THE INFLUENCE OF RADIATION ON THE CHARACTERISTICS OF SUPERINSULATOR FILMS

by

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The paper considers the effect of radiation exposure on the characteristics of the superinsulator phase using Monte Carlo simulation of radiation transport through superinsulator films. The unique physical properties of the superinsulator state are captured by a 2-D array model of Josephson junctions to describe the structure of the material. Simulations were carried out for different film thicknesses, as well as for radiation beams of different geometries. In the assessment of radiation resistance, the Monte Carlo method of simulating the passage of radiation through the material and the deposition of energy gives good results. Using numerical simulation, along with a precise definition of the problem from the point of view of the characteristics of the radiation field and the observed material environment, it is possible to predict the response of thin superinsulator films to ionizing radiation. Based on the obtained results, suggestions were given for the possibility of further application of superinsulator materials in the production of electronic circuits.

Key words: ionizing radiation, superinsulator film, radiation resistance

INTRODUCTION

The increasing degree of miniaturization of electronic components and the increase in the contamination of the natural environment with ionizing radiation actualize the problem of the influence of radiation on electronic components and materials. The study of this problem is necessary to predict the electronic components reliability in the fields of ionizing radiation. The analysis of the impact of ionizing radiation on modern materials is significant because of the insight into their behavior during technological processes [1-4].

The stability of the characteristics of semiconductor components is especially relevant in the case of semiconductor memories in computer technology. Semiconductor memories are mainly made in MOS technology. On this occasion, insulating materials play an important role in the production of vertical separation layers, insulation of semiconductor and conductive layers, and passivation of the entire layer. For this purpose, new insulating materials are being sought to replace traditional materials, the shortcomings of which are becoming an increasing problem with each new generation of integrated circuits [5, 6]. Recently, it has been discovered that superconducting materials, under suitable conditions, go into a state of zero conductivity. Materials in this state are called superinsulators [6, 7].

This work is aims to analyze the effects of exposure of superinsulators to ionizing radiation using a Monte Carlo simulation of the transport of ionizing radiation through superinsulator films. Thus, a conclusion can be reached about the applicability of these materials for the production of electronic components that should work in the fields of ionizing radiation.

EFFECT OF RADIATION ON FILMS OF INSULATING MATERIALS

Different types of interaction of radiation with materials can be divided into: inelastic scattering on electrons in the shell, inelastic scattering on the atomic nucleus, and elastic scattering on the atomic nucleus [8-10]. In the solid state, the interaction of radiation with materials can lead to the transfer of individual electrons to the conduction zone. The electron transferred to the conduction zone participates in the electrical and thermal conduction of the material. Such an electron, however, does not move completely freely since it is acted upon by the electric field of the crystal

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lattice. In the case of insulating materials, for the creation of new free electrons, it is necessary to invest energy greater than about 9 eV (since 8.9 eV is the width of the forbidden zone of amorphous SiO_2). The mass migration of electrons into the conduction zone leads to the breakdown of the insulator. In practice, however, transferring a larger number of electrons into the conduction zone requires a higher radiation energy. This energy is about three times greater than the width of the forbidden zone of the insulator. This amount of radiation energy is a consequence of energy dissipation to other mechanisms (intrazonal excitation of electrons, vibrational excitation of the crystal lattice and scattering on atomic nuclei) [10-12].

Elastic scattering of the incident radiation on the nuclei leads to the ejection of atoms from their positions in the crystal lattice. The influence of the crystal lattice on the process of displacement of atoms determines the energy threshold of this process. The displacement of atoms from their positions in the crystal lattice leads to material damage. The resulting vacant place in the crystal lattice is called a vacancy. When the displaced atom occupies a place in the space between the lattice nodes, the so-called interstitial atom is formed. An interstitial atom and a vacancy form a Frenkel pair.

