SIMULATED RADIATION EFFECTS IN THE SUPERINSULATING PHASE OF TITANIUM NITRIDE FILMS

by

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Scientific paper
UDC: 661.882:544.54:519.245
DOI: 10.2298/NTRP1103254V

This paper investigates possible effects of alpha particle and ion beam irradiation on the properties of the superinsulating phase, recently observed in titanium nitride films, by using numerical simulation of particle transport. Unique physical properties of the superinsulating state are considered by relying on a two-dimensional Josephson junction array as a model of material structure. It is suggested that radiation-induced change of the Josephson junction charging energy would not affect the current-voltage characteristics of the superinsulating film significantly. However, it is theorized that a relapse to an insulating state with thermally activated resistance is possible, due to radiation-induced disruption of the fine-tuned granular structure. The breaking of Cooper pairs caused by incident and displaced ions may also destroy the conditions for a superinsulating phase to exist. Finally, even the energy loss to phonons can influence the superinsulating state, by increasing the effective temperature of the phonon thermostat, thereby reestablishing means for an energy exchange that can support Cooper pair tunneling.

Key words: superinsulator, radiation effects, titanium nitride, Monte Carlo simulation

INTRODUCTION

Titanium nitride has long been known to superconduct below 5.6 K. It is only recently, however, that an infinite-resistance phase of TiN films, appearing at extremely low-temperatures, has been observed and studied [1-3]. This newly observed state, referred to as superinsulating, is thought to be dual to that of a superconductor, vanishing when the magnetic field, temperature or voltage exceed certain critical values. TiN achieves the superinsulating state through another distinct insulating phase, that of a Cooper-pair insulator, in close relation to the superconducting state.

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) act as insulators with thermally activated Cooper pairs as charge carriers [4-8]. Beside TiN, such behavior has been found in InO_x and Be films [7, 9]. It has been proposed, and eventually verified through simulation and experiment, that this insulating phase is characterized by a granular like structure, consisting of superconducting droplets distributed throughout a matrix of normal material [10-13]. It appears spontaneously in the material as a consequence of increased disorder. The phenomenon of superconducting materials turning into insulators is therefore often regarded as a disorder-driven superconductor-insulator transition.

The granular superconducting structure can be modeled by 2-D Josephson junction array (2D JJA) [1,4,14]. It is a system consisting of small superconducting islands, each coupled to its nearest neighbors by Josephson weak links. Each junction is characterized by the Josephson coupling energy, $E_J \quad \hbar I_c/2e$, where I_c is the Josephson critical current, as well as by charging energies E_c and E_{c0}, related to inter-island capacitance and capacitance to ground, respectively. The charging energy E_c is the energy needed for a Cooper pair to be transferred between neighboring islands.

Extensive studies of the conditions under which the stated materials act as either superconductors or insulators at temperatures below 1 K have shown that, in terms of the JJA model, the insulating phase emerges only when the conditions determining the degree of disorder make the charging energies larger than the

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coupling energy (E_c, $E_{c0} > E_J$), with the superconducting gap still exceeding the inter-island charging energy ($\Delta > E_c$) [6, 7]. At the application of an external voltage, in addition to local phase coherence and spatial confinement of Cooper pairs, wave function phase synchronization of all Cooper pairs in the JJA occurs, giving rise to a collective current state. Josephson current couples the phases of adjacent junctions, so as to provide minimal power dissipation in the array. This establishes a global phase-synchronized state, and transport occurs as a simultaneous thermal activation of Cooper pairs through the whole array [4, 8]. Resistance follows an Arrhenius-like temperature dependence

$$R \quad \exp \frac{\Delta_c}{k_B T} \tag{1}$$

where Δ_C is the collective Coulomb barrier of the array, k_B – the Boltzman constant, and T – the absolute temperature. For the 2-D JJA this barrier is

$$\Delta_c \quad E_c \ln \frac{L}{d} \tag{2}$$

where L being the characteristic linear size of the system, and d – the size of an elemental cell in the array [4].

