

## Structural analysis of simulated swift heavy ion tracks in quartz



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### ARTICLE INFO

#### Article history:

Received 17 July 2013

Received in revised form 29 October 2013

Accepted 29 October 2013

Available online 7 February 2014

#### Keywords:

Molecular dynamics

Ion irradiation

Ion tracks

Swift heavy ions

Quartz

### ABSTRACT

Swift heavy ions (SHI), of specific kinetic energies in the excess of 1 MeV/u, can create cylindrical regions of structural transformation in SiO<sub>2</sub> targets, also known as SHI tracks. Recent measurements of the track cross-sections in  $\alpha$ -quartz show significant and consistent discrepancies across different experimental techniques used. In particular, the track radii obtained from channelling experiments based on the Rutherford Backscattering Spectrometry (RBS-c) method increase monotonically with the electronic stopping power, whereas the track radii obtained from the Small Angle X-ray scattering (SAXS) saturate past a certain stopping power threshold. We perform a systematic study of the structure of the  $\alpha$ -quartz tracks obtained from the molecular dynamics (MD) simulations incorporating a time-dependent energy deposition based on the inelastic thermal spike model, which allows us to discuss the possible origins of these experimental discrepancies.

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## 1. Introduction

Swift heavy ion (SHI) projectiles can produce cylindrical regions of structural modification in an irradiated target, also known as ion tracks. These features were first observed in nuclear fission reactors [1] and since then they have been widely studied due to their growing list of technological applications. In particular, predicting the exact structural changes in quartz under SHI irradiation, would find immediate applications in nano-fabrication of optical devices [2], as quartz's refractive index changes in response to irradiation.

Ion tracks can pose a challenging modelling problem as they originate from a transient, highly non-equilibrium state. Due to the high specific kinetic energies of SHIs ( $\gtrsim 1$  MeV/u) they mainly interact inelastically with electrons of a target material, producing a trail of electronic excitations, whilst the probability of a direct projectile–target atomistic collision is very small and often neglected. The two types of interaction can be quantified by the respective energy losses of the projectile per unit distance. These so-called electronic and nuclear stopping powers,  $S_{e,n} = \frac{\partial E_{e,n}}{\partial x}$ , are customarily estimated from SRIM computer code [3]. Since  $S_e \gg S_n$  for SHIs, only the electronic component is typically considered and hereafter referred to as the stopping power.

The electronic excitation energy is initially highly localised around the projectile path and later transferred to the atom lattice. The subsequent thermal relaxation leaves the structural changes effectively quenched in, producing an ion track. Nonetheless, the exact energy transfer mechanisms involved are debated and presumably vary with the class of the target material. Track formation models range from Coulomb explosion [4], variations of the inelastic thermal spike (i-TS) [5] to structural relaxation methods [6].

The i-TS model, a variant of which we adapt here, is based on the two-temperature approach [7], where distinct electronic ( $T_e$ ) and ionic temperatures ( $T_i$ ) are assumed. The spatio-temporal evolution of these is governed by a set of two separate heat diffusion equations, linked by the energy exchange term proportional to the electron–phonon (e–p) coupling [8]. Thus far, the model was used to model SHI interaction with metals [9], alloys [10], semiconductors [11] and insulators [12]. While its application to band-gap materials remains controversial [13], especially in the light of more sophisticated methods being developed for semiconductors [14], it remains widely used. This is due to its relative simplicity and thus a relatively small set of parameters required to be fitted, its success history of reproducing the experimental latent track radii variation with the stopping power, and accounting for the velocity effect [15]. While we acknowledge the limitations of the i-TS model, we use it as an exemplar technique to primarily focus on the structural analysis of the simulated SHI tracks in  $\alpha$ -quartz.

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From an experimental point of view, the track properties measured from RBS-c and SAXS (and referred to as radius) are not the same. RBS-c measures the fraction of the damaged material (the atoms outside their crystalline structure) [16] compared to the pristine matrix while SAXS is sensitive to the electron density fluctuations regardless of any change in the crystallinity.

A comparison of the recent experimental data [17] (see also Fig. 1) shows that the latent track radii in quartz vary significantly above the stopping power of about 17 keV/nm. We aim to relate the experimental discrepancies to the structural analysis of the simulated ion tracks in SiO<sub>2</sub> and provide a set of methods that can be used to determine the simulated ion track radii.

## 2. Method

The simulations were performed with the classical MD code PARCAS [18]. We included the effect of the i-TS model through a time-dependent deposition of kinetic energy in random directions for all the atoms in the simulation cell, using radially symmetric energy deposition profiles. The profiles were based on the i-TS calculations with the model parametrisation detailed in [16]. The simulation cell consists of a 23 nm wide cube of initially relaxed and perfectly crystalline  $\alpha$ -quartz configuration with periodic boundary conditions imposed and edges of the cell cooled with a Berendsen thermostat at 300 K [19]. A computation time of 100 ps with an adaptive timestep (typically about 0.4 fs) was used. The interatomic interactions were calculated with the Watanabe–Samela Si–O mixed system many-body potential [20,21].

To study the structure of the simulated latent tracks, we used the coordination defect number, radial density, and broken bonds criteria. While the coordination defects (i.e. atoms with wrong coordination number) can be identified using the pair correlation function within a cutoff distance [22], the other two quantities are less commonly used and hence described below.

