TEM BASED VERIFICATION OF A COMPUTATIONAL APPROACH TO STUDYING RELAXATION KINETICS AND DAMAGE ACCUMULATION DUE TO SHI IMPACT IN INSULATORS

Jacques O'Connell[1)], Ruslan Rymzhanov[2)], Vladimir Skuratov[2)]
[1)]*CHRTEM, NMU, University Way, Summerstrand, Port Elizabeth 6031, South Africa, jacques.oconnell@gmail.com*
[2)]*FLNR, JINR, 6 Joliot-Curie, 141980 Dubna, Moscow Region, Russia, rymzhanov@jinr.ru, skuratov@jinr.ru*

An attempt was made in order to experimentally verify the accuracy of our developed combined Monte Carlo and molecular dynamics approach to describing SHI impacts in insulators. Good agreement was found between simulated structures produced by 167 MeV Xe in single crystal MgO, Al_2O_3 and YAG (representing non-amorphizable and amorphisable insulators) and experimental TEM images of such structures. It was found that the computational approach can successfully reproduce the observed damage morphology both in the bulk and at the irradiated surface for a variety of materials. This suggests that fundamental mechanisms responsible for the material modification is successfully captured by the model. We may therefore use the model to study the relaxation kinetics and attempt to elucidate those material properties which determine the nature of the damage produced by SHI impacts in a given material.

Keywords: Swift Heavy Ions; Transmission Electron Microscopy; Molecular Dynamics; Relaxation Kinetics.

Introduction

A swift heavy ion (SHI, E >1 MeV/amu) loses the largest part of its energy (>95%, 5–50 keV/nm along the ion trajectory) through excitation of the electronic subsystem of the target material [1,2]. Subsequent relaxation of the excited volume occurs at ultrashort spatial (nanometers) and temporal (femto- to pico-second) scales and cannot be described in the framework of usual macroscopic models [3,4] and is impossible to follow experimentally. As a result, unusual kinetics often produce unusual structure and phase transformations which constitute a latent ion track: structure modified material with a diameter of ~1–10 nm and a length of ~10–100 μm along the trajectory of the projectile. The identification of mechanisms governing these kinetics forms a fundamental field of interest in SHI track effects. SHI penetration trough different insulators have shown quite different manifestations of structure transformations [1]: amorphous tracks (e.g. $Y_3Fe_5O_{12}$, α-quartz), defected crystalline tracks (e.g. Mg_2AlO_4, Al_2O_3, ZrO_2, TiO_2) or production of isolated point defects and color centers (e.g. MgO, alkali halides). This motivates research aimed at the understanding of mechanisms of track (and surface hillock) formation in these materials. As with most multiscale physics problems, track formation cannot be traced accurately within a single model posing formidable challenges for the theoretical description and understanding of the underlying physical mechanisms. In order to determine which of the above-mentioned process is the main governing mechanism of track formation, we have chosen three dielectric materials (Al_2O_3, MgO and $Y_3Al_5O_{12}$) with different lattice structures but comparable energy deposited into the lattice. It has been shown that SHI irradiation of these targets leads to quite different damage structures, despite the fact that the initial damaged regions have similar sizes and structures.

Currently no experimental technique exists with both the spatial and temporal resolution required to study the relaxation kinetics of single SHI tracks. One possibility however is to use a molecular dynamics (MD) based approach to study the transient behavior of a system during relaxation. However, the behavior of such a system is highly dependent on the chosen potential, simulation parameters and initial energy distribution. In this work we used a combination of TEM image simulation (using the resultant MD cell as object) and experimental TEM micrographs in order to verify the accuracy of the model in reproducing the observable final relaxed state. Once such correspondence is achieved, it is reasonable to use the model to study the relaxation kinetics.

Experimental

We compared the responses of single crystalline α-Al_2O_3, MgO and $Y_3Al_5O_{12}$ (YAG) specimens to irradiation with 167 MeV Xe ions at 300 K.

Irradiations were performed at fluences ranging from 10^{10} to 10^{12} cm^{-2} at the IC-100 cyclotron at FLNR JINR (Dubna, Russia). In order to avoid track overlap, the average ion flux was limited to ~10^9 $cm^{-2}s^{-1}$. Ion beam homogeneity better than 5% at the surface of the irradiated specimen was achieved using beam scanning in horizontal and vertical directions.

High resolution transmission electron microscopy (HRTEM) studies were carried out at the Centre for HRTEM at Nelson Mandela University (Port Elizabeth, South Africa). TEM lamellae were extracted using an FEI Helios Nanolab 650 FIB. In order to observe surface hillocks, the irradiated surface was coated with amorphous carbon to a thickness of 0.5 μm using electron beam assisted carbon deposition in the FIB in order to preserve surface hillocks and provide protection from the 30 kV Ga beam which was used for further carbon deposition (up to 2 μm) and lamellae extraction. Standard FIB procedures were used for plan view specimens and the lamellae were extracted within 1 μm of the irradiated surface. Final thinning was performed at 5 kV Ga beam energy with an additional cleaning step using 2 kV Ga. Samples were imaged using a C_s corrected JEOL ARM-200F TEM operating at 200 kV.