When a film made of TiN that behaves as a Cooper-pair insulator is cooled below approximately 0.1 K it undergoes a transition into the zero-conductivity superinsulating state. Transition from thermally activated insulator to superinsulator occurs at the temperature $T_{SI} \sim E_c/k_B$ [1, 2]. A theoretical explanation for this transition has recently been produced [3]. Tunneling through mesoscopic junctions, such as those between superconducting islands in a TiN film, requires an exchange of energy between the tunneling charge carriers and some kind of excitation modes, to compensate for the difference in energy levels at junction banks. At low temperatures characteristic of the Cooper-pair insulator phase, charge transfer is accompanied by a two-stage energy exchange. The tunneling Cooper pairs generate electron-hole pairs that serve as an environment exchanging energy with the tunneling current and then slowly losing it to phonons. At the nano-scale typical of the granular structure in TiN films, the Coulomb interaction acquires a logarithmic dependence on the distance between charges. At extremely low temperatures, below T_{SI}, this Coulomb interaction drives the environment of unbound electrons and holes through a Berezinskii-Kosterlitz-Thouless (BKT) transition. This opens an energy gap in the electron-hole spectrum that prevents energy transfer from Cooper pairs to the electron-hole environment, and thus suppresses the Cooper-pair tunneling current.

In the superinsulating phase, temperature dependence of the resistance becomes double-exponential [2]

$$R \quad \exp \frac{\Delta_c}{E_c} \exp \frac{E_c}{2k_B T} \tag{3}$$

where Δ_c is the collective Coulomb barrier of eq. (2). The low-bias current-voltage characteristic in the superinsulating state also has a double-exponential form

$$I \quad I_c \exp \frac{(\Delta_c \quad eV)^2}{E_c \Delta_c} \exp \frac{E_c}{2k_B T} \tag{4}$$

where I_c is the Josephson critical current, and $e = 1.6 \cdot 10^{-19}$ C is the elementary charge [1].

This paper investigates the effects of alpha particle and ion beam irradiation on the properties of the TiN superinsulating phase. Radiation effects are analyzed by combining results obtained from Monte Carlo simulations of particle transport with the theoretical model describing the superinsulating state. Simulations were performed in the TRIM (transport of ions in matter) part of the SRIM software package [15], which has been proved to provide results in significant agreement with experiments. Its continuous upgrades during the past decade have made it into a foremost tool for numerical analysis of interaction of ions with matter and validation of theoretical predictions [16-18]. TRIM calculates the range of ions in matter, but 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, *i. e.*, histories followed. A calculation for 1000 incident ions will give better than 10% accuracy [15].

SIMULATION OF HEAVY ION TRANSPORT

Monte Carlo simulations of ion beams traversing 5 nm thick plane-parallel TiN films were conducted with ions chosen to represent certain well known radiation fields, such as those encountered in the space environment [19, 20], or beams commonly used in ion implantation processes. Simulations were restricted to monoenergetic unidirectional beams, incident perpendicularly on thin film surface. Beam energy was varied across typical energy spectra of different ion species. Each simulation run followed 10000 ion histories and took between 10 and 15 minutes to complete. Results in this paper are presented selectively for energies and types of beams that resulted in substantial damage within TiN films. That were the results such as these that allowed a meaningful discussion of the possible radiation effects, based on the theoretical model for the superinsulating phase.

Distributions of displaced titanium and nitrogen atoms in a 5 nm thick TiN film, for four ion beam types with different energies, are shown in fig. 1. Volume concentration of displaced Ti or N atoms at a specific

