



Delineating the interplay effects of microstructure topology and residual stresses in ultrafast laser irradiated thin films

Hariprasath Ganesan ^a,* , Stefan Sandfeld ^{a,b}

^a Institute for Advanced Simulations – Materials Data Science and Informatics (IAS-9), Forschungszentrum Juelich, Juelich, Germany

^b RWTH Aachen University, Faculty of Georesources and Materials Engineering, Chair of Materials Data Science and Materials Informatics, Aachen, Germany

ARTICLE INFO

Keywords:

Thin films
Molecular dynamics
Microstructure
Residual stress
Ultrafast laser
Nanomanufacturing

ABSTRACT

Advanced nanodevices require high-precision machining of thin films using ultrafast lasers. However, thin-film fabrications cause variations in microstructure, crystallographic orientation, and residual stresses owing to coating conditions and substrate choice. This work investigates the complex interaction between these factors in ultrafast laser-irradiated gold (Au) thin films using a hybrid Two Temperature Model-Molecular Dynamics simulations. We realized microstructure-informed atomistic models with varying grain topologies (randomized vs. equiaxed), grain sizes, and residual tensile/compressive stress configurations. Our results reveal a clear hierarchy of influence on laser-metal interaction: (1.) Microstructure configuration (2.) Topology (3.) Grain Size (4.) Crystallographic orientations. In fine-grained thin films, grain boundaries act as primary melting precursors, while local crystallographic orientation determines the melting extent in coarser grains. Residual tensile stresses contribute to higher melting and greater laser-induced expansion than unstrained films. Conversely, residual compressive stresses resist deformation, as deposited thermal energy is utilized to overcome lattice compression, leading to reduced expansion. We found that microstructure grain topology and size exert a stronger fingerprint on film expansion than the initial defect density.

1. Introduction

Advanced nanomanufacturing techniques involving ultrafast laser processing [1] of metallic thin films has a range of applications, including nanostructuring, plasmonic/optical device fabrication, surface functionalization, and energy devices. Laser ablation is widely used as a nanoscale processing technique for controlled material removal. At the core of these techniques lies laser-metal interaction [2], which deposits high energy into the target material within ultrafast time (10^{-9} s). In metals, upon excitation of conduction-band electrons, the lattice subsystem is heated via electron–phonon interactions, leading to thermal transport phenomena that influence the thin-film microstructure.

Improving the quality and precision of such nanomanufacturing processes requires a deep understanding of laser-metal interactions [3]. On the one hand, finding optimal laser processing parameters (e.g., laser profile, pulse duration, intensity, etc.) remains challenging. On the other hand, accounting for variations in thin-film microstructure and thermodynamic state poses further bottlenecks. Investigations that oversimplify any of these aspects are prone to bias and lack physical realism.

The current understanding of the complexity of ultrafast laser-metal interaction is mainly possible due to some advancements in experimental imaging and characterization techniques, including time-resolved optical methods [4], ultrafast imaging of dynamics [5,6], ultrafast diffraction and spectroscopy, and transmission electron microscopy [7]. Together, these techniques advanced our knowledge in aspects like electron/lattice dynamics, phase transition, plasma characterization, spallation, subsurface melt flow, temperature fields, laser-induced defects, melting, and material removal. However, a holistic understanding of these processes remains challenging, and modeling efforts [8] have gained significant traction in recent times. In particular, a hybrid approach [9] combining the continuum two-temperature model (TTM) with molecular dynamics is particularly suitable because it can simulate ultrafast processes at atomic resolution.

For a comprehensive review of atomistic simulations of laser-driven nanomanufacturing and coupled TTM modeling works, refer [10,11]. Note that ultrafast laser-metal interaction encompasses both laser (energy) and target film (material) aspects. Until recently, however, several studies [12–15] have focused primarily on energy-related aspects: identifying optimal laser process parameters, such as laser intensity,

* Corresponding author.

E-mail address: h.ganesan@fz-juelich.de (H. Ganesan).

URL: <https://www.fzj.de> (H. Ganesan).

<https://doi.org/10.1016/j.jmrt.2026.07.122>

Received 21 May 2026; Accepted 5 July 2026

Available online 23 July 2026

2238-7854/© 2026 The Authors. Published by Elsevier B.V. This is an open access article under the CC BY license (<http://creativecommons.org/licenses/by/4.0/>).

profile, and pulse duration; thus, the insights remain limited. Also, some studies have focused on femtosecond laser ablation of metals and thin films, in particular [16–20]. Thin-film fabrication often involves physical and chemical vapor deposition techniques, resulting in variations in microstructure, residual stresses, and preferred crystallographic orientation. Thus, a rigorous physical understanding, including these contributions, becomes essential. Our recent work [21] yielded critical insights into how thin-film microstructures and crystallographic orientations influence laser-induced deformation. In particular, we showed that enhanced defects, such as dislocations, stacking faults, and grain boundaries, affect the ablation threshold in thin films. Consequently, the applied laser fluence results in varying degrees of laser-induced deformation [22].

To this end, this work addresses the research question of delineating the interplay between microstructural topology and residual stresses in thin films irradiated by an ultrafast laser, where *interplay* refers to their persistent, multi-stage coupling that is active before, during, and after laser irradiation, rather than their independent contributions considered separately. Atomistic simulation and modeling [23–25] enable efficient evaluation of these factors with and without incurring cross-effects. Therefore, this work encompasses a range of atomistic models with varying microstructural topologies (i.e., grain size and grain shape) and different coupled microstructure - residual stresses reported in as-deposited thin films. Section 2 discusses the Materials & Methods, modeling assumptions, and simulation procedure, and provides them in sufficient detail. Section 3 discusses the results from the TTM-MD simulation, focusing on 3.1 - the standalone variation of the thin-film microstructure and 3.2 - interplay of thin-film microstructure and residual stresses, where the coupled and non-independent nature of these two variables is systematically examined.

2. Materials and methods

TTM-MD approach and Interatomic Potential:

Classical Molecular dynamics (MD) simulates the collective behavior of atoms representing the material, evolving in time under given initial and boundary conditions. MD suits the spatio-temporal context of the laser-metal interaction process, i.e., atomic length (nm) and ultrafast time (fs) scale. However, irradiation of metallic thin films with ultrafast laser pulses results in a nonequilibrium thermal state consisting of hot electrons and cold lattice subsystems. Here, we combined the continuum two-temperature model (TTM) and atomistic MD to systematically simulate both short-time and long-time effects encountered during laser-metal interaction. For specific details on the theory, implementation of TTM-MD and related parameters, we refer the readers to the previous works, including ours [21,26,27].

Accordingly, eq. (1) indicates the continuum TTM part for describing the energy transport within the electron subsystem using the heat diffusion equation.

$$C_e \rho_e \frac{\partial T_e}{\partial t} = \nabla \cdot (\kappa_e \nabla T_e) - \gamma_p (T_e - T_a) + \gamma_s T_a + S \quad (1)$$

where C_e is the specific heat, ρ_e is the electron density, κ_e is the thermal conductivity, γ_p indicates the electron-phonon coupling coefficient, γ_s indicates the electron stopping coupling parameter, and $S = -I(t) \frac{\exp(-\frac{x}{l})}{l}$ refers to the applied laser source [28]. Herein, the electron temperature T_e dependence of both specific heat and thermal conductivity is considered [21]. We discretized the atomistic model spatially with grid cells of $120 \times 8 \times 8$ along the X, Y, and Z directions. The atomic part describes the ion dynamics with modified Newton's equation of motion, including a coupling term $\xi = \frac{\frac{1}{n} \sum_{k=1}^n \gamma_p V(T_e - T_a)}{\sum_i^N m_i v_i^2}$ for the energy exchange between the two subsystems [29].

$$m_i \frac{d^2 \mathbf{r}_i}{dt^2} = \mathbf{F}_i + \xi m_i \mathbf{v}_i \quad (2)$$

where m , $\mathbf{r}$ and $\mathbf{F}$ indicates mass, position, and force of atom i , respectively. In the coupling term ξ , n indicates the finite difference integration step within each MD step, whereas N and V indicate the number of atoms and the volume of the grid cell with fitted parameters and thermo-physical quantities [21,27].

Embedded Atom Method (EAM) potentials [30] are parameterized functions that are fitted to capture the material properties of target metals and alloys. Herein, Au has an electronic configuration with both delocalized s electrons and localized d electrons. For a faithful simulation of the laser ablation process, it is important to capture the electron temperature dependence on the physical properties of the laser ions. To this end, we chose the Norman electron temperature-dependent potential [26] for the target Au thin-film system.

Atomistic modeling of thin films: Thin films refer to solid material layers ranging from a few nanometers to micrometers in thickness, often fabricated using physical or chemical vapor deposition techniques. Here, we realized atomistic Au thin-film models coated on a pseudosubstrate (i.e., vacuum), representing free-standing thin films to capture the sole contribution of material aspects under ultrafast-laser irradiation.

