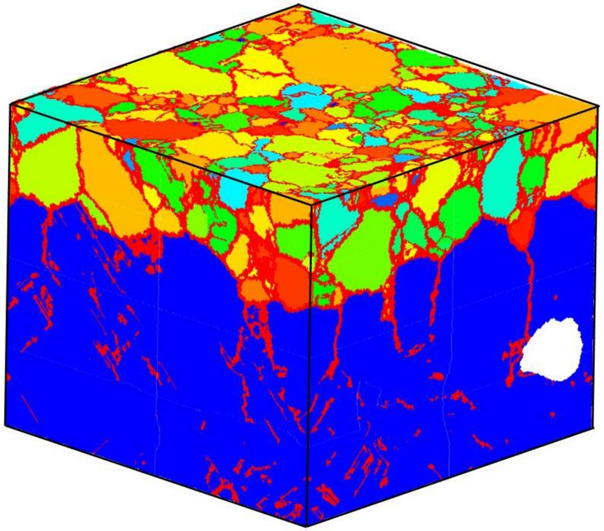
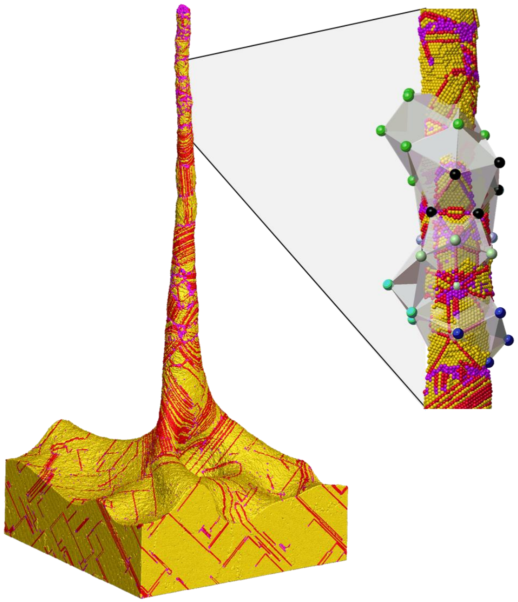
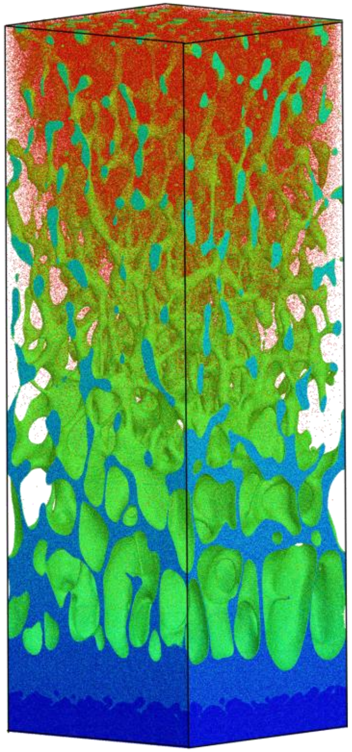
Petascale atomistic simulations of short pulse laser-induced surface nanostructuring

Chengping Wu, Maxim Shugaev, Eaman Karim and Leonid V. Zhigilei
Department of Materials Science and Engineering, University of Virginia







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Material modification with short (ps and fs) laser pulses

short pulse laser irradiation of a metal target

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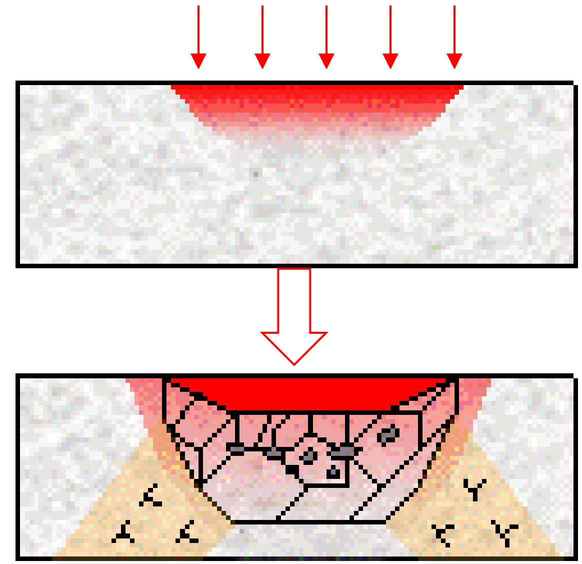
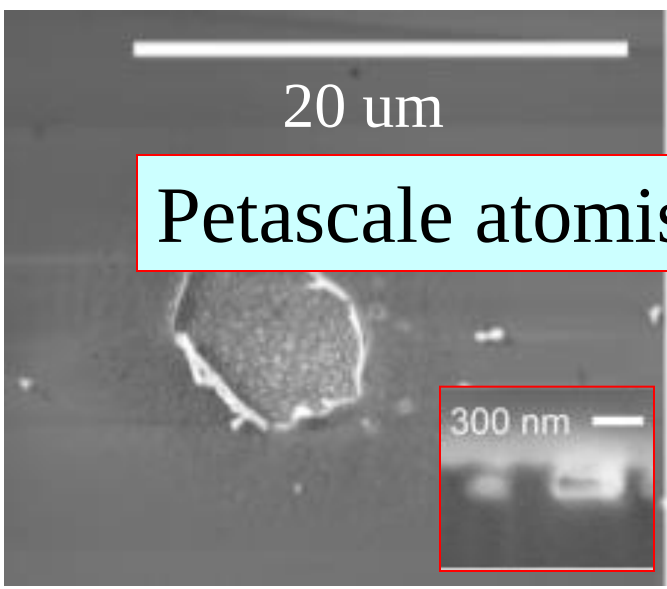
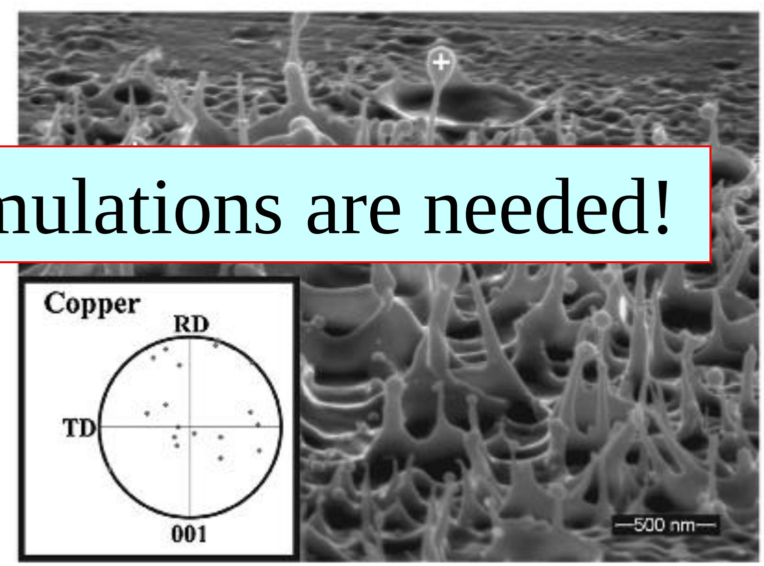
- ultrafast heating/cooling + laser-induced stresses
- ultrahigh density of crystal defects
- nonequilibrium/metastable phases
- complex nano/micro-scale surface structures



Petascale atomistic simulations are needed!

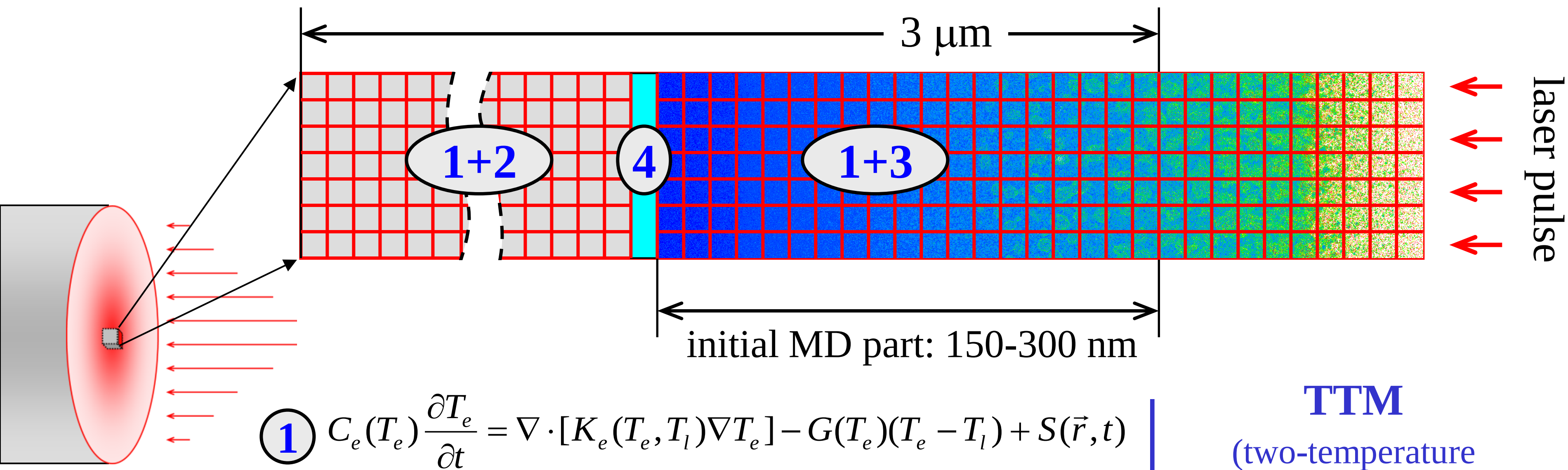


Savolainen, Christensen, Balling
Phys. Rev. B **84**, 193410, 2011



Oboňa et al., *Appl. Surf. Sci.* **303**, 118, 2014

TTM-MD model for laser interaction with metals



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

TTM (two-temperature model)
Sov. Phys. JETP **39**, 375, 1974

3 $m_i\, d^2\vec{r}_i/dt^2 = \vec{F}_i + \xi m_i \vec{v}_i^{\,th}$, $\quad T_l^{cell} = \sum_{cell} m_i \left(v_i^{th}\right)^2 / \left(3k_B N_{cell}\right)$

