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Research Article

Effects of Ion Beam Irradiation on Nanoscale InO_x Cooper-Pair Insulators

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This paper examines the effects of irradiating indium oxide films of nanoscale thickness by ion beams, when these films are in the Cooper-pair insulator state. Radiation effects are predicted on the basis of Monte Carlo simulations of ion transport. Results of numerical experiments are interpreted within the theoretical model of a Cooper-pair insulator. The study suggests that radiation-induced changes in InO_x films exposed to ion beams could significantly alter their current-voltage characteristics and that a transition to a metallic state is possible, due to radiation-induced perturbation of the fine-tuned granular structure. Furthermore, incident and displaced ions can break up enough Cooper pairs in InO_x films to cause dissolution of this specific insulating state.

1. Introduction

Cooper-pair insulators (CPIs) are a newly discovered class of materials, with distinct structural properties and a unique physical mechanism of electrical current conduction. The Cooper-pair insulating phase appears only at very low temperatures, in materials that are otherwise superconducting. The superconductor-insulator transition (SIT) has been studied extensively in the recent years, and a fairly accurate understanding of this process has been acquired [1–6].

During the past decade, a lot of efforts have also been invested in designing resistive random-access memories (RRAMs). The cells of these new memories are expected to have a high on-to-off current ratio, dependent on a large change of ohmic resistance, such as the one that occurs during an SIT. CPIs are, therefore, one of the promising materials for future resistive memories.

Like many other memories, CPI-based RRAMs would also be used in various radiation environments, for example, in radiology and nuclear medicine departments, at nuclear facilities, or aboard space satellites [7, 8]. Consequently, the issue of radiation effects in materials which behave as Cooper-pair insulators is of considerable relevance.

This paper investigates the effects of ion beam irradiation on the properties of nanoscale indium oxide films in the Cooper-pair insulator state, by using numerical simulation of ion transport.

2. Theoretical Foundations

The Cooper-pair insulators are materials that exhibit superconducting behavior, but under specific conditions (regarding film thickness, bias voltage, applied magnetic field, and presence of magnetic impurities) they become insulators with thermally activated Cooper pairs as charge carriers [1–4]. Such behavior has been observed in films of indium oxide with nanoscale thickness [5, 6, 9, 10]. The insulating phase has a granular structure of superconducting islands (regions of localized Cooper pairs) distributed throughout a matrix of normal nonsuperconducting material [11–13]. When the conditions are right, the grouping of Cooper pairs into superconducting islands occurs spontaneously in the material, due to increased disorder. This process is akin to the Anderson localization, which is an absence of diffusion of waves (including quantum waves assigned to particles) in a disordered medium, provided that the degree of randomness of impurities or defects is sufficiently large. One such effect of disorder on electronic systems is the localization of electrons, which transforms an otherwise metallic system into an insulator [1]. The phenomenon of superconducting materials turning into insulators is therefore often regarded as a disorder-driven superconductor-insulator transition [13–17].

A two-dimensional Josephson junction array (2D JJA) is used to model the granular structure of a CPI [2, 18]. A 2D JJA consists of small superconducting islands, each coupled to the nearest neighboring islands by the Josephson weak links. A weak link that forms over the section of non-superconducting material between superconducting islands constitutes a Josephson junction. Each junction is characterized by the following Josephson coupling energy:

$$E_J = \frac{\hbar I_c}{2e}, \tag{1}$$

where I_c is the critical current of the junction, e is the elementary charge, and $\hbar = h/2\pi$ is the reduced Planck constant. Two more energies that characterize each junction are the charging energies E_c and E_{c0}, related to inter-island capacitance and capacitance to the ground (substrate), respectively. The inter-island charging energy E_c is the energy needed for a Cooper pair to be transferred between neighboring islands.

Investigations of the conditions under which indium oxide acts as either a superconductor or an insulator at temperatures below 1 K have shown that, in terms of the JJA model, the Cooper-pair insulating phase emerges only when the degree of disorder makes the charging energies larger than the coupling energy ($E_c, E_{c0} > E_J$), while the superconducting gap still exceeds the interisland charging energy ($\Delta > E_c$) [6, 9].

