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Synergy of inelastic and elastic energy loss: Temperature effects and electronic stopping power dependence ☆

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ABSTRACT

A combination of an inelastic thermal spike model suitable for insulators and molecular dynamics simulations is used to study the effects of temperature and electronic energy loss on ion track formation, size and morphology in $SrTiO_3$ systems with pre-existing disorder. We find temperature dependence of the ion track size. We also find a threshold in the electronic energy loss for a given pre-existing defect concentration, which indicates a threshold in the synergy between the inelastic and elastic energy loss.

Electronic effects are of significant importance in a wide variety of fields where high energy irradiation processes take place, including nuclear applications, the semiconductor industry, material synthesis, modification and characterization [1,2]. The importance of the coupling of electronic and atomic processes in ionic and covalent materials has been emphasized in recent studies [3–15,1], where it has been shown that these effects can have linearly additive [3–8] or competing [9–11] impacts on the defect production. A more recent study by Weber et al. [16] reveals a remarkable synergy between the inelastic energy loss and pre-existing damage, showing that the presence of pre-existing disorder in the system enhances the sensitivity of the material to the electronic energy loss effects. Furthermore, we previously showed [17] that the size of nanoscale ion tracks can be controlled by the concentration of pre-existing disorder in pre-damaged $SrTiO_3$ systems. These results emphasize the importance of the pre-existing damage on the energy dissipation in the system and highlights the need to investigate further the role of the defects and defect excitation in microstructure alterations.

In the present paper, we study the effects of two factors on the synergy between electronic and atomic processes, and consequently on ion track formation in pre-damaged $SrTiO_3$, namely the effects of temperature and the effects of varying electronic energy loss.

We use a combination of an inelastic thermal spike (iTS) model suitable for insulators [18] and molecular dynamics (MD) simulations to model the energy dissipation due to irradiation into the system. The iTS model describes the energy exchange between the electronic and the atomic systems in terms of a set of two coupled heat diffusion equations, one for the electronic (1) and one for the atomic (2) system. The energy transfer from the electronic to the atomic lattice occurs via the electron–phonon interactions.

$$C_e\frac{\partial T_e}{\partial t}=\frac{1}{r}\frac{\partial}{\partial r}\left[rK_e\frac{\partial T_e}{\partial r}\right]-g(T_e-T_\alpha)+A(r,t) \quad (1)$$

$$C_a\frac{\partial T_a}{\partial t}=\frac{1}{r}\frac{\partial}{\partial r}\left[rK_a\frac{\partial T_a}{\partial r}\right]+g(T_e-T_a) \quad (2)$$

C_e and C_a are the specific heat coefficients of the electronic and atomic systems respectively, whereas K_e and K_a are the thermal conductivities of the electronic and the atomic system. The term g is the electron–phonon coupling parameter, and $A(r,t)$ describes the energy deposition from the incident ion to the electrons [19]. The second term on the right side of the Eqs. (1) and (2) represents

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the energy exchange between the electronic and atomic subsystems due to the difference between the electronic temperature T_e and the atomic temperature T_a.

The specific heat coefficients used for the electronic and atomic systems are $C_e = 1\ \mathrm{J\,cm^{-3}\,K^{-1}}$ [20,18,21] and $C_a = 0.544\ \mathrm{J\,g^{-1}\,K^{-1}}$ [22], respectively. To account for the decrease in the thermal conductivity due to the presence of the defects [23,24], we used reduced values of K_e and K_a by an order of magnitude in comparison to the values for the perfect crystalline system. Such a decrease in the thermal conductivity is reasonably consistent with the large changes in thermal conductivity of $SrTiO_3$ due to processing defects [25] or cation non-stoichiometry (a few percent) [26]. For the crystalline system K_e is equal to $K_e = C_eD_e$ as previously suggested [20,18,21], where D_e is the thermal diffusivity, and K_a is equal to $11.2\ \mathrm{W\,m^{-1}\,K^{-1}}$ at 300 K. Here we note that we do not take into account the difference in the thermal conductivity for the calculations at 500 K and 800 K, as the difference is small and the effect in the energy deposition is negligible. The value of g for a crystalline system is, as described elsewhere [18], $4.3 \times 10^{18}\ \mathrm{W\,m^{-3}\,K^{-1}}$. We used a 35% larger value for the pre-damaged system to account for the decrease in the electron–phonon mean free path due to the defects.

The electronic energy loss due to 21 MeV Ni, 20 MeV Ti, 12 MeV and 21 MeV Si ions irradiation are calculated using Stopping and Range of Ions in Matter (SRIM) code [27], and is determined as 9.74 keV/nm, 8.5 keV/nm, 6.9 keV/nm, and 6.18 keV/nm respectively at about 120 nm irradiation depth. Fig. 1 shows the energy deposition profile as a function of the distance from the ion path for these ions in pre-damaged $SrTiO_3$ systems.