MD

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

4 pressure-transmitting, heat-conducting boundary conditions

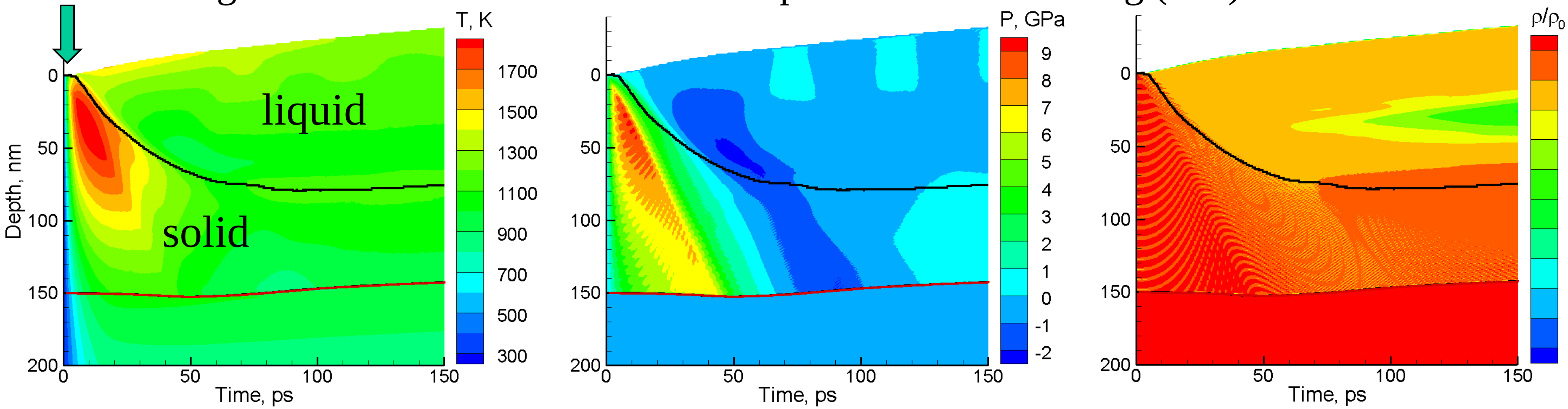
The combined TTM-MD model adds **physics missing in classical MD**

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

Phys. Rev. B **68**, 064114, 2003; *J. Phys. Chem. C* **113**, 11892, 2009; *Appl. Phys. A*, 114, 11-32, 2014

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

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



94 × 94 × 200 nm³, 107M atoms

fast heating → compressive stresses

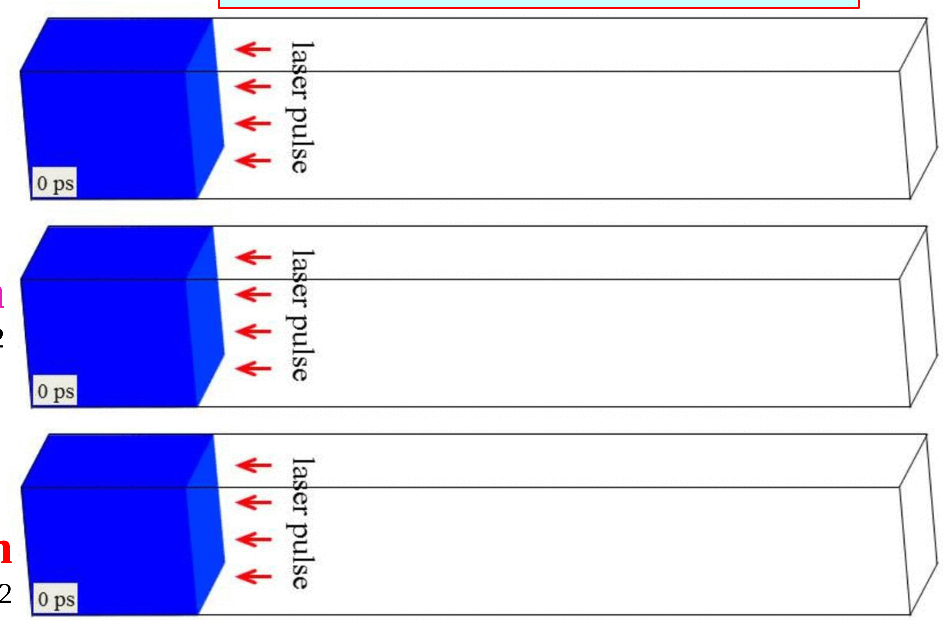
→ tensile (unloading) stress wave

Al (001) target, 100 fs pulse

melting
650 J/m²

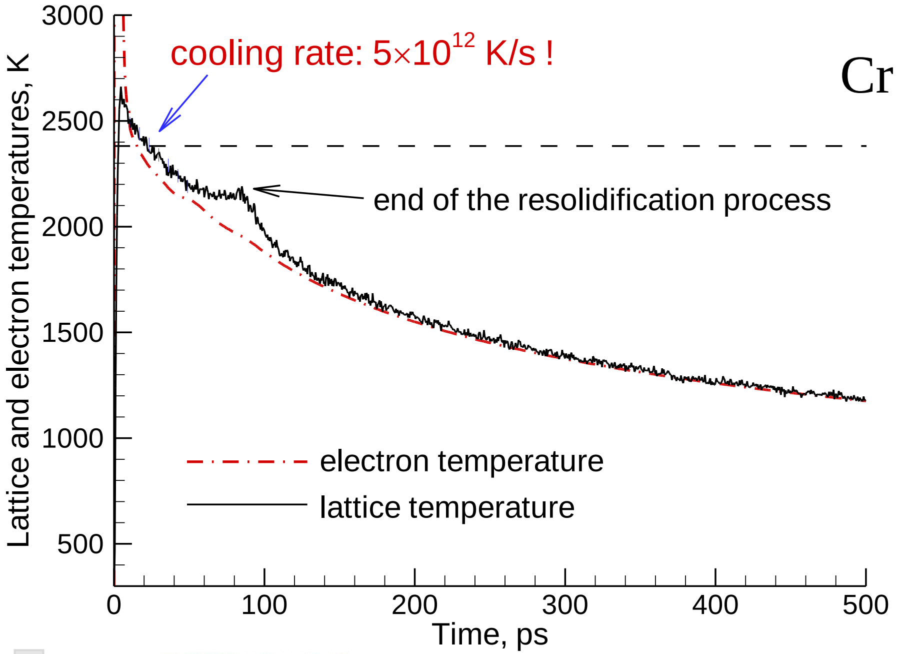
spallation
900 J/m²

phase explosion
2000 J/m²



Wu and Zhigilei, *Appl. Phys. A* **114**, 11, 2014

Ultrafast cooling ➔ Final surface microstructure?



Cr target

cooling rates up to 10^{12} K/s and fast resolidification

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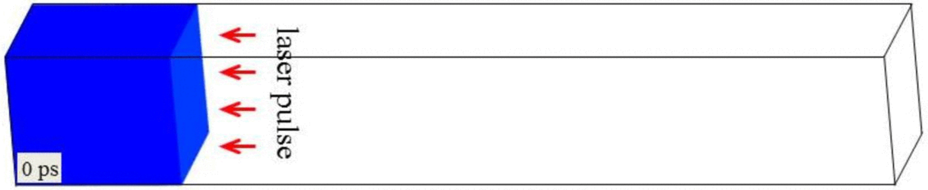
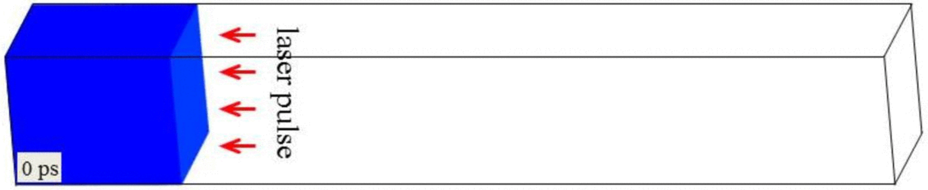
generation of metastable phases and unusual structures ?