Along their paths, recoiled atoms lose energy in two ways: by ionization and by displacement of other atoms. Starting from the primary ejected atom, new displaced atoms appear branching through the material forming a dislocation tree. Non-ionizing displacement interactions dominate near the end of the path (when the last 10 % of the energy of the process is consumed). Thus, at the ends of the branches of the displacement tree, a region with a high concentration of Frenkel defects is formed, which is called a zone cluster. The cascade originating from one primarily ejected atom usually contains two to three zone clusters with longitudinal dimensions of about 5 nm, fig. 1. The entire displacement tree with the final clusters is formed in about 1 ns. Defects created in this way along the path of the displaced atoms form the basis of radiation damage inside the insulator [13, 14].

The first material in which the superinsulator phase was experimentally established was titanium nitride (TiN), made as a nanometer film on a SiO_2 substrate [15]. The SiO_2 layer was formed on the silicon substrate by thermal oxidation, while the TiN layer was obtained by chemical vapor-phase deposition. Figure 2 shows a cross-sectional micrograph of the structure obtained with a high-resolution transmission electron microscope (HRTEM).

The substrate on which the film of the superinsulator material is made, as well as the layers that can be deposited over it, can have an impact on the radiation resistance of the superinsulator when it is exposed to a multidirectional radiation field. If, as in fig. 2, it is a SiO_2/Si structure, the radiation effects related to the interface of these two layers are important. Radiation damage in the oxide layer includes three phenomena: appearance of charge trapped in the oxide, increase in the number of traps on the interface, and increase in the number of traps inside the oxide. The total trapped charge in the oxide after irradiation is, most often, positive. In this case, the surface traps can freely exchange charge with the silicon substrate. As a result, their sign depends on the applied voltage.

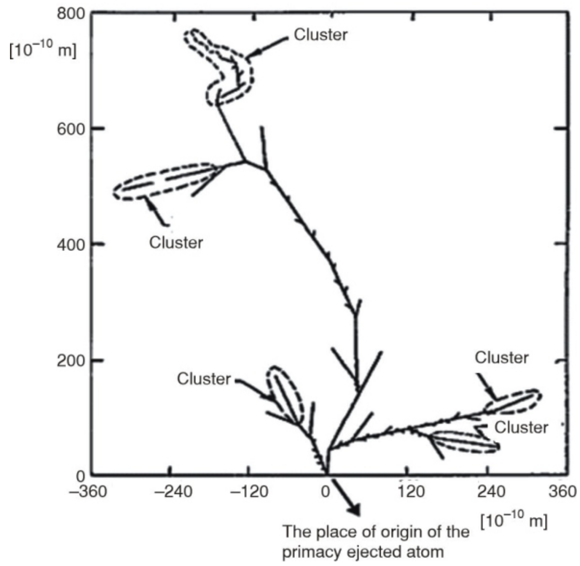

Figure 1. Displacement tree with final clusters [13, 14]

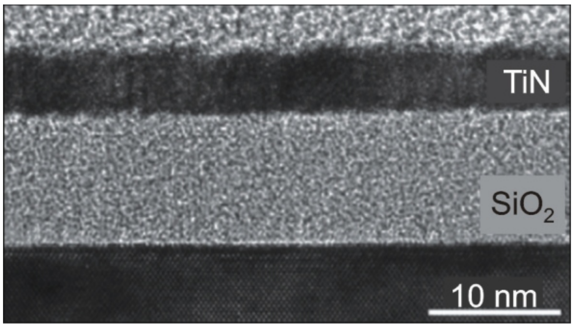

Figure 2. The HRTEM micrograph of the cross-section of the structure used to study the superinsulator phase of the TiN film [15]

By choosing the appropriate material for the layers surrounding the superinsulating film, it is possible to achieve its radiation hardening. This means that the technique of making radiation-resistant superinsulator films should be sought in combination with reducing the thickness of the superinsulator film and the appropriate selection of materials surrounding it.

Nanometer-thick superinsulator films (which enable their insulating properties) should be resistant to radiation damage by displacement of atoms, since these damages are of micron dimensions, which should not effect such thin films.

In the case of massive insulators, displacement trees with terminal clusters are located inside the insulator. The same happens with insulating films (layers) made of classic insulating materials, since their thickness is many times greater than 5 nm to have resistance to the occurrence of overvoltage [15]. However, superinsulator materials perform reliable insulation with films of nanometer dimensions. This characteristic should make them resistant to radiation damage by displacement of atoms, since within their volume, a larger number of displacement trees with terminal clusters cannot be formed.