Cooper pairs spatially confined to a single superconducting island in a JJA exhibit *local* coherence of the wave function phase. When an external voltage is applied to the ends of a JJA, wave function phase synchronization of all Cooper pairs in the JJA occurs. This *global* phase coherence gives rise to a collective current state. The DC Josephson current that runs through the JJA couples the phases of adjacent junctions, so as to provide minimal power dissipation in the array. This establishes a global phase-synchronized state, and the transport of current occurs through simultaneous thermal activation of Cooper pairs throughout the whole array [2, 3]. Due to this activation conduction, resistance of a Cooper-pair insulator follows an Arrhenius-like temperature dependence. This also means that the low-bias ($V \ll \Delta_c/e$) current-voltage dependence in the temperature interval $E_c < k_B T < \Delta_c$ is exponential:

$$I = I_c \exp\left(-\frac{(\Delta_c - eV)^2}{2\Delta_c k_B T}\right), \tag{2}$$

where I_c is the critical current of a Josephson junction in the 2D array, k_B is the Boltzmann constant, T is the absolute temperature, and Δ_c is the collective Coulomb barrier of the array. For the 2D JJA, this barrier is given as follows:

$$\Delta_c = E_c \ln \frac{L}{d}, \tag{3}$$

where L is the characteristic linear size of the array (e.g., film thickness) and d is the size of an elemental cell in it [2].

3. Monte Carlo Simulations of Ion Beam Transport

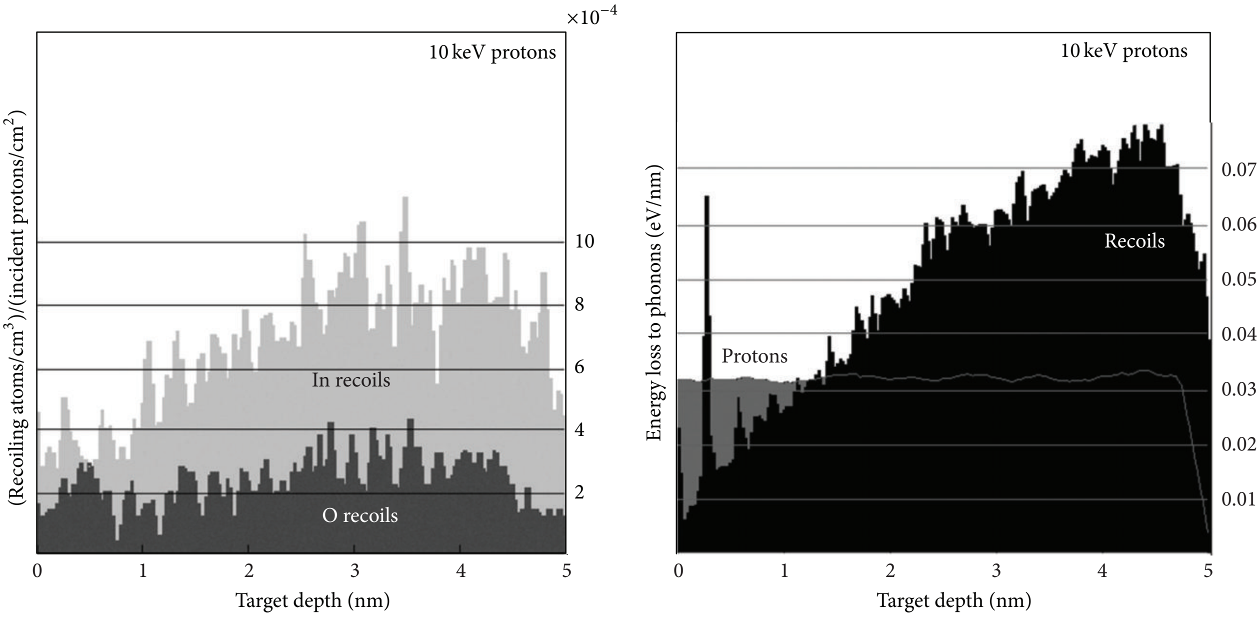
Monte Carlo simulations of ion transport through InO_x films of varying thickness (5 to 15 nm) were performed in the TRIM module of the SRIM software package [19]. Oxygen content was varied from $x = 0.95$ to $x = 1.05$ to reflect the amorphous structure of indium oxide films used in previous experimental studies [5, 6, 9, 10].