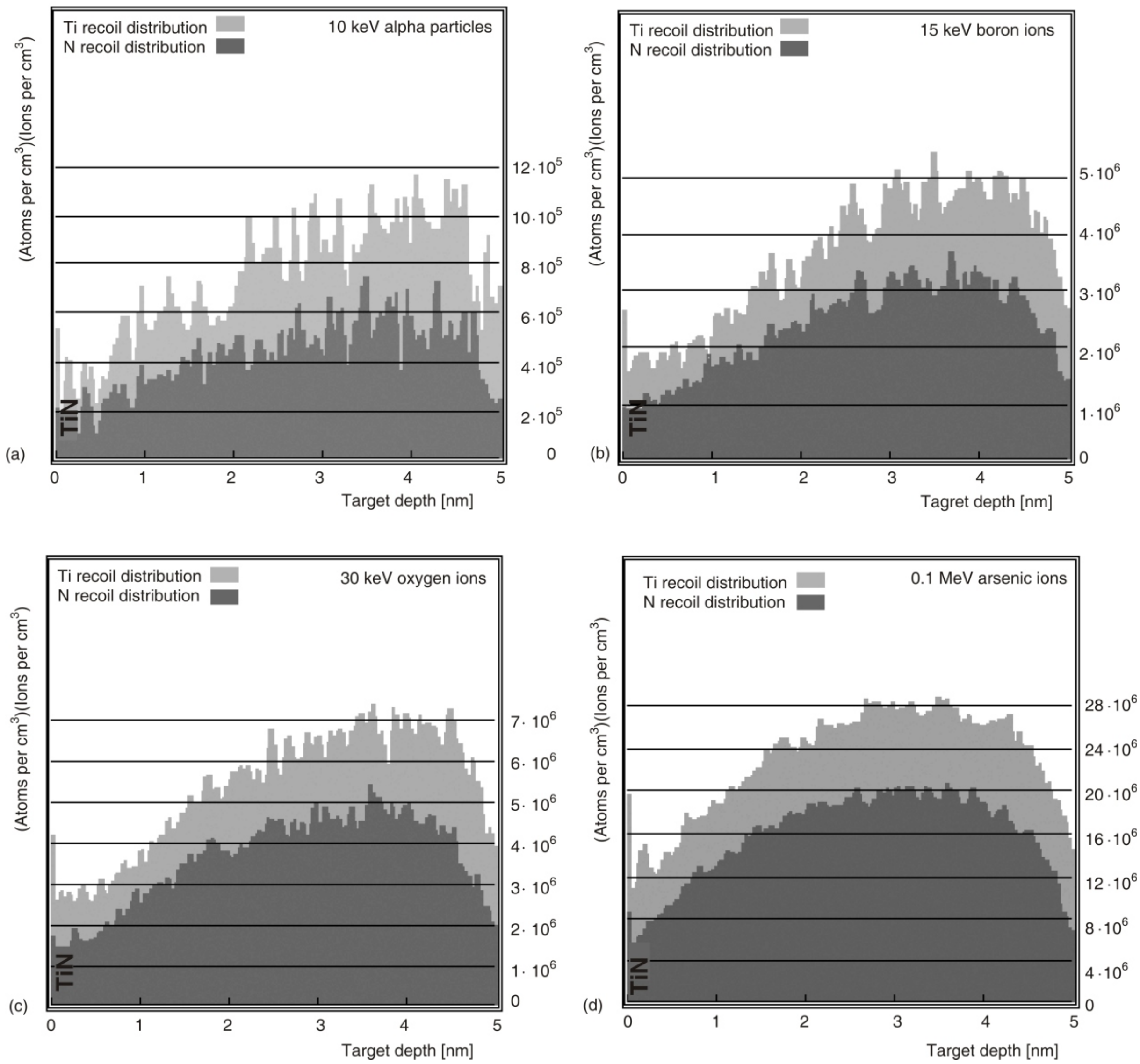

Figure 1. Distribution of displaced Ti and N atoms in a 50 nm thick TiN film irradiated by a beam of ten thousand (a) 10 keV alpha particles, (b) 15 keV boron ions, (c) 30 keV oxygen ions, and (d) 0.1 MeV arsenic ions

depth within the target film is obtained in units of atoms/cm^3 by multiplying the corresponding value from the plot by the ion beam fluence expressed in units of ions/cm^2. Out of the four ion species, arsenic causes most atomic displacements per unit fluence of the incident beam. In all four cases, displacement damage peaks between 3 nm and 4 nm in the film.

Ionization energy losses per unit depth by ions and recoils (displaced Ti and N ions) are shown in fig. 2, for the same four beams as in fig. 1. It is noticeable that ionization losses are mostly due to the incident ions. In case of heavy ion beams, such as those of arsenic or iron, displacement collisions occur with a larger relative energy transfer to recoiling atoms of Ti and N. The displaced atoms then cause more ionization, and possibly cascading atom displacements, resulting in the rise of overall ionization damage toward the back side of the irradiated film, observed in fig. 2(d).

Graphs in fig. 3 present energy losses per unit depth by ions and recoils to phonons. As opposed to ionization losses, which are mostly due to the incident ions, phononic energy losses are almost completely due to the displaced titanium and nitrogen ions.