Volumetric damage in the insulator can also occur as a result of nuclear reactions. Heavy charged particles emitted in such reactions can themselves produce volumetric damage (and can also become impurity atoms). However, the appearance of secondary electrons and impurity atoms due to nuclear reactions has a negligible effect on the characteristics of insulating materials compared to displacement damage.

NUMERICAL SIMULATION OF RADIATION TRANSPORT THROUGH A SUPERINSULATOR FILM

In this work, the Monte Carlo method was used for the numerical simulation of the effects of radiation on superinsulator films. The simulation of the passage of particles through a material is based on the stochastic nature of collision processes. At each collision, a particle loses energy which the Monte Carlo procedure is determined from the appropriate distribution. On its way, the particle loses energy up to some threshold energy at which it is considered absorbed. The energy absorbed in a given volume of material changes its characteristics, and the resulting changes affect the passage of particles in subsequent histories of the overall process [16, 17].

The Monte Carlo method is based on the numerical simulation of random variables based on their known distributions. The process of obtaining a random variable from its distribution function, or probability density function, is called *sampling the value of a random variable*. For this purpose, either the function inversion method the rejection method or the compensation method is used [18]. Sampling a random variable using the method of inversion of the distribution function is the most advantageous, but it is practically possible only when the analytical form of the inverse function is known. Under this condition, the value of the random variable is obtained by replacing the corresponding distribution obtained by the random number generator with the inverse value of the function [19]. If the distribution function is not invertible, the von Neumann rejection method is applied [20].

In this paper, a standard pseudorandom number generator was used. The used generator met the conditions: generation of a series of pseudorandom numbers uniformly distributed in the interval (0, 1), reproducibility, portability and high speed. The TRIM module (transport of ions in matter) of the program package SRIM (Stopping and range of ions in matter) was used, which enabled the calculation of the energy loss of incident radiation through ionization, phonon excitation of the lattice and displacement of material atoms [21]. When calculating the SRIM cascade damage of displaced atoms, four types of events are distinguished: collision with displacement, production of vacancies, collision with replacement of atoms and appearance, of interstitial atoms. The SRIM assumes that the material is at absolute zero temperature before radiation exposure, which makes this program suitable for considering radiation effects in superinsulators since they exist only at extremely low temperatures. The analysis of breaking up of Cooper pairs in superinsulator islands during the passage of radiation was performed by replacing the ionization energy of material atoms with a value corresponding to the dissociation energy of a Cooper pair. The ion beams in the simulations were chosen to match the radiation fields in which electronic components are often found. For materials in spacecraft and satellites, hydrogen, helium and iron ion beams are the most important, while phosphorus, boron and arsenic are most often used for the technological procedures of implanting ions with the aim of doping.

RESULTS

Figure 3 shows the distribution of displaced Ti and N atoms (recoil distribution) in a 5 nm thick TiN superinsulator film irradiated with beams of boron, phosphorus, arsenic and iron ions.

Figure 4 shows the ionization energy losses of incident ions and recoils (displaced Ti and N atoms) in a 5 nm thick TiN superinsulator film irradiated with beams of boron, phosphorus, arsenic and iron ions.

Figure 5 shows the particle tracks for an incident beam of 10^5 iron ions with an energy of 10 keV in a TiN superinsulator film with a thickness of 20 nm.

Figure 6 shows the results for an incident beam of 10^4 protons with an energy of 10 keV in a TiN superinsulator film with a thickness of 5 nm.

Simulation of radiation transport shows, however, that for certain types and energies of ions in irradiated superinsulator films, a large number of displaced atoms occurs. The number of displaced atoms is directly proportional to the incident radiation flux, *i.e.*, to the number of ions whose transport is monitored in the Monte Carlo simulation. Ionization losses of incident and displaced ions correspond to the breaking of Cooper pairs in superinsulating islands. The simulation results also show that part of the radiation energy is converted into the energy of lattice phonon excitations.