Simulations were conducted with ions chosen to represent certain well-known radiation fields, such as those encountered in the space environment (hydrogen, helium, and lead) [20, 21] or beams commonly used in ion implantation processes (phosphorus, boron, and arsenic). Simulations were restricted to monoenergetic unidirectional beams, incident perpendicularly on the film's surface. Beam energy was varied across typical energy ranges of the considered ion species.

SRIM has been proved to provide results in significant agreement with experiments [19]. Its continuous upgrades during the past decade have made it a foremost tool for numerical analysis of interaction of ions with matter and validation of theoretical predictions [22–24]. Transport of ions in matter (TRIM) not only calculates the range of ions in matter, but it also details many other aspects of the damage done to the target during the ion beam slowing-down process, such as cascades of displaced target atoms. It makes calculations for one ion at a time, in order to make precise evaluations of the physics of each encounter between the ion and a target atom. The accuracy of a simulation run is determined by the number of ions, that is, histories followed. A calculation for 1000 incident ions will give better than 10% accuracy [19].

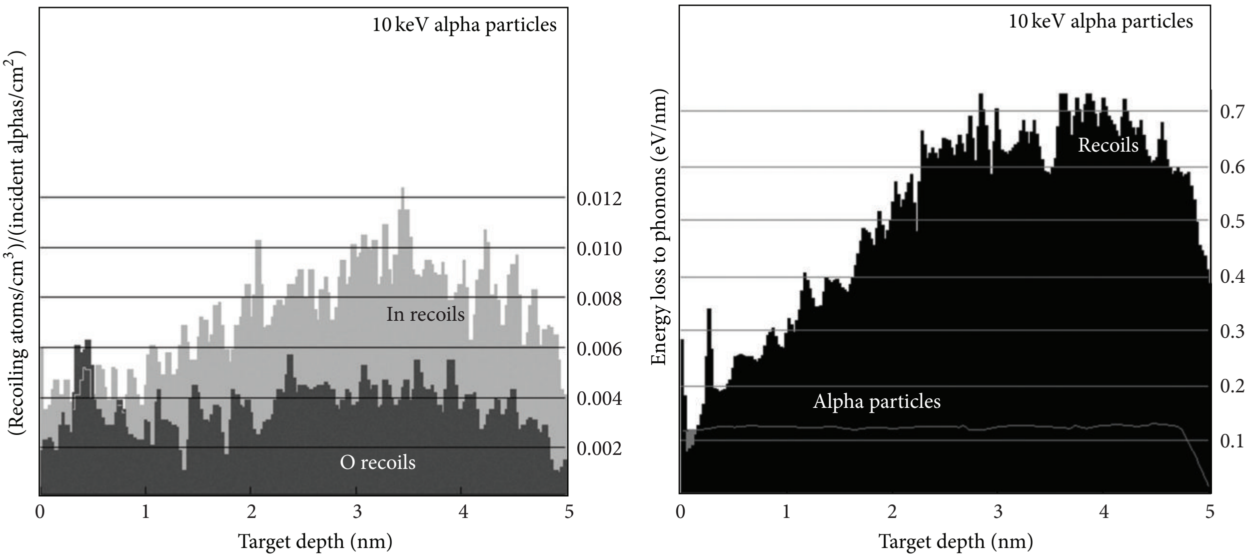
The fact that TRIM calculations assume target temperature to be absolute zero makes it an appropriate tool for modeling radiation effects in the Cooper-pair insulating state, which exists only at very low temperatures anyway, so that thermal diffusion and annealing of displaced atoms in the film can be neglected. Another approximation inherent in TRIM is that there is no buildup of radiation damage in the target; that is, for each new incident ion, the film is assumed to contain no damage produced by the previous ions. Any assessment of the degree of radiation damage in the film is therefore an underestimation of the damage that a real ion beam would have caused.

Ionization energy losses calculated by TRIM were taken as an estimate of the extent of the Cooper-pair dissociation in superconducting islands within the granular structure of InO_x films. Assessment of pair dissociation can be performed by comparing ionization potentials of indium and oxygen



(a) (b)

Figure 1: Results of irradiating a 5 nm thick InO_x film by a beam of 10^5 protons with 10 keV energy. (a) Distribution of the concentration of the displaced indium and oxygen atoms (recoils) per unit proton fluence. (b) Energy loss to phononic excitations per unit depth by protons and recoils.



