



Full length article

Amorphization due to electronic energy deposition in defective strontium titanate



Haizhou Xue^a, Eva Zarkadoula^b, Peng Liu^{a, c}, Ke Jin^b, Yanwen Zhang^b, William J. Weber^{a, b, *}

^a Department of Materials Science & Engineering, University of Tennessee, Knoxville, TN, 37996, USA

^b Materials Science & Technology Division, Oak Ridge National Laboratory, Oak Ridge, TN, 37831, USA

^c School of Physics, State Key Laboratory of Crystal Materials and Key Laboratory of Particle, Physics and Particle Irradiation (MOE), Shandong University, Jinan, 250100, China

ARTICLE INFO

Article history:

Received 19 September 2016

Received in revised form

19 January 2017

Accepted 24 January 2017

Available online 27 January 2017

Keywords:

SrTiO₃

Energy dissipation

Stopping power

Ion track

Amorphization

ABSTRACT

The synergistic interaction of electronic energy loss by ions with ion-induced defects created by elastic nuclear scattering processes has been investigated for single crystal SrTiO₃. An initial pre-damaged defect state corresponding to a relative disorder level of 0.10–0.15 sensitizes the SrTiO₃ to amorphous track formation along the ion path of 12 and 20 MeV Ti, 21 MeV Cl and 21 MeV Ni ions, where Ti, Cl and Ni ions otherwise do not produce amorphous or damage tracks in pristine SrTiO₃. The electronic stopping power threshold for amorphous ion track formation is found to be 6.7 keV/nm for the pre-damaged defect state studied in this work. These results suggest the possibility of selectively producing nanometer scale, amorphous ion tracks in thin films of epitaxial SrTiO₃.

© 2017 Acta Materialia Inc. Published by Elsevier Ltd. All rights reserved.

1. Introduction

Ion irradiation is an important and routinely utilized tool to modify the band structure and charge density of semiconductors. On the other hand, it is also well known that ion irradiation has been proven to be a highly efficient, low cost method to simulate defect production, volume swelling and phase transformations in nuclear materials due to radioactive decay, nuclear fission, and neutron irradiation. With regard to these applications, it is important to understand and predictively model the response of electronic and nuclear materials to ion irradiation in order to develop the next generation of electro-optic-magnetic devices and more radiation tolerant nuclear materials. Therefore, a comprehensive understanding of ion-solid interactions at the level of atoms and electrons is urgently needed. Because of the complexity of ion-solid interactions, a well-accepted approach is to separate the energy deposition from ions into (a) nuclear stopping power (S_n), which is the elastic energy loss to atomic nuclei; and (b) electronic stopping

power (S_e), which is the inelastic energy loss to electrons. The understanding of S_n is relatively well established by an elastic scattering or collision model. It is known that S_n transfers energy to target nuclei that directly results in the displacement of target atoms, which leads to the production of point defects and defect clusters. The energy loss to the electrons, its dissipation and the effects on atomic processes are, however, relatively much less understood, especially, at intermediate ion energies ranging from several hundreds of keV to tens of MeV. Recent studies have demonstrated that S_e can couple complexly with the defects induced by S_n , which can lead to modification of the target materials in different ways [1]. For materials such as CeO₂ and ZrO₂ [2], ZrSiO₄ [3] and probably α -SiO₂ [4], the electronic energy loss leads to damage production that is linearly additive to the damage produced by elastic nuclear collisions. On the other hand, electronic energy loss can induce a localized thermal spike via the diffusion of hot electrons and electron-phonon coupling that can cause damage recovery, such as that reported for SiC [5–7], Gd₂TiZrO₇ [1], and (Ca,Sr)₂(La,Nd)₈(SiO₄)₆O₂ [8]. Recently, a substantial synergistic effect has been reported for single crystal SrTiO₃ [5,9–11]. Both experimental and computer simulation results confirm that the presence of a small level of pre-existing damage in single crystal

* Corresponding author. Department of Materials Science & Engineering, University of Tennessee, Knoxville, TN, 37996, USA.

E-mail address: wjweber@utk.edu (W.J. Weber).

SrTiO₃, created by nuclear collision processes, leads to the formation of amorphous ion tracks by 21 MeV Ni ions; whereas no tracks form in the pristine material. The value of S_e (9.9 keV/nm) for the 21 MeV Ni ions is below the calculated threshold of ~12 keV/nm for ion track formation in pristine SrTiO₃ due to track overlap [12].

In the present study, this synergistic effect in SrTiO₃ is further investigated in order to determine the electronic energy loss threshold for this phenomenon and the dependence on initial level of pre-damage. The motivation, on one hand, is to improve the understanding of the coupled effects of electronic and nuclear stopping powers. A practical concern is that SrTiO₃ and similar materials are considered for the immobilization of nuclear wastes [13,14], and a synergistic effect may dramatically accelerate damage accumulation from radioactive decay. On the other hand, there may be beneficial applications for this synergistic effect in ion beam modification and processing, such as the selective production of nanoscale amorphous tracks (i.e., cylindrical columns) in thin film devices, with the cross-section of the amorphous track controlled by the electronic energy loss and initial defect concentration. In addition, SrTiO₃ is a critical foundational material in micro-electronics [15–17], and the interface it forms with other dielectric oxides and selenides, such as LaAlO₃ [18–20] and FeSe [21], or even with its amorphous phase [22], results in unique electronic and ferromagnetic properties of interest. By taking advantage of the synergistic effect, ion beam modification is a potential tool for creating concentric nanometer diameter interfaces with unique functionalities for device applications.