DISCUSSION OF THE POSSIBLE RADIATION EFFECTS

Owing to their small thickness (10 nm), titanium nitride films are immune to ions with energies above 1 MeV. Non-ionizing energy loss of high energy ions is very low, and they traverse the films without deflection, causing only slight damage [21-23]. Simula-

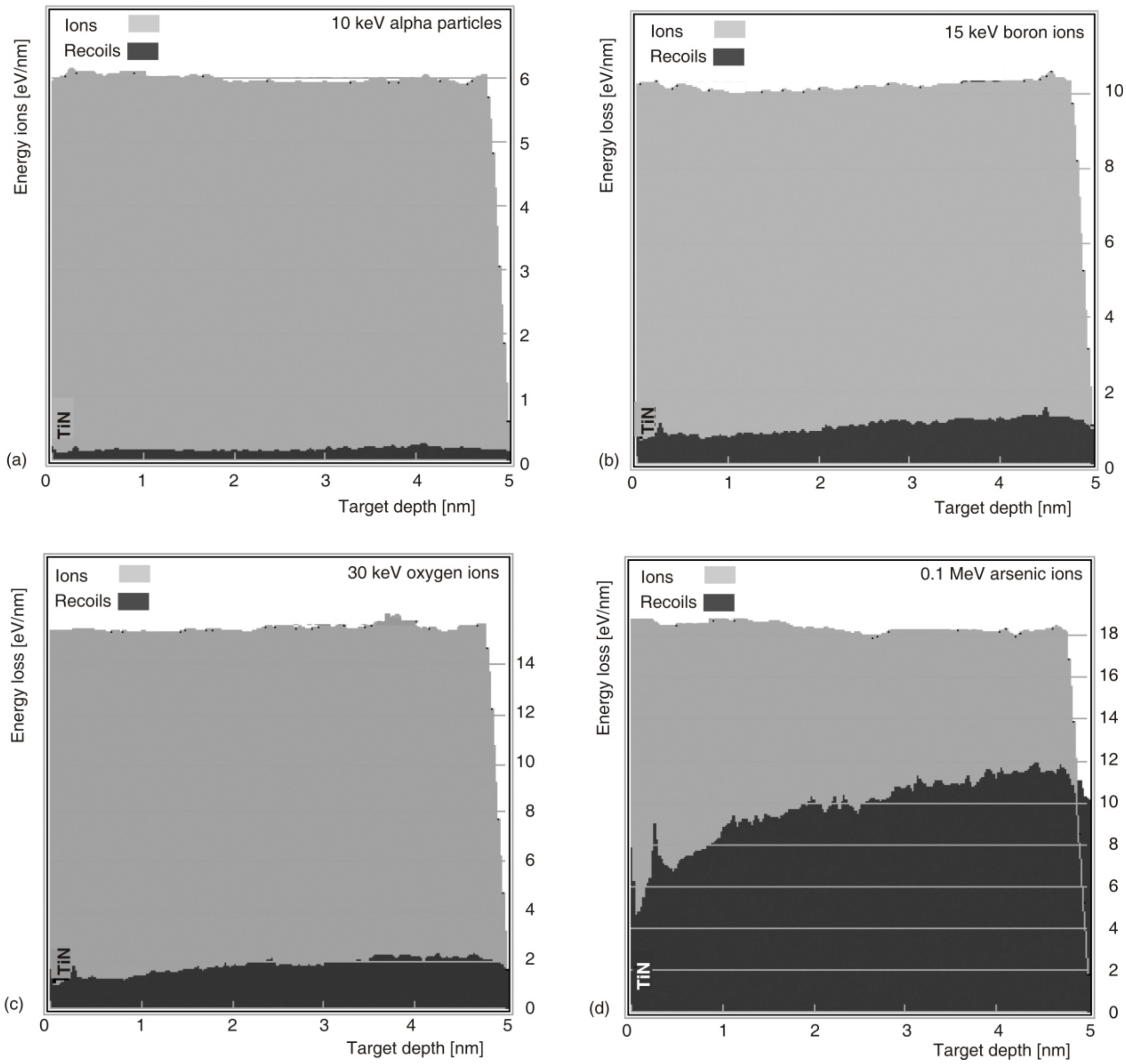

Figure 2. Ionization energy loss by ions and recoils (displaced Ti and N ions) in a 50 nm thick TiN film for an incident beam of ten thousand (a) 10 keV alpha particles, (b) 15 keV boron ions, (c) 30 keV oxygen ions, and (d) 0.1 MeV arsenic ions