(a) (b)

Figure 2: Results of irradiating a 5 nm thick InO_x film by a beam of 10^4 alpha particles with 10 keV energy. (a) Distribution of the concentration of the displaced indium and oxygen atoms (recoils) per unit alpha particle fluence. (b) Energy loss to phononic excitations per unit depth by alphas and recoils.

atoms with the depairing energy (i.e., the energy for breaking up a Cooper pair) in indium oxide. First ionization energies of indium and oxygen are 5.786 eV and 13.618 eV, respectively. The energy for breaking up a Cooper pair at absolute zero can be assessed as $2\Delta \sim 3.5 k_B T_C$, which for InO, with the critical temperature of $T_C \approx 3.3$ K, yields the value of ≈0.001 eV.

4. Results and Discussion

Plots in Figures 1, 2, 3, 4, and 5 demonstrate some of the most relevant results obtained from simulations. These plots show the distributions of ionizing events (commensurate to the Cooper-pair dissociations) and nonionizing events

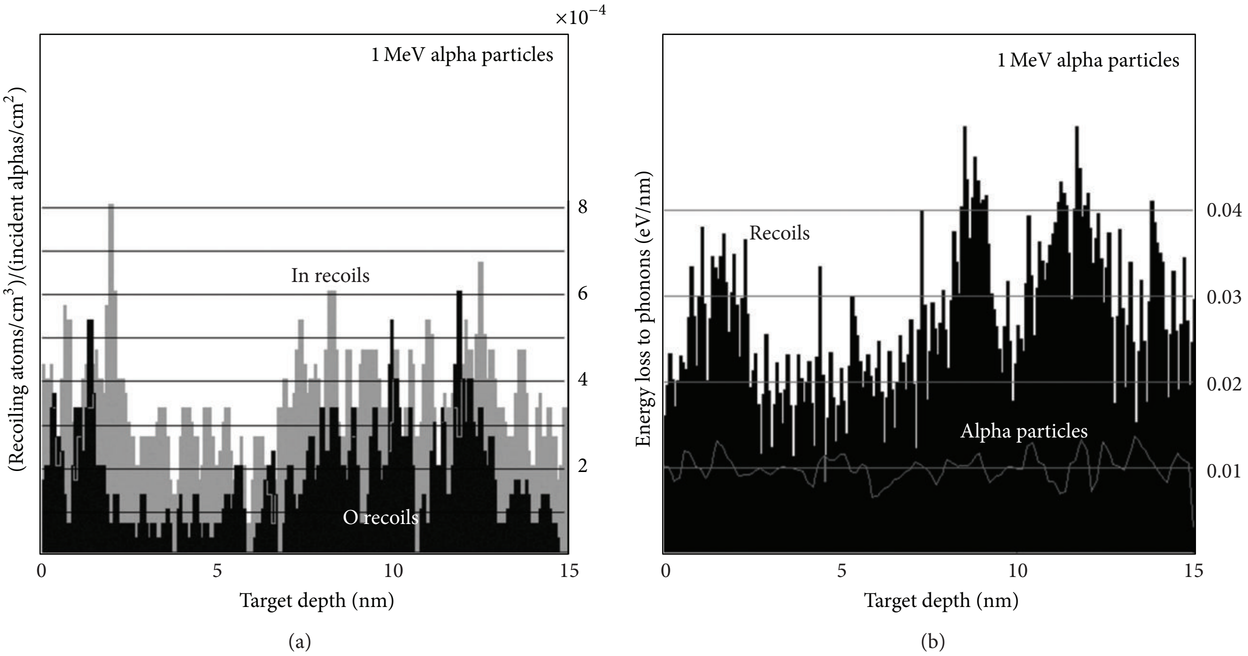


Figure 3: Results of irradiating a 15 nm thick InO_x film by a beam of 10^4 alpha particles with 1 MeV energy. (a) Distribution of the concentration of the displaced indium and oxygen atoms (recoils) per unit alpha particle fluence. (b) Energy loss to phononic excitations per unit depth by alphas and recoils.

