TRACK INTERFERENCE IN SWIFT HEAVY ION IRRADIATED Al_2O_3

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The morphology of latent ion tracks induced by high energy heavy ions in Al_2O_3 have been studied using a combination of high resolution TEM (HRTEM), exit wave reconstruction, geometric phase analysis and numerical simulations. Single crystal α-Al_2O_3 crystals were irradiated with 167 MeV Xe ions along the c-axis to fluences between 1 x 10^{10} and 1 x 10^{13} cm^{-2}. Planar TEM lamella were prepared by FIB and geometrical phase analysis was performed on the phase image of the reconstructed complex electron wave at the specimen exit surface in order to estimate the latent strain around individual track cores. In addition to the experiments, the material excitation in a SHI track was numerically simulated combining Monte-Carlo code describing the excitation of the electronic subsystem with classical molecular dynamics of the lattice atoms. Experiment and calculation both demonstrated that the relaxation of the excess lattice energy results in the formation of a cylinder-like disordered region of about 4 nm in diameter consisting of an underdense core surrounded by an overdense shell. Modeling of the passage of a second ion in the vicinity of this disordered region revealed that this damaged area can be restored to a near damage free state. The estimation of a maximal effective distance of recrystallization between the ion trajectories yields values of about 6 - 6.5 nm which are of the same order of magnitude as those estimated from the saturation density of latent ion tracks detected by TEM.

Introduction

In this paper we continue the TEM study of the morphology of latent tracks induced by high energy heavy ions in Al_2O_3 started in [1], and complement it with results on the size and shape of the latent strain field around single ion tracks using high resolution TEM (HRTEM) and classical molecular dynamics (MD) modeling. Recently, it was shown that the kinetics of valence band holes in insulators plays an important role in SHI track creation [2]. It was demonstrated that coupling of only the excited electrons to the lattice does not provide sufficient energy for a detectable structure transformations in SHI tracks. Only when the excess energy of valence band holes was included, the track radii formed in the modeling were similar to those found in experiments [1]. In the present work we compare the MD results to experiment and try to estimate the effective radius of the dynamic ion track produced by 167 MeV Xe ions representing the area over which the structure of existing latent tracks could be affected.

Experimental

Single crystal α-Al_2O_3 crystals were irradiated with 167 MeV Xe ions to fluences in the range 10^{10} – 10^{13} cm^{-2}. TEM lamellae were prepared using an FEI Helios Nanolab 650 FIB. Milling and initial thinning was performed with 30 keV Ga ions and final polishing was performed using 1 kV Ga ions. Lamellae were imaged using a third order image corrected JEOL ARM-200F TEM operating at 200 kV. Electron exit wave reconstruction was performed on through focal series HRTEM micrographs using the FTSR plugin for Digital Micrograph by HREM Research inc [3]. Local strain maps were computed from the phase component of restored electron exit waves using the GPA plugin for Digital micrograph from HREM Research inc [4]. In addition to the experiments, the excitation of α-Al_2O_3 in a SHI track was simulated with the help of classical molecular dynamics using the classical MD code LAMMPS [5]. The velocity distribution of the Al and O atoms in proximity around the ion trajectory was obtained from a Monte-Carlo model describing the excitation of the electronic subsystem of a target inside a SHI track [6].

Results and Discussion

Figure 1 shows a BF TEM micrograph recorded along the c-axis of an Al_2O_3 crystal irradiated with 1 x 10^{13} cm^{-2} Xe ions. Some of the tracks are clearly visible while older tracks appear as faint background spots.

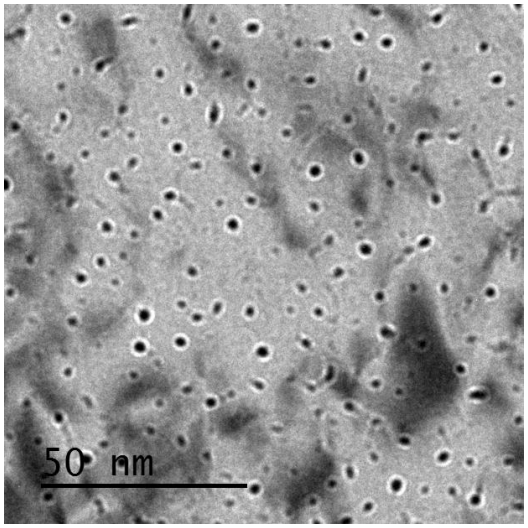

Fig. 1. Plan view BF TEM micrograph of 167 MeV Xe ion latent tracks along the c axis in Al_2O_3.

Figure 2 shows the result of counting individual tracks in micrographs such as these for fluences between 2 x 10^{12} and 1 x 10^{13} cm^{-2}. The data suggests a saturation track density around 1.1 x 10^{12} cm^{-2} which corresponds to an effective radius of influence within which previous tracks will be recovered of about 5.4 nm.

