858
doi:10.1017/S1431927622003816
Microsc. Microanal. 28 (Suppl 1), 2022

Atomistic Modeling of Ultrashort Pulse Laser-Induced Generation of Crystal Defects

Miao He,[1,2] Eaman T. Karim,[1,3] Maxim V. Shugaev,[1] Cheng-Yu Shih,[1,4] and Leonid V. Zhigilei[1*]

[1.] Department of Materials Science and Engineering, University of Virginia, Charlottesville, VA, United States.
[2.] Department of Mechanical Engineering, University of Michigan, Ann Arbor, MI, United States.
[3.] American Dental Association Science and Research Institute, Gaithersburg, MD, United States.
[4.] Longterm Concept International Pte Ltd, Singapore.
* Corresponding author: lz2n@virginia.edu

Rapid advancements in the development of accessible sources of ultrashort (pico- and femtosecond) laser pulses open up new possibilities for surface modification with high accuracy and spatial resolution. The shallow depths of the laser energy deposition and steep temperature gradients, produced by the ultrashort pulse laser irradiation, can lead to the cooling rates of more than 10^{12} K/s. Resolidification of a transiently melted surface region, occurring under conditions of rapid quenching and dynamic relaxation of laser-induced stresses, creates the conditions for generation of highly nonequilibrium densities and configurations of crystal defects, which can drastically alter the physical, chemical, and mechanical properties of surface layers processed by ultrashort laser pulses [1,2].

Detailed characterization of microstructural changes produced by ultrashort pulse laser irradiation has revealed the presence of high densities of dislocations [3,4], nanoscale twinned domains [5,6], and nanograins [6] in the surface regions of the irradiated targets. The small size of the laser-modified zone, however, makes the characterization of laser-induced nanostructure challenging and, at the same time, increases the importance of understanding of the nucleation, mobility, interactions and stability of individual crystal defects. Large-scale atomistic modeling of laser-materials interactions [2,5,7-11] can help in the physical interpretation of experimental data and, eventually, in the development of new reliable methodology for experimental probing of laser-modified zone with nanometer structural and compositional resolutions. Several examples of the application of atomistic modeling to investigation of the generation of crystal defects in ultrashort pulse laser interactions are provided below.

The mechanisms of the generation of vacancies, dislocations, stacking faults, and twin boundaries in Ni-based single-phase solid-solution alloys ($Ni_{50}Fe_{50}$, $Ni_{80}Fe_{20}$, and $Ni_{80}Cr_{20}$) are investigated in the irradiation regime of melting and resolidification [8]. The decrease in the thermal conductivity and strengthening of the electron-phonon coupling due to the intrinsic chemical disorder in the solid-solution alloys are found to have important implications on localization of the energy deposition and generation of thermoelastic stresses. The interaction of the laser-induced stress waves with the melting front is found to play a key role in roughening of the crystal-liquid interface and generation of dislocations upon the solidification, *e.g.*, Figure 1a. A common feature revealed in the structural analysis of all irradiated targets is the presence of high vacancy concentrations exceeding the equilibrium values at the melting temperature by about an order of magnitude, *e.g.*, Figure 1b. The analysis of the first atomic shell surrounding the vacancy sites in Ni-Fe alloys uncovers the preference for the vacancy sites to be surrounded by Fe atoms, Figure 1c, and suggests that atomic-scale chemical heterogeneities may play an important role in defining the properties of the single-phase concentrated solid-solution alloys.

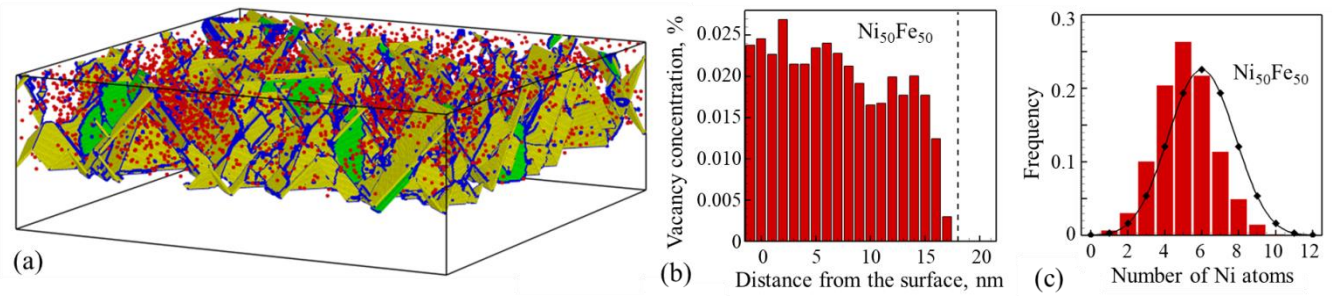

Figure 1. Results of a simulation of a $Ni_{50}Fe_{50}$ target irradiated by a 100 fs laser pulse at an absorbed fluence of 400 J/m^2, in the regime of melting and resolidification [8]. (a) Snapshot of the top 30-nm-thick surface region of the target, with atoms with local fcc structure blanked and the remaining atoms colored by their local structural environment, so that atoms in dislocation cores are blue, atoms surrounding vacancies are red, stacking faults are yellow, and twin boundaries are green. (b) Distribution of vacancy concentration with respect to the depth under irradiated surface, with the dashed line marking the maximum melting depth reached after the laser irradiation. (c) Distribution of the number of Ni atoms in the first nearest neighbor shell of individual vacancies, with the black line showing the distribution expected for a random arrangement of Ni and Fe atoms.