Fig. 1(a) shows the schematic of the generalized atomistic model setup for the free-standing Au thin film considered in this work to investigate ultrafast laser-metal interactions. Herein, the applied ultrafast laser pulse irradiates the whole surface of the thin film, indicated by red arrow markers. Furthermore, we considered two categories of Au thin film models: (b.) effects of varying microstructure (i.e., **grain topology** and **granularity**) and (c.) effects of combined variation in microstructure and residual stresses. Herein, the initial unstrained atomistic models have comparable dimensions of $60 \text{ nm} \times 28 \text{ nm} \times 28 \text{ nm}$ (along the X, Y, and Z) made of $\approx 1.6 \times 10^6$ Au atoms existing in a face-centered cubic (fcc) configuration with $a = 4.08 \text{ \AA}$ (see Fig. 1 a). Unlike typically assumed rod-shaped atomistic models, we considered topologically affine Microstructure-Informed Atomistic Models (MIAMs) [21,31,32] to incorporate microstructure variations in sufficient detail. However, to keep the computational load tractable, we applied periodic boundary conditions in the lateral directions Y and Z, with a free boundary condition in the X direction to allow film expansion. The immobile zone at the bottom film/vacuum interface represents a region with restricted atomic mobility to mimic the substrate's physical constraint. Owing to the choice of substrate and coating conditions, the deposited thin films are susceptible to variation in microstructure, crystallographic orientations, and residual stresses. Accordingly, we considered thin-film [33,34] with microstructure variation (see Fig. 1 b) with **grain topology**: Single Crystalline (SC), i.e., $(-1 \ 1 \ 0)$, poly-nanocrystalline with randomized and equiaxed configurations and **granularity**: (coarser (12 Grains), moderate (24 Grains), and fine-grained variants (48 Grains) of both randomized and equiaxed configurations. Here, we started with a modeled thin film rather than simulating the deposition process; thus, the roughness parameter is not significant. All the model constructions were realized using Atomsk [35] and in-house codes. To this end, the constructed free-standing atomistic Au thin-film models encompass a range of microstructures, including grain topology and grain-size distributions.

Preparation Simulation: We performed energy minimization of these atomistic thin-film models, followed by pressure and thermal equilibration at 0 bar and 300 K. A short microcanonical dynamic simulation of such equilibrated models ensured the right starting conditions. Later, to systematically capture the contributions of residual stress, we numerically pretrained thin films (i.e., as pre-tensioned and pre-compressed models as shown in Fig. 1 c) to various extents under uniaxial tension and compression along the X-direction at a strain rate of $10^9/\text{s}$. It is worth noting that such a applied strain rate is a physical necessity of the MD methodology, arising from the femtosecond timestep constraint that confines the accessible simulation time to the picosecond-to-nanosecond scale, and falls well within the range of 10^8 to $10^{10}/\text{s}$ consistently reported in peer-reviewed atomistic studies of

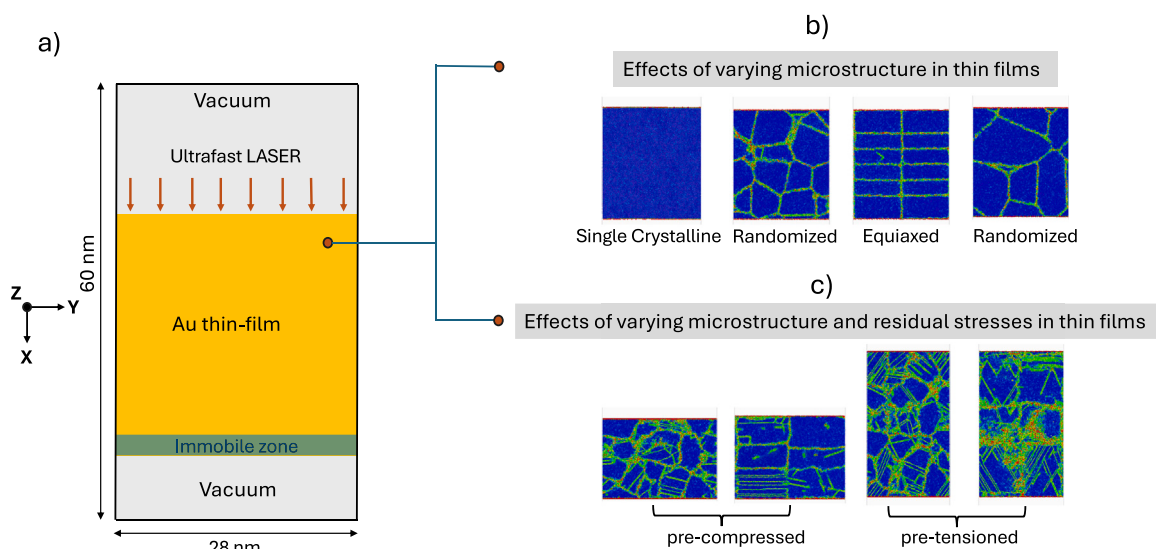


Fig. 1. (a.) Schematic of the atomistic model setup of Au thin-film irradiated by the ultrafast laser pulse. **Consideration of two categories of thin films (only selected models are shown here for brevity):** (b.) Effects of varying microstructure: grain size (coarser vs. finer) & topology (Single Crystalline, equiaxed, randomized), (c.) Effects of varying microstructure and residual stresses.

mechanical deformation [24,31,36]. Note that such prestrained models represent thin films with varying degrees of residual stress (i.e., tension or compression), as typically reported in experiments along the lateral directions. Our simulation models take the normal direction (i.e., X) for residual stress consideration. We assumed periodic boundary conditions in all directions for both equilibration and prestraining simulations along with a timestep of 1 fs.

Process Simulation: To simulate ultrafast laser irradiation and microstructure evolution in thin films, we adopted a two-stage approach following the electron-ion equilibration time [26,37] with further justification following the earlier works [21,38], i.e., stage-1 with TTM-MD and stage-2 with MD, along with chosen parameters. Accordingly, in stage-1, we simulated uniform ultrafast laser irradiation using a hybrid TTM-MD approach (see Fig. 1 a) for 30 ps with the following laser parameters (pulse duration, $\tau = 100$ fs, wavelength, $\lambda = 400$ Å, and laser penetration depth, $l_{skin} = 20$ Å) [21]. Then in stage-2, we simulated microstructure/thermodynamic evolution using MD for another 70 ps to capture heat transfer, melting, and phase transformation.

Herein, we applied the open-source software LAMMPS [28] (stable version August 2023) for all classical MD and hybrid TTM-MD simulations. We visualized the atomistic configurations and performed relevant characterization/analysis (i.e., Centro-symmetry parameter (CSP), phase transformation, and displacement analysis) using OVITO [39]. Herein, CSP measures the local lattice distortion of each atom's neighborhood with reference to the defect-free fcc crystalline Au. Consequently, unperturbed atoms have lower CSP values close to zero (i.e., blue), and atoms undergoing lattice distortion deviate significantly from the reference and thus exhibit moderate (turquoise) to high (red) CSP values. Thus, CSP aids in characterizing whether the atom is part of a perfect lattice, a local defect (e.g., a dislocation or stacking fault), or located at a surface, and remains an effective tool for tracking deformation in crystalline thin films. In this work, the term 'high CSP volume fraction' refers to the instantaneous percentage of atoms with CSP above 2.95, indicating localized loss of crystallinity.

3. Results and discussions

This section discusses the results of Au thin films with varying microstructure, residual stresses, and their interaction during ultrafast laser metal interaction, following the two-stage simulation protocol described in Section 2.

3.1. Effects of varying microstructure in thin films

To this end, we irradiated different thin film atomistic models with varying microstructural features, namely, grain topology (random vs. equiaxed) and granularity (coarser vs. finer), all evolving under a comparable applied laser fluence. We computed the centrosymmetry parameter (CSP) for all atoms in unstrained thin-film models with varying microstructure during and after irradiation with an ultrafast single-pulse laser with an applied fluence of 1.28 J/cm^2 , as we are focusing below the ablation threshold [21].

At 0 ps, the defect-free SC thin-film with $(-1 \ 1 \ 0)$ orientation showed 98% of atoms with zero CSP values except the surface (see Fig. 2). Following laser irradiation, the thin-film underwent heating and expansion around 50 ps, with increasing lattice distortion resulting in stacking faults around 100 ps. Eventually, such defect-starved thin films exhibited minimal heterogeneous melting confined to the surface, as most of the laser-deposited thermal energy was utilized to overcome the configuration's cohesive energy, resulting in negligible laser-induced deformation.

To capture thin-film grain-size sensitivity, we considered a poly-NC configuration with varying granularity of 12 (coarser), 24 (moderate), and 48 (finer). Unlike the SC thin film, the poly-NC configuration showed relatively more grain-boundary-mediated melting. In particular, the fine grain configurations (i.e., 48 grains) exhibit more melting regions owing to high grain boundary volume fraction. However, for coarser grains, the influence of local crystallographic orientation becomes dominant, leading to spatially varying melting extents. In comparison, equiaxed fine grained thin films recorded extensive melting than randomized poly-NC counterpart, emphasizing the combined role of grain topology and grain size.

Fig. 3 shows the temporal evolution of atoms with high CSP¹ and FCC volume fraction for unstrained Au thin-film with varying microstructure. Herein, the SC configuration started from nearly 0% and rose to 50% and fluctuated slightly owing to tension and compression stages, whereas all poly-NC configurations showed a nonlinear increase in high CSP volume fraction, among which equiaxed configurations with (finer grains) recorded relatively higher CSP volume

¹ for calculation, only atoms with CSP > 2.95 were considered, indicating loss of crystallinity.

