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Medium-energy ion-beam simulation of the effect of ionizing radiation and displacement damage on SiO_2-based memristive nanostructures

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A B S T R A C T

The principles of ion-beam simulation of the effect of fast (fission) neutrons and high-energy protons based on medium-energy ion irradiation have been developed for the $Au/Zr/SiO_2/TiN/Ti$ capacitor-like memristive nanostructures demonstrating the repeatable resistive switching phenomenon. By using the Monte-Carlo approach, the irradiation fluences of H^+, Si^+ and O^+ ions at the energy of 150 keV are determined that provide the ionization and displacement damage equivalent to the cases of space protons (15 MeV) and fission neutrons (1 MeV) irradiation. No significant change in the resistive switching parameters is observed under ion irradiation up to the fluences corresponding to the extreme fluence of 10^{17} cm^{-2} of space protons or fission neutrons. The high-level radiation tolerance of the memristive nanostructures is experimentally confirmed with the application of 15 MeV proton irradiation and is interpreted as related to the local nature of conducting filaments and high concentration of the initial field-induced defects in oxide film.

1. Introduction

Recently, the phenomenon of resistive (memristive) switching has became a subject of intensive research in connection with the fact that, according to the existing forecasts, it can be used for the development of emerging memory or logic devices [1–3] and, in prospect, the systems of neuromorphic computing and artificial intelligence [4–6] approaching to the capabilities of a human brain. The Resistive Random Access Memory (RRAM) is of interest due to a number of advantages, such as scalability and simplicity of the fabrication technology, the attainability of fast, low-energy, high-endurance operation [3,7]. The fabrication of RRAM devices on the basis of SiO_2 thin films is especially attractive due to their best integrability with the current CMOS technology. In contrast to those types of memory, in which the information storage is provided by the storage of charge (e.g. flash memory), the information write/erase process in RRAM is relied on the presence of at least two states of a material with essentially different electric resistances. Such a memory type is assumed to be less sensitive to the radiation impact. This ensures good prospects for the application of RRAM in electronic devices operating in the conditions of radiation exposure, particularly in space environment and in nuclear industry. A number of works devoted to the study of radiation effect on memristive devices on the basis of transition metal oxides [8–14] have been published so far, however there are only few reports on similar studies for the SiO_2-based memristive devices [15].

Direct testing of the devices under the actual conditions of space or reactor radiation is usually inaccessible for the researchers. However, the evaluation of radiation tolerance can be done by applying the physical simulation. It has been already shown [16,17] that the tolerance of thin heterophase structures to fast neutron radiation can be proved by the simulating ion irradiation. Analogously, to simulate the ionizing effect of high-energy (space) protons, it is possible to use the irradiation with H^+ ions of medium energies [18]. This approach is accessible for the users of ion implanters available in many research centers and in microelectronic industry.

In the present work, the given approach is applied for the ion-beam simulation of the effect of space protons (with typical energy of ~15 MeV) and fission neutrons (with typical energy of ~1 MeV) on the $Au/Zr/SiO_2/TiN/Ti$ capacitor-like devices demonstrating the repeatable bipolar resistive switching related to the formation

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and local oxidation of conducting paths (filaments) in oxide material [19]. The obtained results allow predicting the high radiation tolerance of the studied memristive devices.

2. Principles of ion-beam simulation

Let's consider first the basic principles of medium-energy ion-beam simulation of the irradiation of memristive nanostructures with fast (fission) neutrons and high-energy protons. The effect of irradiation on memristive devices of "metal–insulator–metal" type is assumed to be caused predominantly by the radiation damage in the switching medium, in our case – SiO_2 film.