The simulations of laser-material interactions are complemented by systematic analysis of the generation of crystal defects at a rapidly advancing solidification front in molecular dynamics simulations of solidification occurring at fixed levels of undercooling. The generation of defects is correlated with the velocity of the solidification front, and the processes responsible for creating the strong vacancy supersaturation [11] and nanoscale twinned domains [5] are revealed.

At higher laser fluences, above the ablation threshold, the generation of crystal defects is intertwined with processes responsible for the material ejection (phase explosion, spallation), which affect the cooling rates and solidification kinetics. In particular, the generation of high densities of dislocations and vacancies in frozen surface features produced by spatially-modulated laser ablation of Cr targets is predicted in atomistic simulations illustrated by Figure 2 [9,10]. The mechanical and chemical properties of the frozen protrusions generated by the ablation in vacuum (Figure 2a) and in water environment (Figure 2b) can be expected to be strongly affected by the very high vacancy concentrations exceeding 10^{-3} and dislocation densities on the order of 10^{15} m^{-2}. [12]

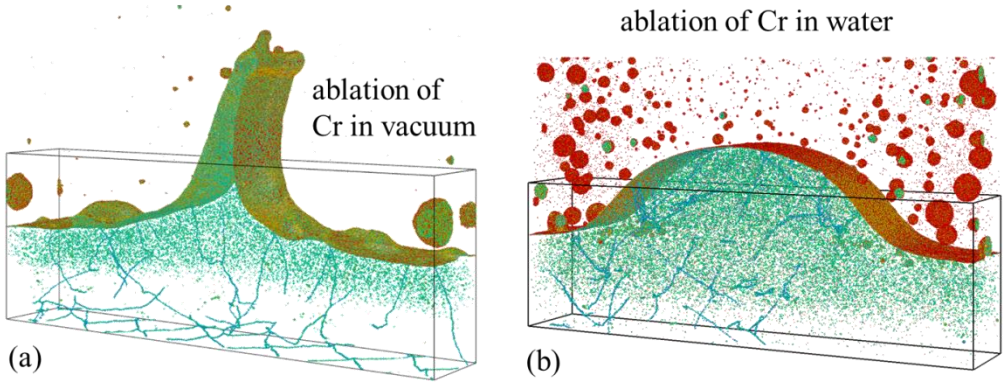

Figure 2. Dislocations and vacancy clusters present in and below the frozen protrusions generated in simulations of spatially-modulated laser ablation of Cr targets in vacuum [9] and in water environment [10]. The defects are exposed by blanking the atoms with local bcc coordination.

https://doi.org/10.1017/S1431927622003816 Published online by Cambridge University Press

References:

[1] R. Vilar, in Handbook of Laser Micro- and Nano-Engineering, Edited by K. Sugioka (Springer, 2021 (Springer, Cham, Switzerland, 2021), pp. 213-272. doi: 10.1007/978-3-030-63647-0_14
[2] M. V. Shugaev et al., in Handbook of Laser Micro- and Nano-Engineering, Edited by K. Sugioka (Springer, Cham, Switzerland, 2021), pp. 83-163. doi: 10.1007/978-3-319-69537-2_11-1
[3] X. Sedao et al., Appl. Phys. Lett. **104**, 171605 (2014). doi: 10.1063/1.4874626
[4] M. P. Echlin et al., Acta Mater. **124**, 37-46 (2017). doi: 10.1016/j.actamat.2016.10.055
[5] X. Sedao et al., ACS Nano **10**, 6995-7007 (2016). doi: 10.1021/acsnano.6b02970
[6] J. V. Oboňa et al., Appl. Surf. Sci. **303**, 118-124 (2014). doi: 10.1016/j.apsusc.2014.02.104
[7] M. V. Shugaev et al., in Advances in the Application of Lasers in Materials Science, Springer Series in Materials Science, Vol. **274**, Edited by P. M. Ossi (Springer International Publishing Switzerland, 2018), pp. 107-148. doi: 10.1007/978-3-319-96845-2_5
[8] M. He et al., J. Phys. Chem. C **123**, 2202-2215 (2019). doi: 10.1021/acs.jpcc.8b09922
[9] M. V. Shugaev et al., Phys. Rev. B **96**, 205429, 2017.
[10] C.-Y. Shih et al., Nanoscale **12**, 7674-7687 (2020). doi: 10.1039/D0NR00269K
[11] M. He et al., Acta Mater. **203**, 116465 (2021). doi: 10.1016/j.actamat.2020.11.007
[12] Financial support for this work was provided by the National Science Foundation (NSF) through Grant No. DMR-1610936.