The consequences of radiation for superinsulating films are manifold. The space charge created by displaced ions occupying internodal (interstitial) positions

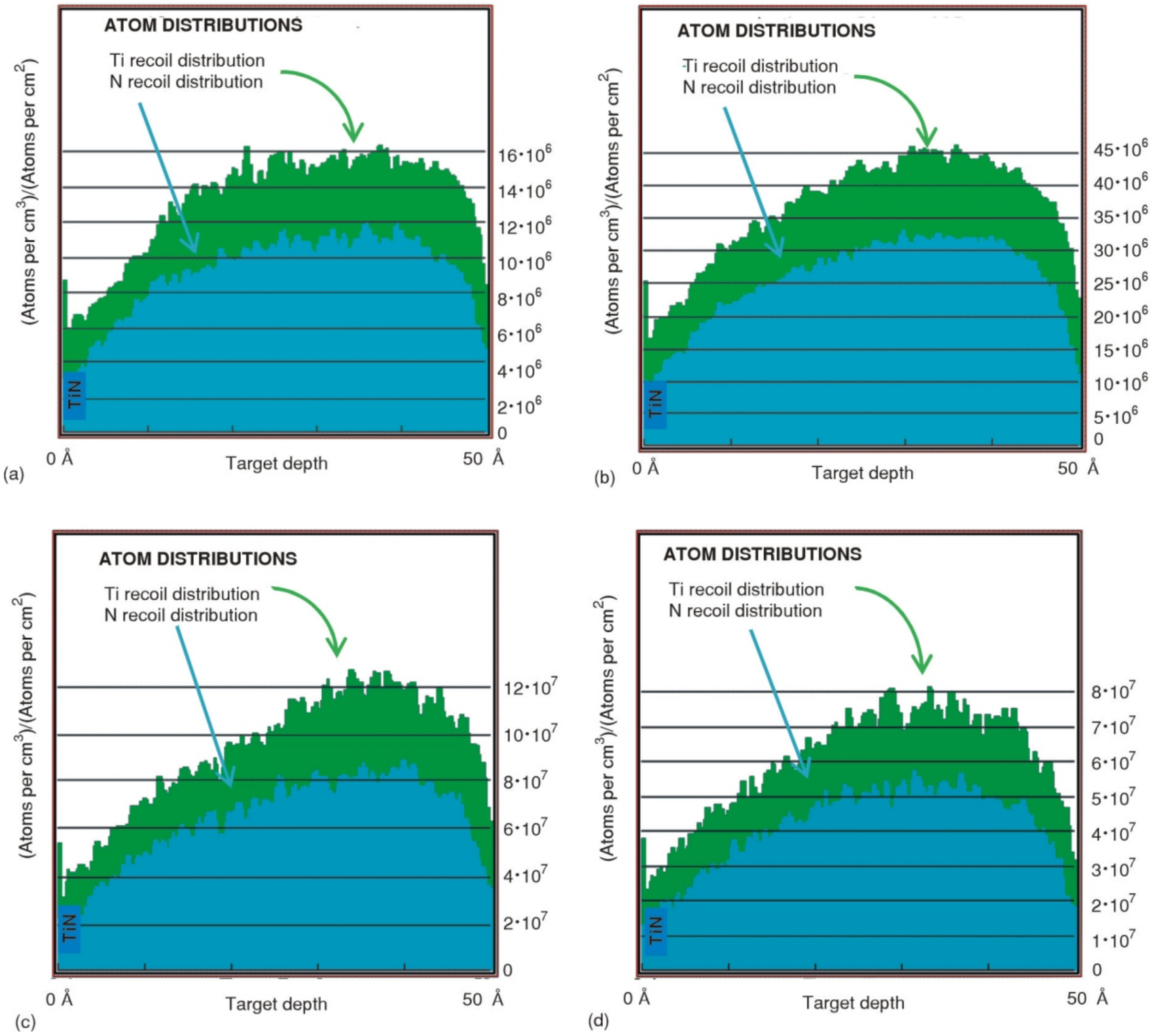

Figure 3. (a)-(d) Distribution of displaced T and N atoms in a 5 nm thick TiN film irradiated with a beam of (a) 10^4 boron ions of energy 10 keV, (b) 10^4 phosphorus ions of energy 50 keV, (c) 10^3 arsenic ions of energy 50 keV, and (d) 10^3 iron ions of energy 100 keV. The unit along the abscissa, which corresponds to the depth inside the film, is the angstrom (1 Å = 0.1 nm)

affects the charge energy of Josephson junctions, which exist between superinsulating islands in the granular structure of superinsulators. Such a change in the charge energy of Josephson junctions results in a change in the collective Coulomb barrier.