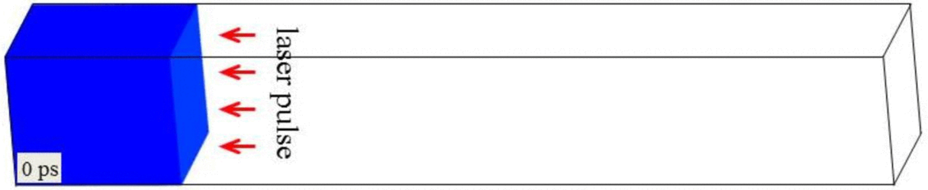
melting
650 J/m²



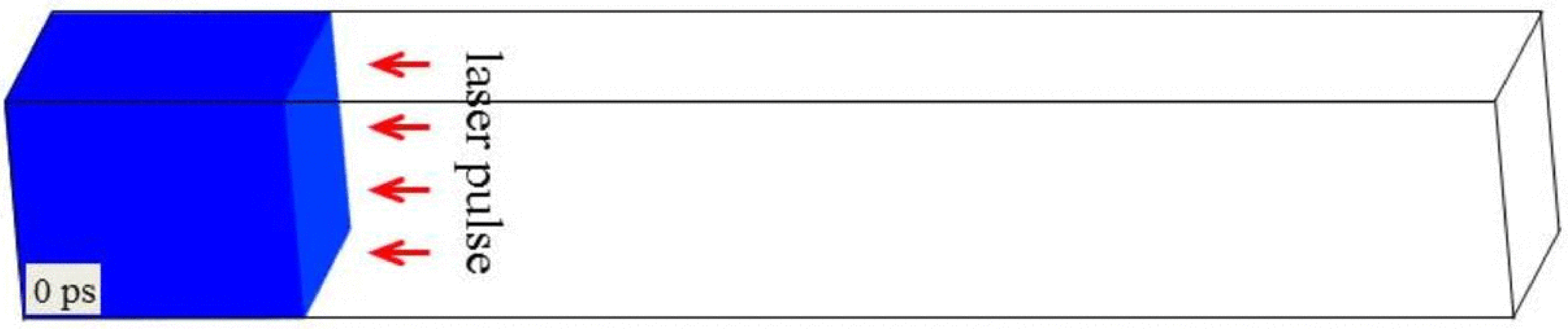
spallation
900 J/m²



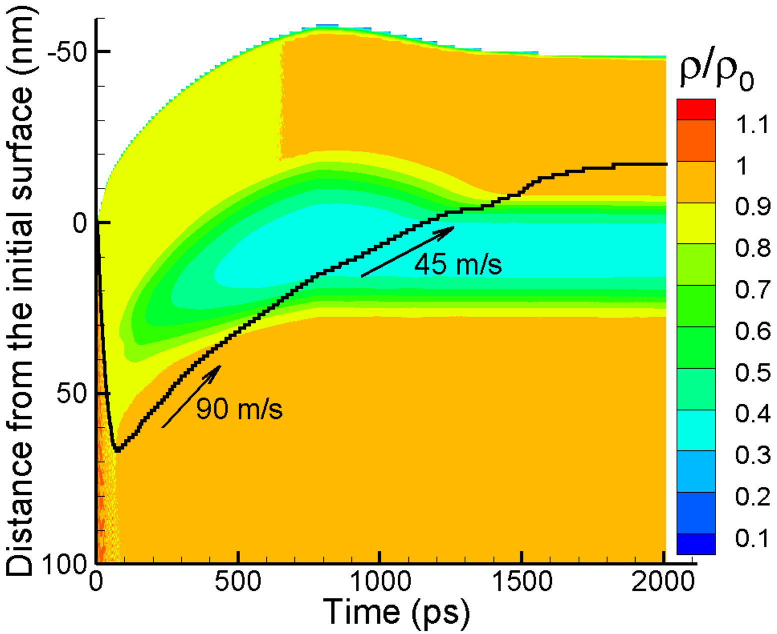
phase explosion
2000 J/m²



Melting and Resolidification



Void nucleation, growth, and capture by resolidification: surface swelling

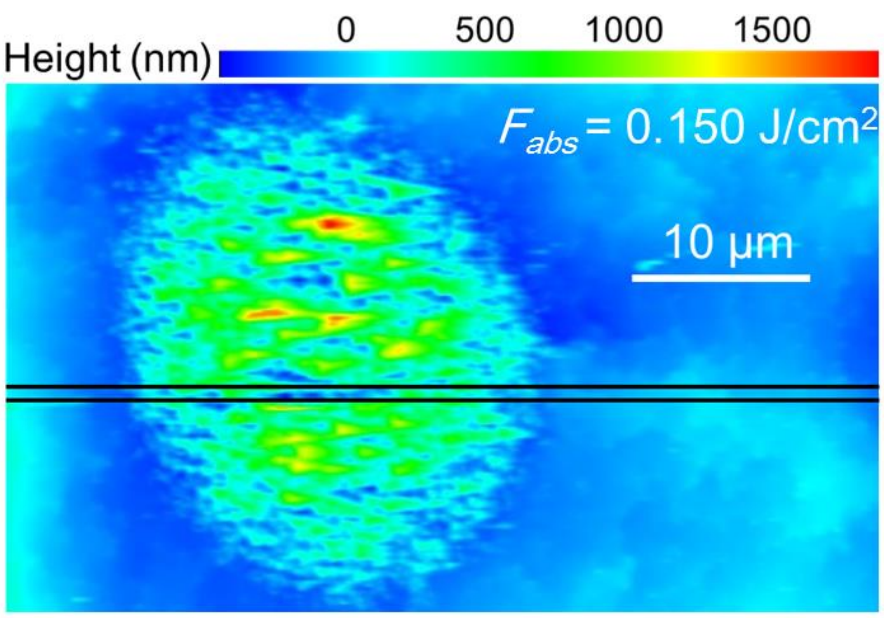


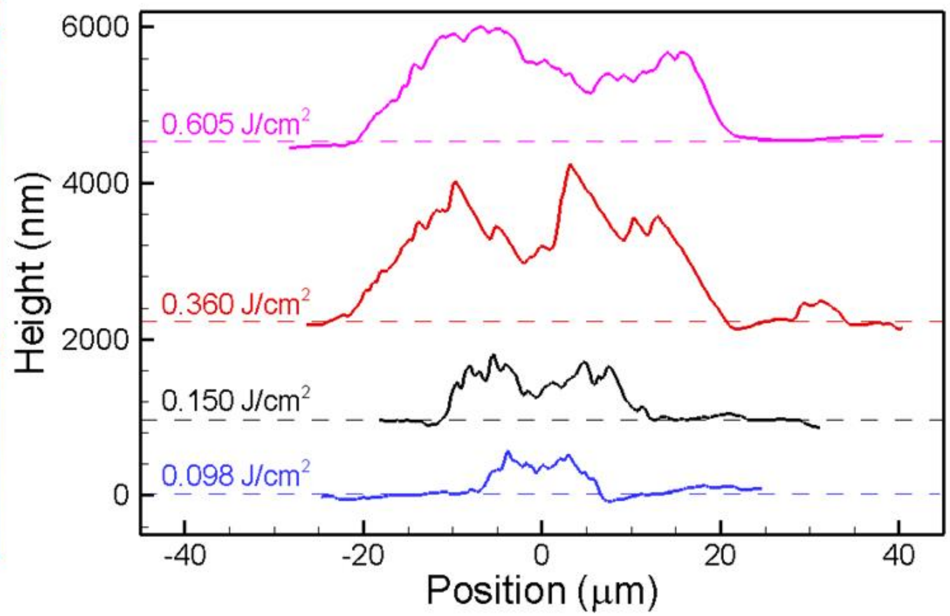
TTM-MD simulation of Ag (001) target irradiated by 100 fs pulse at an absorbed fluence of 850 J/m^2: $100 \times 100 \times 150$ nm^3, 84M atoms

voids generated in the melted region can be captured by rapidly advancing solidification front

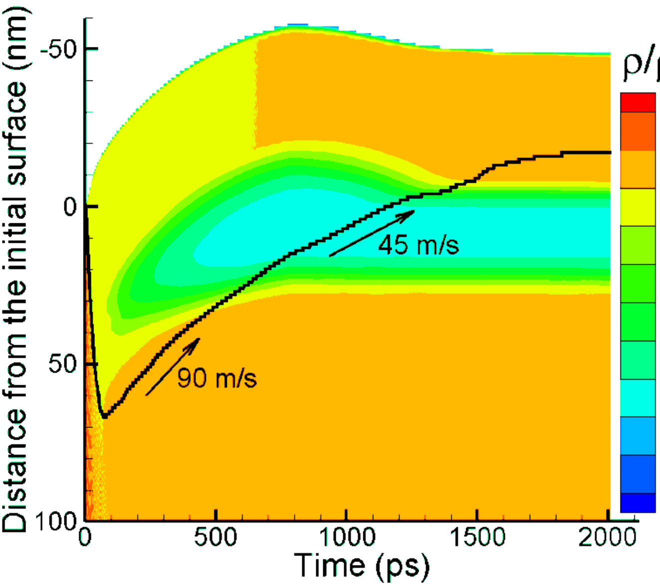
- generation of sub-surface porous region
- swelling of the irradiated target

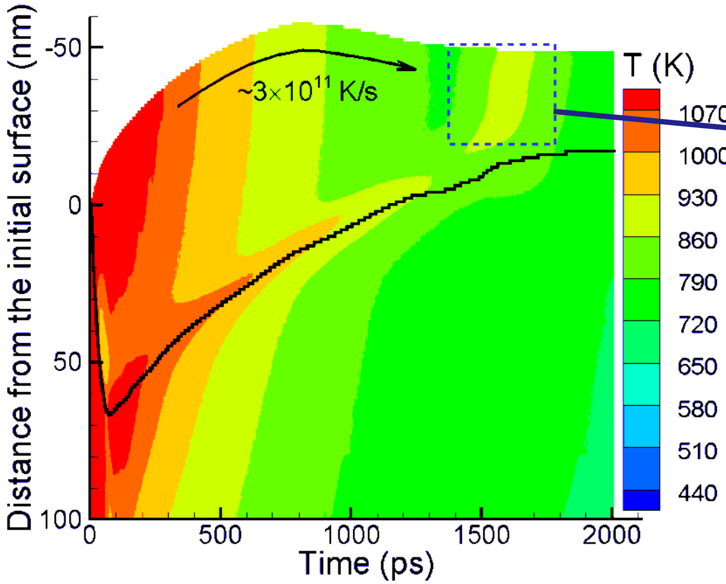
Wu et al., *Phys. Rev. B* **91**, 035413, 2015





Deep undercooling → generation of nanocrystalline surface layer

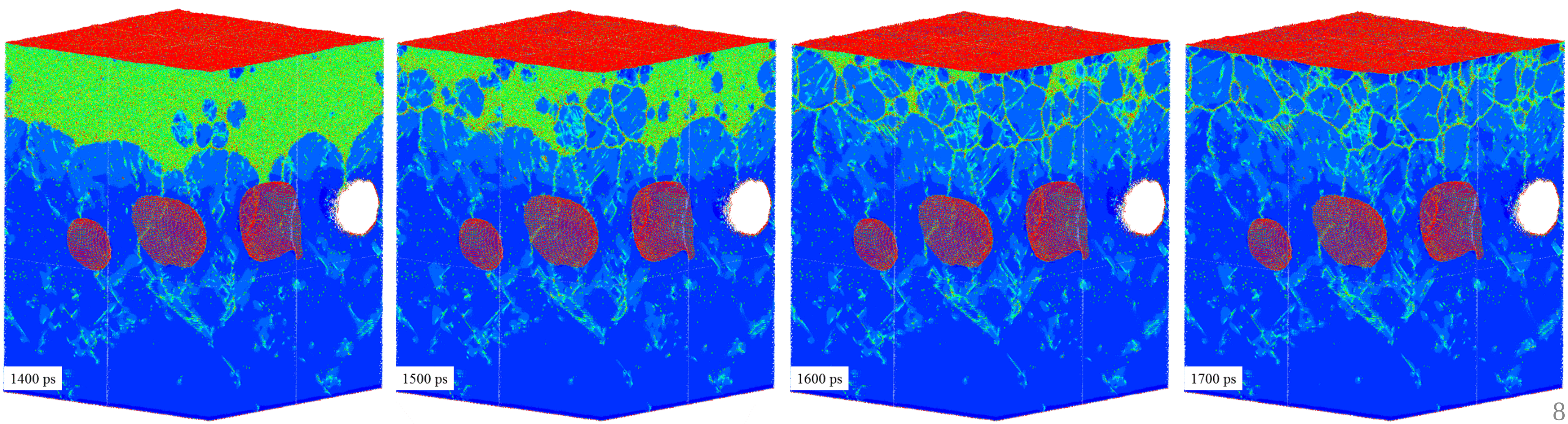




massive homogeneous nucleation and growth of crystallites at $T \approx 0.69T_m$

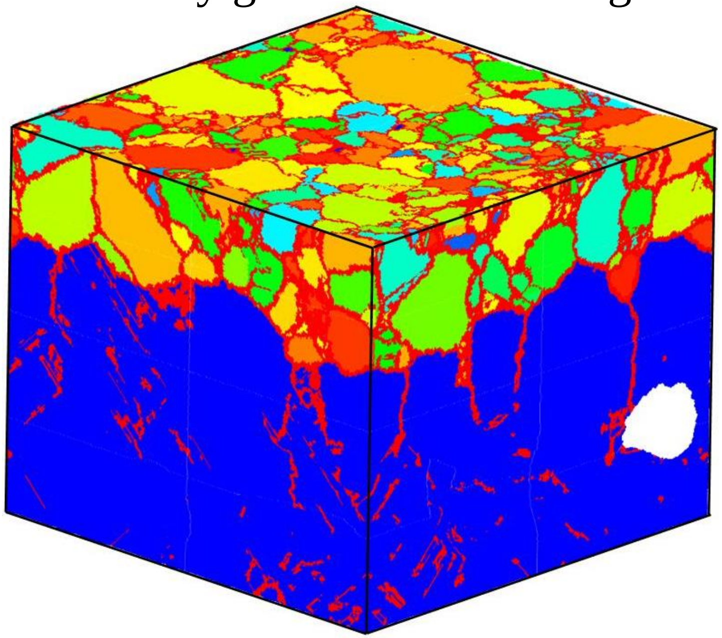
undercooling down to ~0.69 T_m, ➔ homogeneous nucleation ➔ ~30 nm nanocrystalline layer