Simulation

A hybrid simulation approach used in this work consists of two models: Monte Carlo simulation (MC code TREKIS [5, 6] of the electron kinetics, and Molecular Dynamics model of atomic dynamics. TREKIS describes the temporal evolution of excited electrons generated by a SHI as well as the interaction of primary and secondary electrons with the target lattice in an ion track. The resulting distribution of energy transferred to the ionic subsystem of the target is inserted into classical MD code LAMMPS [7] which is used to simulate lattice energy relaxation and further structure transformations in the vicinity of the ion trajectory. No a-posteriori fitting parameters are used in the model. For S/TEM image simulation the freely available MuSTEM [8] code was employed.

Results

Despite comparable energy deposition into the lattice (especially for radii >1 nm), the passage of 167 MeV Xe produced notably different damaged structures in the investigated oxides. Figure 1 shows the simulation results for track formation in MgO, Al_2O_3 and YAG with experimental TEM insets (bottom right) for Al_2O_3 and YAG as well as simulated TEM images with comparable observation conditions as the experimental images. MgO showed no experimentally observable track which is consistent with simulation results. This is due to only a few isolated point defects remaining after relaxation. For Al_2O_3, only a slight reduction in contrast is observed at the imaging conditions used but the effect is visible in both the simulated and experimental images near the center. In this case the remaining damage is similar in nature to that of MgO but at significantly higher concentration. In the case of YAG, the simulated and experimental ABF STEM images have almost perfect correspondence apart from the relatively larger track observed experimentally. Despite this size discrepancy however, the general structure of the amorphous track is accurately reproduced by simulation: an almost perfectly circular region of amorphous material with a relatively sharp boundary with no intermediate phase around the edge. It should be noted that the simulation cells had thickness of 14.5 nm, 19.4 nm and 16 nm for MgO, Al_2O_3 and YAG respectively while the experimental images were acquired over regions of about 30-40 nm as measured by EFTEM thickness maps. The discrepancy in size is due to the computational requirements for larger simulation cells and the difficulty of producing extremely thin TEM foils of good quality.

Figure 2 shows simulation results of a 167 MeV Xe ion impacting the surface of a YAG crystal (top) representing an amorphizable insulator and an MgO crystal (bottom) representing a non-amorphizable insulator. An experimental TEM micrograph (thermally colored to improve contrast) is inset for both systems at the same scale. It is clear that the simulation accurately reproduces the final size and structure of the surface hillock in both cases. In the case of YAG, the hillock is amorphous and exhibits a mound-like structure. In contrast, the hillock in MgO has a more spherical shape and is both crystalline and epitaxial to the bulk crystal.

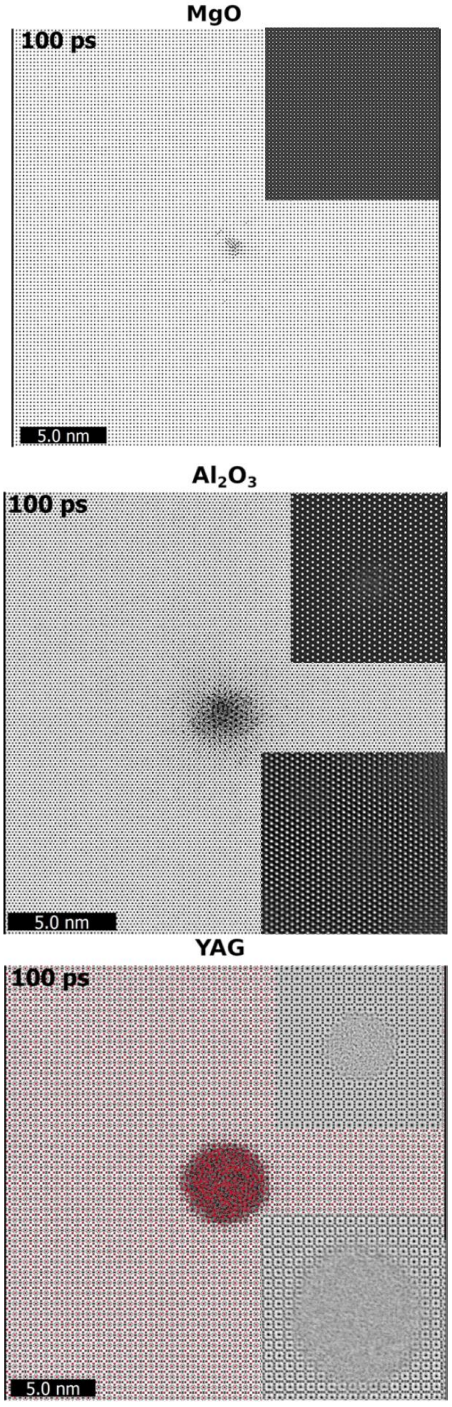
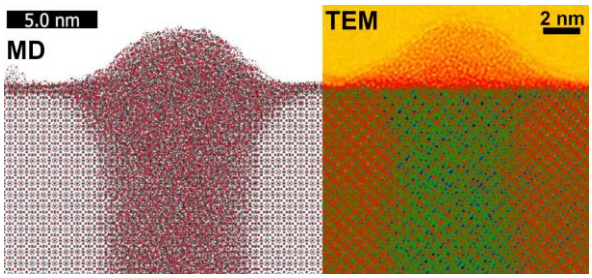