(phononic excitations and atomic displacements) within InO_x films.

Distributions of displaced indium and oxygen atoms in a 5 nm thick InO_x film irradiated by a beam of 10^5 protons with 10 keV energy are shown in Figure 1(a). Energy losses to phononic excitations for the same irradiation conditions are shown in Figure 1(b).

For comparison with proton results, Figure 2 presents atomic displacements and energy losses to phonons in a 5 nm thick InO_x film irradiated by a beam of 10 keV alpha particles (10^4 histories). Figure 3, on the other hand, shows the results for high-energy alphas (1 MeV) in a 15 nm thick InO_x film.

Results for 0.1 MeV iron ions, one of the few heavy-ion species with considerable fluences in the primary cosmic rays that reach the Earth's atmosphere, are presented in Figure 4 for three different thicknesses of the InO_x film: 5 nm, 10 nm, and 15 nm.

Finally, plots in Figure 5 offer a comparison of the effects caused by different ion species (phosphorus, boron, and arsenic) with the same beam energy (0.1 MeV) in a 15 nm thick InO_x film.

Since the investigated InO_x films are less than 15 nm in thickness, they are immune to the passage of high-energy ions. Both ionizing and nonionizing energy losses of high-energy ions are low, and they traverse the thin InO_x films without deflection or notable ionization, producing only slight effects [25–30].

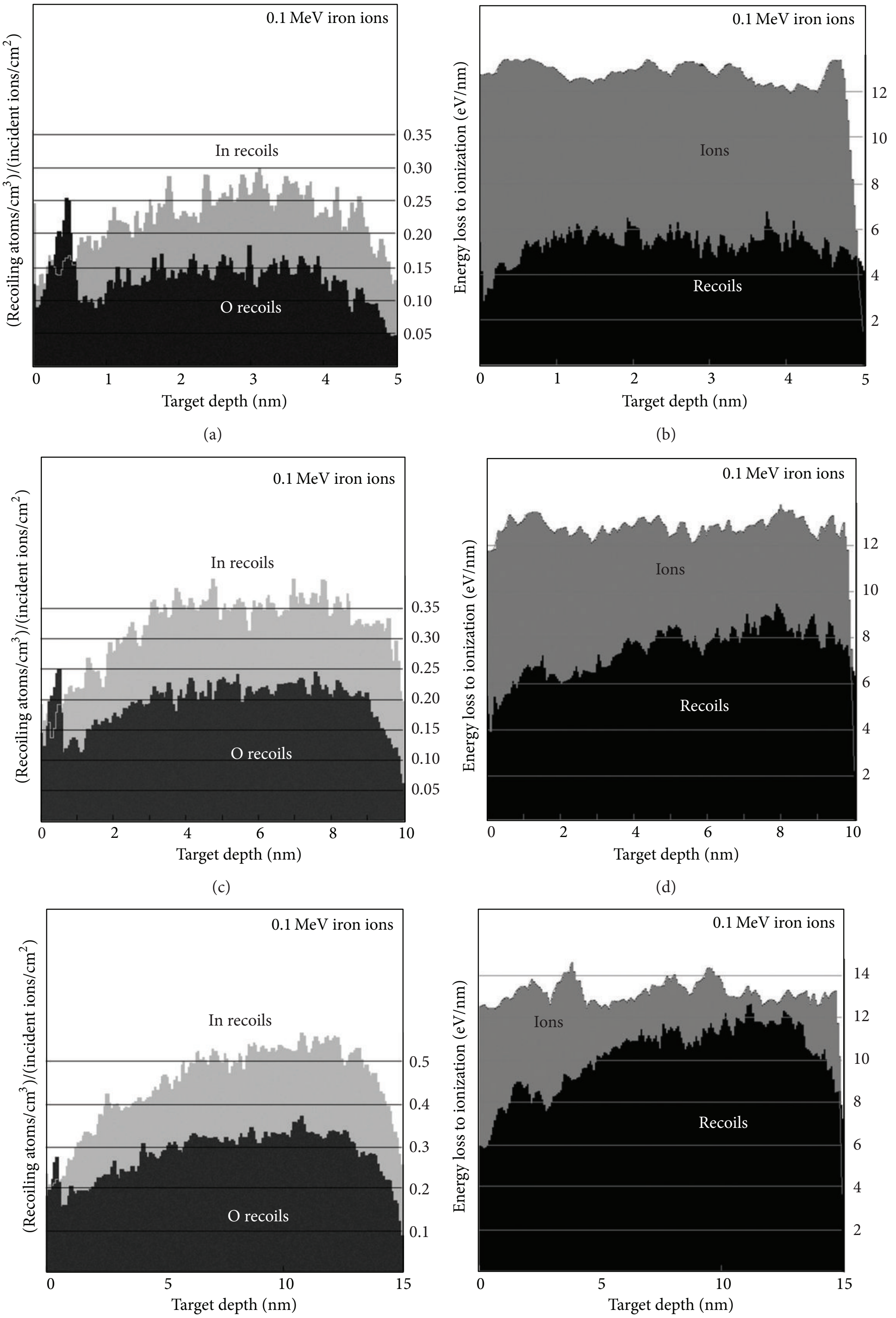
The selected results presented in Figures 1–5 suggest, however, that for certain ion species there exist energy ranges in which a great number of atom displacements, phonon excitations, and ionization events would occur in irradiated InO_x films.

