MICROSCOPICS OF ION TRACK FORMATION IN THE BULK AND AT THE SURFACE

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The formation of ion tracks in the bulk and near-surface regions of various dielectrics have been studied using a multiscale approach combining Monte-Carlo code TREKIS and molecular dynamics simulations. It was demonstrated that the morphology of swift heavy ion tracks in non-amorphizable oxides is determined by recrystallization of the initially disordered region, which, in turn, is governed by the kinetics of the metallic and oxygen sublattices at the crystallization surface. The presence of a free surface can significantly suppress the lattice recovery rate, producing larger structure modified tracks in the near-surface regions. Matching of simulation results with TEM images of crystalline hillocks and tracks in Al_2O_3 revealed that the recrystallization of sub-surface tracks (~20 nm depth) in this solid requires much longer times in comparison to bulk simulations. The recrystallization processes strongly manifest themselves in the overlapping tracks regime. Overlapping of tracks in non-amorphizable solids could realize in several modes depending on the distance between the tracks: from almost complete structure recovery, to the formation of connected or isolated tracks. In contrast, track interaction in amorphizable targets has almost no effect on the defective region morphology.

Keywords: electronic excitation; surface damage; swift heavy ion; molecular dynamics; dielectrics.

Introduction

Electronic stopping of a swift heavy ion (SHI) in a solid and further relaxation of the excess energy of excited electrons and lattice cause the formation of nanometric structure-modified regions around the ion trajectory. Such transformations of the structure of a material can significantly change the physical, chemical and mechanical properties of a solid, as well as its radiation resistance to other kinds of irradiations [1].

SHI irradiation is used in a wide range of applications such as creation of nanodots, production of track-etched membranes and nanostructuring of solids [1], etc. Cosmic ray effects on electronics and biological tissues, is also modeled through irradiations with SHIs.

A fundamental interest in SHI irradiation effects arises from an extremely high level of material excitation and the ultrashort spatial and temporal scales of the subsequent kinetics. Such extreme conditions restrict an applicability of models based on macroscopic conceptions to track problems and requires appropriate microscopic approaches [2, 3].

In the present work, we review such an approach, which describes all the successive stages of SHI track formation, to shed light on the main processes of excitation of bulk dielectrics and their surface regions.

Methods

Single crystalline α-Al_2O_3, MgO and $Y_3Al_5O_{12}$ (YAG) specimens were irradiated with 167 MeV Xe and 710 MeV Bi ions at 300 K. The irradiations were performed at fluences ranging from 10^{10} to 10^{13} cm^{-2} at the cyclotron complex of FLNR JINR (Dubna, Russia). High-resolution transmission electron microscopy (HRTEM) studies were carried out at the Centre for HRTEM at Nelson Mandela University (Port Elizabeth, South Africa).

An original multiscale model was applied to study the excitation and relaxation kinetics of the electronic and the ionic systems of a target exposed to swift heavy ions [4]. The approach couples the asymptotic trajectory Monte Carlo (MC) code TREKIS [5] used to determine the initial parameters of solid excitation with the classical molecular dynamics code LAMMPS [6] simulating structure transformations in the vicinity of the ion trajectory.

Results and discussion

It was shown that 167 MeV Xe ion irradiations induces notably different damaged structures in considered targets, despite similar energy deposition and almost identical size of the initially disordered region after ion impacts [7].

MgO demonstrate no clear SHI track formations, which was also observed experimentally [8]. Only a few point defects near the ion trajectory are found. Ion passage in Al_2O_3 forms discontinuous crystalline tracks of ~2±0.3 nm in diameter, which coincide well with the HRTEM studies of irradiated samples. This was discussed in detail in Ref. [9]. A track in YAG is a cylindrical amorphous region with the size of ~5.1±0.2 nm, in a reasonable agreement with TEM.

This difference can be attributed to the strong recrystallization ability of MgO and Al_2O_3 to, whereas YAG has no such behavior.

The recrystallization of a damaged region after an SHI impact can be very fast (~20-70 ps), which does not allow detection of this effect directly through existing experimental techniques. One of the possible ways of an indirect study of such processes is a comparison of track morphology at different fluences: individual tracks ($<10^{11}$ cm^{-2}), overlapping tracks ($\sim10^{12}$ cm^{-2}) and multiple overlapped tracks ($>10^{13}$ cm^{-2}). One could expect a strong effect of newly produced tracks on the existing defects in the case of high recrystallization efficiency [4].

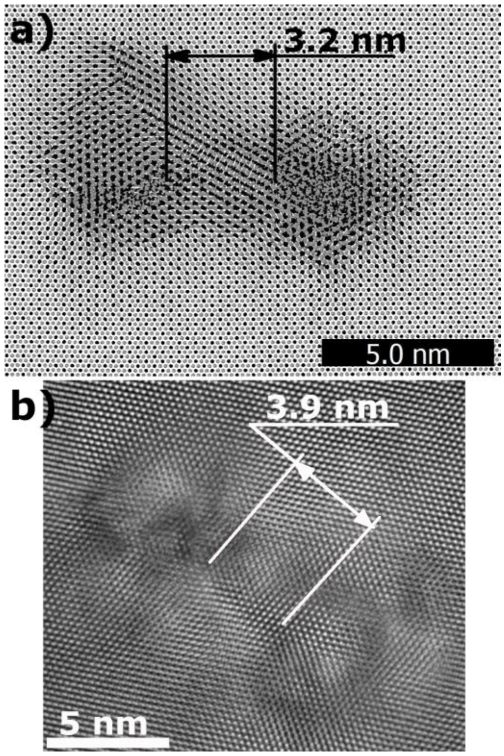

Fig. 1. (a) Results of a simulation of two subsequent impacts of Bi 700 MeV ions in Al_2O_3 at 6.5 nm between trajectories. (b) TEM image of 700 MeV Bi ion tracks in Al_2O_3

