Atomistic modeling of atomic mixing and structural transformations in Ag film - Cu substrate systems irradiated by femtosecond laser pulses

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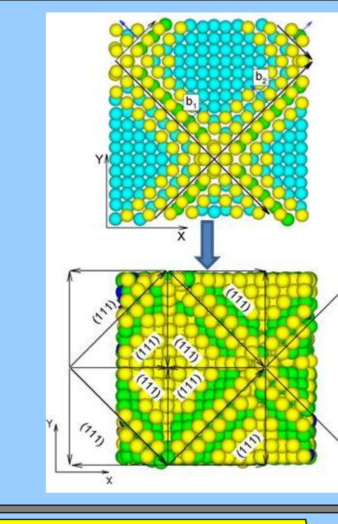
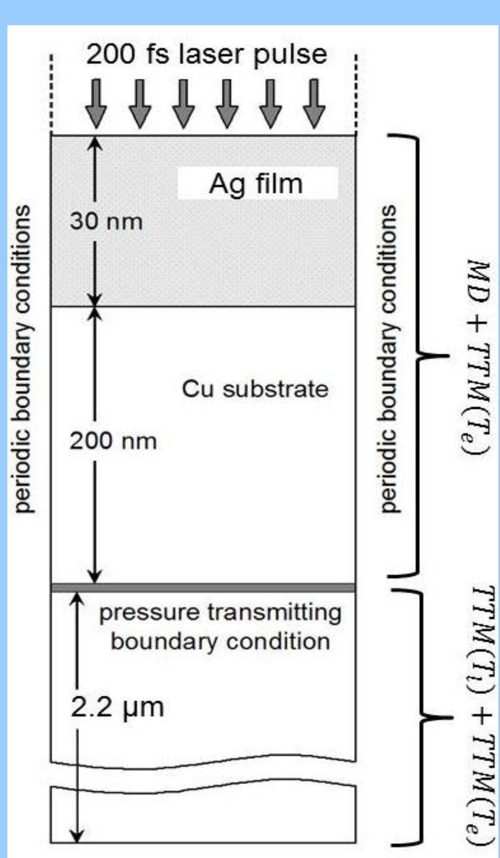


Introduction

The small size of the laser-modified zone (from 10s of nm to 100s of nm) and the short time of the melting-resolidification cycle (from 100s of ps to 10s of ns at fluences below the permanent damage threshold, e.g. [1-3]) makes experimental characterization of laser-induced structural changes challenging, but suggest a possibility for direct atomic-level modeling of processes involved in short pulse laser processing. In this poster, we report results of a series of molecular dynamics simulations aimed at investigation of the laser-induced phase transformations and atomic mixing in a target composed of a Cu substrate and a 30 nm Ag layer.

Computational model



- combined atomistic-continuum model: Classical molecular dynamic (MD) + two-temperature model (TTM) [4] is applied in the top 230 nm part of the system. Continuum model (TTM) is applied in deeper substrate to account for the heat conduction.

$$m_i \frac{d^2\vec{r}_i}{dt^2} = \vec{F}_i + \varepsilon m_i V_i^T, \qquad MD$$

$$C_e(T_e)\frac{\partial T_e}{\partial t} = \frac{\partial}{\partial z}\left(K_e(T_e)\frac{\partial}{\partial z}T_e\right) - G(T_e - T_l) + Laser\ energy\ deposition, \qquad TTM(T_e)$$

$$C_l(T_l)\frac{\partial T_l}{\partial t} = G(T_e - T_l), \qquad TTM(T_l)$$

- interatomic interaction: Embedded Atom Method (EAM) [5] gives good description of Ag-Cu system. The calculated phase diagram based on this potential agrees semi-quantitatively with experiment [8].
- electron temperature dependences of the electron-phonon coupling factor, electron heat capacity, and heat conductivity account for the contribution from the thermal excitation of d-band electrons [6].
- Pressure transmitting boundary condition is applied to avoid artifacts due to reflection of laser-induced pressure wave from the boundary of TTM-MD computational cell.

Melting and resolidification in Cu-Ag system

White dashed lines separate the film from the substrate

- Melting front
- Region shown in snapshots
- Pressure transmitting boundary

Note that $T_m(Ag) < T_m(Cu)$!