Monte Carlo simulation results for TiN films show that, almost always, displacements of Ti atoms are dominant over displacements of N atoms. Interstitial Ti atoms form a positive volume charge, so their presence reduces the charge energy. This is extremely important since the superinsulator state can only exist if the charge energy is greater than the thermal energy ($k_B T$, where k_B is the Boltzmann constant and T is the absolute temperature), which is a condition that is violated in case of greater radiation damage. By violating the ratio of charge energy and thermal energy, $k_B T$, the conditions are created for the superinsulator film to go into a state with thermal excitation of charge carriers, which translates their current-voltage characteristic from double exponential to exponential.

Ionization losses calculated based on of Monte Carlo simulation refer to the generation of electron-hole pairs in the normal phase of the material, but indicate the possibility of a significant breaking of Cooper pairs, when the energy of ionization is replaced by the dissociation energy of a Cooper pair. Incident and displaced ions can lead to a sufficiently large number of dissociations of Cooper pairs to violate the basic condition of superinsulator behavior, that is, the existence of global phase coherence of pairs throughout the material sample.

The inverse phenomenon of superinsulators is the phenomenon of superconductivity. In the literature, the phenomenon of superconductivity is explained by Cooper pairs of electrons. Such an interpretation is acceptable and is reduced to paired electrons of opposite spin at the minimum potential energy. Such a pair of electrons cannot enter into any interaction that would have to result in a change of energy since their quantum mechanical state does not allow