tions of ion transport suggest, however, that for certain ion species there are energy ranges in which a great number of atom displacements would occur in irradiated TiN films. The number of atomic displacements is in direct proportion to the fluence of incident radiation, *i. e.*, the number of particle histories followed in the Monte Carlo simulation. Ionization energy losses by both the incident ions and the recoil ions of titanium and nitrogen can cause the breaking of Cooper pairs in superconducting islands. Certain amount of ion energy is also lost to phononic excitations of the lattice, as evidenced by simulation results.

There are several possible ways in which radiation effects produced by ions could affect the superinsulating TiN films [24-26].

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 according to eq. (1). Simulation results show that Ti ions would suffer much more displacements than N atoms for all used ion beams. This would make the space charge positive, causing a decrease of the charging energy. Nevertheless, the double-exponential dependences of eqs. (3) and (4) are insensitive to any changes in the charging energy that could possibly be caused by the traversing ions and secondary particles they dislodge. The current remains nearly zero for all changes of E_c that could be expected to arise from radiation effects, while the relative change of film resistance is negligible.

The stability of the superinsulating state is, however, critically dependent on the value of E_c, and only exists when $E_c > k_B T$. If the radiation damage produced by the ion beams is large enough to disrupt this condition, the thin film may revert to thermally activated behavior and the plain exponential *i-v* dependence.