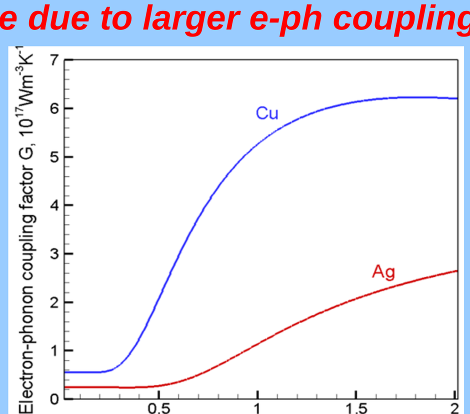
1100 J/m²

melting at the interface is mostly on the film side

homogenous nucleation of liquid region in the substrate, assisted by tensile stresses

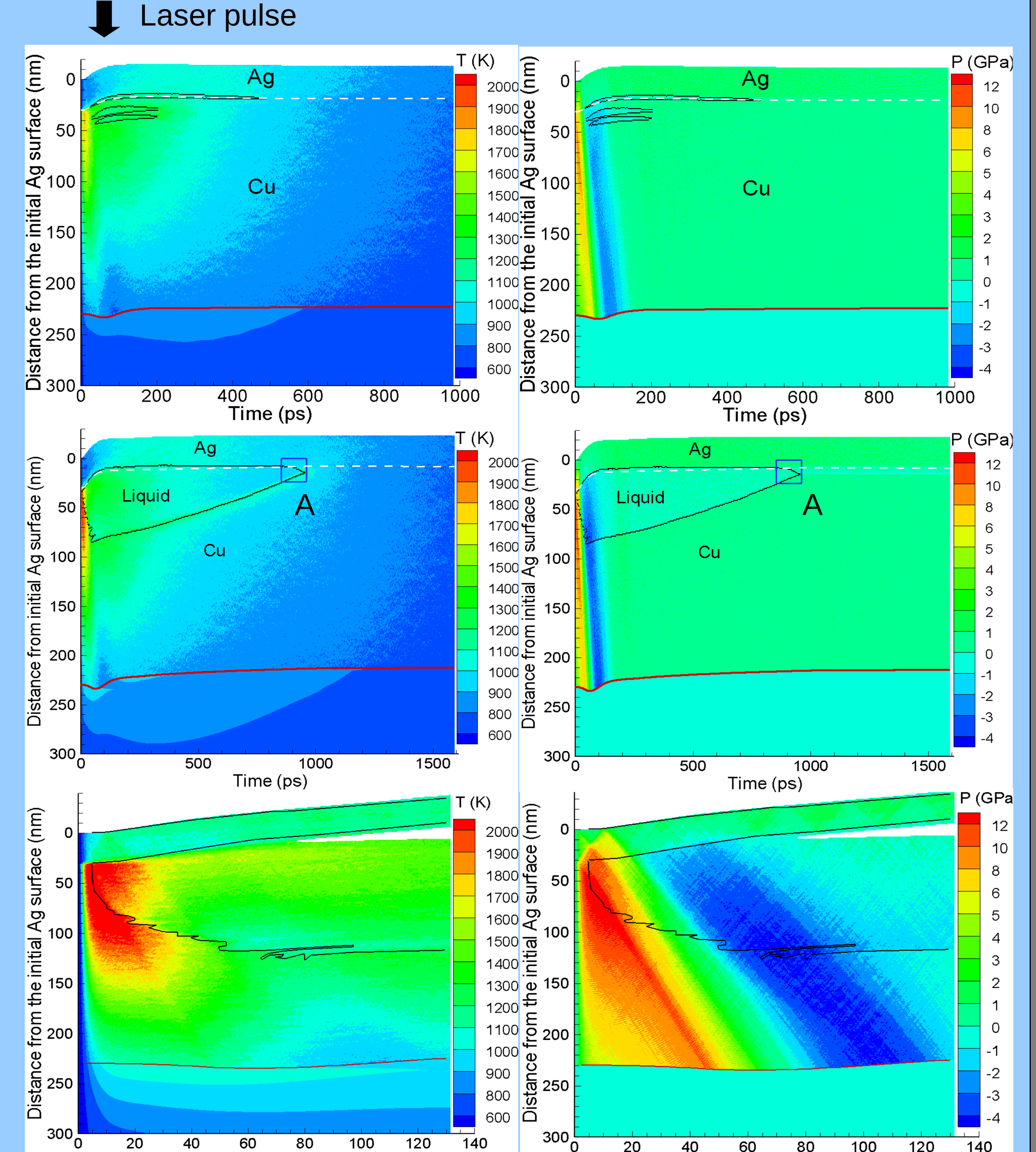
1300 J/m²

subsurface heating and melting of Cu substrate due to larger e-ph coupling in Cu



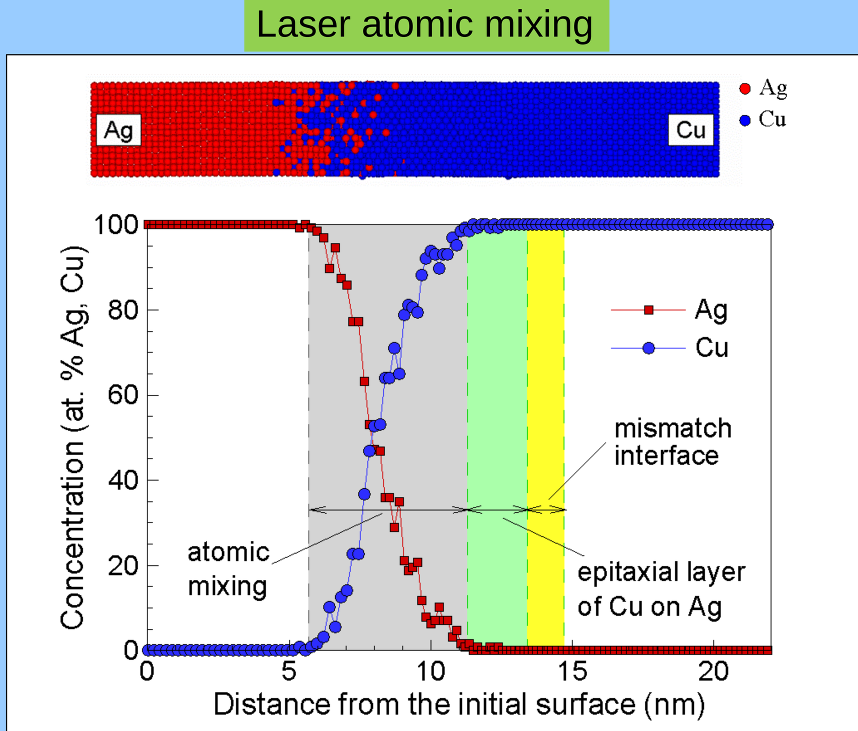