Simulation reveals that impacts of Bi ions in Al_2O_3 at a short separation distance cause almost perfect annealing of the existing defective structure. Existing tracks recover only partially at a distance of ~6.5 nm between track centers (3.2 nm between edges, Figure 1). A damaged region also appears, connecting the two tracks. The damaged structure between close tracks is also observed experimentally in the TEM micrograph, confirming the model predictions (4), as shown in Figure 1. Increasing of the distance between tracks up to ~8 nm results in formation of isolated tracks.

The influence radius of the second track in YAG is smaller than that in alumina. It was demonstrated numerically and experimentally (4) that the inter-track connection forms at distances of ~1 nm between their edges, which seems mainly due to material expansion after transient melting rather than the recrystallization during cooling. At a distance of 2 nm between edges, tracks in YAG become isolated.

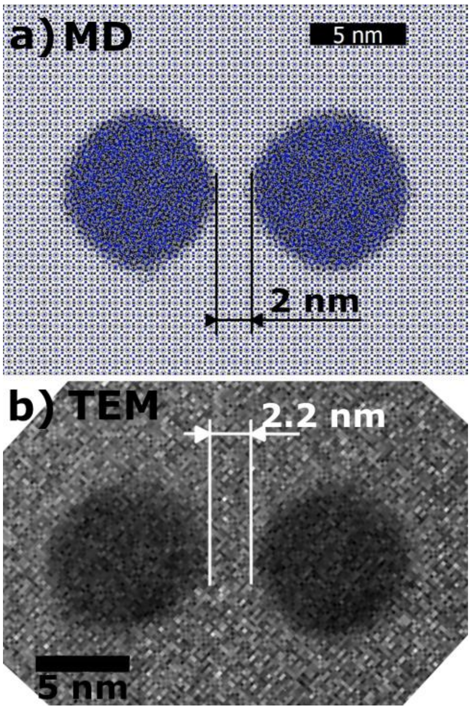

Fig. 2. (a) Results of a simulation of two subsequent impacts of Bi 700 MeV ions in YAG at 10 nm. (b) TEM image of 700 MeV Bi ion tracks in YAG

The small value of the radius of track interaction in YAG may be interpreted as an indirect experimental evidence of the absence of, or very low recrystallization rate of YAG in SHI tracks.

We have shown that recrystallization ability depends on several factors including lattice structure, mobility of atoms etc. (7). In this work, we also demonstrate that a free surface can strongly suppress recrystallization in the near-surface region. Fig. 3 shows the formation of a conical crystalline track in alumina after an ion impact. Surface effects can be seen up to a depth of ~25 nm, below which the track has a discontinuous morphology. The subsurface damaged region has a polycrystalline structure. TEM images show a reasonable agreement in the size, shape and structure of a track (10). It should be noted that according to MD results, recrystallization in the near-surface region is possible only at elevated temperatures, which should last at least 600-800 ps, much longer in comparison to the bulk recovery typically lasting for 50-70 ps. The question arises whether there is any source of track heating that could last for such long times, which forms the topic for future dedicated studies.

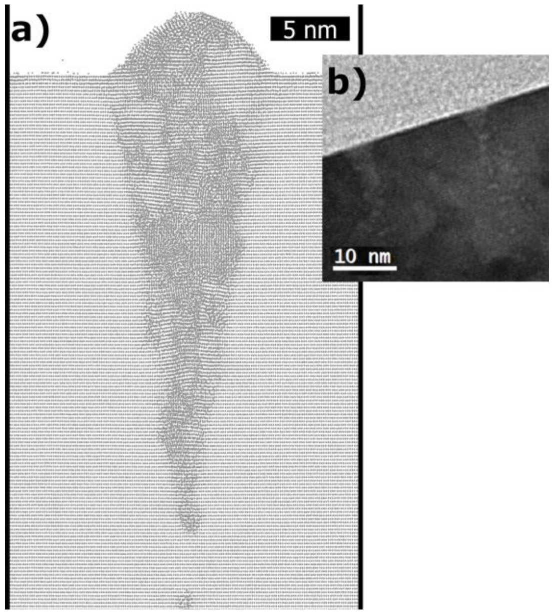

Fig. 3 (a) MD simulation of a 167 MeV Xe ion impact in Al_2O_3 (2 nm slice of oxygen sublattice at 1 ns). (b) TEM image of conical tracks in Al_2O_3

Conclusions

We showed that in bulk non-amorphizable materials, a damaged structure in a SHI track can recover to a nearly perfect crystalline state. Amorphizable materials show almost no recovery, and the size of the transient molten region coincides with the final track. A correlation between the crystal structure and the efficiency of the material recovery in SHI tracks was revealed.

Overlap of two consequent tracks may anneal the first one, if the second track is sufficiently close. At larger distances, overlap of transient molten zone with existing track results in a connecting damaged structure between the two tracks.

The presence of a free surface strongly affects the recrystallization process producing conically shaped tracks. The crystallization of such near-surface tracks can take much longer times in comparison with the bulk.

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