Figure 3 shows high resolution image of single Xe ion track in alumina. Using a reconstructed phase image of the complex electron wave at the exit surface of α-Al_2O_3 crystal irradiated with 2 x 10^{10} cm^{-2} Xe ions we computed lattice dilation in the basal plane. It was found that track region consists a low

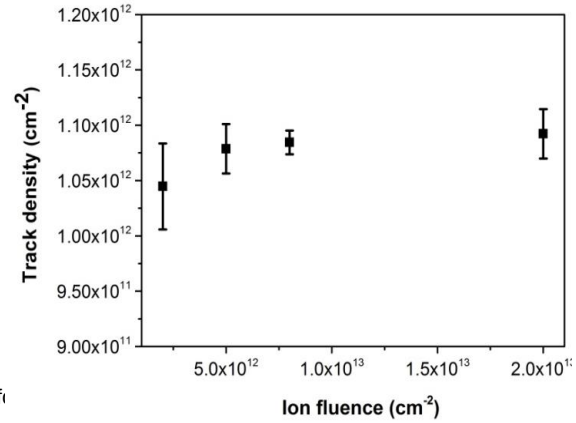

Fig. 2. TEM observed track density versus ion fluence for 167 MeV Xe in Al_2O_3.

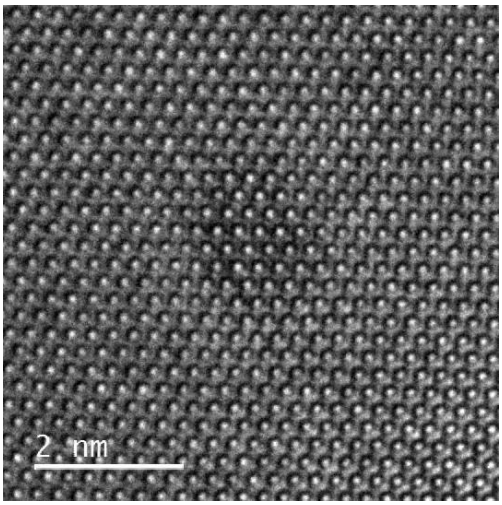

Fig. 3. HRTEM image of 167 MeV Xe ion track in Al_2O_3.

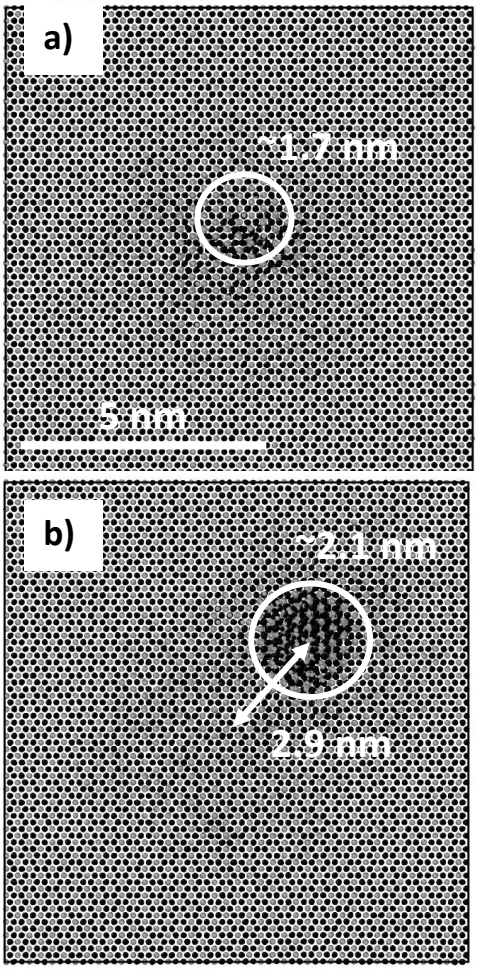

Fig. 4. (a) Simulated ion track in Al2O3 after passage of a 167 MeV Xe ion. (b) The same system as in (a) after passage of a second 167 MeV Xe ion 2.9 nm from the first. The first track is almost completely recovered and a significant size difference exists between the two tracks. Al atoms are blue dots and O atoms are red dots.

density core with a radius of about 1 nm surrounded by a ~2 nm radius shell of slightly higher than average density.