1600 J/m²

subsurface melting of Cu substrate and separation (spallation) of Ag film



Laser atomic mixing in Cu-Ag system

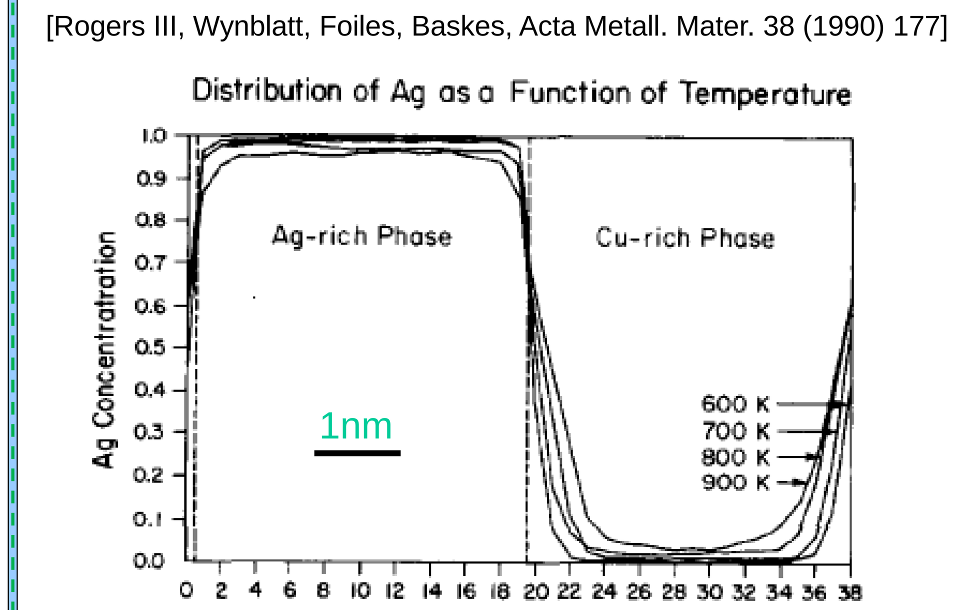
Laser atomic mixing



Equilibrium mixing

Monte Carlo simulation of atomic mixing at Cu-Ag (001) interface

[Rogers III, Wynblatt, Foiles, Baskes, Acta Metall. Mater. 38 (1990) 177]