The energy profile calculated with the iTS model is used as input to the MD simulations, for which we use the DL_POLY [28] code. The pre-damaged systems were created by introducing Frenkel pairs (FP) randomly in the systems, which were consequently relaxed under constant pressure and temperature. The total number of atoms in each system is about 7 million. The irradiation of the systems is along the z direction under the constant-energy, constant-volume (NVE) ensemble at the target temperature, with a variable timestep, in a depth of about 26 nm. The atoms contained in the x and y boundary of the MD box, in a layer of about 1 nm thickness, are connected to a Langevin thermostat at 300 K to emulate the effect of energy dissipation into the sample. The defects are identified using the sphere criterion [29], with a cut-off radius of 0.75 Å. We use empirical potentials by McCoy et al. [30] joined to the ZBL [31] repulsive potentials for short distances. The ZBL potential was used for all pair interactions.

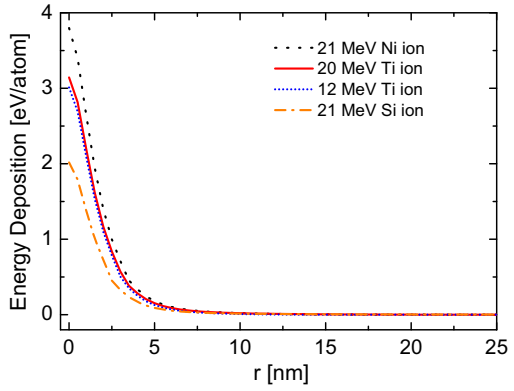

Fig. 1. Energy deposition profiles for 21 MeV Ni, 20 MeV Ti, 12 MeV Ti, and 21 MeV Si ions in pre-damaged $SrTiO_3$.

To study the effects of temperature on the synergy of the elastic and inelastic energy loss, we used two systems containing about 0.5% FP and 1% FP, where we applied the energy deposition that corresponds to 21 MeV Ni ions at 300 K, 500 K and 800 K. To study the effects of the coupling of different electronic energy loss and the pre-existing disorder, we used two systems containing about 2% and 12% FP, where we applied the energy profiles shown in Fig. 1 at 300 K simulation temperature.

Fig. 2(a) shows the cross sections of systems containing 0.5% and 1% FP at the end of the simulation time at different temperatures. We see that the ion track diameter increases for higher temperature. Fig. 2(b) illustrates perpendicular slices (parallel to z axis) of the same systems, where we see that for a given concentration of pre-existing defects the resulting ion tracks become more continuous.

Higher temperature means that the energy needed to cause melting of the material is less, therefore the damage efficiency is higher as the temperature increases, and the resulting ion tracks are larger for higher temperatures. Such behavior has been observed in experiments in pyrochlore for 40 keV/nm energy loss at 8 K and 300 K [32] and in bismuth for energy loss about 30–38 keV/nm for a range of temperatures from 20 K to 300 K [33]. The non-continuous morphology of the ion tracks for low disorder level systems, as well as differences in the track diameter size, in different depth for larger disorder level systems, as discussed elsewhere [17], can be attributed to the inhomogeneity of the defect distribution along the ion path.

Irradiation of systems with 2% pre-existing FP with 9.74 keV/nm, 8.5 keV/nm and 6.9 keV/nm electronic stopping power results in the formation of an ion track only for 9.74 keV/nm electronic energy loss. For the lower values of stopping power, the damage created during the irradiation has recovered, with only a few point defects and point defect clusters surviving after the system relaxation. However, irradiation of systems that contains 12% FP results in formation of ion tracks in all three cases of electronic stopping power. For higher electronic energy loss, the ion track is larger, more homogeneous and more continuous. Fig. 3 shows the cross sections of the higher defect density (12% FP) systems, where the ion track diameter ranges from about 1.8 ± 0.4 nm to 2.6 ± 0.5 nm for energy loss from 6.9 to 9.74 keV/nm respectively. The boundaries of these ion tracks are difficult to distinguish due to the higher level of disorder in the surrounding material. The ion track diameter through the samples that contain 12% FP initial damage as a function of the electronic energy loss is shown in Fig. 4, along with experimental data for samples with 11% initial disorder. The similar trends seen here suggest a good agreement between the computational and experimental results, and both indicate a linear relationship between average track diameter and the electronic stopping power. Irradiation with 21 MeV Si ions (6.18 keV/nm electronic energy loss) did not produce ion tracks in either system.

The presence of the pre-existing damage means that less energy is needed in order for the defects to start percolating. With increasing electronic energy loss, more energy is deposited to the lattice, including the pre-existing defects, and the ion tracks become more continuous and larger in size. The fact that no ion track is produced for 6.18 keV/nm energy loss indicates that despite the presence of the defects, the energy deposited is not sufficient to enhance the disorder in the vicinity of the ion path and create a track. Therefore these results suggest that there is a threshold for the synergy of the inelastic energy loss and the atomic defects, which depends on the combination of the pre-existing disorder and the stopping power.

In summary, our findings show that there is a temperature dependence of the ion track size and morphology in pre-damaged systems of $SrTiO_3$; higher irradiation temperature