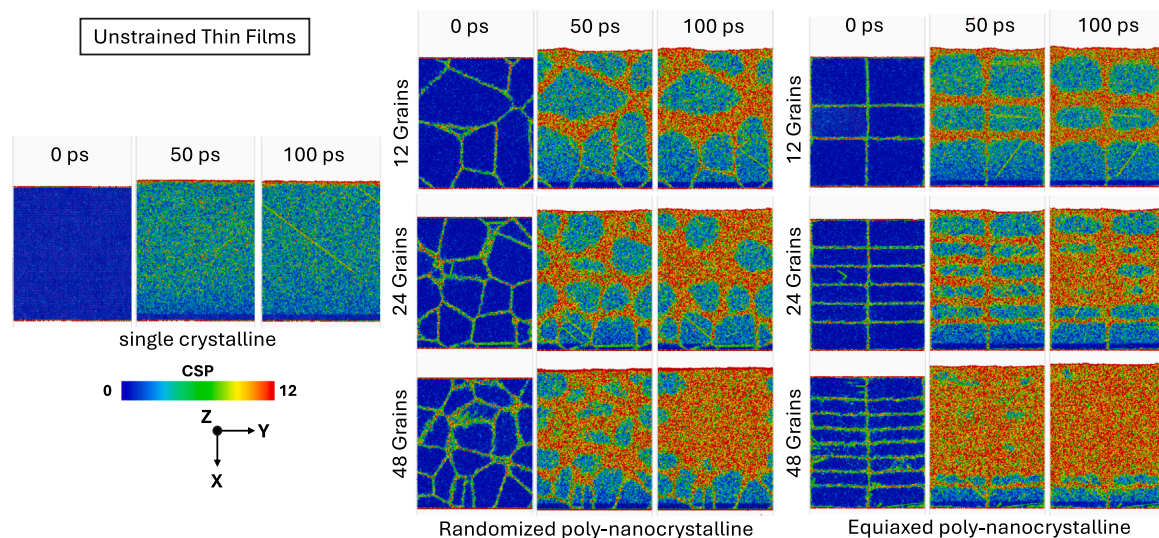


Fig. 2. Comparison of Au thin-film microstructure effects: single crystalline and poly-nanocrystalline models following ultrafast laser irradiation at selected time steps. Atoms are color-coded after the centro-symmetry parameter (CSP), indicating that the atom with zero CSP is part of a perfect lattice, with non-zero CSP as a part of a defect (e.g., a dislocation, stacking fault, surface).

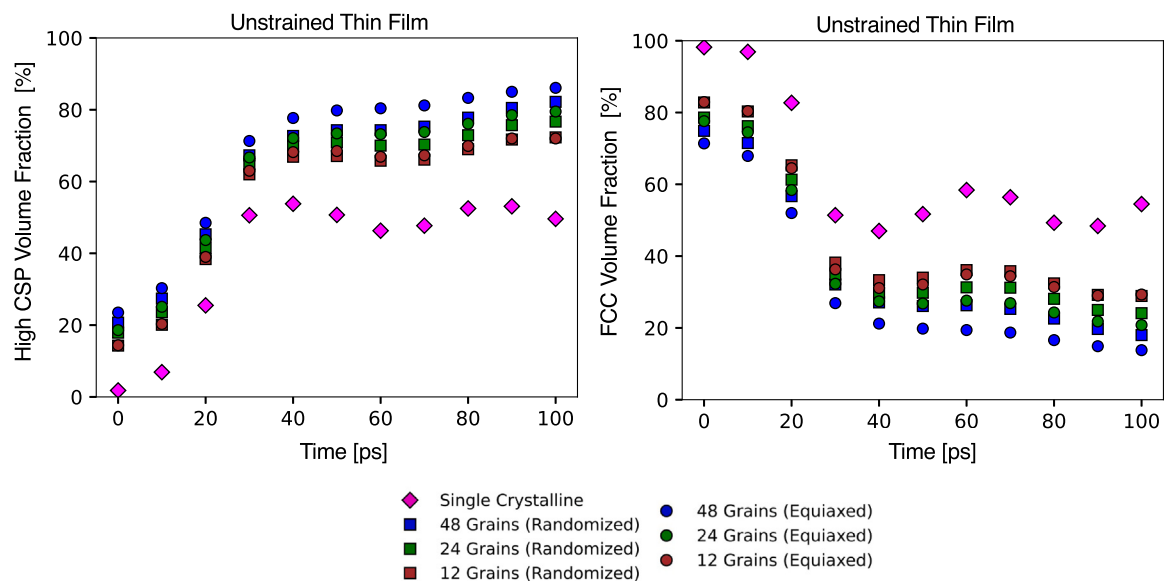


Fig. 3. High CSP and FCC volume fraction evolution for unstrained thin-film with varying microstructure irradiated by ultrafast laser pulse.

fraction throughout in comparison to the randomized configuration. The FCC volume fraction decreased continuously, confirming loss of crystallinity due to melting. For both high CSP and FCC volume fraction, single-crystalline thin films showed considerable fluctuations due to the laser-induced pressure wave, resulting in compression and subsequent tension. In contrast, both randomized and equiaxed poly-NC configurations with different granularity showed nearly monotonous increase (high CSP) and decrease (FCC) trend due to faster melting. However, coarser 12 grains poly-NC thin films showed almost negligible fluctuations. These results emphasize the critical role of initial grain boundary volume fraction vs. crystalline region in accommodating laser-induced stresses and the corresponding phase volume fraction change. Thus, for consistent ablation behavior, a fabricated thin film with fine-grained microstructure is deemed more suitable, owing to its high grain-boundary volume fraction.

Fig. 4 shows the spatio-temporal distribution of atomic pressure for unstrained thin films measured following ultrafast laser irradiation. Herein, SC thin-film showed an extensive pressure-free region

measuring around 0 GPa, whereas the thin-film with randomized poly-NC microstructure showed some islands of mild compression (red) and tension (blue) regions. For brevity, only fine-grained poly-NC configurations as representations are shown here. Comparing the initial distribution (at 0 ps) captures the spatial pressure-gradient pattern arising from the sensitivity of underlying microstructural changes prior to laser irradiation. Consequently, equiaxed poly-NC thin films result in regions of extreme compression and tension. Our results revealed that variation in thin-film microstructural features (i.e., grain topology and grain size) strongly influenced the initial pressure distribution, thereby delineating the microstructural fingerprint's contribution to subsequent pressure evolution following ultrafast laser-metal interaction. In general, for SC thin films, the lack of defects and grain boundaries leads to slower compression and subsequent heating. In contrast, poly-NC thin films exhibit faster compression and tension cycles, characterized by large variations in field pressure magnitude (between 30 and 100 ps) and by global pressure plots.

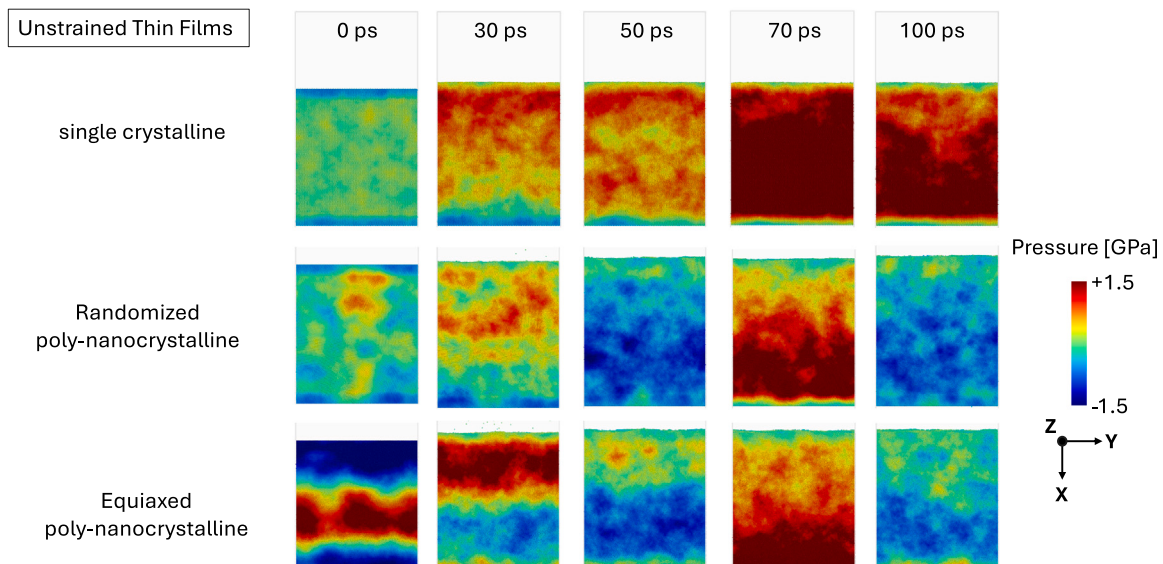


Fig. 4. Spatio-temporal pressure plot comparison for unstrained thin films. For the representation of poly-nanocrystalline configurations, we considered only thin films with 48 grains (i.e., finer).