Colored by energy



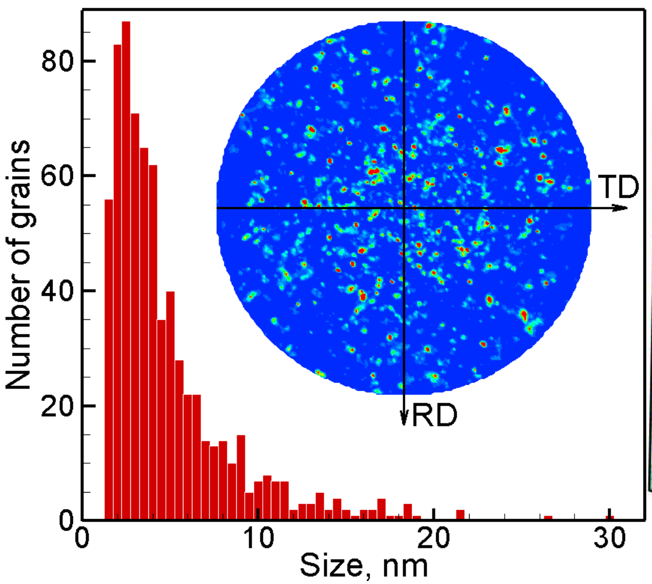
Nanocrystalline structure of the surface region

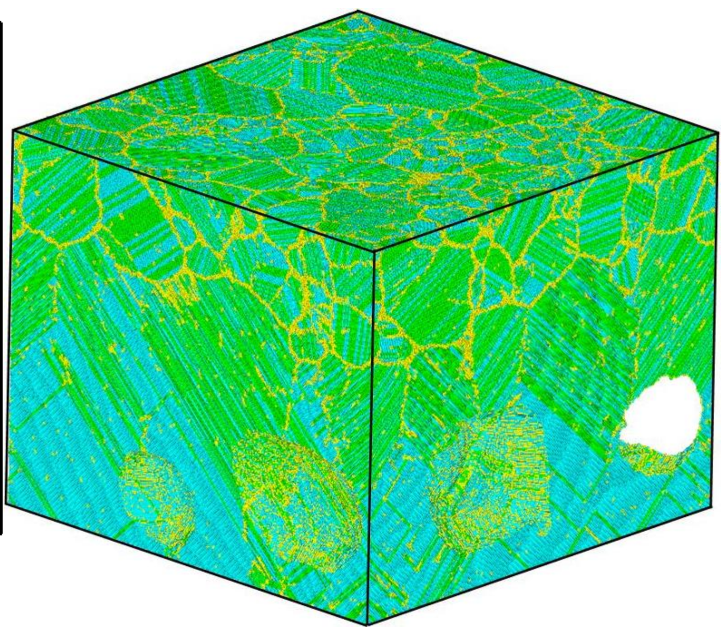
colored by grain orientation angle

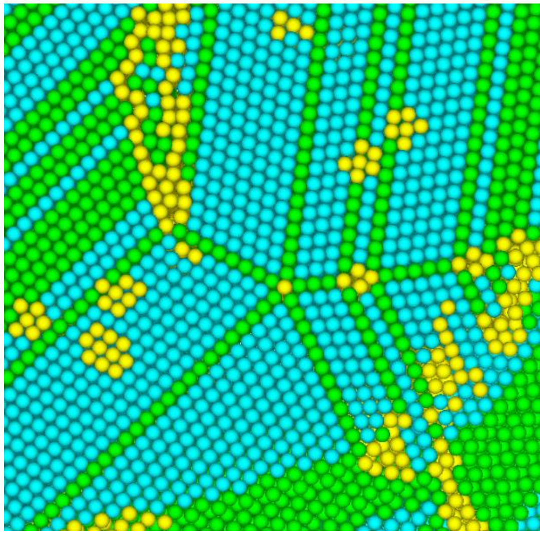


- nano-grains with random orientation
- high density of grain boundaries, twins, and stacking faults
- Nanoscale twinning structures with 5-fold symmetry
- high hardness of the surface can be expected

FCC atoms HCP atoms defects

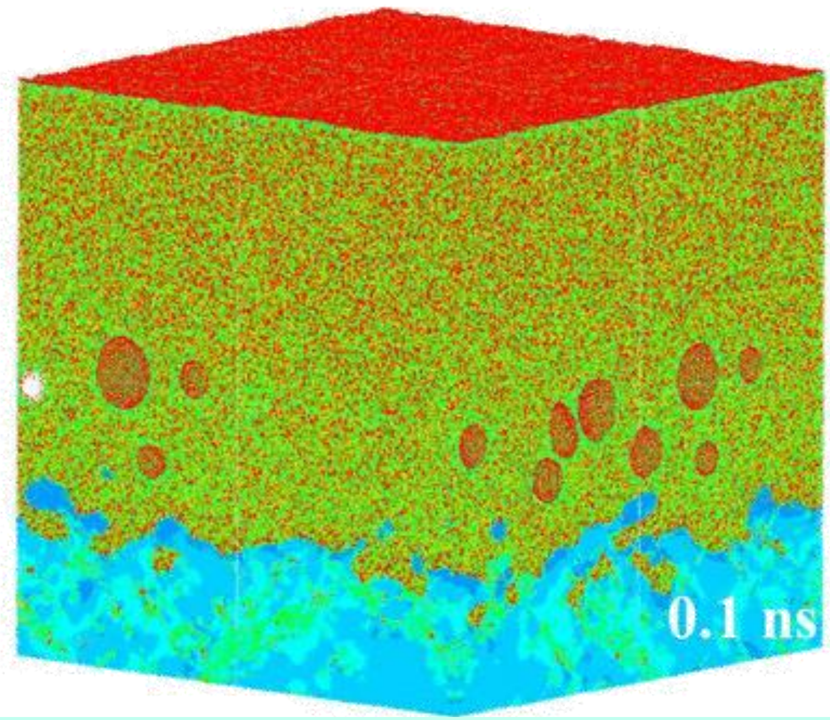






Melting and Resolidification: materials dependence

Ag (001)



Fast cooling

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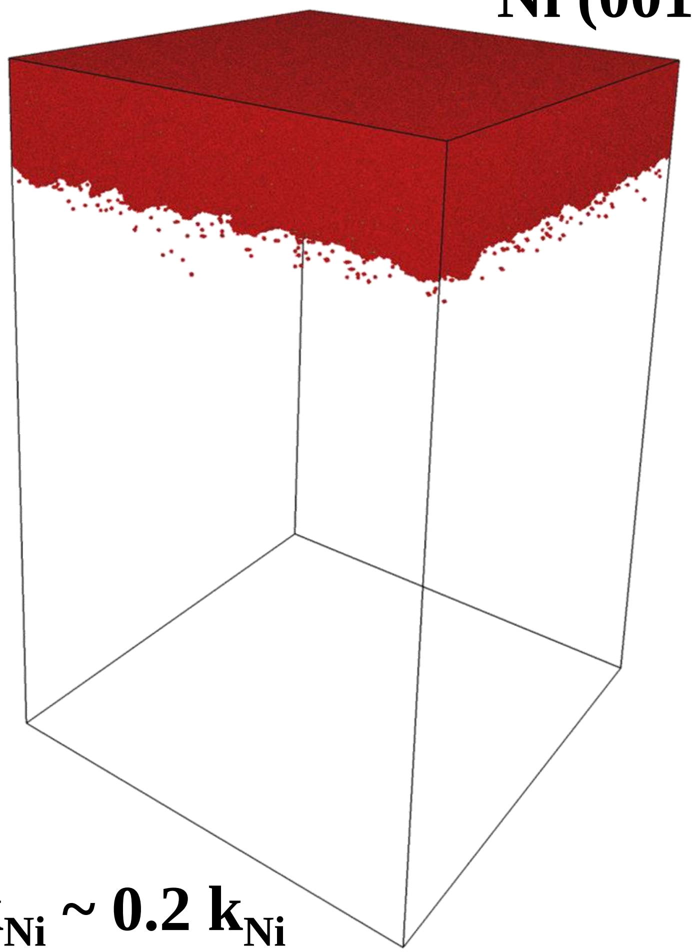
Competition between epitaxial regrowth and homogeneous nucleation

⇩

Nanocrystalline surface layer generation

Ni (001)



$k_{Ni} \sim 0.2\ k_{Ni}$

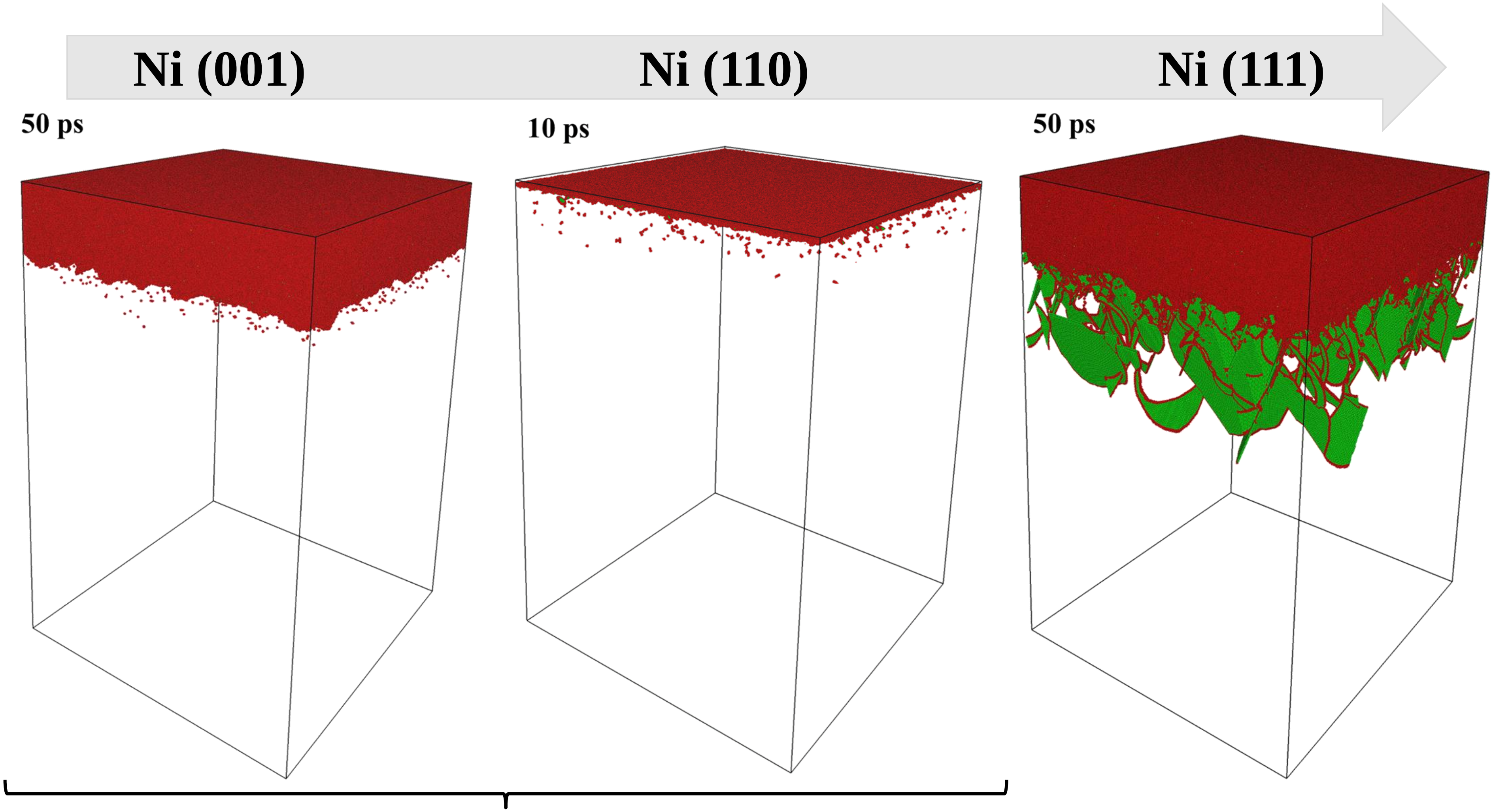
<u>Epitaxial regrowth only!</u>

FCC Ni is blanked

red – liquid, vacancies and dislocation lines

green – HCP Ni (stacking faults and twin boundaries)

