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Generation of nanocrystalline surface layer in short pulse laser processing of metal targets under conditions of spatial confinement by solid or liquid overlayer

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ABSTRACT

The effect of spatial confinement by a solid or liquid overlayer on short pulse laser-induced surface microstructure modification is investigated in a series of large-scale atomistic simulations performed for Ag targets irradiated in the regime of melting and resolidification, below the thresholds for laser spallation and ablation. For Ag targets with free surfaces, the formation of a nanocrystalline region with random crystallographic grain orientation is observed under irradiation conditions leading to the generation of numerous sub-surface voids that slow down the solidification process. When no voids are generated, the resolidification produces grains misoriented with respect to the bulk of the target by just several degrees and separated from each other by low angle grain boundaries or dislocation walls. The presence of a liquid or solid overlayer suppresses nucleation of sub-surface voids, provides an additional pathway for cooling through the heat conduction to the overlayer, and facilitates the formation of nanocrystalline structure in a region of the metal target adjacent to the overlayer. Moreover, the stabilizing effect of the solid overlayer may result in an incomplete melting of metal in the vicinity of the interface, making it possible for grains growing from the interface to retain "memory" of the target orientation and to produce nanocrystalline interfacial region with small misorientation of grains with respect to the bulk of the target. In all simulations, the nanocrystalline layers generated by laser processing of single crystal Ag targets are characterized by a high density of stacking faults, twin boundaries, and point defects produced in the course of the rapid resolidification.

1. Introduction

Laser processing of metal surfaces in liquid environment and in the presence of a solid overlayer has a number of distinct characteristics that may be beneficial for practical applications. The confining effect of optically transparent solid or liquid layers covering irradiated targets can alter the dynamics of material decomposition and ejection in the ablation regime [1–5], thus directly affecting the surface morphology of laser-modified targets. Moreover, the interaction of melted metal with an overlayer can contribute to cooling of the molten layer, thus creating conditions for stronger, as compared to laser processing in air or vacuum, undercooling and formation of highly nonequilibrium surface microstructure. Indeed, a number of experimental studies have demonstrated that the presence of liquid environment [6–14] or a solid overlayer [15–17] can have major impact on the morphology of laser-processed surfaces. The effect of the spatial confinement on the microstructure of the irradiated targets has also been demonstrated in laser shock peening experiments performed in the ablation regime [18–20].

More "gentle" microstructure modification at lower laser fluences, in the regime of melting and resolidification, however, has not been investigated in the presence of liquid or solid overlayers so far. For irradiation in vacuum or in air, the utility of short laser pulses for selective nanoscale engineering of surface microstructure has been demonstrated for various material systems. In particular, the formation of nanoscale twinned domains has recently been observed in femtosecond laser processing of Ni and explained by the onset of growth twinning under conditions of strong undercooling realized in the course of the rapid resolidification of a transiently melted surface region [21]. Even stronger undercooling of the melted region, sufficient to trigger massive nucleation of new crystallites and generation of nanocrys-

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talline surface layer with high density of twin boundaries and stacking faults present in the nanograins, has been predicted in recent atomistic simulations of laser processing of Ag targets [22]. The possibility of the generation of thin nanocrystalline layers with high density of twins is highly attractive, as such layers can exhibit an unusual combination of high strength and ductility [23–26], as well as enhanced catalytic activity [27,28] and tunable surface reflectance spectra [29]. The results of the simulations, however, suggest that the laser-induced generation of nanocrystalline layer on the surface of a pure Ag target is only possible in a narrow range of laser fluences close to the spallation threshold, when the generation sub-surface voids slows down the propagation of the resolidification front from the bulk of the target [22]. This leads to an intriguing question on whether the presence of liquid or solid overlayer could broaden the range of irradiation conditions that yield surface nanocrystallization and result in the formation of a nanocrystalline surface layer without the generation of sub-surface voids.

In this paper, we investigate the specific mechanisms of overlayer-assisted surface nanostructuring by performing a series of large-scale atomistic simulations of short pulse laser irradiation of Ag targets in water environment and under confinement by a silica glass overlayer, and contrasting the results of these simulations to the ones obtained for similar irradiation conditions in vacuum. The main focus of the analysis of the simulation results is on revealing the irradiation conditions and mechanisms responsible for the generation of nanocrystalline surface layer and establishing the processes that control the structural characteristics of the nanocrystalline layer. A brief description of the computational model is provided below, in Section 2, and is followed by presentation of the simulation results in Section 3 and summary in Section 4.