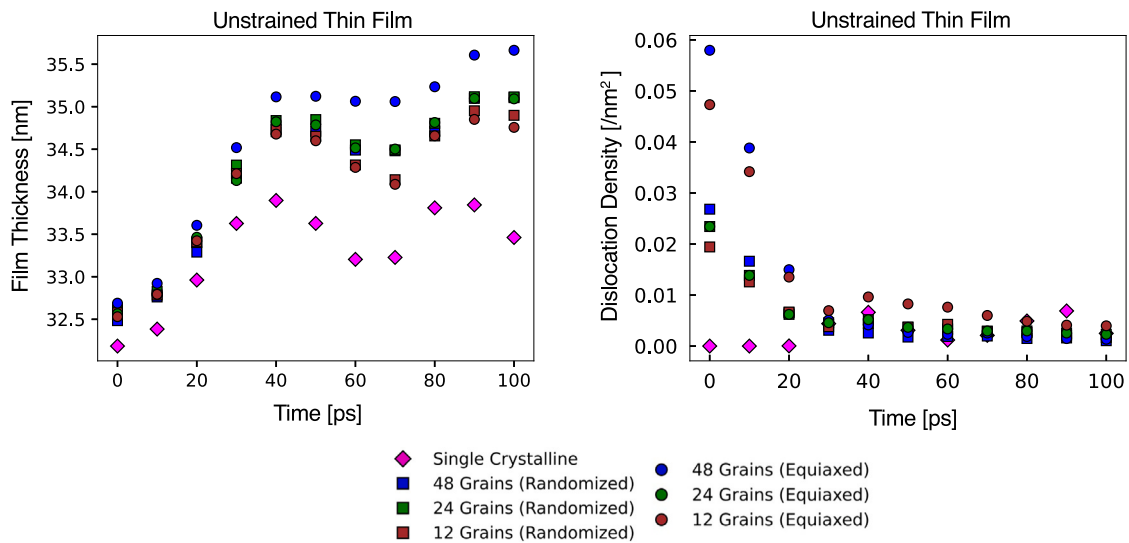


Fig. 5. Unstrained Film thickness and dislocation density evolution under the fluence of 1.281 J/cm².

Furthermore, we quantified geometric variation by characterizing the evolution of film thickness (see Fig. 5) for different thin-film models with varying microstructures irradiated by an ultrafast laser pulse. For SC models, the thin-film thickness increased smoothly up to 40 ps and then fluctuated strongly due to compression and tension cycles, reaching a maximum thickness of 1.9 nm. On the other hand, poly-NC thin films showed relatively higher expansion with mild fluctuation (≈ 0.5 nm). Altogether, fine-grained (i.e., 48 grains) equiaxed poly-NC thin films showed greater film thickness expansion (≈ 3.1 nm), suggesting that the combination of grain topology and size/granularity plays a decisive role. Additionally, in only extreme cases did preexisting defects make a weak contribution to the thin-film expansion following laser irradiation. For instance, the SC thin film, which initially had zero dislocation density, resulted in the lowest thin-film expansion compared to the equiaxed thin-film poly-NC, which started with the highest dislocation density ($0.058/\text{nm}^2$). Most importantly, the distribution of initial dislocation density for other poly-NC thin films did not show a strong spread ($\approx 0.04/\text{nm}^2$) across final film thicknesses, indicating that microstructure topology and size have a stronger fingerprint than initial defect density.

Thus, from our extensive simulations, we propose the following hierarchy of influence for thin-film microstructure:

1. Microstructure configuration (SC vs. Poly-NC)
2. Microstructure topology (Random/Equiaxed)
3. Granularity (coarser vs. finer)
4. Crystallographic orientations

It is worth emphasizing that in our earlier work [21], we clearly demonstrated the effects of grain-boundary-mediated melting (exhibiting a significant heterogeneous melting profile) in Au thin films.

3.2. Interplay of thin-film microstructure and residual stresses

Experimental investigation [40] characterized and validated the existence of residual stresses originating from the tensile or a compressive state of thin films. To delineate such residual stress contributions in thin films, we mechanically prestrained thin-film models and compared them with the unstrained (as-deposited) configuration.

It is worth noting that the pre-straining procedure for thin films modifies the microstructural state prior to laser irradiation by nucleating stacking faults, altering the average grain size due to lateral compression, and increasing the initial dislocation density at grain boundaries. Therefore, microstructure and residual stress are not independent variables in this study, as the imposed stress state reshapes the microstructure, analogous to what occurs during thin-film fabrication processes. Consequently, the thin-film configurations entering laser irradiation in this subsection exhibit a coupled microstructure and stress fingerprint, distinct from the unstrained baseline discussed in Section 3.1. Importantly, this coupling is not confined to the pre-irradiation stage alone, but continues to govern how the deposited laser energy is utilized, how defects nucleate and evolve, how the pressure field distributes and relaxes, and ultimately how much the thin film expands or resists deformation. It is this persistent, multi-stage coupling between microstructure and residual stress, active before, during, and following laser irradiation, that we refer to as *interplay*.

For an SC thin-film, the tensioning procedure led to the nucleation of stacking faults (see white arrow markers in Fig. 6), characterized by several activated parallel slip planes. Following ultrafast laser irradiation, around 50 ps, we observed considerable thin-film expansion, with inhomogeneous melting confined to the surface. Locking of cross-gliding planes results in high local lattice distortions, which facilitate melting. Consequently, at 100 ps, some melted regions (see white circle in Fig. 6) in isolation existed deeper inside the thin-film away from the surface. In contrast, during pre-tension, both randomized & equiaxed poly-NC thin films resulted in relatively fewer stacking faults (see Fig. 6 at 0 ps). To a large extent, both grain size and local crystallographic orientation determine the extent of stacking faults, as grain boundaries can hinder their propagation. Unlike unstrained poly-NC thin films (see Fig. 2), around 50 ps, we observed a slightly higher film expansion even for coarser grains characterized by grain boundary mediated melting. Evidently, around 100 ps, equiaxed poly-NC thin-film showed a greater extent of melting than a randomized microstructure. It is worth noting that prestraining resulted in a smaller average grain size due to lateral compression, besides nucleating new dislocations from the grain boundaries, thus, high initial defect density. In contrast to unstrained cases, residual tensile stresses in all thin films with varying microstructures showed higher melting and laser-induced deformation. This observation emphasizes the complex combined contribution of grain topology, grain size, residual stresses, and initial defect concentration.

In unstrained thin films, the initial high CSP volume fraction stems solely from microstructural features, whereas in tensioned thin films, deformation-induced defects also contribute to high CSP. Eventually, the SC thin films start from slightly high CSP values (27%) and remain around 70% after 40 ps. In contrast, finer equiaxed poly-NC thin films showed a continuously increasing high CSP volume fraction reaching a maximum of slightly under 100%. Altogether, the poly-NC thin films showed a tight spread, indicating that the pretensioning procedure contributed significantly to laser-induced lattice distortion. In general, the FCC volume fraction showed a monotonically decreasing trend (*consistent inverse trend when compared with high CSP*) for all tensioned thin films (see Fig. 7). In particular, the fine-grained equiaxed microstructure resulted in nearly complete melting, whereas the SC thin film fluctuated around 30% after the initial drop.

Pre-compressed thin-film with SC microstructure yielded moderate defect densities, with several short glided slip planes that led to locking due to cross gliding. Such locking junctions served as hotspots, leading to lattice distortions characterized by high CSP values. Following ultrafast laser irradiation, these thin films exhibited negligible thin-film expansion and heterogeneous surface melting. However, the randomized poly-NC thin-film exhibited a slightly modified grain topology, with some grains exhibiting relatively more stacking faults due to favorable crystallographic orientations (see coarser equiaxed Poly-NC thin film in Fig. 6). Across different pre-compressed fine-grained polycrystalline thin films (both randomized and equiaxed), grain boundary-mediated

melting was more pronounced. In our previous investigation [21] of ultrafast laser irradiation on unstrained thin films with varying microstructures, we observed an initial thin film compression to resist laser-induced lattice straining, followed by tension as a precursor to melting. Consequently, pre-compressed thin films tend to further reduce laser-induced thin-film expansion, as the laser-deposited thermal energy gets utilized to overcome lattice compression. Our results reveal that residual compressive stresses in thin films strongly reduced the extent of laser-induced deformation relative to thin-film microstructural features, namely, grain size and topology.

The pre-compressed poly-NC configurations showed a comparable initial high CSP volume fraction (0 ps) that increased gradually following laser irradiation, reaching a maximum ranging from 65–80%, whereas the SC film saturated around 50% after 30 ps. Despite starting from high FCC volume fractions (at 0 ps), the poly-NC thin films gradually declined, reaching a minimum of 23%. Altogether, these observations reveal that residual stresses in pre-compressed thin films exhibit strong resistance to laser-induced deformation; thus, optimization of nanostructure ablation process parameters with ultrafast lasers should account for any residual stresses in thin films.

In Fig. 8, after pre-tension, both SC and poly-NC thin films exhibited a distribution of strong and moderate tension (blue) regions, along with some islands of compression (red) regions. Following ultrafast laser irradiation, both poly-NC thin films underwent compression and tension cycles for up to 50 ps, after which the pressure intensities decreased considerably. Dedicated experimental investigations [41–43] on laser irradiation of metallic thin films agree with our observations of laser-induced compressive stresses due to ultrafast heating, followed by structure change accompanied by pressure relaxation resulting in breathing of thin films. Specifically, our previous work [21] on Au thin films revealed that defect nucleation mechanisms like cavitation, dislocations, and planar defects act as pressure-relieving mechanisms based on microstructure-dependent pressure threshold, resulting in pressure drop. At 100 ps, the randomized poly-NC thin film exhibited slightly higher compressive values than the equiaxed counterpart due to incomplete melting. Following precompression, different thin films with varying microstructures (at 0 ps) showed a similar distribution of high- and moderately compressed regions. Interestingly, SC thin films retained predominantly high compression regions following laser irradiation. On the other hand, randomized/equiaxed poly-NC thin films showed a weakly fluctuating tension and compression regions until (50 ps), followed by a moderate distribution of compressive stresses around 100 ps in thin films. Unlike pre-tensioned thin films, no strong pressure intensities were observed herein. These observations emphasize how thin-film residual stresses influence laser-induced pressure and targeted machinability.