The number of atomic displacements is in direct proportion to the fluence of incident radiation, that is, the number of particle histories followed in the Monte Carlo simulation. Space charge created by the displaced ions that finally take interstitial positions could affect the size of the Josephson junction-charging energy E_c, which then changes the collective Coulomb barrier Δ_c. The change in the I-V curve of an indium oxide film, resulting from the radiation-induced change of E_c, is illustrated in Figure 6. The curves in this figure were obtained from expressions (2) and (3) for three different values of E_c, with $L = 15$ nm, $d = 1$ nm, $I_c = 10\,\mu$A, and $T = 0.3$ K.

The Cooper-pair insulating state depends critically on the value of E_c, existing only when $E_c > E_J$. If the radiation damage produced by ion beams is large enough to disrupt this condition, InO thin film may revert to the ordinary metallic state.

Ionization energy losses by incident ions and recoils, observed in simulations, indicate that appreciable breaking of Cooper pairs in superconducting islands is possible. Decrease of Cooper-pair concentration could destroy the insulating state during irradiation. With other conditions unchanged, this effect is expected to be transient. Once the InO_x film is no longer exposed to ions, Cooper pairs may reform, and the insulating state could be restored.

Results of the simulations also indicate that one part of ion beam energy is converted to phononic excitations. Energy losses to phonons increase the effective temperature



(a)

(b)

(c)

(d)

(e)

(f)

FIGURE 4: Results for 10^3 iron ions with 0.1 MeV energy. Distribution of the concentration of the recoils per unit ion fluence and ionization energy losses per unit depth for various thicknesses of the InO_x film: 5 nm (a and b), 10 nm (c and d), and 15 nm (e and f).