laser atomic mixing >> mixing in the equilibrium configuration

References

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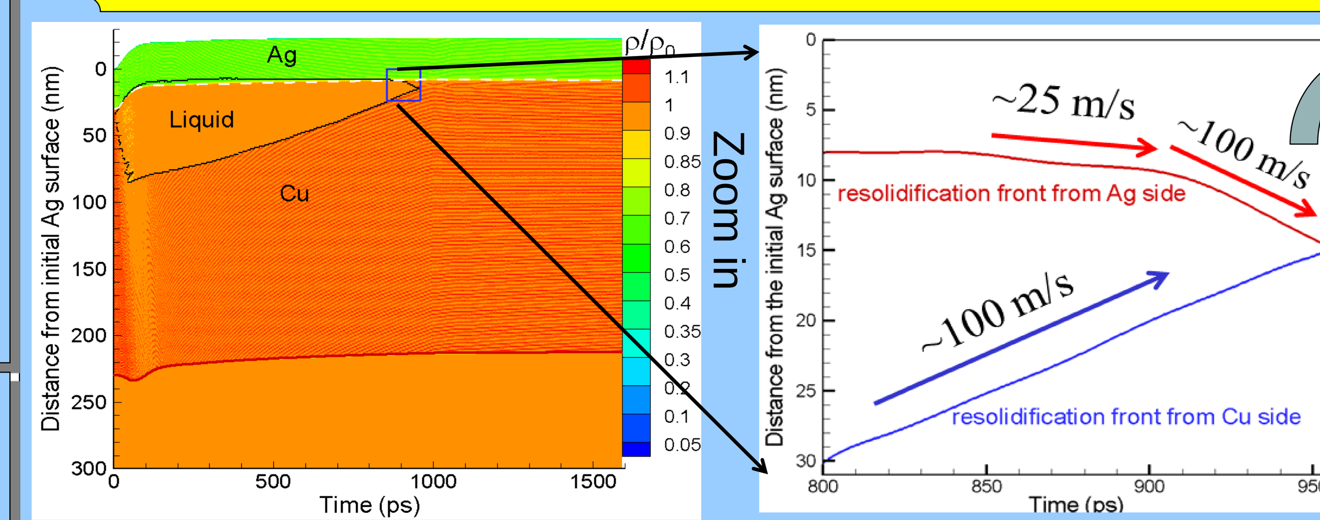
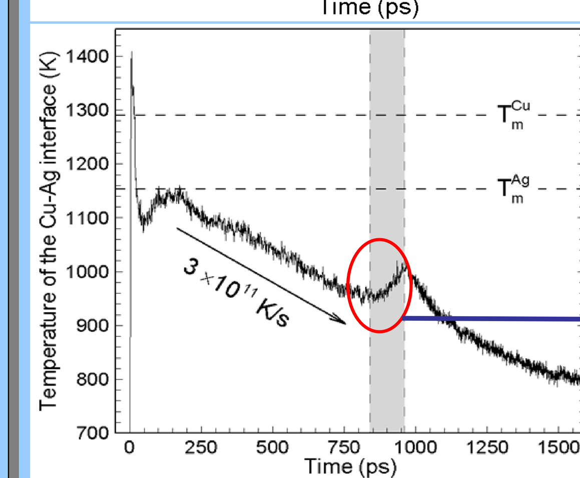
Epitaxial growth of Cu on top of Ag, 1300 J/m²

