EXPLORING SURFACE EXCITATION KINETICS IN DIELECTRICS UNDER SWIFT HEAVY ION IRRADIATION

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Swift heavy ions (SHIs) that penetrate a solid primarily lose their energy through electronic stopping. The high energy deposited by the incoming ion leads to significant electronic excitation on a femtosecond timescale, which is followed by the relaxation of the disturbed electron ensemble. This process is followed by the acceleration of target atoms on sub-picosecond timescales, resulting in the formation of a nanometer-scale damaged region within hundreds of picoseconds after the ion's passage. These structural alterations can have profound effects on the physical, chemical, and mechanical properties of the irradiated material. Due to these characteristics, accelerated ion beams are utilized as versatile tools for patterning and modifying nanometer-sized materials [1, 2], as well as for studying radiation stability in the context of cosmic rays and fission fragments [3-5]. Despite extensive experimental research, the mechanisms underlying surface damage induced by SHI irradiation remain incompletely understood.

This study reviews our findings from simulations of swift heavy ion (SHI) damage in near-surface regions of both amorphizable and non-amorphizable dielectrics irradiated with high-energy heavy ions. To investigate the structural changes in insulators induced by SHI irradiation, we employ a multiscale hybrid model [6-9]. This model integrates the Monte Carlo (MC) TREKIS code [8], which simulates the excitation of the electron subsystem and energy transfer to the target lattice, with molecular dynamics (MD) simulations that model the subsequent relaxation of the atomic subsystem [10-12].

Figure 1 illustrates the simulation results for surface modifications in Al_2O_3, WO_3 [13] and CeO_2 [14] under normal SHI irradiation conditions. The materials studied exhibited distinct relaxation kinetics of lattice energy deposited by the SHI [12]. Understanding the underlying factors driving these differences is crucial for controlling nanofeature formation, which is essential for various nanostructuring technologies. The simulations demonstrate that WO_3 and CeO_2, with their lower viscosities and surface tensions, allow for a more pronounced extrusion of molten material in the early stages, resulting in the formation of spherically shaped crystalline hillocks or even through nanochannels in the case of tungsten oxide. Conversely, the higher surface tension and viscosity of Al_2O_3 restrict the protrusion of a liquid droplet from the surface, leading to the transient formation of semi-spherical hillocks [12], which requires additional post-irradiation annealing for recrystallization, aligning with experimental observations.

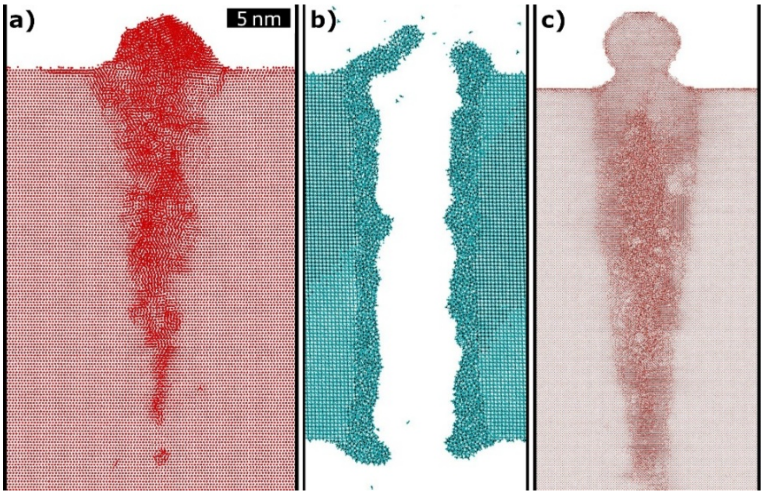

Fig. 1. The images of MD simulated supercells of (a) oxygen sublattice of Al_2O_3, (b) WO_3 [13] and (c) CeO_2 [14] after a passage of a swift heavy ion

The simulation of grazing ion irradiation on CeO_2 (fig. 2 *a, b, c*) reveals the formation of a rift at depths of approximately 1-3 nm, while at 5 nm, the protruded area exhibits a uniform in lenght structure. The kinetics of groove formation, surrounded by hillocks (1 nm depth), closely resemble those observed for CaF_2 in Ref. [15], involving a significant protrusion of molten material followed by the emission of atoms and atomic clusters. However, unlike the observations in CaF_2 (fig. 2 *d, e, f*), where chains of hillocks typically cover the rift at intermediate depths (~3-4 nm), no transition region was identified in CeO_2.

We hypothesize that the difference in surface track formation between CeO_2 and CaF_2 is due to the lower surface tension in CeO_2. As discussed in [15, 16], materials with lower surface tension tend to form grooves more readily. High surface tension inhibits the strong expulsion of molten material, resulting in the formation of a single protruding structure, which may then split into hillocks. In contrast, when surface tension is low, the molten material is expelled more rapidly, leading to the formation of two jets moving in opposite directions.

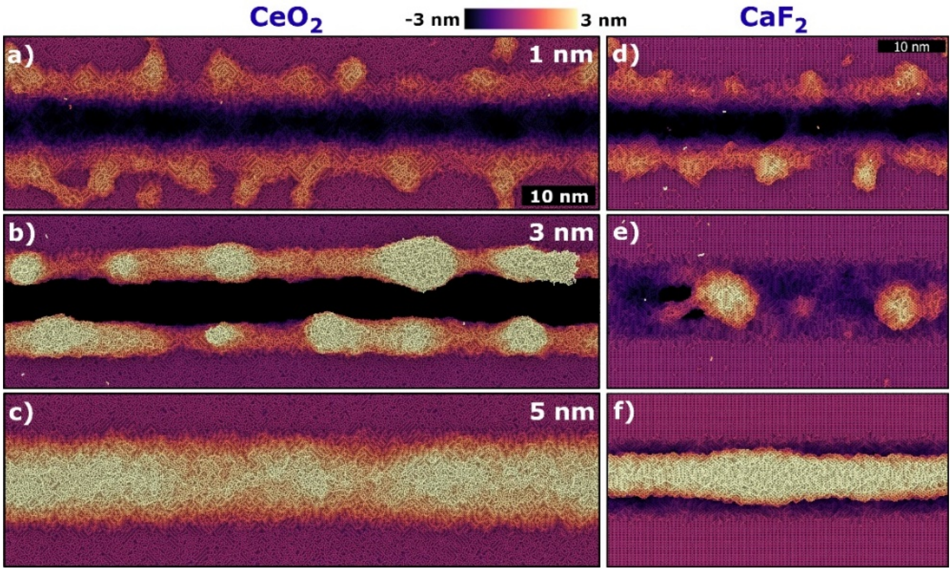

Fig. 2. Results of MD simulation of 200 MeV Au ion impacts parallel to the surface of CeO_2 at (a) 1 nm, (b) 3 nm and (c) 5 nm depth. (d), (e), (f) shows similar simulations for 100 MeV Pb ion in CaF_2. Atoms are colored according to their *Z* coordinates (counted from the surface) to reflect the height of the nanostructures

To sum up, our research has examined how the high-energy heavy ion irradiation affects the surfaces of various dielectrics. We found that the normal irradiation result in formation of spherical and semi-spherical nanostructures 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. Our findings are consistent with recent experiments, and shed light on the mechanisms of extreme excitation of surface and interface regions.

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