For an unstrained SC thin film, the peak film thickness expansion was 2.0 nm. In contrast, the pre-tensioned and pre-compressed SC thin films showed considerably higher and lower peak expansions of roughly 4.5 nm and 1.6 nm (see Fig. 9), respectively. Note that for the pre-tensioned case, both poly-NC thin films started at slightly lower values than in the SC case (at 0 ps) and then reached a peak around 60 ps, after which they declined moderately. Among the poly-NC thin films, the fine-grained equiaxed case recorded a peak film expansion of nearly 5 nm in contrast to the fine-grained randomized case showing 3.4 nm. Following precompression, both randomized and equiaxed poly-NC thin films showed a gradual increase in thickness, with mild fluctuations, eventually reaching an average peak of 1.5 nm. The distribution spreads over time after 40 ps. Comparing the two pre-strained samples, the finer equiaxed polycrystalline thin films (i.e., 48 grains) exhibited the highest film thickness, underscoring the role of grain topology and grain-size contributions in thickness evolution. Note that the prestraining procedure resulted in different initial thicknesses (± 10 nm of unstrained film thickness as reference) of thin films that accommodate residual stresses, irradiated by an ultrafast laser pulse at a comparable applied laser fluence. [44] performed experimental and

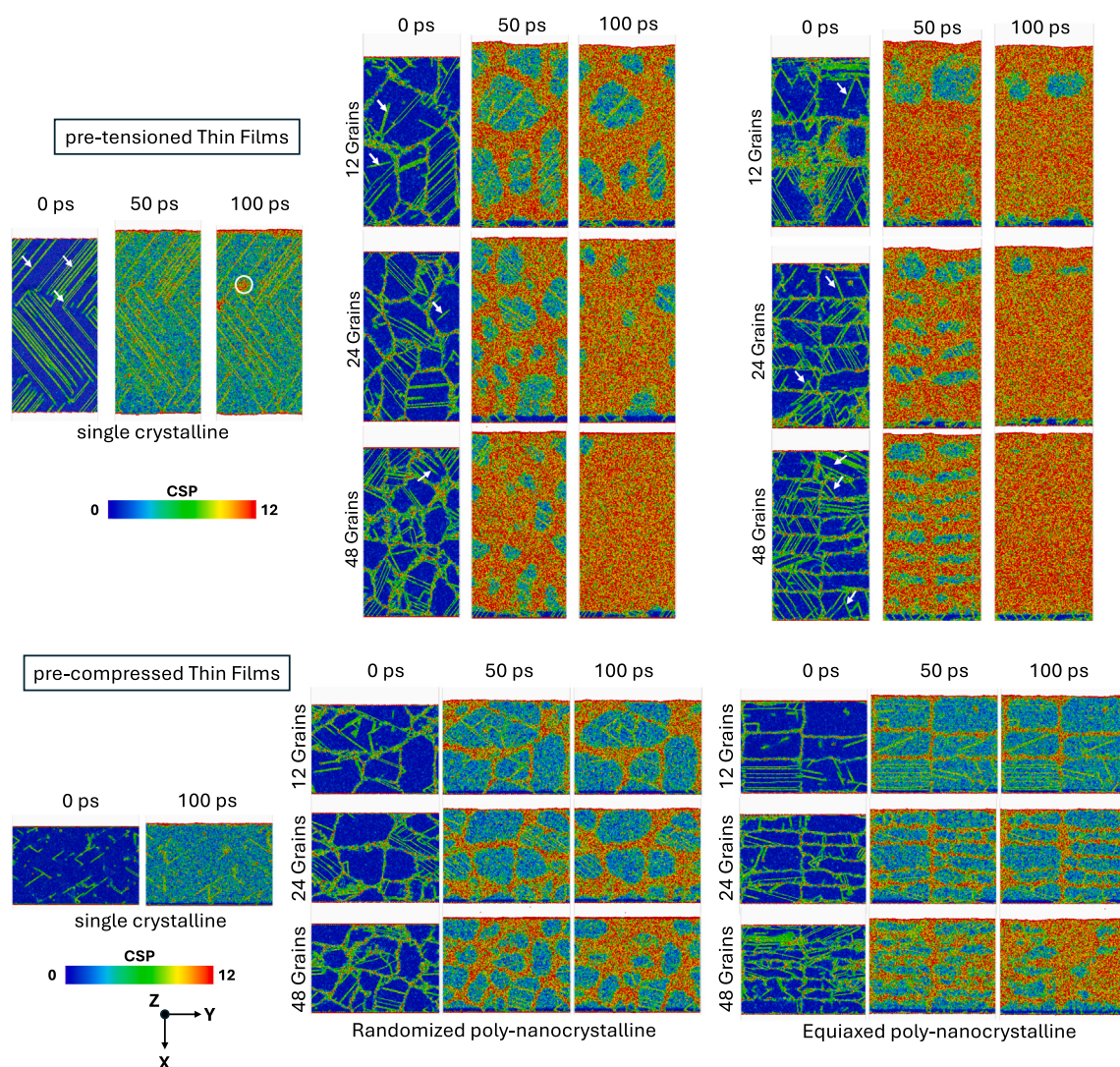


Fig. 6. Comparison of prestrained (Tension vs. Compression) single crystalline and poly-nanocrystalline microstructure models irradiated with applied laser fluence. Atoms are color-coded after the centro-symmetry parameter (CSP), indicating that the atom with zero CSP is part of a perfect lattice, with non-zero CSP as a part of a defect (e.g., a dislocation, stacking fault, surface). The white arrows indicate stacking faults, and the white circle denotes several isolated melting regions.

numerical TTM-MD simulations of gold films with different thicknesses and reported a strong correlation between the separation threshold and increasing film thickness. However, these findings did not account for residual stresses in thin films. Often, ultrafast laser irradiation of metallic thin film results in a spatial distribution of varying absorbed laser energy intensities (i.e., low to high) [34,45]. Our previous study on the Au thin-film system revealed that for the lower applied fluence (1.281 J/cm^2 (lowest) laser-induced lattice distortions reduced crystal stability, a precursor to grain boundary mediated melting, and for the higher applied laser fluence (8.01 J/cm^2), showed laser-induced pressure resulting in breathing of the thin film with cavitation bubble [21]. Thus, our investigations reveal that the inclusion of microstructure and residual stresses contributions becomes necessary for obtaining physically informed threshold parameters at lower applied fluence, whereas laser-induced pressure effects dominate at higher applied fluence.

Furthermore, we computed the evolution of the dislocation density (ρ_{Dis}) in different thin-film models irradiated by the applied laser fluence studied herein. In prestrained thin films, defects originate from both original misfit dislocations at grain boundaries and reactions induced by the applied strain. For the pre-tensioned case, the equiaxed

poly-NC thin films with 48 grains showed the highest initial dislocation density ($\approx 0.160/\text{nm}^2$), followed by 24 grains. Relatively, coarser equiaxed thin films exhibited a higher initial dislocation density, owing to consistent, uniform activation volumes available across all grains. However, randomized grain topology showed a slightly different trend in starting dislocation densities, underscoring the role of local crystallographic orientation in determining activated slip planes and available activation volumes. Although the SC thin-film underwent comparable straining, the starting dislocation density was the lowest. In all cases studied herein, the dislocation densities decline sharply due to the loss of crystallinity caused by melting. In contrast, for the precompression case, the finer randomized thin-film with 48 grains showed a higher dislocation density of $\approx 0.140/\text{nm}^2$, followed by SC with $\approx 0.09/\text{nm}^2$ and finally randomized thin-film with 24 grains showing $\approx 0.06/\text{nm}^2$. Note that some crystalline regions exhibit more stacking faults and dislocations under different prestraining conditions, primarily due to tension-compression asymmetry in the material's deformation mechanisms, arising from differences in slip systems and dislocation behavior.

Our investigations reveal that critical factors such as microstructure, residual stresses and their interplay in thin films warrants consideration