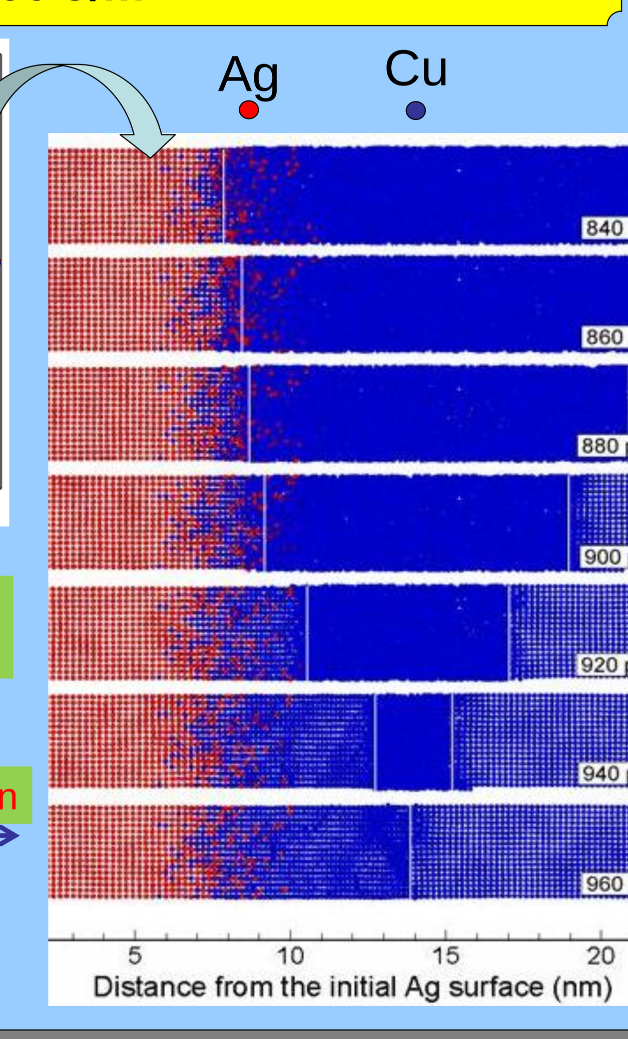




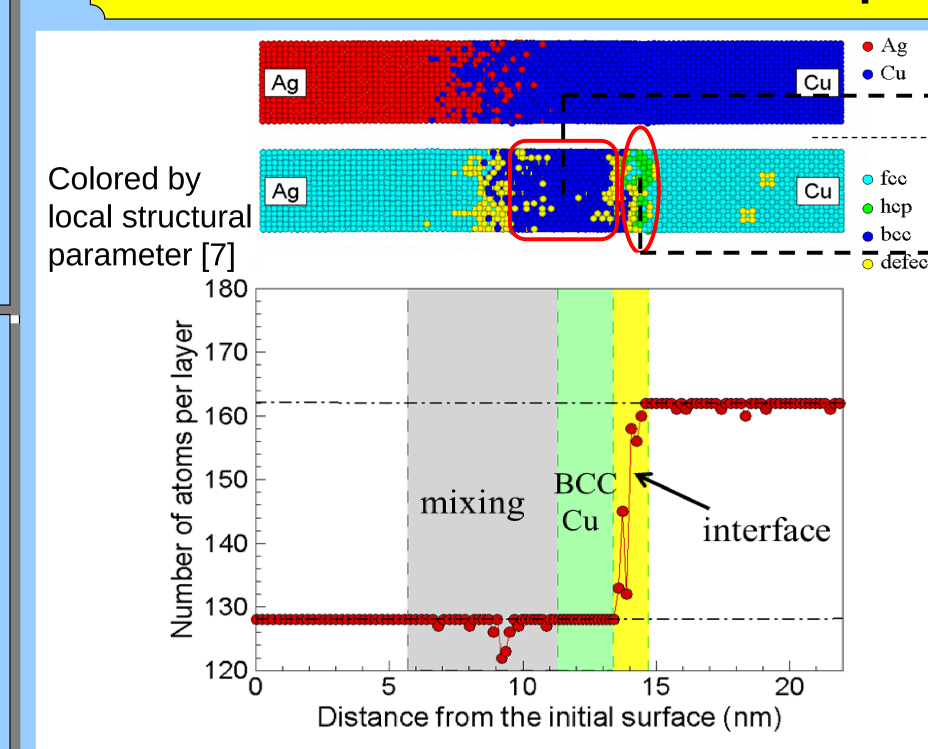
strong undercooling (down to eutectic temperature, ~950K, at the interface)