Melting and Resolidification: surface orientation dependence

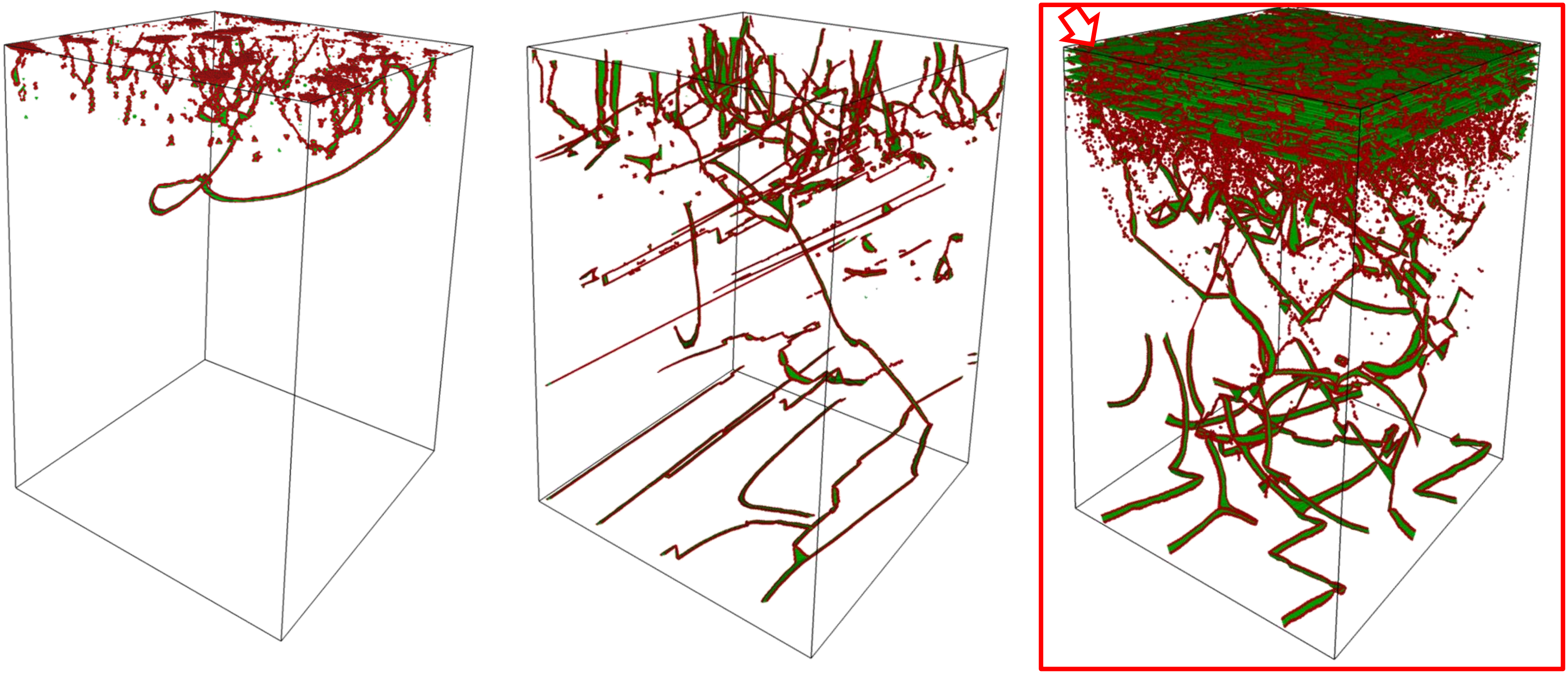


No twins, but growth dislocations

Growth twinning:
Nanotwinned layer

Melting and Resolidification: surface orientation dependence

Ni (100), 400 ps Ni (011), 500 ps Ni (111), 720 ps



vacancies are blanked

No twins, but growth dislocations

Growth twinning:
Nanotwinned layer

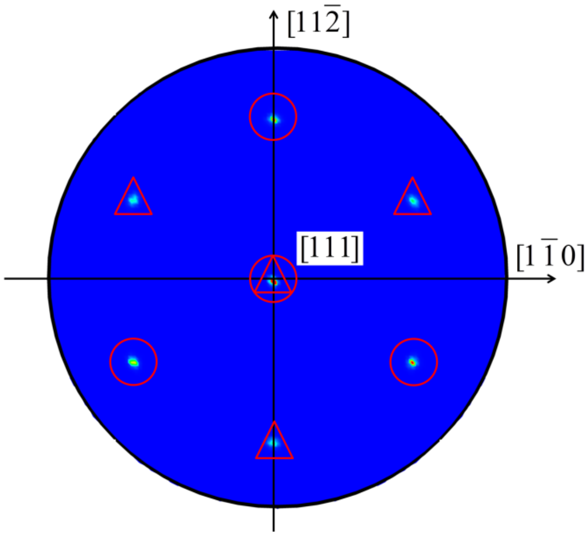
Experimental evidence of growth twinning

MD simulation

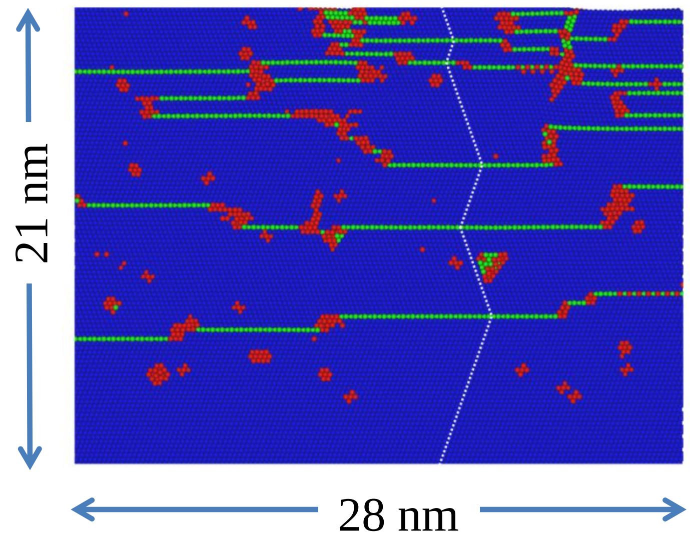
⟨111⟩ pole figures of
top 20 nm layer:

Ni (111) →

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



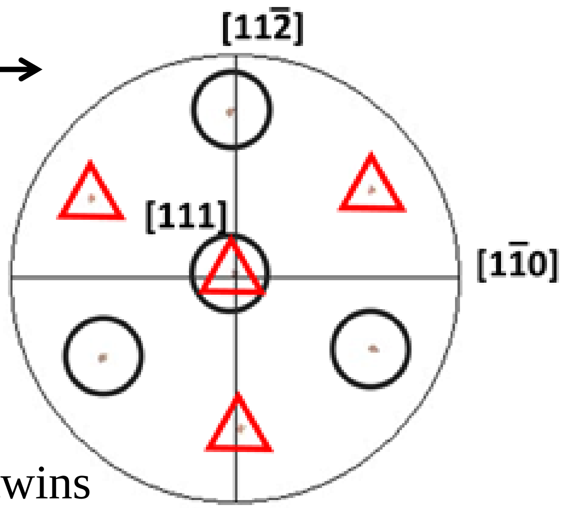
microstructure



Experiment

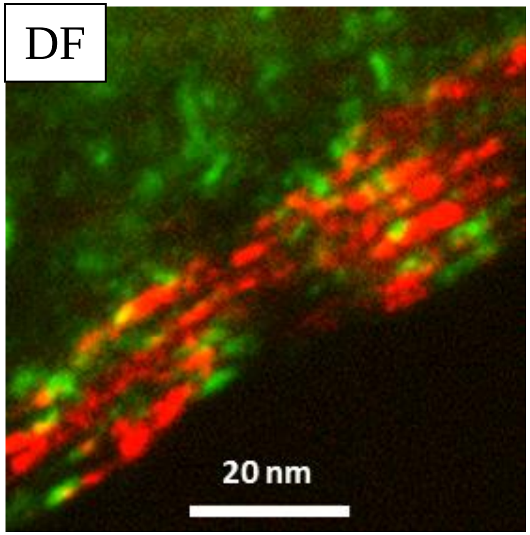
Ni (111), 1900 J/m^2 →
(absorbed ~ **600 J/m^2**)

No twins in Ni (100)



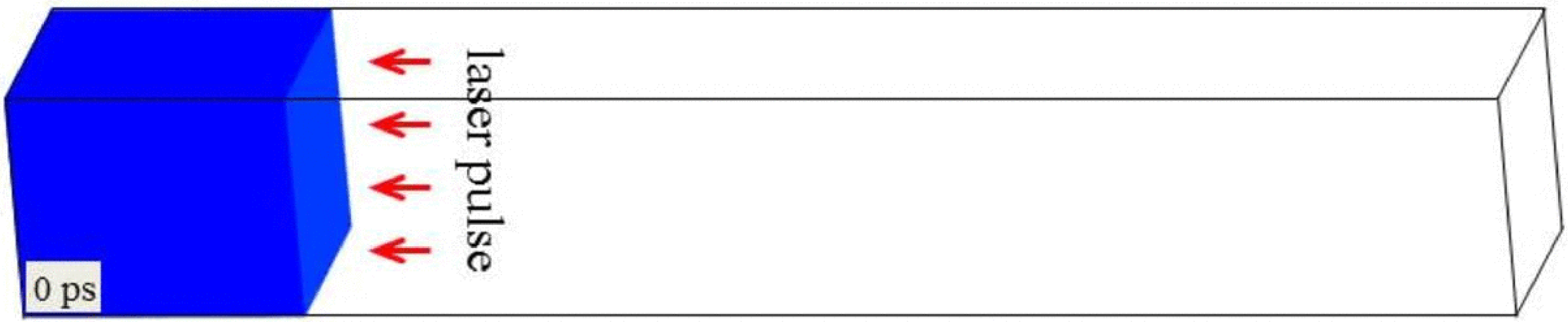
○ – original {111}

△ – {111} generated by twins



X. Sedao, M. V. Shugaev, C. Wu, T. Douillard, C. Esnouf, C. Maurice, S. Reynaud, F. Pigeon, F. Garrelie, L. V. Zhigilei, and J.-P. Colombier, Under Review