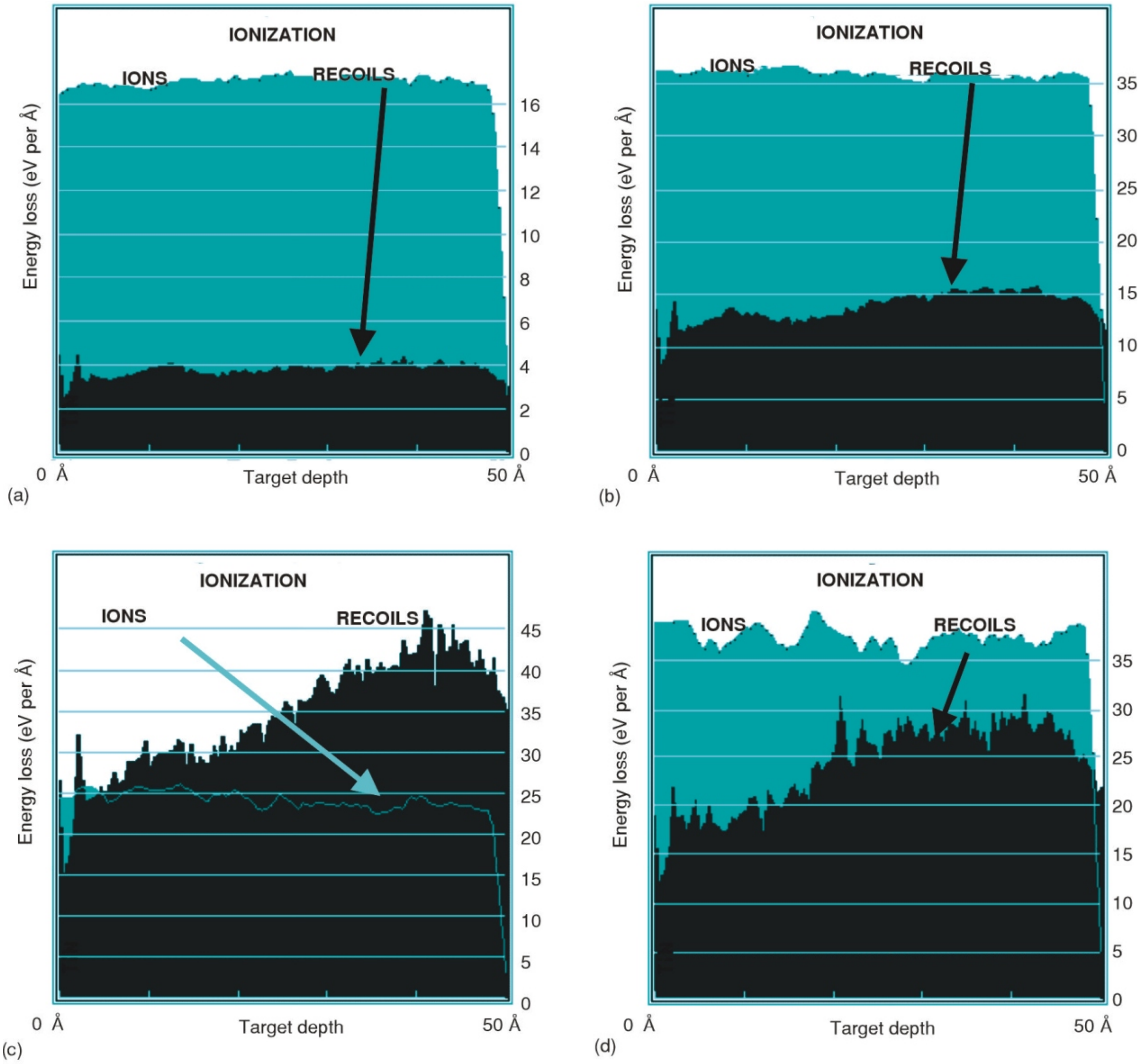

Figure 4. (a)-(d) Ionization losses of incident ions and displaced Ti and N atoms in a 5 nm thick TiN film irradiated with a beam of (a) 10^4 boron ions of energy 10 keV, (b) 10^4 phosphorus ions of energy 50 keV, (c) 10^3 arsenic ions energy 50 keV, and (d) 10^3 iron ions energy 100 keV. The unit along the abscissa, which corresponds to the depth inside the film, is the angstrom (1 Å = 0.1 nm)

any process of changing any quantum number because they are fixed at a minimum value. This interpretation of superconductivity is just a special case of the Pauli principle. This indicates that the appearance of superinsulators can also be explained by the Pauli principle. Such an interpretation would start from the idea that in a superinsulator there is an extremely small number of paired electrons that cannot exchange quantum numbers with other electrons that are also tightly paired with opposite spins. From such a model follows the conclusion that superconductors and superinsulators represent a direct consequence of the Pauli principle of the prohibition of interaction, *i. e.*, prohibiting energy exchange with changes in conductivity and resistance [22, 23].

The effect of phonon excitation of the material increases the effective temperature of the phonon environment. Excited phonon modes, which are suppressed at the low temperatures necessary for the existence of the superinsulator state, are re-established by the effect of radiation. Such phonon excitations mediate the energy exchange processes that accompany the tunneling of Cooper pairs between islands, which results in the appearance of a tunneling current and the breaking of the superinsulator state.

The obtained results show that the breaking of Cooper pairs and the displacement of material atoms become more pronounced with the increase in the thickness of the superinsulator film. The breaking up of Cooper pairs is mostly contributed by recoiled atoms (more than incident ions). Displaced atoms significantly affect the phonon excitation of the crystal lattice of the superinsulator film material.