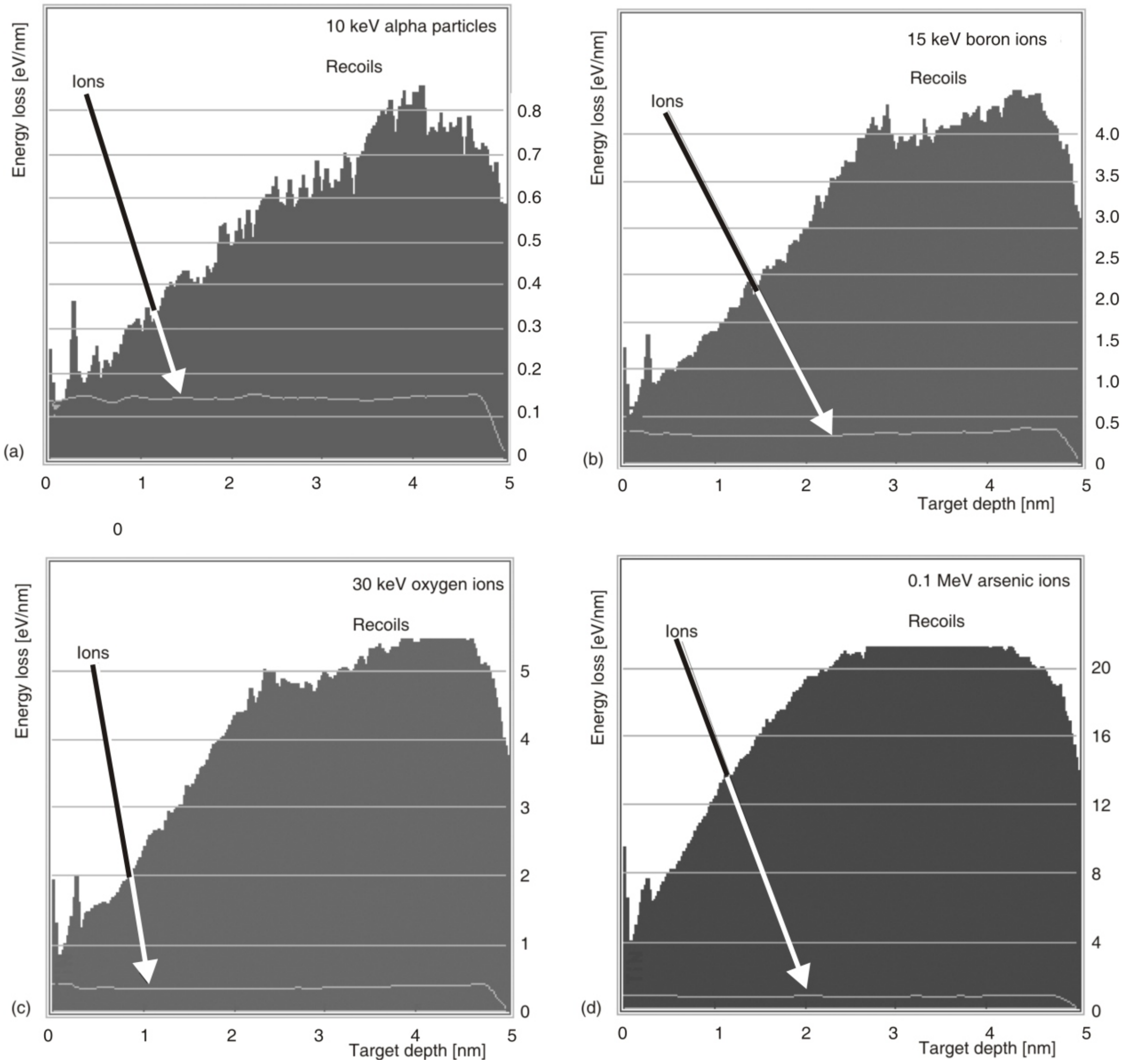

Figure 3. Energy loss to phonons by ions and recoils (displaced Ti and N ions) in a 50 nm thick TiN film for an incident beam of ten thousand (a) 10 keV alpha particles, (b) 15 keV boron ions, (c) 30 keV oxygen ions, and (d) 0.1 MeV arsenic ions

Ionization calculated from the simulations refers to the generation of electron-hole pairs in the normal phase, but points to the possibility of substantial Cooper pair dissociation in the supeinsulating state. Ionization losses of both the incident and the displaced ions could cause enough Cooper pairs to break, that the basic condition for superinsulating behavior, that of global phase coherence of these pairs across the whole sample, would vanish. Radiation induced dissociation of Cooper pairs has already been linked to the reduction of the critical temperature in superconducting materials [27].

Energy losses to phonons increase the effective temperature of the phonon thermostat. Phononic excitations that are normally suppressed at low temperatures are thereby reestablished. Energy exchange processes that accompany Cooper pair tunneling can then be mediated by these phononic excitations, and the tunneling current can reappear.

CONCLUSION

Although TiN 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 some energies, fluencies, and types of ions encountered in cosmic rays and used for implantation processes. Displacement damage, affecting primarily the value of inter-island charging energy in the 2-D Josephson junction array that represents the material in the superinsulating phase, has little influence on either the current-voltage characteristics or the resistance, due to their double-exponential dependences on this energy. The conditions for the superinsulating state to subsist may, however, be disrupted by irradiation, through the decrease of the charging energy, the breaking of Cooper pairs, or the reemergence of phononic excitation modes.

ACKNOWLEDGMENT

The Ministry of Education and Science of the Republic of Serbia supported this work under contract 171007.

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Received on June 6, 2011
Accepted on July 7, 2011

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СИМУЛАЦИЈА ДЕЈСТВА ЗРАЧЕЊА НА СУПЕРИЗОЛАТОРСКУ ФАЗУ ФИЛМОВА ТИТАНИЈУМ НИТРИДА

У раду се, применом нумеричке симулације транспорта честица, разматра дејство које озрачивање алфа честицама и јонским снопoвима може да има на карактеристике суперизолаторске фазе, недавно уочене у филмовима титанијум нитрида. Јединствене физичке особине суперизолаторског стања обухваћене су ослањањем на модел дводимензионалног низа Џозефсонових спојева за опис структуре материјала. Показује се да промене енергије наелектрисавања Џозефсоновог споја изазване зрачењем не би значајно утицале на струјно-напонску карактеристику суперизолаторског филма. С друге стране, теоријска разматрања указују да нарушавањем прецизне грануларне структуре услед дејства зрачења може да дође до повратка у изолаторско стање са термички активираном отпорношћу. Раскидање Куперових парова дејством упадних и измештених јона такође може да уништи услове постојања суперизолаторске фазе. Коначно, чак и фононски механизам губитка енергије може да утиче на суперизолаторско стање, повећањем ефективне температуре фононског окружења, чиме се поново успостављају услови за размену енергије која омогућава тунеловање Куперових парова.

Кључне речи: суперизолатор, ефекти зрачења, титанијум нитрид, Монте Карло симулација