Photomechanical Spallation



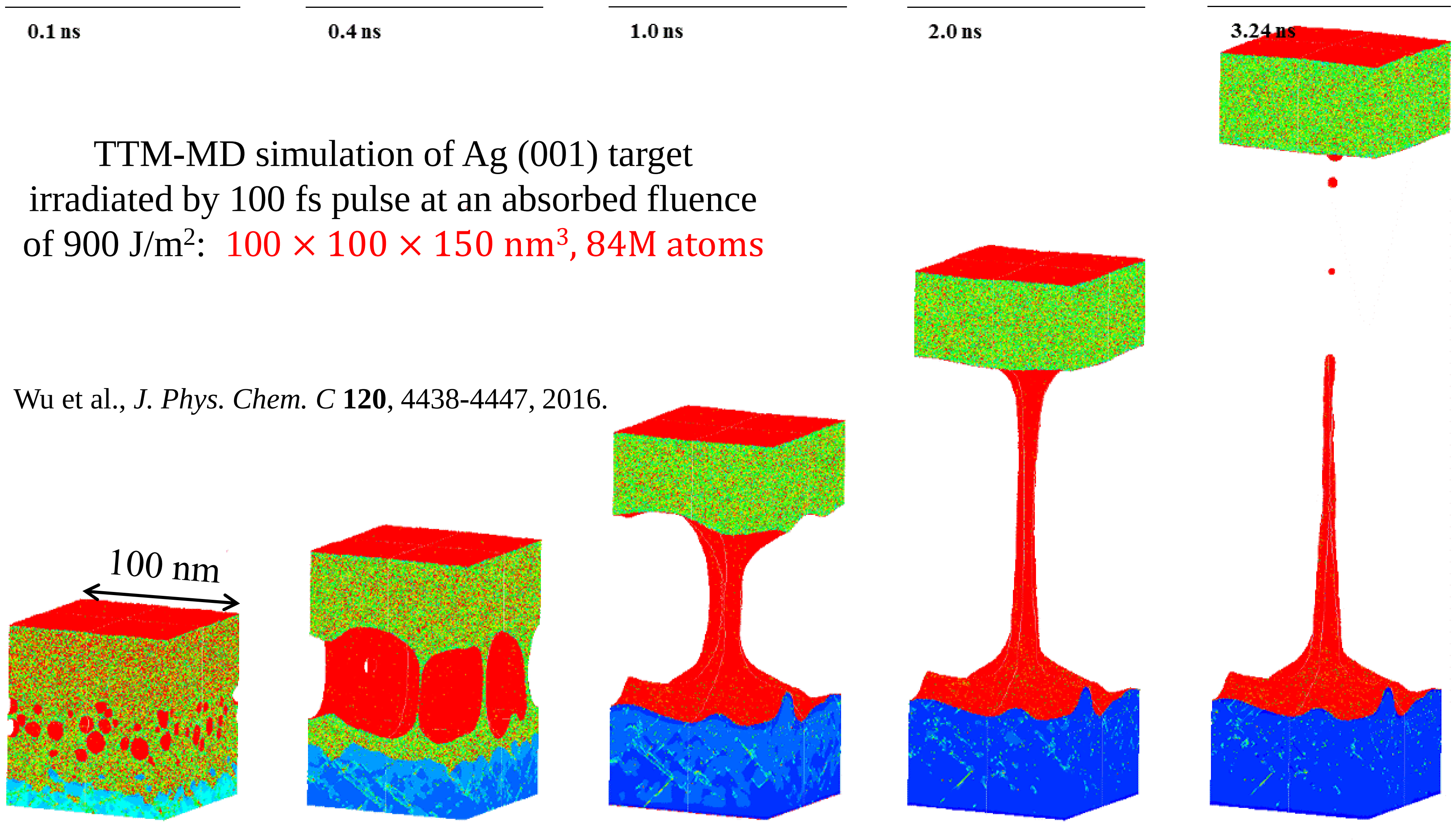
Spallation ➔ Nanospike formation

Void nucleation, growth, and coalescence spallation of a melted layer

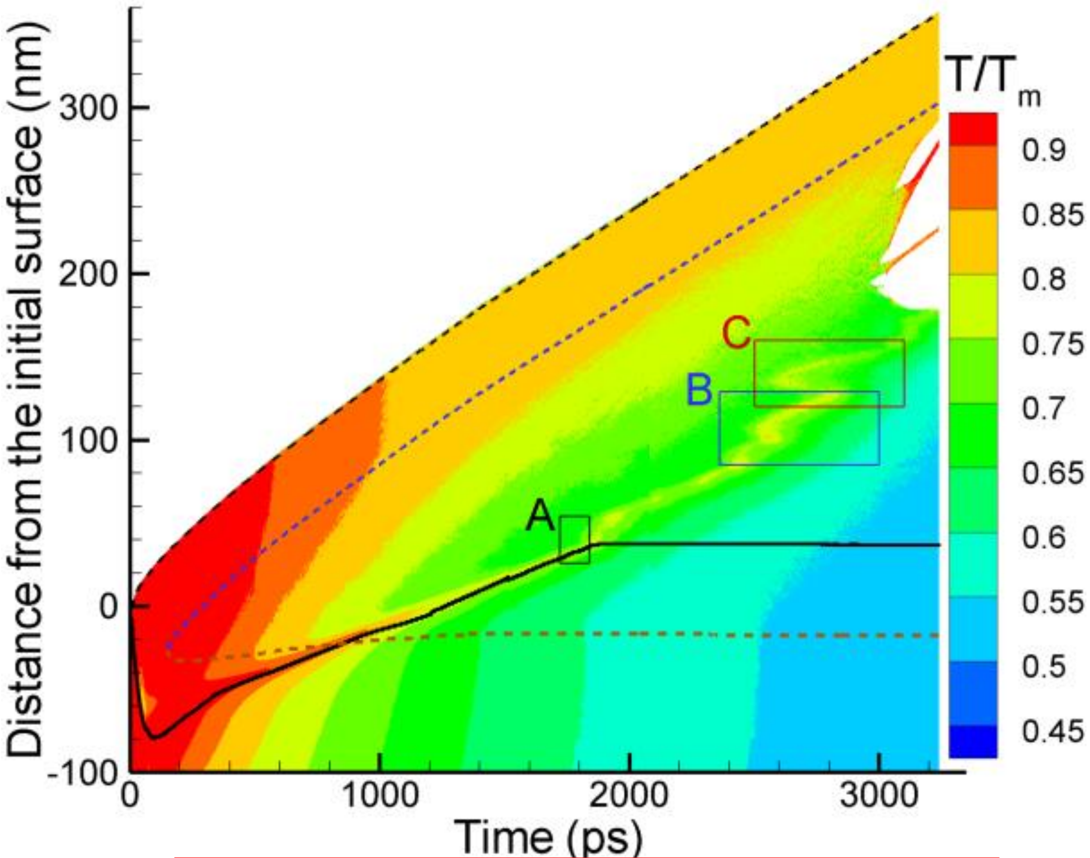
- long bridge between the substrate and the top layer breaks at 3.0 ns
- nanospike (~6 nm in diameter) freezes shortly after the break

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

Wu et al., *J. Phys. Chem. C* **120**, 4438-4447, 2016.



Spallation ➔ Nanospike formation



undercooling down to ~0.69 T_m

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homogeneous nucleation

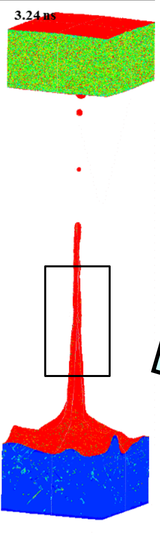
⇩

nanospike formation

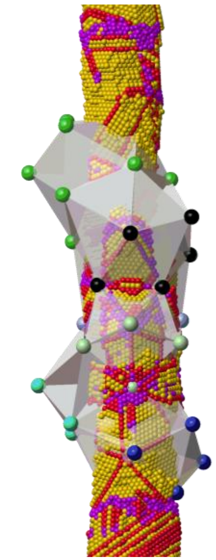
Wu et al., *J. Phys. Chem. C* **120**, 4438-4447, 2016.

- liquid/surface/defects
- FCC
- HCP

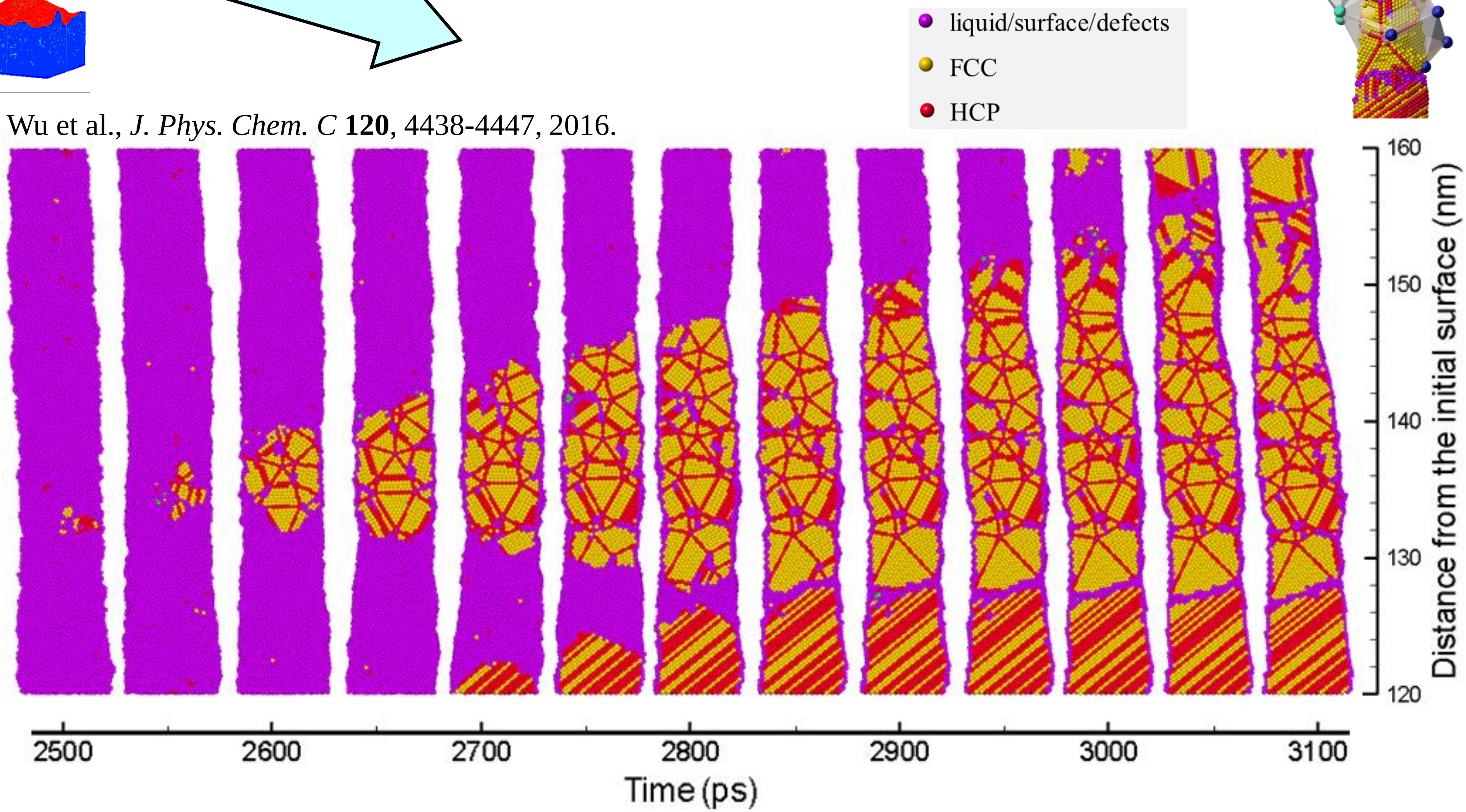
Nano-crystalline structure of the nanospike