2. Experimental

Single crystal SrTiO₃ samples used in this work were (100) oriented wafers obtained from the MTI Corp. The ion irradiations and Rutherford Backscattering Spectrometry in channeling geometry (RBS/C) measurements were performed using a 3 MV tandem accelerator and facilities in the Ion Beam Materials Laboratory (IBML) located at the University of Tennessee [23]. In order to produce initial pre-damaged regions with different levels of disorder in the pristine SrTiO₃, low energy irradiations using either 600 keV O ions at an incident angle of 60° or 900 keV Au ions at near normal incidence were performed. The RBS/C measurements revealed relatively low initial damage profiles in the SrTiO₃, with maximum disorder fractions ranging from 0.10 to 0.15 for 600 keV O ion irradiation to a fluence of $6.8 \times 10^{14} \text{ cm}^{-2}$ and 900 keV Au ion irradiation to a fluence of $3.9 \times 10^{13} \text{ cm}^{-2}$. In addition, a relatively higher initial peak disorder fraction of 0.35 was produced by Au ion irradiation to a fluence of $5.4 \times 10^{13} \text{ cm}^{-2}$. Both pristine samples and these pre-damaged samples were subsequently irradiated with 21 MeV Ni, 12 and 20 MeV Ti, 21 MeV Cl and 18 MeV Si ions, which have different ratios of electronic to nuclear energy loss, as summarized in Table 1. While the 600 keV O irradiation was performed at 60° relatively to the surface normal direction, all the other irradiations were performed with the incident ion beam only several degrees off the surface normal to avoid channeling effects. For the pre-damaged samples, multiple spots were irradiated with high-energy ions to fluences ranging from 10^{11} to 10^{14} cm^{-2} in order to obtain both isolated and overlapping ion paths. Before and after each ion irradiation, RBS/C measurements using 3.5 MeV He ions were performed in situ to determine the change in disorder profile of each irradiated spot with the sample remaining in place and retaining its orientation. All the ion irradiations and RBS/C measurements were carried out at room temperature in a high vacuum.

3. Thermal spike model

An inelastic thermal spike model suitable for insulators [24,25]

describes the energy exchange between the electronic and atomic subsystems due to energy deposition from a high-energy single ion. The interactions can be described in terms of a set of two heat diffusion equations, one describing the evolution of the electronic temperature T_e (Eq. (1)), and the other one describing the evolution of the atomic temperature T_a (Eq. (2)).

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

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

Here, C_e and C_a are the heat capacities of the electronic and the atomic systems, respectively; K_e is the electronic thermal conductivity; and K_a is the thermal conductivity of the atomic system. The energy exchange between the electronic and the atomic systems depends on the temperature difference $T_e - T_a$ between them, and g , which is the electron-phonon coupling parameter, determines the strength of the interaction. The term $A(r, t)$ describes the energy deposition from the incident ion to the electrons [26].

For the specific heat, we used $C_e = 1.0 \text{ J cm}^{-3} \text{ K}^{-1}$ [24,27] and $C_a = 0.544 \text{ J cm}^{-3} \text{ K}^{-1}$ [28]. For the crystalline pristine sample, $K_e = C_e D_e$ [24,27], where D_e is the thermal diffusivity and has a value of $1.0 \text{ cm}^2 \text{ s}^{-1}$ [24,27], K_a is $11.2 \text{ W m}^{-1} \text{ K}^{-1}$ at 300 K, and the electron-phonon coupling parameter g is $4.3 \times 10^{18} \text{ W m}^{-3} \text{ K}^{-1}$ [24]. To account for the decreased electron mean free path due to the presence of the irradiation defects in the pre-damaged systems [29], we assumed the values of K_e and K_a are reduced by an order of magnitude relative to the values for the perfect crystalline system, as in our previous studies [9–11]; likewise, we assumed that the value of g is 35% larger than the one estimated for the pristine crystalline SrTiO₃ [9,11]. The electronic energy loss for each ion was calculated using the SRIM code [30] at an irradiation depth of 100 nm (180 nm for the case of 21 MeV Ni ion irradiation of the pre-damaged state from 600 keV O ions).

4. Results and discussion

For the low-energy 600 keV O ions and 900 keV Au ions, their S_e values are relatively low (<1.3 keV/nm) and far below the estimated threshold value (~12 keV/nm) for amorphous track formation in pristine SrTiO₃ [12]. Under these irradiation conditions, the damage production is mainly attributed to atomic displacements induced by S_n , and the kinetics are described by a direct-impact/defect-stimulated model [31]. The high energy ions, however, were chosen with S_e values that range from 6.2 to 9.9 keV/nm, as predicted by the SRIM2008 code [30] at the peak of pre-damage states. Under these high-energy irradiation conditions, the dominance of S_e over S_n produces intensive ionization that results in a highly local, radially dependent thermal spike along the ion path due to electron-phonon coupling. As shown in Table 1, the values of S_n at these high energies are negligible, and the ratios of S_e/S_n are large. Consequently, few defects are expected to be induced along the first 500 nm of ion path for the ion fluences utilized in this work. Therefore, any significant damage production must be due to the electronic energy loss. As shown in the inset of Fig. 1a, 12 and 20 MeV Ti ions do not produce significant damage in the pristine single crystal SrTiO₃, similar to what has been previously reported for 21 MeV Ni ions [10]. On the other hand, the RBS/C spectra in Fig. 1a clearly show damage accumulation under 20 MeV Ti irradiation when a pre-existing damage state (relative disorder peak of 0.14) is present. The backscattering yield increases rapidly and a fully amorphous

Download English Version:

<https://daneshyari.com/en/article/5436343>

Download Persian Version:

<https://daneshyari.com/article/5436343>

[Daneshyari.com](https://daneshyari.com)