resolidification across atomic mixing region

epitaxial growth of Cu on top of Ag

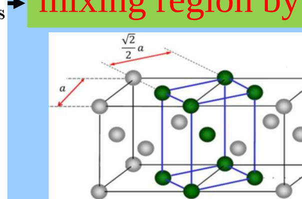


Structure of epitaxial growth region

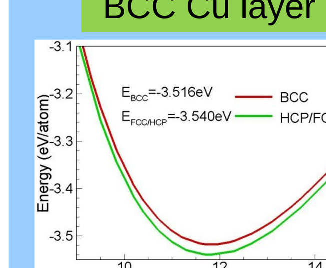


Due to epitaxial growth of Cu on top of Ag, 2 nm wide BCC Cu layer is generated

The new lattice mismatch interface is separated from atomic mixing region by the intermediate BCC Cu layer



BCC Cu layer



Good match between the lattice parameter of BCC Cu structure and the 1st neighbor distance in the FCC Ag structure:

BCC Cu: $a \sim 2.90$ Å

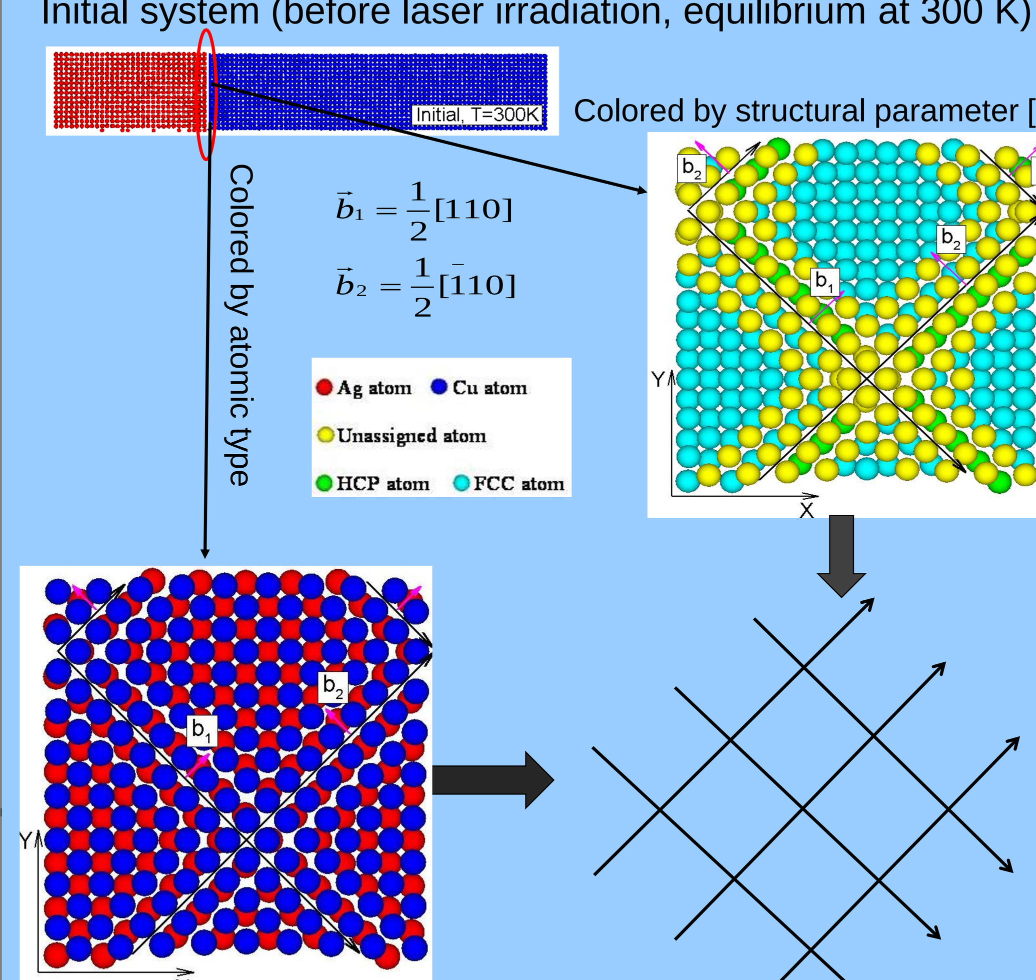
FCC Ag: 1st neighbor distance $= \frac{\sqrt{2}}{2}a \sim 2.89$ Å

Small energy difference between BCC Cu and FCC Cu:

The difference in cohesive energies predicted with FBD EAM potential is small, 22 meV/atom