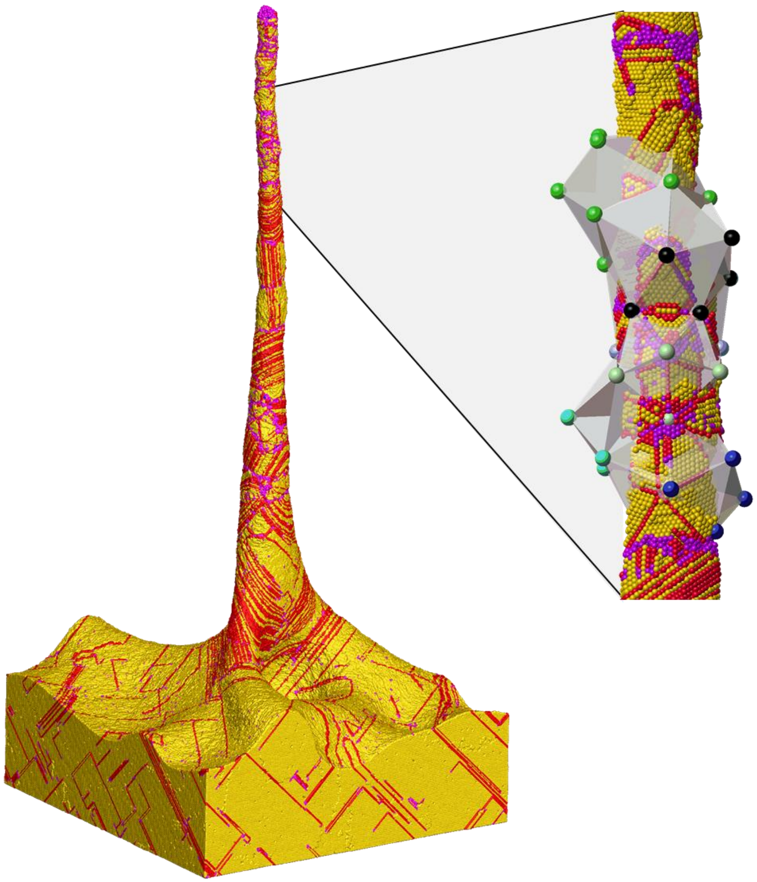
Homogeneous nucleation and growth ➔ 6 icosahedra interpenetrating with each other



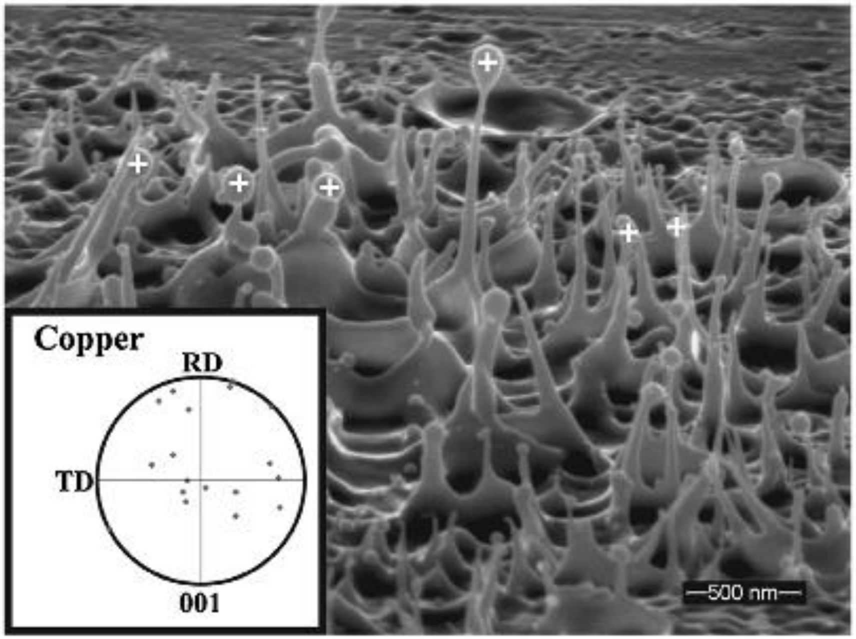
Wu et al., *J. Phys. Chem. C* **120**, 4438-4447, 2016.



Nanospike formation – connections to experiment



Oboňa et al., *Appl. Surf. Sci.* **303**, 118, 2014



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

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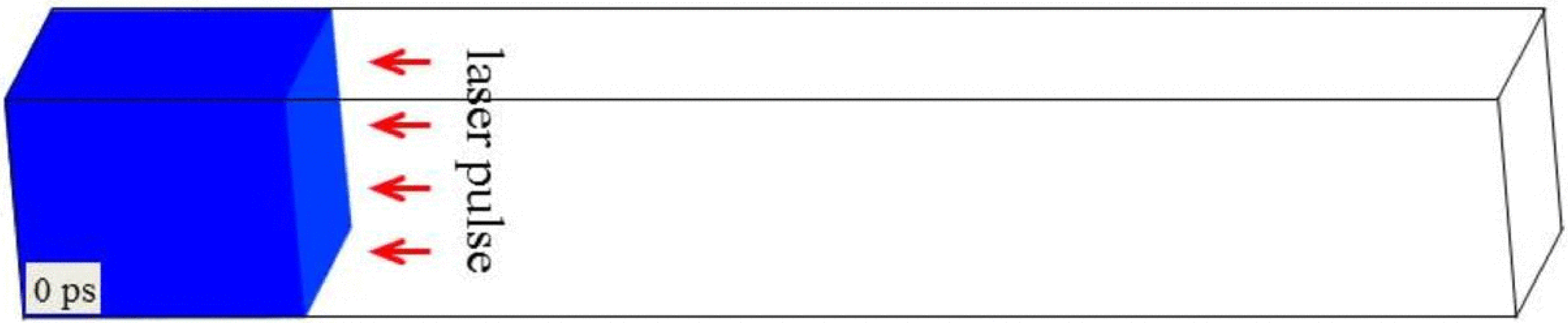
(001) pole figure of EBSD measurement showing random crystal orientation of the head of the spikes

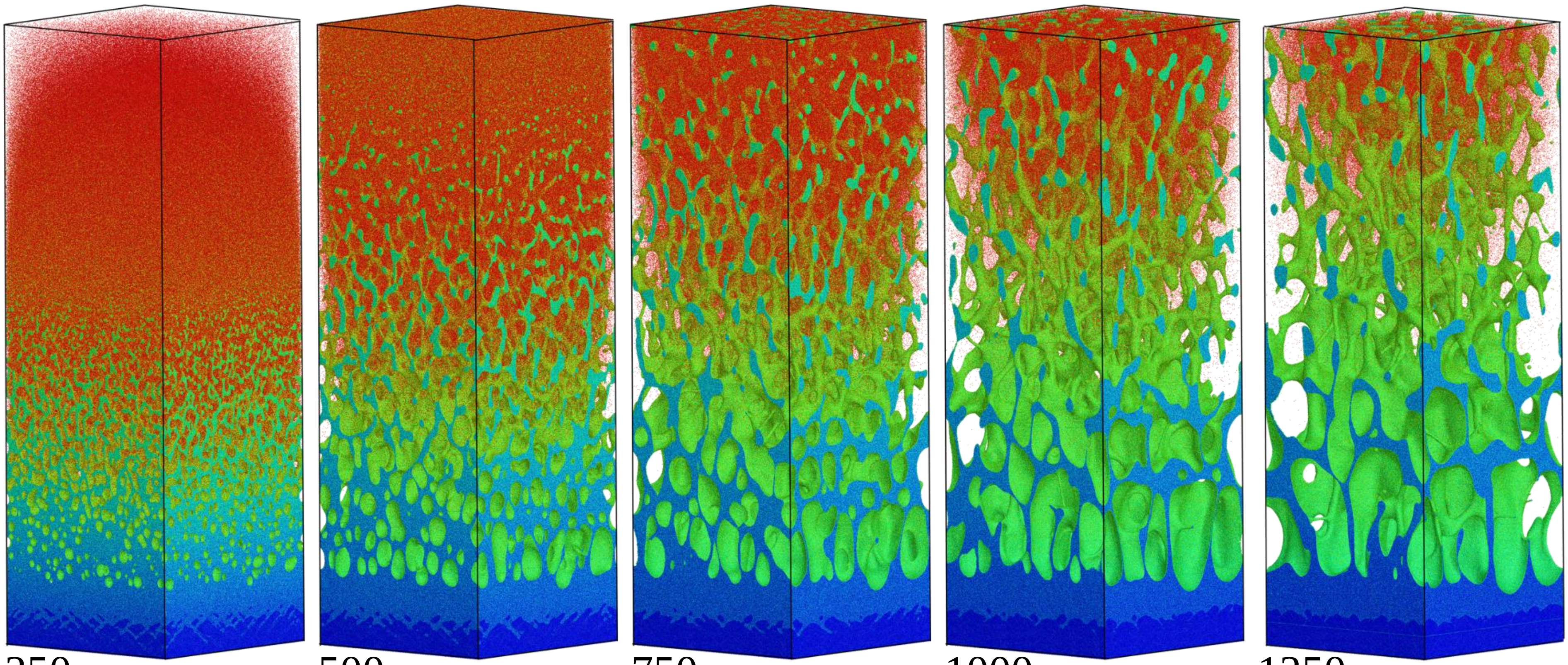
Phase Explosion



Higher laser fluence ➔ phase explosion ➔ 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