Unlike the effects produced by the displacement of the atoms of the material, which are, for the most part, of a permanent character, dissociation of Cooper

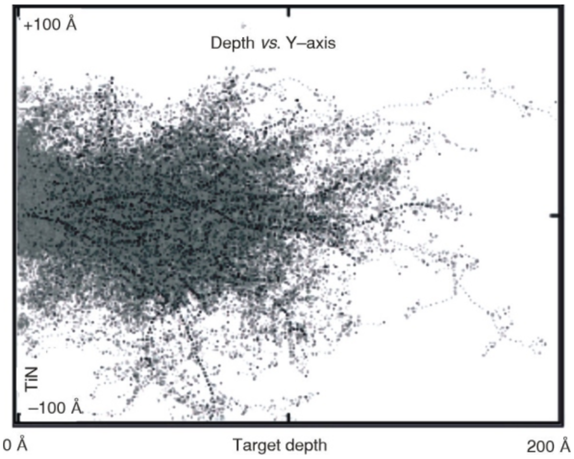

Figure 5. Particle tracks for an incident beam of 10^5 iron ions with an energy of 10 keV in a TiN film with a thickness of 20 nm. The darker traces correspond to Fe ions, while the lighter traces are the trajectories of displaced Ti and N ions. The unit along the abscissa axis, which corresponds to the depth inside the film, is angstrom (1 Å = 0.1 nm)

pairs and phonon excitation are transient effects and they affect the properties of superinsulator films only while the radiation lasts.

DISCUSSION AND CONCLUSION

The paper analyzed the effect of exposure of the superinsulator phase on radiation transport through superinsulator films using Monte Carlo simulation. The procedure was successful. It has been shown that nanometer-thick superinsulator films are generally immune to the passage of high-energy ionizing radiation. In addition, numerical simulations of the passage of radiation through films of superinsulating material show that for certain energy ranges, fluxes and types of radiation, significant effects can be expected even at very small material thicknesses. Theoretically, it proved possible to explain how radiation effects affect the properties of superinsulator films. The space charge formed by ions displaced by the passage of radiation can affect the charge energy of Josephson junctions, and thus the collective Coulomb barrier of the array. The stability of the superinsulator state depends on the value of the charge energy. If in the irradiated superinsulator this energy becomes less than the thermal excitation energy, the conditions for the survival of the superinsulator phase are violated and the superinsulator film passes into a state of thermal active resistance with an exponential dependence.

Radiation can also lead to a significant breaking of Cooper pairs in the superinsulator phase. The dissociation of a larger number of Cooper pairs can disrupt the phase coherence that is established in superinsulators throughout the material sample. In this way, the basic condition for the existence of a superinsulator phase is lost. Phonon excitations of the lattice of the material have no effects at low temperatures, but when radiation passes through the superinsulator film, they can become apparent. In this way, the processes of energy exchange are re-