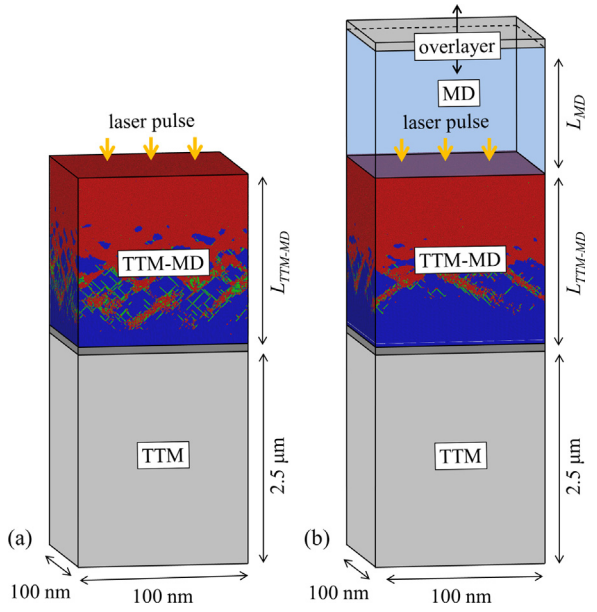

Fig. 1. Schematic sketches of the computational setups used in MD simulations of laser interaction with metal targets in vacuum (a) and in the presence of an optically transparent solid or liquid overlayer (b). The top part of the target is represented by TTM-MD model, whereas the temperature evolution in the deeper part is described by TTM equations. Molecular dynamics representation is used for parts of the solid or liquid overlayers adjacent to the target, as shown by the blue rectangular parallelepiped in (b). At the bottom of the TTM-MD and on the top of the MD regions the pressure wave transmitting boundary conditions are imposed. The boundary conditions mimic the non-reflective propagation of laser-induced pressure waves in the Ag target and the overlayer, which are assumed to be sufficiently thick to ensure that any effects caused by the reflection of the waves from the outer surfaces of the target and overlayer can be neglected. Periodic boundary conditions are applied in the lateral directions, parallel to the surface of the target. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

2. Computational model

The computational setups used in the simulations of laser interactions with bulk Ag targets in vacuum and in the presence of liquid or solid overlayer are schematically illustrated in Fig. 1. In all simulations, the top part of the Ag target is simulated with a hybrid atomistic–continuum model [30] that combines the classical molecular dynamics (MD) method [31] with the continuum-level two-temperature model (TTM) [32]. The model accounts for the laser excitation of conduction-band electrons, electron-phonon coupling, and the fast electron heat conduction from the hot surface region of the irradiated target to the bulk of the target. At the same time, the atomistic representation of the surface region of the target enables detailed analysis of the kinetics and mechanisms of laser-induced structural and phase transformations. The electronic heat transfer in the deeper part of the target, where no structural changes take place in response to the laser irradiation, is described by the conventional TTM. The depth covered by the TTM is chosen to be 2.5 μm to ensure a negligible temperature change at the bottom of the computational system by the end of the simulation. The absorption of laser energy is represented through a source term added to the TTM equation for the electron temperature [30]. The source term has a temporal Gaussian profile and includes a description of the ballistic energy redistribution by the excited electrons through an appropriate modification of the effective energy deposition depth [30,33]. The optical absorption depth, 12 nm at laser wavelength of 800 nm [34], and the effective depth of the ballistic energy transport, estimated to be about 56 nm for Ag [22,35] are used in the simulations reported in this paper.

A dynamic pressure wave transmitting boundary condition [36,37] is applied at the bottom of the TTM-MD part of the model to ensure nonreflecting propagation of the laser-induced stress wave into the bulk of the target. Periodic boundary conditions are applied in the lateral directions, parallel to the irradiated (001) surfaces of the Ag targets, and the dimensions of the computational systems in these directions are about 100 nm × 100 nm. The interatomic interactions are described by the embedded atom method (EAM) potential parametrized for Ag [38]. Before applying laser irradiation, all computational systems are thermalized at 300 K for 150 ps. A complete description of the combined atomistic-continuum model is provided elsewhere [22,30], and all the parameters of the model for simulation of laser interaction with a Ag target are given in Ref. [22]. Thus, below we only delineate parameters of the computational setups specific for the simulations reported in this paper and provide a brief description of the model representations of the liquid and solid overlayers.

Since the goal of this study is to explore the possibility of laser-induced surface nanocrystallization, the irradiation conditions in all simulations are chosen to produce transient melting and resolidification of a surface region of the target. The simulations of laser interaction with Ag targets in vacuum are performed for two sets of irradiation parameters (laser pulse duration τ_p and absorbed laser fluence F_{abs}). The first set, $\tau_p = 100$ fs and $F_{abs} = 850$ J/m^2, corresponds to the formation of a porous sub-surface region covered by a nanocrystalline surface layer, as discussed in detail in Ref. [22]. The second set is performed with $\tau_p = 10$ ps and $F_{abs} = 900$ J/m^2 and results in the melting and resolidification of ~60 nm surface layer without nucleation of sub-surface voids. The depth of the surface part of the Ag target represented with atomistic resolution, L_{TTM-MD} in Fig. 1, is 150 nm for the first set and 103 nm for the second set of irradiation parameters, which corresponds to 84.2 and 62.5 million

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