrafast cooling \(\rightarrow\) Final surface microstructure?

- Cooling rates up to \(10^{12}\) K/s and fast resolidification
- Generation of metastable phases and unusual structures?

**Cr target**

- Cooling rate: \(5 \times 10^{12}\) K/s!
- End of the resolidification process

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**Thermodynamic States**

- **Melting:** 650 J/m²
- **Spallation:** 900 J/m²
- **Phase Explosion:** 2000 J/m²

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**Time, ps**

0 100 200 300 400 500

**Lattice and Electron Temperatures, K**

- Electron temperature
- Lattice temperature

**Graphical Representation**

- Graph showing temperature changes over time.
- Diagrams illustrating melting, spallation, and phase explosion.
Melting and Resolidification
Void nucleation, growth, and capture by resolidification: surface swelling

TTM-MD simulation of Ag (001) target irradiated by 100 fs pulse at an absorbed fluence of 850 J/m²: $100 \times 100 \times 150$ nm$^3$, 84M atoms

- voids generated in the melted region can be captured by rapidly advancing solidification front
- generation of sub-surface porous region
- swelling of the irradiated target

Wu et al., *Phys. Rev. B* 91, 035413, 2015
massive homogeneous nucleation and growth of crystallites at $T \approx 0.69T_m$

undercooling down to $\sim 0.69 T_m$, $\rightarrow$ homogeneous nucleation $\rightarrow$ $\sim 30$ nm nanocrystalline layer
Nanocrystalline structure of the surface region colored by grain orientation angle

- nano-grains with random orientation
- high density of grain boundaries, twins, and stacking faults
- Nanoscale twinning structures with 5-fold symmetry
- high hardness of the surface can be expected

![Diagram showing nanocrystalline structure with FCC and HCP atoms, defects, and grain orientation statistics.](image)
Melting and Resolidification: materials dependence

Ag (001)

Fast cooling

\[ k_{\text{Ni}} \sim 0.2 \ k_{\text{Ni}} \]

Epitaxial regrowth only!

FCC Ni is blanked

red – liquid, vacancies and dislocation lines

green – HCP Ni (stacking faults and twin boundaries)

Ni (001)

Competition between epitaxial regrowth and homogeneous nucleation

Nanocrystalline surface layer generation
Melting and Resolidification: surface orientation dependence

Ni (001)  
No twins, but growth dislocations

Ni (110)  
10 ps

Ni (111)  
50 ps
Growth twinning: Nanotwinned layer
Melting and Resolidification: surface orientation dependence

Ni (100), 400 ps
No twins, but growth dislocations

Ni (011), 500 ps

Ni (111), 720 ps
Growth twinning: Nanotwinned layer

vacancies are blanked
Experimental evidence of growth twinning

(111) pole figures of top 20 nm layer:

Ni (111) → 

No twins in Ni (100) and Ni (011)

Ni (111), 1900 J/m² → 

(absorbed ~ 600 J/m²)

No twins in Ni (100)

Ø – original {111}

△ – {111} generated by twins

Photomechanical Spallation
Spallation $\Rightarrow$ Nanospike formation

Void nucleation, growth, and coalescence of a melted layer

- long bridge between the substrate and the top layer breaks at 3.0 ns
- nanospike (~6 nm in diameter) freezes shortly after the break

TTM-MD simulation of Ag (001) target irradiated by 100 fs pulse at an absorbed fluence of 900 J/m$^2$: $100 \times 100 \times 150$ nm$^3$, 84M atoms

Spallation $\Rightarrow$ Nanospike formation

undercooling down to $\sim 0.69 \, T_m$

homogeneous nucleation

nanospike formation

Nano-crystalline structure of the nanospike

Homogeneous nucleation and growth $\rightarrow$ 6 icosahedra interpenetrating with each other

Nanospike formation – connections to experiment


Nanocrystalline structure of the spike generated by homogeneous nucleation under deep undercooling

(001) pole figure of EBSD measurement showing random crystal orientation of the head of the spikes
Phase Explosion
Higher laser fluence $\Rightarrow$ phase explosion $\Rightarrow$ surface morphology

TTM-MD simulation of Ag (001) target irradiated by 100 fs pulse at an absorbed fluence of 3000 J/m$^2$: $400 \times 400 \times 300 \text{ nm}^3$, 2.8 billion atoms

Great thanks to Titan (OLCF)!

Resolidification of the foamy structure $\Rightarrow$ complex surface morphology
Higher laser fluence $\Rightarrow$ phase explosion $\Rightarrow$ surface morphology?

TTM-MD simulation of Ag (001) target irradiated by 100 fs pulse at an absorbed fluence of 3000 J/m$^2$: $400 \times 400 \times 300$ nm$^3$, 2.8 billion atoms

Resolidification of the foamy structure $\Rightarrow$ complex surface morphology
“Big picture”

“mosaic” approach to mapping the processes occurring at the scale of the whole laser spot

Ionin et al., JETP Lett. 94, 753, 2011
Zhao et al., Optics Express 15, 15741, 2007
Summary

Short pulse laser irradiation

→ thermal spike + laser-induced stresses

Surface nanostructuring: nanocrystalline layer, nanospike, high density of crystal defects (dislocations, twins, stacking faults),
Other work ongoing

laser pulse
laser pulse
laser pulse
laser pulse

liquid
transparent
overlayer
gas

Thanks to OLCF and all for your attention!