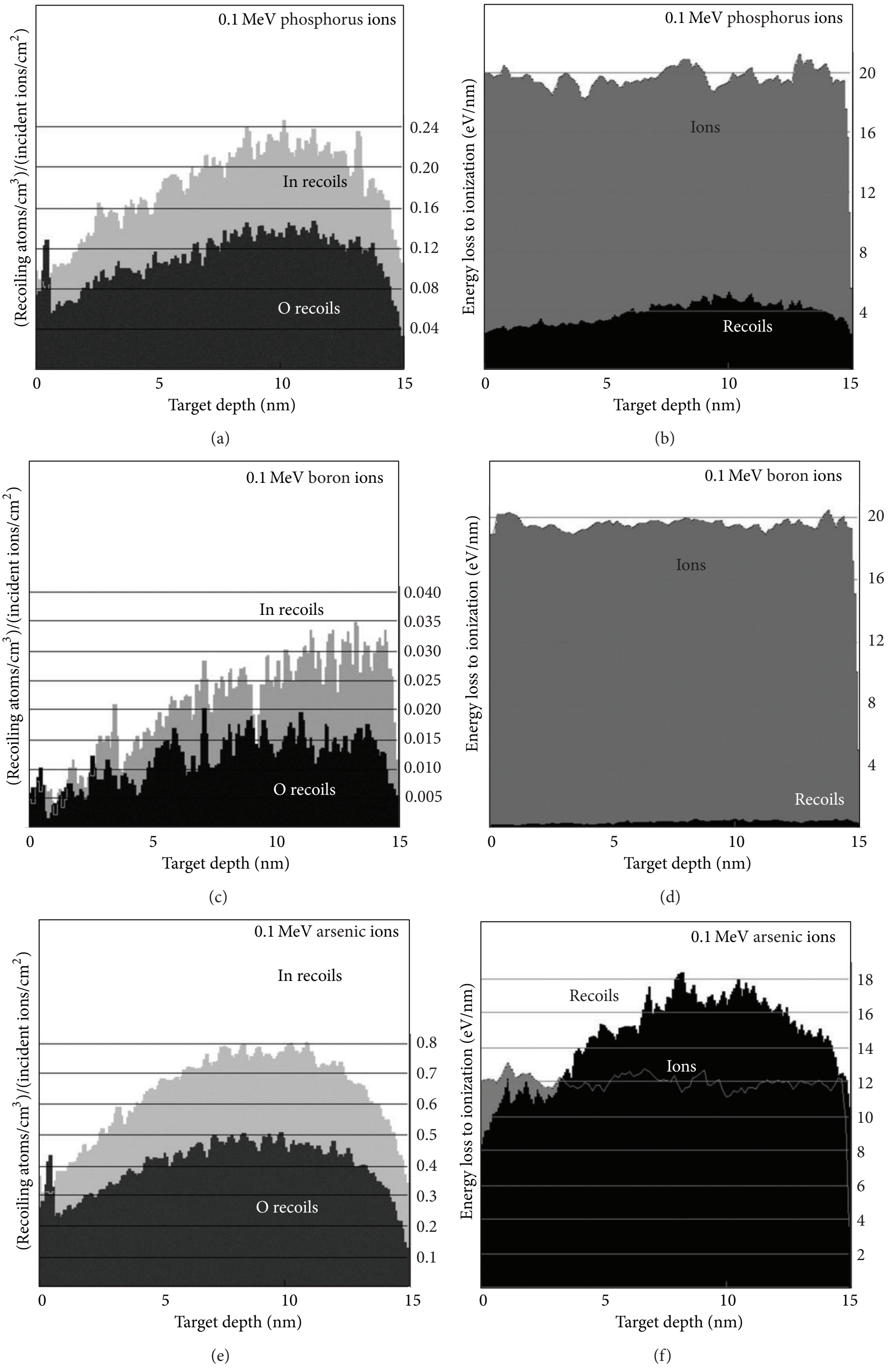


FIGURE 5: Distribution of the concentration of the recoils per unit ion fluence and ionization energy losses per unit depth in a 15 nm thick InO_x film for a 0.1 MeV incident beam of 10^3 phosphorus ions (a and b), boron ions (c and d), and arsenic ions (e and f).

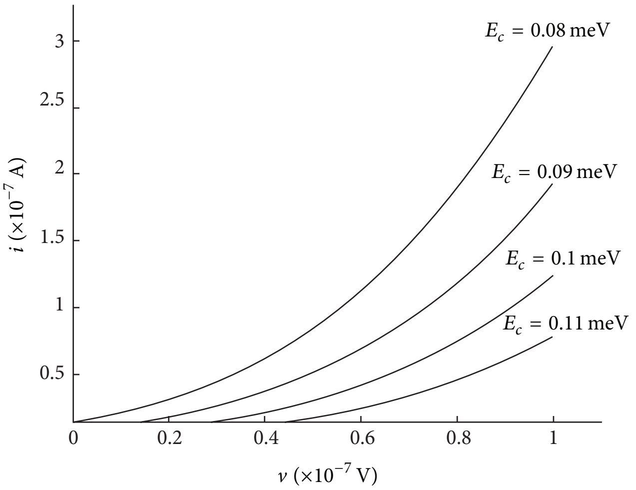


Figure 6: InO_x film I-V curves for three values of the Josephson junction-charging energy E_c. The change in E_c is brought about by the space charge of the displaced In and O ions (recoils) which become interstitials in the irradiated film.

of the InO_x film. Eventually, film temperature could become large enough for the Cooper-pair insulating phase to collapse ($T > \Delta_c/k_B$), or even for the superconducting state to become unsustainable ($T > \Delta/k_B$).