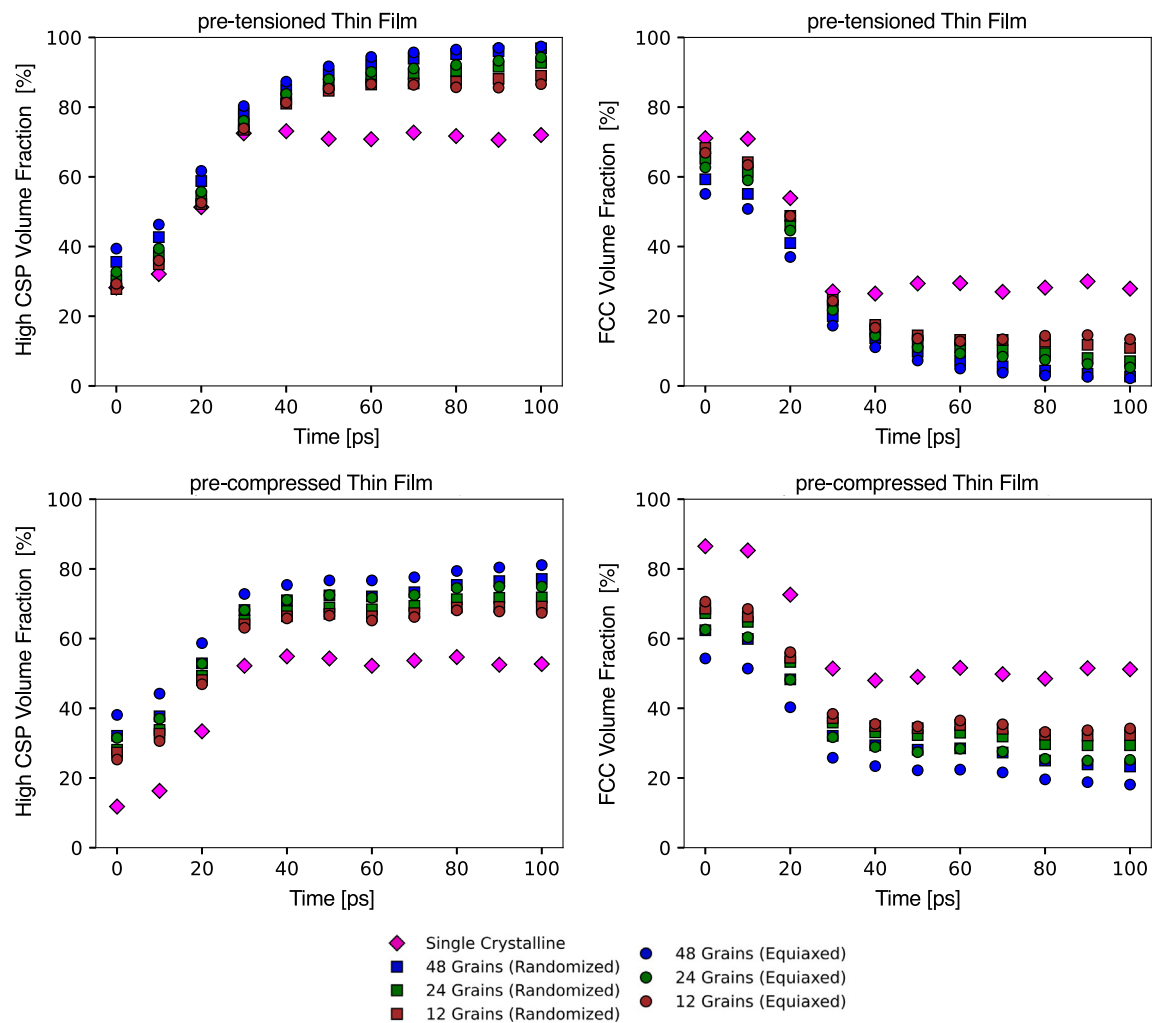


Fig. 7. Comparison of CSP and FCC volume fraction for pre-tensioned and pre-compressed thin films with varying microstructure (i.e., single crystalline, randomized, and equiaxed poly-nanocrystalline).

in the development of physically informed threshold parameters at lower applied fluence (as evidenced by the distinctly differentiated film thickness evolution and pressure responses in Figs. 8 and 9 at 1.281 J/cm²), whereas laser-induced pressure effects dominate at higher applied fluence (as demonstrated in [21] across four fluence regimes from 1.28 to 8.01 J/cm²). Grain boundary scattering reduces the electron mean free path in polycrystalline thin films; the grain boundary density inherent to the microstructure directly controls the spatial extent of electron energy deposition, thereby coupling microstructural topology to the subsequent mechanical stress evolution [46]. This fluence-regime-dependent transition is consistent with experimental observations reported by Chen et al. [41] on femtosecond-irradiated Au thin films, where crystallographic-orientation-dependent defect formation and coherent pressure wave behavior were observed at near-threshold fluences, while complete homogeneous melting irrespective of initial microstructure characterized the higher fluence response. Furthermore, Shugaev et al. [2] corroborate this interpretation at the field level, explicitly identifying that structural and microstructural factors govern phase transformation near the ablation threshold, while pressure-driven mechanisms, including spallation and phase explosion, become dominant above it. Regarding the role of residual stresses, Li et al. [47] experimentally confirmed on Au thin films that tensile residual stress promotes enhanced structural modification under laser irradiation. Furthermore, key factors, such as activated deformation

mechanisms, critical resolved shear stress, and Schmid factors, are sensitive to loading direction.

4. Conclusion

This work advances the understanding of optimal laser process parameter selection by systematically delineating the roles of thin-film microstructure, residual stresses, and their interactions under ultrafast laser irradiation. Microstructural features in thin films govern ultrafast laser-metal interaction in a specific order of importance: Microstructure configuration (Single Crystalline vs. Polycrystalline) > Topology (Random/Equiaxed) > Granularity > Crystallographic orientation. Residual tensile stresses contributed to higher melting (characterized by high CSP volume fraction) and greater laser-induced expansion (i.e., increase in film thickness of 5 nm for thin film with equiaxed 48 grains) than unstrained films (i.e., 2 nm). On the other hand, residual compressive stresses resisted laser-induced deformation, as laser-deposited thermal energy was utilized to overcome lattice compression, leading to reduced lattice straining and lower film expansion (i.e., ≈1.5 nm). The grain topology and granularity are more influential than the initial defect density in determining thin film expansion. For fine-grained thin films, grain boundaries (GBs) act as the primary sites for melting initiation, whereas, in coarser grains, the local crystallographic orientation determines the spatial extent of that melting. At the lower

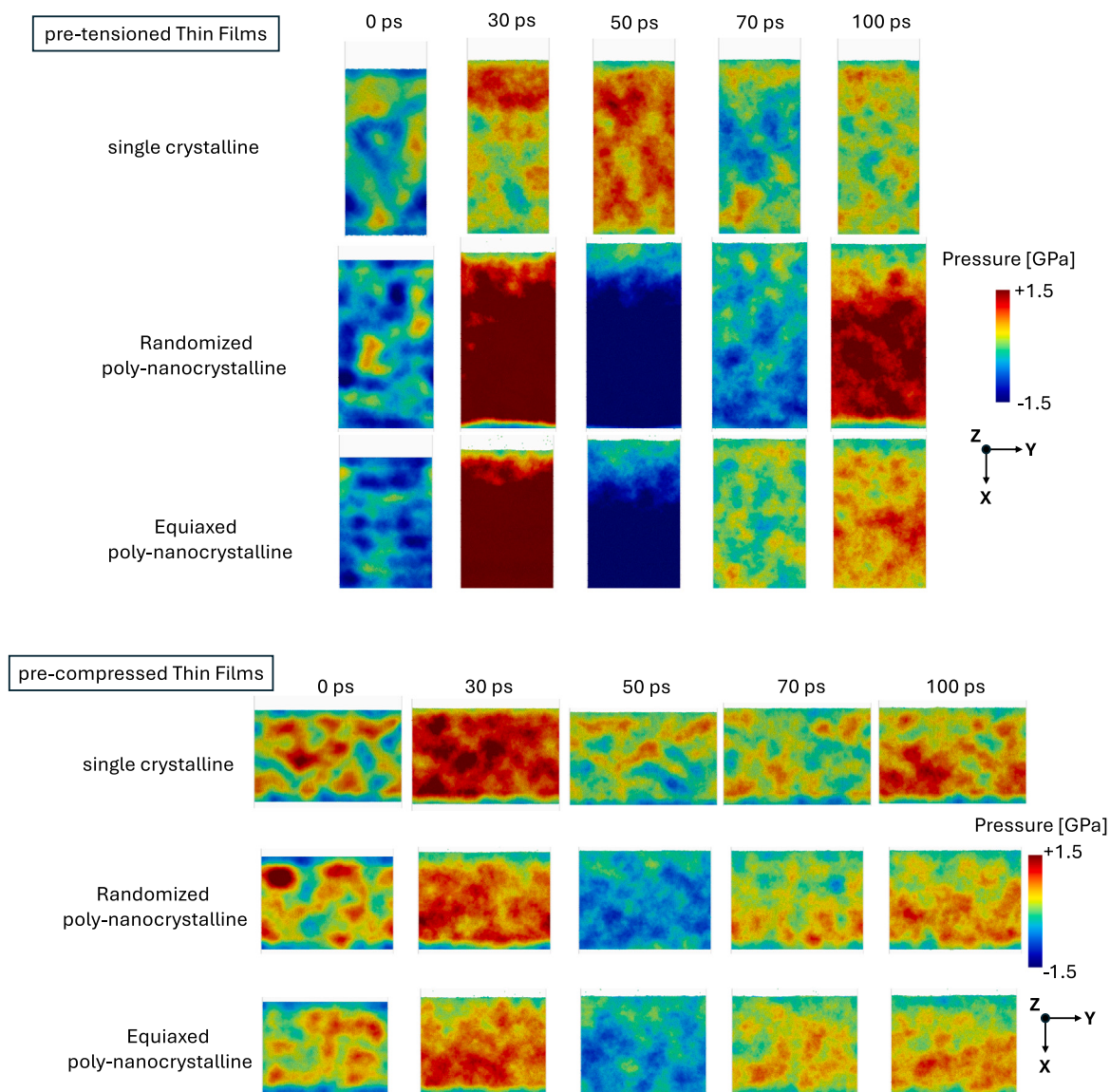


Fig. 8. Spatio-temporal pressure field plot comparison for pre-tensioned and pre-compressed thin films.

applied fluence considered herein (1.281 J/cm^2), the interplay between microstructure and residual stress dominates the deformation of thin films, characterized by CSP field plot of defects, whereas laser-induced pressure effects become dominant at higher fluences [21]. Our results demonstrate that neglecting either microstructural topology or the residual stress state, and their subsequent interaction, in process models increases uncertainty in predicting ablation thresholds that affect machinability precision. Advancing ultrafast laser-metal processing toward industrially relevant nanomanufacturing requires physically rigorous models that integrate microstructure, residual stresses, and deposited energy profiles to determine optimal processing parameters.

