Mechanical response to swift ion irradiation-induced nano-tracks in silica


Ion irradiation on dielectric materials produces several processes, such as ionization and defect formation followed by a decay governed by thermal processes such as heat diffusion and atomic rearrangement. Finally in the irradiated region the mechanical properties are altered, strain and stress fields appear, a densification takes places and other properties such as the refractive index are affected.

In order to simulate the mechanical response of silica to swift ion irradiation we use a methodology based on molecular dynamics (MD) and finite element methods (FEM). We use information from MD to obtain the local densification generated by an incoming swift ion. Finally we calculate the densification in the ion track using FEM. This method provides information on the strain and stress field along the material as a function of ion irradiation fluence.

For this work an experimental campaign using Br ions from 5 to 50 MeV has been done at CMAM accelerator (Madrid). We measured the refractive index and we observe that for high fluences the refractive index decreases. The effect of the strain field on the density could explain the decrease in the refractive index. We check this hypothesis using our methodology coupling MD and FEM.

1. Introduction

Heavy ion irradiation in silica with energies >0.1 MeV/amu is the origin of several effects of technological relevance. On one hand, it leads to material modification, usually, with detrimental results (as in Nuclear fusion facilities [1]) and on the other hand, it can be used to optimize or tailor the physical properties, e.g., as in waveguide fabrication [2,3].

In this type of irradiation, energy is transferred from the projectile mainly to electrons that subsequently produce significant impact ionization. As a result, in a nm-sized region around the ion trajectory a high electronic excitation density is achieved. Due to several processes the stored energy is transferred to the lattice in ps timescales. The consequence of the sudden energy enhancement is strong atom disorder, bond breaking and temperature raise followed by a fast atom rearrangement and cool-down. Permanent effects appear in nm-sized tracks with modified density, stoichiometry, composition and defect level that seriously affect the physical properties.

Effects of ion irradiation in silica have been already studied in different works [4–7]. In these works the track morphology, stresses and strains under irradiation are explained. However no study shows the variation of the refractive index with the ion fluence. Therefore we study how the optical properties of silica are modified upon ion irradiation. We try to correlate the observed changes in the refractive index to variations in the stress/strain fields originated by the irradiation. The conclusion is that the overall mechanical properties affect marginally to the refractive index variation.

2. Experiments

Br ion irradiation (5–40 MeV) of silica was done at CMAM [8]. In situ optical reflectance measurements were carried during the irradiations as depicted in Fig. 1. The measurements show that the reflectance increases with fluence in all cases. It is straightforward to obtain the near-surface refractive index variation during
irradiation from the reflectance (25 MeV Br irradiation in Fig. 2). Br ions produce tracks with higher density than the virgin material in the silica surface. Initially, when tracks do not overlap the refractive index variation is linear. However, when track overlapping becomes important the refractive index tends to saturate. An interesting effect occurs for fluences >10^{15} cm^{-2}, because, unexpectedly, the refractive index decreases.

We have compared the experimental results with a parametric model. In the parametric model we relate the refractive index with the near-surface density using the Lorentz–Lorenz formula [9],

\[ n = \frac{2}{b} \frac{\rho}{8.0324} \delta n \]

where \( \rho \) is the density and \( n \) is the refractive index. In order to calculate the refractive index in a heterogeneous medium (tracks and substrate), the Bruggeman expression is used,

\[ f_A \frac{2^n}{n^2} \frac{\rho_f}{n_f^2} + f_B \frac{2^n}{n^2} \frac{\rho_B}{n_B^2} = 0, \]

where \( f \) is the volume fraction in the near-surface region and subscripts \( A \) and \( B \) refer to tracks and substrate, respectively. The volume fraction of tracks of radius \( r \) is related to the fluence \( \phi \) through a Poisson distribution,

\[ f_A \frac{4}{3} 1 - e^{-\phi r^2}; \]

In Fig. 2 we compare the experimental results to the parametric model. Good agreement is achieved at fluences \( \phi < 10^{15} \) cm^{-2} for track radius \( r = 3 \) nm and a density variation, \( D\rho/q = 3.5\% \). However, at high fluences, \( \phi > 10^{15} \) cm^{-2} the experimental refractive index decreases whereas the model predicts saturation. Therefore, the model cannot explain correctly the observed behaviour at high fluencies when track overlapping occurs and a continuous layer is formed.

The question is why the refractive index decreases when the continuous layer is formed. In the next section we address this question and study the effect of strain relaxation when the continuous layer is formed on the refractive index.

3. Simulations

We used a finite element method (FEM) using Ansys APDL to study the strain field and relaxation at high fluences. The irradiation effect is taken into account in the FEM model through density variation at different fluences. By means of MD simulations (details published elsewhere) we estimate the track density for different electronic stopping power (\( S_e \)). In order to model the thermal evolution of the track region, we have used atomistic molecular dynamics with the Feuston–Garofalini interatomic potential [10]. The simulation boxes have 30 \( \times \) 30 \( \times \) 14 nm\(^3\) in order to simulate

\[ > 8 \times 10^{15} \text{ atoms in a time domain that exceeds 100 ps}. \]

The simulation code was MDCASK [11] run in 256–512 cores at CESVIMA-MAGERIT. Ion irradiation in the simulation box is characterised by an energy deposition \( (S_e) \) in a hot cylinder. The thermal evolution is studied and the final state (see Fig. 3 for \( S_e = 6 \text{ keV/nm} \)) shows the effects of ion irradiation in silica.

From the MD simulation for different \( S_e \) and the \( S_e \) profile along the ion track depth, we can reconstruct the track shape. In Fig. 4a the density variation of an isolated track is shown as a function of depth and axial coordinates in a 2D colour map. As the fluence increases tracks overlap, see Fig. 4b. Finally, a continuous layer is formed, Fig. 4c. According to the MD simulations, 25 MeV Br ion irradiation \( (S_e = 6 \text{ keV/nm at the surface}) \) leads to the formation of tracks with a radius at the surface, \( r > 3 \) nm and a density increase \( D\rho/q \approx 3\% \). This type of tracks agree with the parametric model used to reproduce the experimental results in Fig. 2 (\( r = 3 \) nm, \( D\rho/q = 3.5\% \)).

The track shape shown in Fig. 4 is used as an input for the FEM model to obtain the strain and stress fields in the 25 MeV Br ion irradiated silica for any fluence. Isolated tracks do not substantially

Fig. 1. Schematic representation of the setup for in situ reflectance measurements. A sample can be irradiated with ions and at the same time illuminated with white light (arrows). The reflected light is detected by a spectrometer.

Fig. 2. Variation of refractive index in silica as a function of fluence for 25 MeV Br irradiation. For comparison, results from the parametric models with \( D\rho/q = 3.5\% \) and \( r = 3 \) nm are shown.

Fig. 3. Densified Track after irradiation with an ion of 6 keV/nm. In the track the green arrows shows the displacement vector for the atoms, the average displacement towards the track centre leads to densification of the track.
We have focused on the possibility of a density decrease due to the strain relaxation at high fluxes. In fact, the effect occurs but the density decrease is significantly smaller than the density increase due to track accumulation. We conclude that the strain relaxation is not responsible for the decrease in the refractive index at high fluxes.

Once this a priori obvious assumption is discarded, we are working on an alternative explanation to the effect based on reflectance changes due to enhanced rugosity.

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References