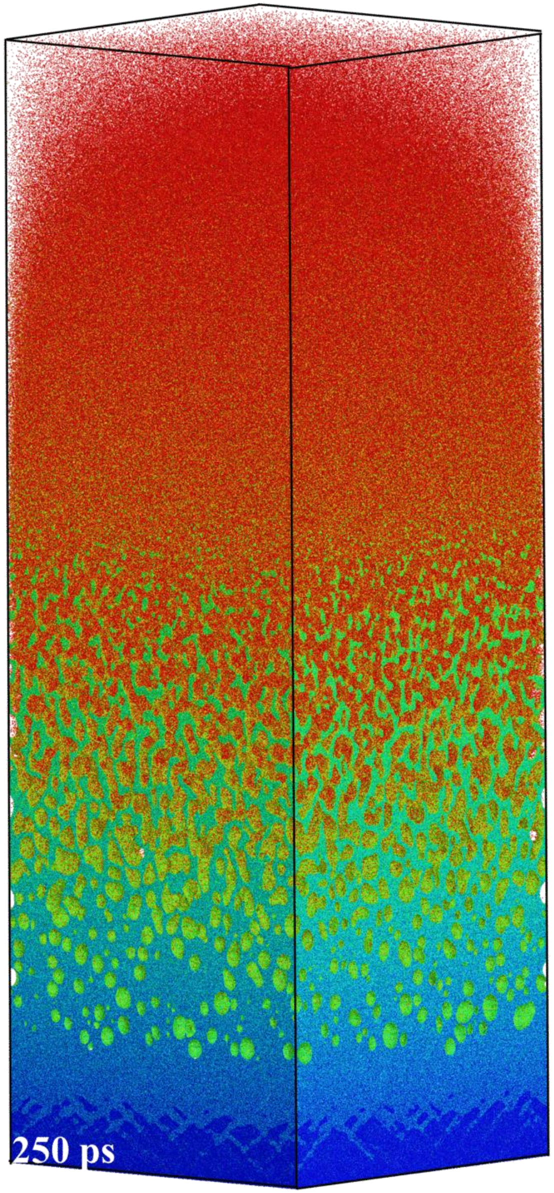
Great thanks to Titan (OLCF)!

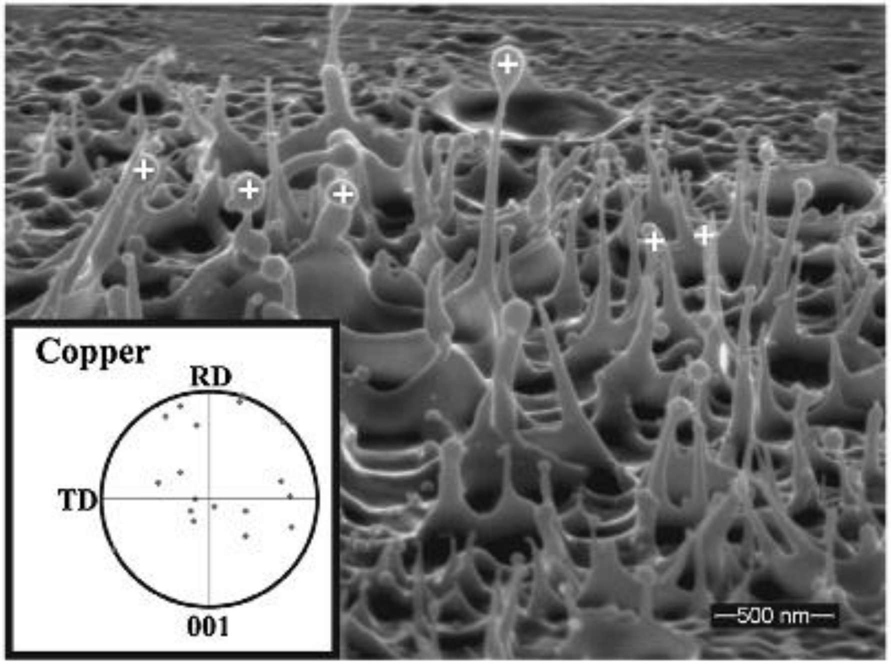


Resolidification of the foamy structure ➔ complex surface morphology

Higher laser fluence ➔ phase explosion ➔ surface morphology?

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

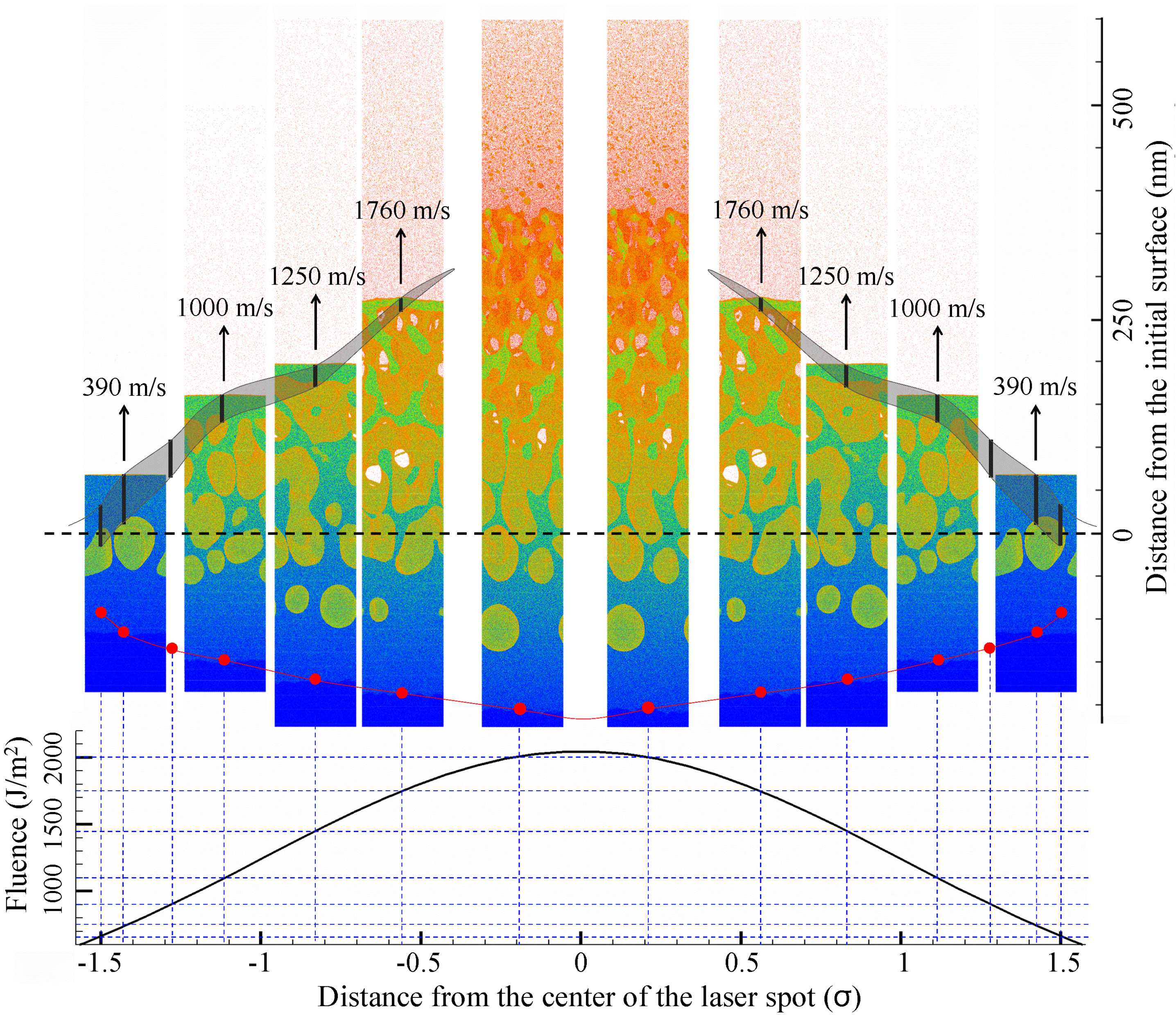




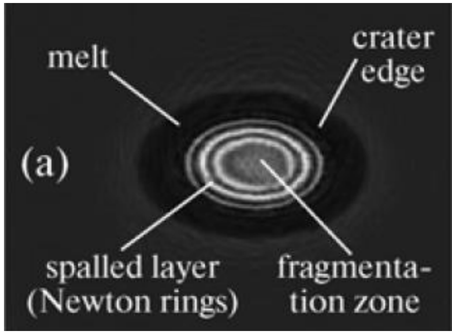
Resolidification of the foamy structure ➔ complex surface morphology

“Big picture”

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



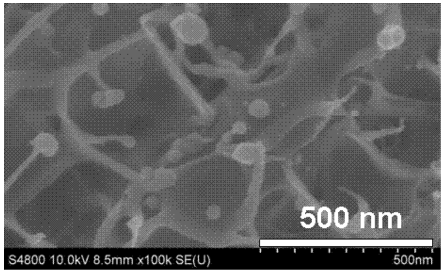
mechanisms of ablation & spallation



Ionin et al., JETP Lett. **94**, 753, 2011

cluster/nanoparicle size distributions

surface morphology



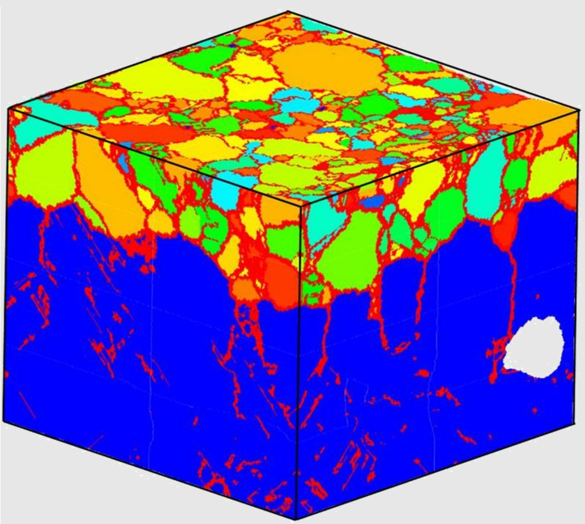
Zhao et al., *Optics Express* **15**, 15741, 2007

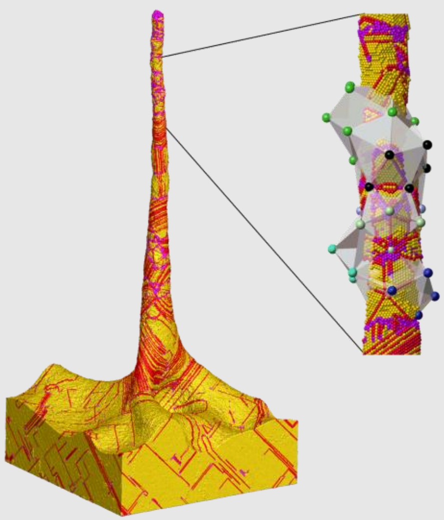
Summary

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

Thanks to OLCF
and all for your attention!