CRediT authorship contribution statement

Hariprasath Ganesan: Conceptualization of this study, Methodology, Software, Writing – original draft preparation, Review. **Stefan Sandfeld:** Funding acquisition, Project administration, and Review.

Declaration of generative AI use

The authors declare that **NO** generative AI tools were used to prepare the text or images in this work.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

The authors gratefully acknowledge funding from the German Research Foundation (DFG) - Grant No. 469106482 (SA2292). Furthermore, fruitful discussions with Gnanavel Vaidyanathan and Markus Olbrich are gratefully acknowledged.

Data availability

The raw/processed data required to reproduce these findings cannot be shared at this time, as the data also forms part of an ongoing study.

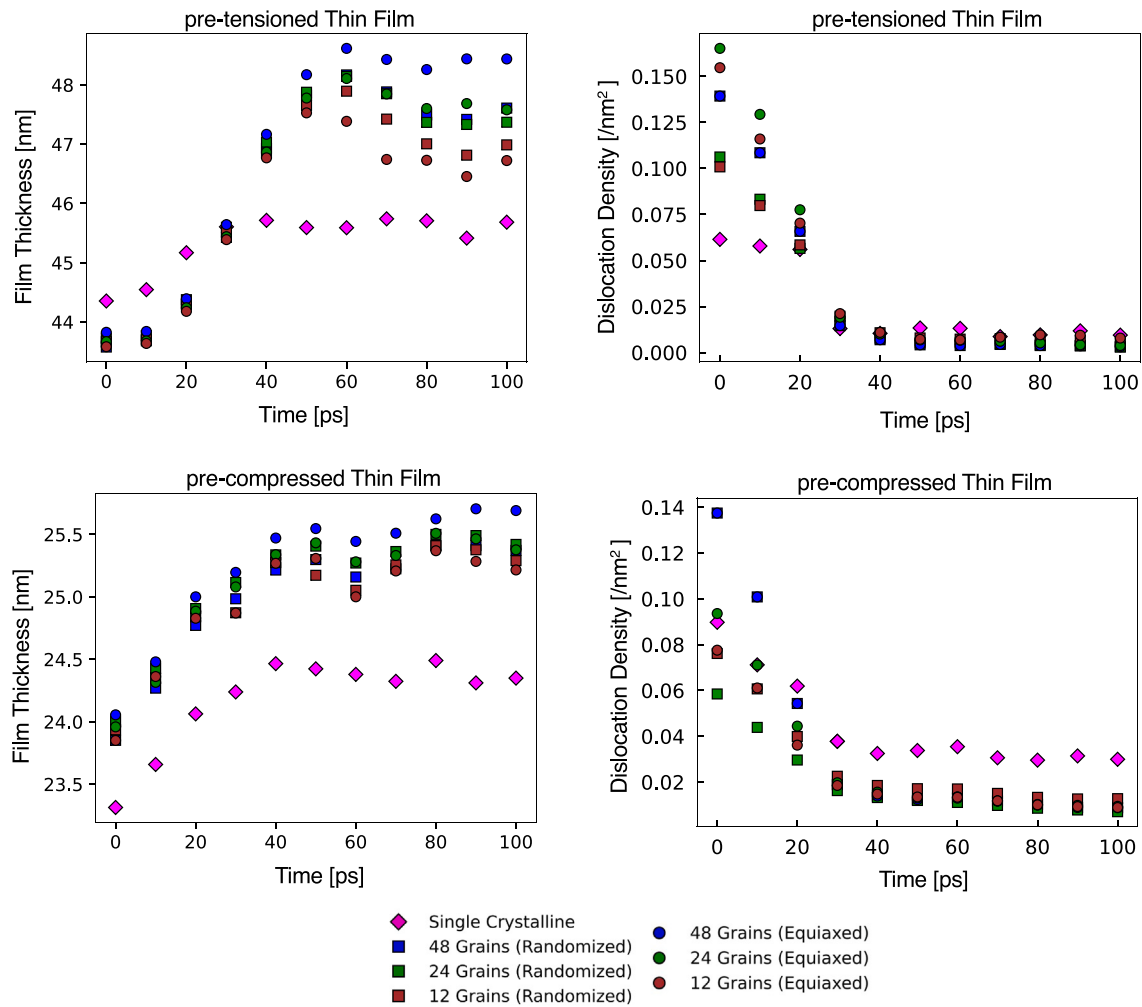


Fig. 9. Prestrained thin-film samples - Film thickness and dislocation density evolution under the fluence of 1.281 J/cm^2 .

References

- [1] Sugioka K. Progress in ultrafast laser processing and future prospects. *Nanophotonics* 2017;6(2):393–413. <http://dx.doi.org/10.1515/nanoph-2016-0004>.
- [2] Shugaev MV, Wu C, Armbruster O, Naghilou A, Brouwer N, Ivanov DS, Derrien TJ-Y, Bulgakova NM, Kautek W, Rethfeld B, et al. Fundamentals of ultrafast laser–material interaction. *MRS Bull* 2016;41(12):960–8. <http://dx.doi.org/10.1557/mrs.2016.274>.
- [3] Lin Z, Ji L, Hong M. Advancing manufacturing limits: Ultrafast laser nanofabrication techniques. *Engineering* 2025. <http://dx.doi.org/10.1016/j.eng.2025.03.017>.
- [4] Guo B, Sun J, Lu Y, Jiang L. Ultrafast dynamics observation during femtosecond laser–material interaction. *Int J Extrem Manuf* 2019;1(3):032004. <http://dx.doi.org/10.1088/2631-7990/ab3a24>.
- [5] Wang D, Wei S, Yuan X, Liu Z, Weng Y, Zhou Y, Xiao T-H, Goda K, Liu S, Lei C. Ultrafast imaging for uncovering laser–material interaction dynamics. *Int J Mech Syst Dyn* 2022;2(1):65–81. <http://dx.doi.org/10.1002/msd2.12024>.
- [6] Martin AA, Caltá NP, Hammons JA, Khairallah SA, Nielsen MH, Shuttlesworth RM, Sinclair N, Matthews MJ, Jeffries JR, Willey TM, et al. Ultrafast dynamics of laser–metal interactions in additive manufacturing alloys captured by in situ X-ray imaging. *Mater Today Adv* 2019;1:100002. <http://dx.doi.org/10.1016/j.mtadv.2019.01.001>.
- [7] Andreev SV, Aseev SA, Bagratashvili VN, Vorob'ev NS, Ishchenko AA, Kompanets VO, Malinovsky AL, Mironov BN, Timofeev AA, Chekalin SV, et al. Ultrafast transmission electron microscope for studying the dynamics of the processes induced by femtosecond laser beams. *Quantum Electron* 2017;47(2):116. <http://dx.doi.org/10.1070/QEL16276>.
- [8] Miloshevsky G. Ultrafast laser matter interactions: modeling approaches, challenges, and prospects. *Modelling Simul Mater Sci Eng* 2022;30(8):083001. <http://dx.doi.org/10.1088/1361-651X/ac8abc>.
- [9] Zhigilei LV, Lin Z, Ivanov DS. Atomistic modeling of short pulse laser ablation of metals: connections between melting, spallation, and phase explosion. *J Phys Chem C* 2009;113(27):11892–906. <http://dx.doi.org/10.1021/jp902294m>.
- [10] Liu H, Xie W, Ding Y, Chen K, Wang S, Huo H, Yang L. Review of molecular dynamics simulations in laser-based micro/nano-fabrication. *Nanoscale* 2024;16(46):21189–215. <http://dx.doi.org/10.1039/D4NR03305A>.
- [11] Song S, Lu Q, Zhang P, Yan H, Shi H, Yu Z, Sun T, Luo Z, Tian Y. A critical review on the simulation of ultra-short pulse laser–metal interactions based on a two-temperature model (TTM). *Opt Laser Technol* 2023;159:109001. <http://dx.doi.org/10.1016/j.optlastec.2022.109001>.
- [12] Yin CP, Zhang ST, Dong YM, Ye QW, Li Q. Molecular-dynamics study of multi-pulsed ultrafast laser interaction with copper. *Adv Prod Eng Manag* 2021;16(4):457–72. <http://dx.doi.org/10.14743/apem2021.4.413>.
- [13] Ivanov DS, Terekhin PN, Kudryashov SI, Klimentov SM, Kabashin AV, Garcia ME, Rethfeld B, Zavestovskaya IN. The atomistic perspective of nanoscale laser ablation. In: *Ultrafast laser nanostructuring: The pursuit of extreme scales*. Springer; 2023, p. 65–137. http://dx.doi.org/10.1007/978-3-031-14752-4_2.
- [14] Valavanis AS, Chen C, Grigoropoulos CP, Eliceiri M, Li J, Zhigilei LV. Laser fluence and film thickness dependence of the mechanisms of femtosecond laser ablation of Ag films from atomistic simulations and optical imaging. In: *High-power laser ablation VIII*. SPIE; 2024, PC129391E. <http://dx.doi.org/10.1117/12.3012635>.
- [15] Xie L, Li Y, Wang F, Yao C, Yuan X. Molecular dynamics simulation study on the ablative copper forming process at the atomic level by femtosecond laser. *J Manuf Process* 2025;150:38–47. <http://dx.doi.org/10.1016/j.jmapro.2025.06.043>.
- [16] Amoroso S, Nedyalkov NN, Wang X, Ausanio G, Bruzzese R, Atanasov PA. Ultrashort-pulse laser ablation of gold thin film targets: Theory and experiment. *Thin Solid Films* 2014;550:190–8. <http://dx.doi.org/10.1016/j.tsf.2013.10.165>.
- [17] Lian Y, Jiang L, Sun J, Lin G, Liang M. Atomistic insight on temperature-dependent laser induced ultrafast thermomechanical response in aluminum film. *Int J Heat Mass Transfer* 2024;231:125809. <http://dx.doi.org/10.1016/j.jheatmasstransfer.2024.125809>.
- [18] Rouleau CM, Shih C-Y, Wu C, Zhigilei LV, Poretzky AA, Geoghegan DB. Nanoparticle generation and transport resulting from femtosecond laser ablation of