5. Conclusion

Although the investigated nanoscale indium oxide CPI films are immune to the passage of high-energy ions, simulations of ion transport reveal that significant ionization, phononic excitation, and production of displaced atoms can be expected for certain energies, fluences, and types of ions. Indium and oxygen recoils that occupy interstitial positions affect the film's current-voltage characteristics. Moreover, conditions for the Cooper-pair insulating state to persist in InO_x may be disrupted by the ion beam irradiation, through the decrease of the charging energy (between superconducting islands in the 2D Josephson junction array that represents the material in the CPI phase), by the breaking of Cooper pairs in the islands, or by the increase of the film's temperature due to energy losses to phonons.

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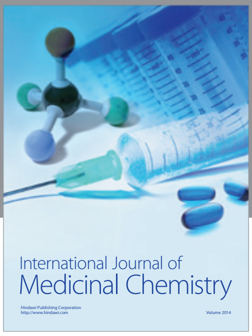
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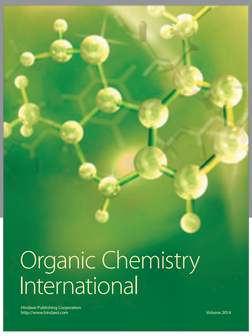
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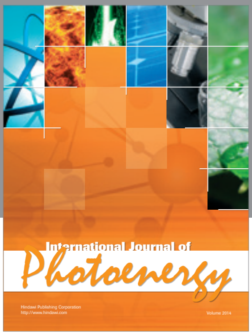
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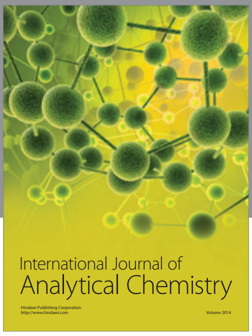
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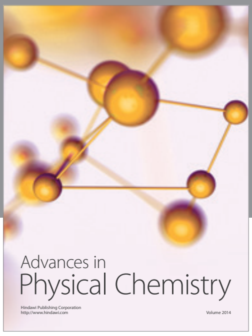
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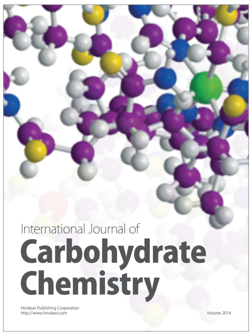
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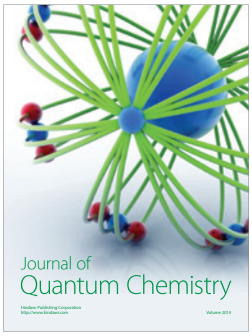
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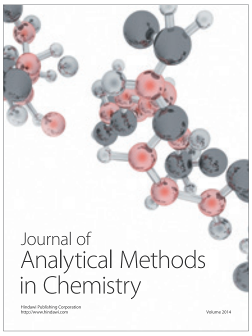
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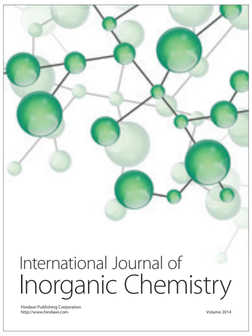
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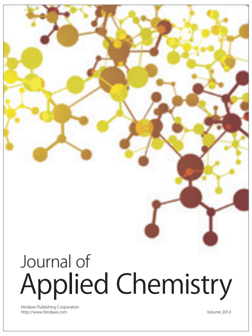
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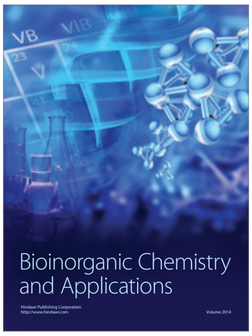
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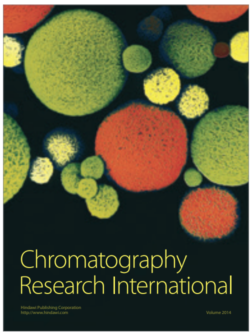
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