Fig. 1. Snapshots of modeled 167 MeV Xe tracks in three materials at 100 fs after ion passage with the experimental (bottom right) and simulated (top right) TEM images as insets. The scales of MD images and TEM insets are the same

These results suggest that the employed interatomic potentials as well as initial energy distribution due to SHI passage reasonably accurately reproduce the relaxed structures observable by TEM. Based on this observation, it is reasonable to consider the relaxation kinetics within the MD simulation as representative of the actual system. Full interrogation

of the dynamic processes and relaxation kinetics is beyond the scope of this paper and will be reported at a later stage.



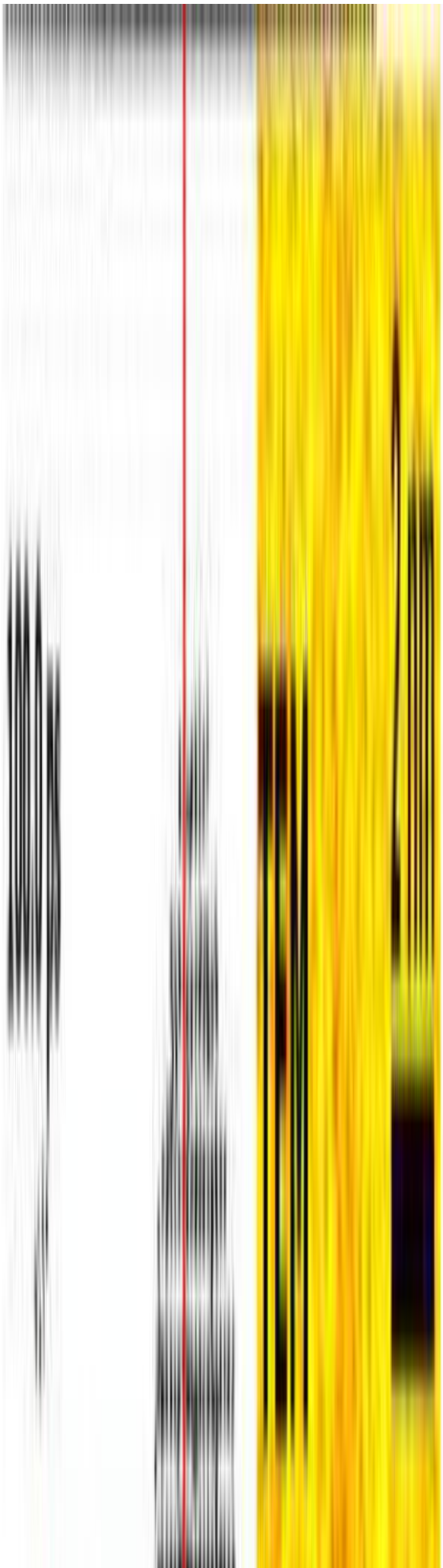


Fig. 2. Snapshots of modeled 167 MeV Xe tracks at the surface of amorphizable (YAG) and non-amorphizable (MgO) insulators during mass ejection and after relaxation. The thermal colored TEM micrograph insets are at the same scale as that of the simulation cell

Conclusions

It was found that our combined MC/MD theoretical approach could sufficiently reproduce the observed structures formed in due to 167 MeV Xe impact in MgO, Al_2O_3 and YAG representing non-amorphizable and amorphizable insulators. Good agreement was found between the structures produced by simulation in the bulk and on the irradiated surface with those observed experimentally. This suggests that the model effectively reproduces the most important aspects of the relaxation process and could be used to interrogate the material parameters responsible for determining the material response to excitation by a SHI.

References

1. Komarov F.F. *Physics-Uspekhi* 2017; (60): 435–471.
2. Martynenko Y.V, Yavlinskii Y.N. *Sov. At. Energy* 1987; (62): 93–97.
3. Toulemonde M., Dufour C., Meftah A., Paumier E. *Nucl. Inst. Meth. B* 2000; (903–912): 166–167.
4. Duffy D.M., Daraszewicz S.L., Mulroue J. *Nucl. Inst. Meth. B* 2012; (277): 21–27.
5. Medvedev N.A., Rymzhanov R.A., Volkov, A.E. J. Phys. D. Appl. Phys. 2015; (48) 355303.
6. Rymzhanov, R.A., Medvedev, N.A., Volkov A.E. *Nucl. Inst. Meth. B* 2016; (388): 41–52.
7. Plimpton S. Comput. Phys. 1995; (117): 1–19.
8. Allen L.J., D'Alfonso A.J., Findlay S.D. Ultramicroscopy 2015; (151): 11-22.