To calculate the radial densities, we used the Voro++ [23] library to first calculate the atomic volumes of each atom. We then used hollow cylinders centered at the ion track and of equal volumes to calculate the average volume for each atom type, i.e.  $\langle V_O \rangle$  and  $\langle V_{Si} \rangle$ . The final density was obtained from  $\rho_{SiO_2} = (m_{Si} + 2m_O) / (\langle V_{Si} \rangle + 2\langle V_O \rangle)$ . This procedure cancels out the numerical noise considerably when compared to the method

of simply dividing the mass within each cylinder by its volume. The procedure is especially useful in crystalline samples, since the cylinder will intersect the unit cells at the borders only partially, leaving an excess of silicon or oxygen in. The Voronoi method ensures that the atoms near the borders are given the correct weight, enabling the fine structure in the density profile to be seen without using an averaging stencil.

We have also identified broken bonds within the track. The criterion for a broken bond was taken as an increase of the separation of a two initially bonded atoms to more than the second nearest neighbour distance ( $\sim 0.3$  nm). We emphasise here that the broken bonds are not dangling bonds, i.e. an atom can have the correct coordination number although several of its original bonds are broken.

## 3. Results and discussion

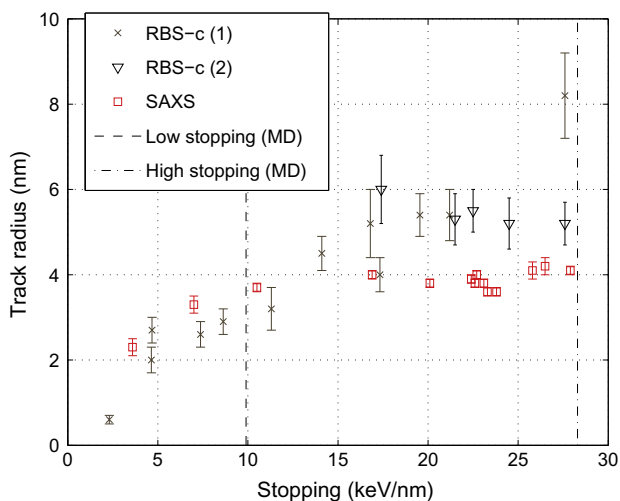
The behaviour of the experimental RBS-c and SAXS data can be seen in Fig. 1. The track radii from SAXS measurements saturate beyond the stopping power of around 10 keV/nm, whereas the first RBS-c radii (dataset (1) in Fig. 1) increase monotonically. A clear distinction between the SAXS and RBS-c data can only be seen after about 17 keV/nm. The second RBS-c data also shows saturation, however, in both cases the track radii are systematically larger than the ones measured with SAXS.

To investigate the qualitative differences in the tracks structure in both low and high stopping power regimes, we consider two limiting cases, i.e. a low energy impact (89 MeV Au ion), and a high energy impact (2.2 GeV Au). These projectile energies translate to the peak stopping powers of 9.9 keV/nm, and 28.3 keV/nm in  $\alpha$ -quartz, respectively [3], and are shown as the vertical lines in Fig. 1.

For a comparison with the SAXS track radii, radial density distributions of the tracks from the MD simulations must be examined, which are shown in the right panel of Fig. 2. Both cases show an underdense core and an overdense shell structure (a “corona”) in the density profile, similar to that seen in amorphous silica [24]. However, the relative changes in the densities are much smaller in quartz. For instance, the overdense shell consists of only 0.5% (89 MeV) and 0.7% (2.2 GeV) relative change in the density. This region is not directly observed in the SAXS measurements [17]. Therefore, the question of how the track radii should be compared remains open. The radii extracted from the underdense area are  $(1.9 \pm 0.1)$  nm and  $(4.3 \pm 0.1)$  nm for the 89 MeV and 2.2 GeV ions, respectively. The corresponding radii from the end of the overdense region are  $(3.0 \pm 0.1)$  nm and  $(6.0 \pm 0.1)$  nm. Both ways to determine the track radius from the densities, however, show a significant increase of the track radius with the stopping power. This is in contrast to the SAXS experiments, which indicate “a saturation” with only minor differences in the track radii.

To compare the simulation results with the RBS-c track radii, defect distributions must be considered. We first attempted to detect the defects by observing the occupation number of Voronoi cells as calculated from the initial MD configuration [25]. However, a small radial strain was observed which shifted the Voronoi cells, which, in turn, led to artifacts in the defect distribution. This strain is due to the ion hammering effect [26], but it does not affect the detection of coordination defects whose distributions are plotted in Fig. 2. We observe in both cases that 10% of the atoms in the amorphous and underdense track core do not have the coordination of quartz and that the concentration of defective material drops down close to zero at the overdense shell. The furthest individual coordination defects are produced relatively far away from the region of density fluctuations (at a distance of 1–2 nm).

However, the group of atoms with coordination defects does not include all atoms that are displaced to the channels, i.e. the



**Fig. 1.** Experimentally measured latent track radii as a function of stopping power. Dataset (1) is reproduced from [16,27], (2) from [28] and (3) from [17]. The stopping powers on the sample surface for the data from Refs. [16,28,27] have been recalculated using a more recent version of SRIM (2008) for consistency. The vertical lines indicate the stopping powers used in the current MD simulations.

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