MODIFICATION OF SURFACES AND INTERFACES WITH SWIFT HEAVY IONS

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Swift heavy ions (SHI) penetrating through a solid lose main part of the energy to the electronic stopping. The extremely high level of the electronic excitation generated by incoming ion at the femtosecond scale is followed by relaxation of the perturbed electron ensemble. Subsequently, the acceleration of target atoms occurs at sub-picosecond times, resulting in the formation of a nanometric damaged region within hundreds of picoseconds after the ion passage. These structural changes can affect its physical, chemical, and mechanical properties of the irradiated target. Due to these features, accelerated ion beams serve as a versatile tool for the patterning and modification of nanometric materials [1,2], as well as the investigation of radiation stability against cosmic rays and fission fragments [3–5].

Despite active experimental investigations, the mechanisms of surfaces and interfaces damage by SHI irradiation are not yet entirely comprehended. Nanohillocks appearing at the surface under irradiation turned out to have different internal structures, dependent on the material and projectile properties [6–9]. Recent research has demonstrated that grazing angle incidence of SHIs can effectively create extended periodical chains of hillocks [10] or grooves [11] on the surfaces of dielectrics.

Nanocrystalline materials are of significant interest nowadays, since it consist of grains with sizes ranging up to several tens of nanometers, resulting in a higher volumetric fraction of interfacial boundaries or grain boundaries [12]. This can change properties and radiation stability of the materials in comparison with conventional polycrystalline targets. Recent studies demonstrated various effects of swift heavy irradiation on grains and their boundaries: grain coarsening [13], improved radiation resistance [14], lowering of amorphization threshold [15] etc.

This work reviews our results on simulations of swift heavy ions damage in near-surface and interface regions in amorphizable and non-amorphizable dielectrics irradiated with high-energy heavy ions. To analyze the changes in the structure of insulators under the influence of swift heavy ions we use a multiscale hybrid model [16–19]. This model integrates the Monte Carlo (MC) TREKIS code [18] describing excitation of the electron subsystem and energy transfer to the target lattice, with molecular dynamics (MD) simulations of further relaxation of the atomic subsystem [20–22].

Figure 1 presents the experimental and theoretical results of surface modifications of MgO, CaF_2 and $Y_3Al_5O_{12}$ (YAG) under normal SHI irradiations. The studied materials demonstrated different relaxation kinetics of the lattice energy deposited by an SHI [22]. Understanding the reasons for these differences can help in managing of a nanofeature formation, which is important for various nanostructuring technologies. The simulations reveal that amorphizable MgO, CaF_2 have lower viscosities and surface tensions allowing molten material to be extruded stronger at initial times and lead to formation of spherically shaped crystalline hillocks. In contrast, the higher surface tension and viscosity in YAG limits protrusion of a liquid droplet from the surface, transiently forming semi-spherical amorphous hillock [22].

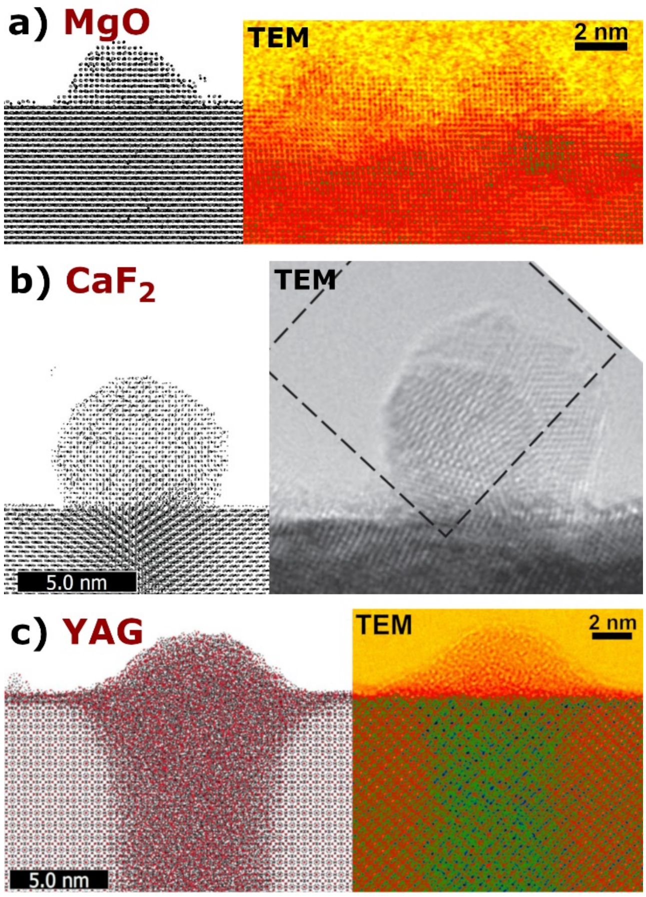

Figure 1. The MD simulated supercells of (a) MgO, (b) CaF_2 and (c) YAG after a passage of a swift heavy ion [22]. Simulation results are accompanied by TEM images. TEM micrograph of surface hillock in CaF_2 was taken from [6].