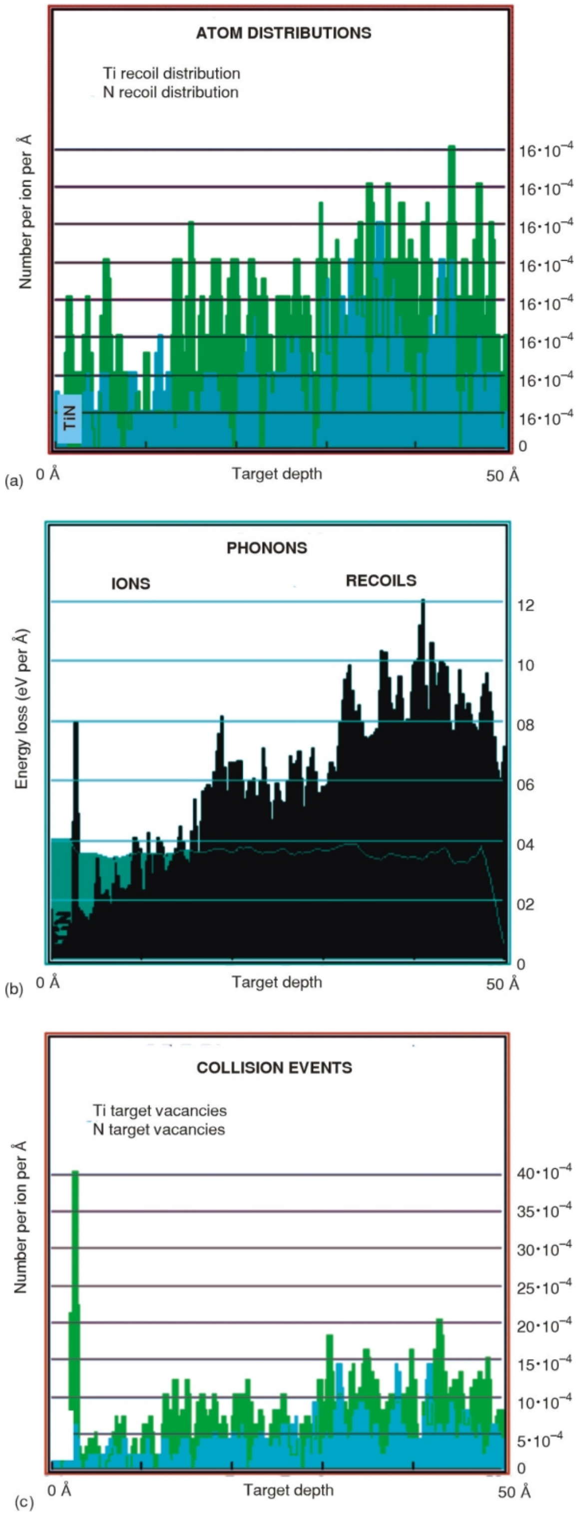

Figure 6. (a)-(c) Results for an incident beam of 10^4 protons of energy 10 keV in a 5 nm thick TiN film; (a) distribution of displaced titanium and nitrogen atoms, (b) phonon energy losses of incident ions and displaced Ti and N atoms, and (c) distribution of vacancies created by displacement of Ti and N atoms. The unit along the abscissa axis, which corresponds to the depth inside the film, is the angstrom (1 Å = 0.1 nm)

stored, which enables the tunneling of Cooper pairs, which leads to the loss of the superinsulator phase. In contrast to the effects produced by displacement of lattice atoms that lead to stable damage, dissociation of Cooper pairs and phonon excitation are transient effects that affect the super-insulating properties of the material only while the irradiation lasts.

Compared to insulators that are mainly used for making planar circuits, for making components in microelectronics and for polarizing microelectronic circuits, superinsulator materials exhibit a higher degree of radiation resistance. Considering that the application of superinsulator materials could solve the problem of leakage current, which is the limiting effect of further miniaturization of integrated circuits, superinsulator films should gain wide application. As in the case of superconductors, the main problem is to reach the superinsulator phase at higher temperatures. A more detailed understanding of the physical characteristics of the superinsulator phase, including radiation effects, contributes significantly to the achievement of this goal.

Further research on radiation effects in superinsulators should be focused on the characteristics of more complex structures in which superinsulator films would be found. Also, a technique for hardening superinsulator films should be developed, to enable their application under conditions of exposure to radiation that could damage the conditions of the superinsulator phase or completely abolish them.

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AUTHORS' CONTRIBUTIONS

N. M. Kartalović – conceptualization, methodology investigation, resources; T. M. Stojić – formal analysis, writing original draft, writing – review and editing, visualization, supervision; U. D. Kovačević – investigation, formal analysis, visualization.

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УТИЦАЈ ЗРАЧЕЊА НА КАРАКТЕРИСТИКЕ СУПЕРИЗОЛАТОРСКИХ ФИЛМОВА

У раду је анализирано дејство излагања зрачењу на карактеристике суперизолаторске фазе применом Монте Карло симулације транспорта зрачења кроз суперизолаторске филмове. Јединствене физичке особине суперизолаторског стања обухваћене су моделом 2-D низа Џозефсонових спојева за опис структуре материјала. Симулације су спроведене за различите дебљине филмова, као и за снопове зрачења различитих геометрија. У процени радијационе отпорности, Монте Карло метод симулације проласка зрачења кроз материјал и депоновања енергије даје добре резултате. Нумеричком симулацијом, уз прецизно дефинисање проблема са становишта карактеристике поља зрачења и посматране материјалне средине, могуће је извршити предикцију одзива танких суперизолаторских филмова на јонизујуће зрачење. На основу добијених резултата дате су сугестије за могућност даље примене суперизолаторских материјала у изради електронских кола.

Кључне речи: јонизујуће зрачење, суперизолаторски филм, радијациона отпорност