Lattice mismatch interface (before and after laser irradiation)

Initial system (before laser irradiation, equilibrium at 300 K)

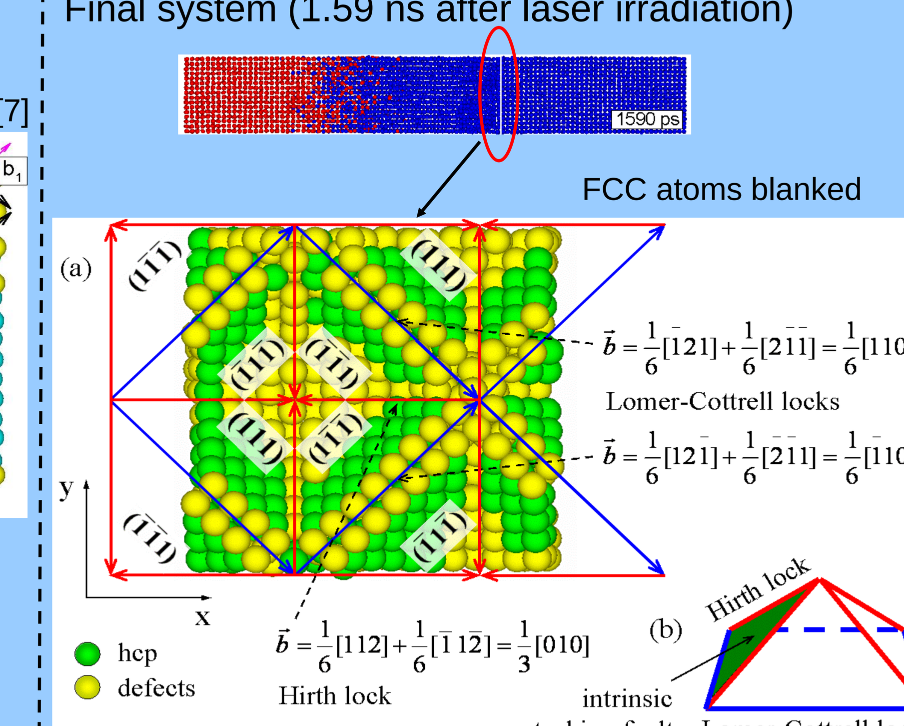


$\vec{b}_1 = \frac{1}{2}[110]$

$\vec{b}_2 = \frac{1}{2}[\bar{1}10]$

$8a_{Ag} \approx 9a_{Cu}$ → 2-dimensional misfit dislocation network at the interface

Final system (1.59 ns after laser irradiation)



The "runaway" lattice-mismatch interface: complex three-dimensional corrugated structure consisting of a periodic array of stacking fault pyramids outlined by stair-rod partial dislocations

The runaway lattice-mismatch interface and BCC Cu layer create a strong barrier for dislocation propagation, resulting in the effective hardening of the layered structure

Conclusions

- The relative strength of electron-phonon coupling of the Ag film and the Cu substrate defines the initial energy redistribution and the depth of the region undergoing transient melting/resolidification → sub-surface heating and melting of the Cu substrate.
- At low fluences, most melting occurs in the film part at the interface due to the lower melting temperature of the film material. At higher laser fluences, a separation (spallation) of the film from the substrate is observed for the Ag-Cu system and related to the interaction of the laser-induced stress wave with the weak film-substrate interface.
- Laser atomic mixing can occur in the region of ~4 nm for Ag-Cu system, much wider than the equilibrium mixing (<1 nm) at the interface.
- Due to strong undercooling (down to eutectic melting temperature at the mixing region), we observe resolidification across the atomic mixing region and thus epitaxial growth of Cu on top of Ag.
- The epitaxial growth of Cu on top of Ag results in the generation of a thin film (~2 nm) of BCC Cu layer, and the shift of lattice-mismatch interface deeper into Cu substrate.
- The "runaway" lattice-mismatch interface (between BCC Cu and FCC Cu) is found to have a complex 3-D corrugated structure consisting of a periodic array of stacking fault pyramids.
- The laser-induced BCC Cu layer and 3-D runaway lattice-mismatch interface are likely to present a strong barrier for dislocation propagation, resulting in the effective hardening of the layered structure.
- The detailed analysis of this work has been accepted to publish recently [9].