Grazing ion impacts into MgO and Al_2O_3 were studied by means of the simulation of the ion trajectories at 1 nm depth parallel to the sample surface. The research discovered that the characteristics of these materials in the molten state play a vital role in shaping tracks on the surface. The differences in atomic mobilities and surface tensions between MgO and Al_2O_3 accounted for the distinct track morphologies observed. While ion track formation in Al_2O_3 was facilitated by the suppression of recrystallization at the material surface, resulting in an usual nanohillock sequence morphology, MgO's lower viscosity and surface tension caused material loss, leading to the formation of groove-like ion track morphology, as indicated by simulations and experimental findings [23].

Studying amorphizable and non-amorphizable nanocrystalline insulators, we found that the recrystallization processes in MgO and Al_2O_3 strongly and very fast (within 100 ps) affects the morphology of the grains and grain boundaries, activating the grain enlargement resulting from the disorder of the track core, further recrystallization and epitaxial growth of neighboring grains. The fragmentation of grains due to strong heating and expansion of a material in the track periphery is also observed in relatively large nanoclusters. On the other hand, SHI impacts in the amorphizable nanocrystalline YAG exhibits a weaker interaction with grain boundaries due to the lack of recrystallization. This material also tend to have larger track sizes compared to their bulk counterparts, which may be caused by elevated stresses within the nanograins [24].

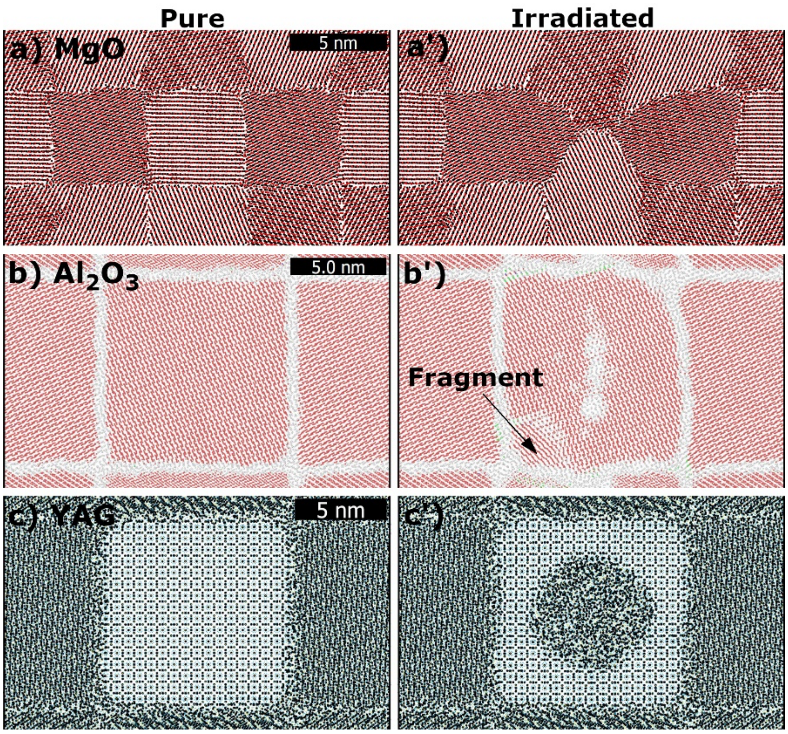

Figure 2. MD cells of nanocrystalline (a) MgO, (b) Al_2O_3 and (c) YAG with cubic grains of 5-10 nm in size before ion impact. (a'-c') shows the same MD snapshots of the same cells after swift heavy ion passage.

To sum up, our research has examined how the high-energy heavy ion irradiation affects the surfaces and interfaces of amorphizable and non-amorphizable dielectrics. We found that the normal irradiation result in formation of spherical and semi-spherical bumps on the surfaces. When an SHI hits the surface at a grazing incidence angle, it causes the formation of a roll-like structure or a groove-like structure surrounded by hillocks. The recrystallization processes observed in non-amorphizable nanocrystalline MgO and Al_2O_3 strongly affects the grain boundaries that may result in association and enlargement of nanoparticles. In contrast, swift heavy ion passage almost does not affect the grains morphology forming amorphous tracks in the amorphizable nanocrystalline sample. Our findings are consistent with recent experiments, and shed light on the mechanisms of extreme excitation of surface and interface regions. These insights offer potentially better control of production of nano-objects by manipulating the parameters of targets and ion beams.

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