- ultrathin metal films: Time-resolved measurements and molecular dynamics simulations. *Appl Phys Lett* 2014;104(19). <http://dx.doi.org/10.1063/1.4876601>.
- [19] Zhang Z, Yang Z, Wang C, Zhang Q, Zheng S, Xu W. Mechanisms of femtosecond laser ablation of Ni3Al: Molecular dynamics study. *Opt Laser Technol* 2021;133:106505. <http://dx.doi.org/10.1016/j.optlastec.2020.106505>.
 - [20] Hayder MM, Moumita TM, Chowdhury S, Rahman KA. Effective electronic properties and coupling for two-temperature model-molecular dynamics simulation of ultrafast laser ablation of nickel. *Mol Simul* 2024;50(14):1140–51. <http://dx.doi.org/10.1080/08927022.2024.2385499>.
 - [21] Ganesan H, Sandfeld S. Capturing thin-film microstructure contributions during ultrafast laser-metal interactions using atomistic simulations. *Mater Des* 2025;114224. <http://dx.doi.org/10.1016/j.matdes.2025.114224>.
 - [22] Olbrich M, Punzel E, Lickschat P, Weißmantel S, Horn A. Investigation on the ablation of thin metal films with femtosecond to picosecond-pulsed laser radiation. *Phys Procedia* 2016;83:93–103. <http://dx.doi.org/10.1016/j.phpro.2016.08.017>.
 - [23] Ganesan H, Teijeiro C, Sutmann G. Parallelization comparison and optimization of a scale-bridging framework to model cottrell atmospheres. *Comput Mater Sci* 2018;155:439–49. <http://dx.doi.org/10.1016/j.commatsci.2018.08.055>.
 - [24] Ganesan H, Scheider I, Cyron CJ. Quantifying the high-temperature separation behavior of lamellar interfaces in γ -titanium aluminide under tensile loading by molecular dynamics. *Front Mater* 2021;7:602567. <http://dx.doi.org/10.3389/fmats.2020.602567>.
 - [25] Ganesan H, Sutmann G. Modeling segregated solutes in plastically deformed alloys using coupled molecular dynamics-Monte Carlo simulations. *J Mater Sci Technol* 2025;213:98–108. <http://dx.doi.org/10.1016/j.jmst.2024.06.030>.
 - [26] Norman GE, Starikov SV, Stegailov VV. Atomistic simulation of laser ablation of gold: effect of pressure relaxation. *J Exp Theor Phys* 2012;114:792–800. <http://dx.doi.org/10.1134/S1063776112040115>.
 - [27] Yao J, Qi D, Liang H, He Y, Yao Y, Jia T, Yang Y, Sun Z, Zhang S. Exploring femtosecond laser ablation by snapshot ultrafast imaging and molecular dynamics simulation. *Ultrafast Sci* 2022. <http://dx.doi.org/10.34133/2022/9754131>.
 - [28] Thompson AP, Aktulga HM, Berger R, Bolintineanu DS, Brown WM, Crozier PS, Veld PJ In't, Kohlmeyer A, Moore SG, Nguyen TD, et al. LAMMPS-a flexible simulation tool for particle-based materials modeling at the atomic, meso, and continuum scales. *Comput Phys Comm* 2022;271:108171. <http://dx.doi.org/10.1016/j.cpc.2021.108171>.
 - [29] Rutherford AM, Duffy DM. The effect of electron-ion interactions on radiation damage simulations. *J Phys: Condens Matter* 2007;19(49):496201. <http://dx.doi.org/10.1088/0953-8984/19/49/496201>.
 - [30] Daw MS. Embedded-atom method: Derivation and application to impurities, surfaces, and other defects in metals. *Phys Rev B* 1984;29(12):6443. <http://dx.doi.org/10.1103/PhysRevB.29.6443>.
 - [31] Ganesan H, Scheider I, Cyron CJ. Understanding creep in TiAl alloys on the nanosecond scale by molecular dynamics simulations. *Mater Des* 2021;212:110282. <http://dx.doi.org/10.1016/j.matdes.2021.110282>.
 - [32] Chandran A, Ganesan H, Cyron CJ. Studying the effects of Nb on high-temperature deformation in TiAl alloys using atomistic simulations. *Mater Des* 2024;237:112596. <http://dx.doi.org/10.1016/j.matdes.2023.112596>.
 - [33] Grover CRM, Hentzell HTG, Smith DA. The development of grain structure during growth of metallic films. *Acta Metall* 1984;32(5):773–81. [http://dx.doi.org/10.1016/0001-6160\(84\)90150-0](http://dx.doi.org/10.1016/0001-6160(84)90150-0).
 - [34] Olbrich M, Pflug T, Wüstefeld C, Motylenko M, Sandfeld S, Rafaja D, Horn A. Hydrodynamic modeling and time-resolved imaging reflectometry of the ultrafast laser-induced ablation of a thin gold film. *Opt Lasers Eng* 2020;129:106067.
 - [35] Hirel P. AtomsK: A tool for manipulating and converting atomic data files. *Comput Phys Comm* 2015;197:212–9. <http://dx.doi.org/10.1016/j.cpc.2015.07.012>.
 - [36] Sun Xuelian, Fan Shanming, Peng Mingjun, Ma Lishi, Shen Li, Qi Huarong, Zhao Yanbo, Li Mengnie. Classical molecular dynamics simulation of atomic structure transitions in FeSiCuMgAl high-entropy alloys under biaxial stretching. *Mater Today Commun* 2024;40:109716.
 - [37] Ivanov DS, Zhigilei LV. Combined atomistic-continuum modeling of short-pulse laser melting and disintegration of metal films. *Phys Rev B* 2003;68(6):064114. <http://dx.doi.org/10.1103/PhysRevB.68.064114>.
 - [38] Iabbadin D, Amodeo J, Fusco C, Garrelieand F, Colombier J-P. Molecular dynamics simulation of structural evolution in crystalline and amorphous CuZr alloys upon ultrafast laser irradiation. *Phys Rev Mater* 2022;6(12):126001. <http://dx.doi.org/10.1103/PhysRevMaterials.6.126001>.
 - [39] Stukowski A. Visualization and analysis of atomistic simulation data with OVITO—the open visualization tool. *Modelling Simul Mater Sci Eng* 2009;18(1):015012. <http://dx.doi.org/10.1088/0965-0393/18/1/015012>.
 - [40] Huff M. Residual stresses in deposited thin-film material layers for micro-and nano-systems manufacturing. *Micromachines* 2022;13(12):2084. <http://dx.doi.org/10.3390/mi13122084>.
 - [41] Chen J, Chen W-K, Tang J, Rentzepis PM. Time-resolved structural dynamics of thin metal films heated with femtosecond optical pulses. *Proc Natl Acad Sci* 2011;108(47):18887–92. <http://dx.doi.org/10.1073/pnas.1115237108>.
 - [42] Wu J, Tang M, Zhao L, Zhu P, Jiang T, Zou X, Hong L, Luo S-N, Xiang D, Zhang J. Ultrafast atomic view of laser-induced melting and breathing motion of metallic liquid clusters with MeV ultrafast electron diffraction. *Proc Natl Acad Sci* 2022;119(4):e2111949119. <http://dx.doi.org/10.1073/pnas.2111949119>.
 - [43] Liubchenko O, Antonowicz J, Sokolowski-Tinten K, Zalden P, Minikayev R, Milov I, Albert TJ, Bressler C, Chojnacki M, Dłużewski P, et al. Laser-induced ultrafast structural transformations in thin Fe layer revealed by time-resolved X-ray diffraction. 2025. <http://dx.doi.org/10.48550/arXiv.2506.18730>, arXiv preprint arXiv:2506.18730.
 - [44] Zhou S, Shen H. Experimental and numerical studies on micro-bumps without melting of gold films with different thicknesses induced by ultrafast laser. *Opt Commun* 2022;514:128178. <http://dx.doi.org/10.1016/j.optcom.2022.128178>.
 - [45] Colombier Jean-Philippe, Combis Patrick, Rosenfeld Arkadi, Hertel Ingolf V, Audouard Eric, Stoian Razvan. Optimized energy coupling at ultrafast laser-irradiated metal surfaces by tailoring intensity envelopes: Consequences for material removal from Al samples. *Phys Rev B—Condens Matter Mater Phys* 2006;74(22):224106.
 - [46] Suzana Ana F, Koch Robert, Wu Longlong, Assefa Tadesse A, Kim Sungwon, Choi Sungwook, Lee Heemin, Kim Sunam, Lee Jae Hyuk, Park Sang-Youn, et al. Compressive effects in melting of palladium thin films studied by ultrafast x-ray diffraction. *Phys Rev B* 2023;107(21):214303.
 - [47] Zhou Shujun, Wu Wei, Huang Xiao, Shao Tianmin, Shao Meng. Effect of pulsed laser irradiation on microstructure and evolution of residual stress for gold film. *Adv Eng Mater* 2023;25(6):2201249.