Figure 4a shows the result of simulation after the passage of a 167 MeV Xe ion along the c axis in an Al_2O_3 crystal. The highly disordered region has a diameter of about 1.7 nm. Figure 4b shows the same system after passage of a second 167 MeV Xe ion at a distance of 2.9 nm from the original trajectory. It is clear from the figure that the original track has been almost fully recovered by the energy transfer of the second ion. Also noticeable is the size difference between the first and second ion track with the second track being significantly larger. Recent results on $SrTiO_3$ [7] shows enhanced track formation in the presence of existing defects caused by low energy ion implantation. The authors suggest that this is due to a reduction of thermal conductivity and an increase in electron-phonon coupling due to the defects which consequently enhances the local thermal spike due to the passage of a swift heavy ion. A similar effect might in part be responsible for the larger track diameters of the second ions observed our simulations. However, our simulation box is relatively small and the second ion passes relatively close to the cooled edges of the simulation box as compared to the first ion. This can affect the size and shape of ion tracks far from the center of the simulation box. Whether the increase in size is due purely to this effect or a combination of the effect of pre-existing damage and the proximity to the simulation box edge is not clear at present but will be reported on in a follow up article. This simulation of the second ion passage was repeated several times with increasing separation between trajectories in order to estimate the effective radius of influence outside of which the initial track was not significantly affected. This distance was found to be approximately 6 – 6.5 nm. The corresponding maximal density of ion tracks is ~ 8.8×10^{11} cm^{-2}, which is in a good qualitative agreement with the experimentally observed saturated ion track density of 1.1×10^{12} cm^{-2}. Difficulty in identifying faint tracks from TEM micrographs probably leads to a slight under estimation of the saturation track density and thus over estimation of the radius of influence through this method. The proximity of the second ion track to the cooled edges of the simulation box will lead to a higher rate of energy removal from the system and could possibly reduce the radius of influence inferred from this technique.

Figure 5a shows a plot of normalized density as a function of radial distance from the track center for the MD simulation shown in figure 4. The curve was computed by calculating the average density of cylindrical layers of a given radius centered on the track and thus represents full 3D density. Figure 5b shows a plot of 1 – mean basal plane dilation as a function of radial distance from the estimated track center for two experimentally observed Xe tracks. This data contains information only in the basal plane and thus the y axes in the two plots are not directly comparable. However, the general shape and size of the density fluctuation of simulation and experiment are in good agreement. The simulated track core size falls within the variation of the experimentally observed and computed core sizes, as does that of the overdense shell. The lower density nature of the core and higher density shell is also reflected in both (a) and (b).

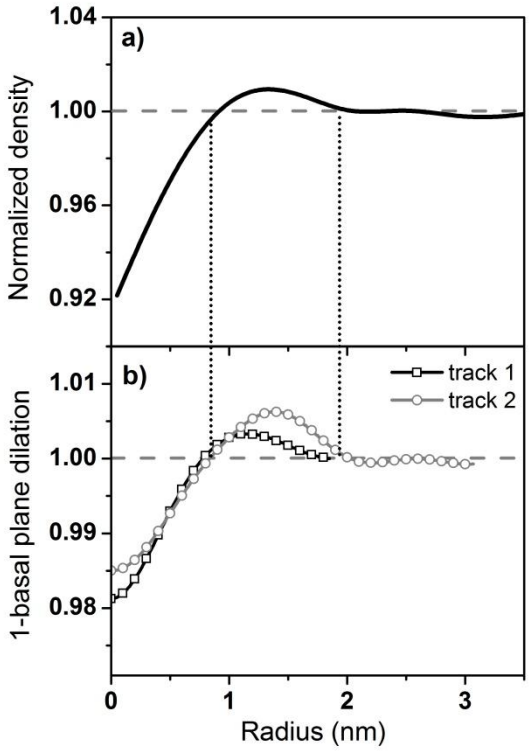

Fig. 5. (a) Normalized density variation as a function of radial distance from the track center as calculated from MD simulation. The values at the very center of the track (distance ~ 0) are not statistically accurate due to the low number of atoms. (b) Two experimental plots of 1 – mean basal plane lattice dilation as a function of radial distance from the track center.

Conclusion

The results of this investigation suggest that He The size and shape of the strain field around latent tracks of 167 MeV Xe ions in Al_2O_3 was studied using HRTEM and compared with the results of MD simulation. Good coincidence was observed between simulation and experiment indicating an applicability of used theoretical approach. TEM and MD agree well on the radii of 167 MeV Xe latent track cores (1.7–2.1 nm) and shell diameters of 3.7 – 5 nm, as well as on the magnitude of the strain inside track cores (~1.5 – 2 %). This underdense core is surrounded by an overdense shell where the increase in density is less than 1% above that of unstrained material. The synergy of the experimental and simulation studies of the structure of 167 MeV Xe ion tracks revealed a probable reason for the observed saturation of the track density. This effect arises from partial annealing of the overlapping ion tracks. The effective distance of track recrystallization is about 6 - 6.5 nm (from simulation) is in a qualitative agreement with the experimentally deduced value of ~ 5.4 nm.

Acknowledgments

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References

1. *Skuratov V.A. et al.* // Nucl. Instr. Meth. B. 2014. V. 326. P. 223.
2 *Terekhin P.N. et al*. // Nucl. Instr. Meth. B. 2015. V. 354. P. 200.
3. *Meyer R.R., Kirkland A.I., Saxton W.O*. // Ultramicroscopy. 2002. V. 89. P. 109.
4. *Hytch M.J., Snoeck E., Kilaas R.* // Ultramicroscopy. 1998. V. 74. P. 131.
5. *Plimpton S.* // J. Comput. Phys.1995. V. 1177. P. 1.
6. *Rymzhanov R.A. et al*. // Nucl. Instr. Meth. B. 2014. V. 326. P. 238.
7. *Weber W.J.* // Scientific Reports, DOI: 10.1038/srep07726.