The interaction of fast neutrons with solids produces displacement damage mainly as a result of elastic atomic collisions leading to the generation of recoils with a continuous energy spectrum. Therefore, it is possible to simulate the fast neutron effect by using the irradiation with ions of chemical species composing the target. In the case of ion irradiation with a certain initial energy, the required fluences of ions must be chosen taking into account both the elemental composition of a structure and the ion energy losses, which determine the concentration of displaced atoms and energy density transmitted to electronic system. The realization of ion-beam simulation of neutron irradiation requires the calculation of ion irradiation fluences that provide the same degree of radiation damage in SiO_2 layer as for the fast neutrons. Recoils of two types – Si and O – are produced in the SiO_2 layer. Then, the fast neutron irradiation can be simulated by the successive irradiation with Si^+ and O^+ ions ($Si^+ + O^+$). As the cross section of recoil formation under the elastic collision with fast neutron depends weakly on the target atom mass [20], and the number of oxygen atoms in SiO_2 is two times higher than the number of silicon atoms, at the first sight, the fluence of O^+ ions should be two times higher than the fluence of Si^+ ions for the simulation. However, it should be also taken into account that incident ions loose certain energy (or even stopped) in the Au electrode before penetrating into the active SiO_2 layer.

The SRIM software [21] was used for the Monte-Carlo calculation of total number of displacements and ionization energy losses created by the irradiation with ($Si^+ + O^+$) ions of the Au (40 nm)/Zr (3 nm)/SiO_2 (40 nm)/TiN (25 nm)/Ti (25 nm) nanostructure on the oxidized silicon substrate, with the geometrical parameters corresponding to the experimentally tested nanostructure [19].

The original Monte-Carlo algorithm described in [18] was applied to calculate the depth profiles of recoils in the memristive structure irradiated by fast neutrons. Briefly, the essence of this algorithm consists of the following. The positions, energy and escape directions of primary recoils under the elastic collisions of neutrons with target atoms are determined. Then, the full displacement cascades induced by each primary recoil are simulated. After the certain number of tests required for obtaining the sufficiently low dispersion, the distributions of vacancies in the structure are obtained.

In the case of irradiation with protons of medium or high energies, the main source of damage are the ionization energy losses. Their profiles (as well as the profiles of elastic losses) were calculated by the conventional SRIM software [21].

3. Calculation results

The depth profiles of ionization energy losses for the irradiation with high-energy protons (15 MeV) and medium-energy H^+ ions (150 keV) are shown in Fig. 1a. It is possible by using these data to choose the fluence of 150 keV H^+ ions to match the total ionization energy losses for the given fluence of 15 MeV protons. It is found that the fluence of $8 \cdot 10^{13}$ cm^{-2} of H^+ ions at the energy of

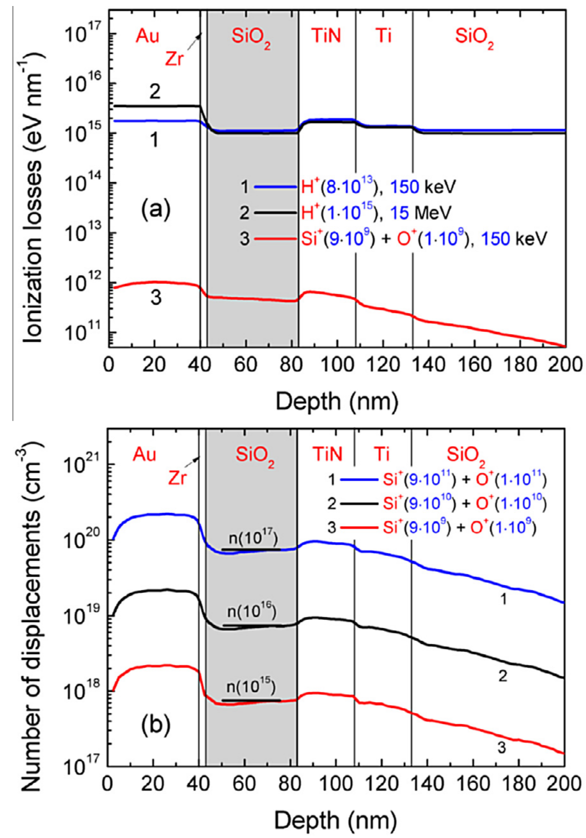

Fig. 1. The calculated depth profiles of total ionization energy losses in the memristive structure irradiated with 150 keV and 15 MeV protons with equivalent fluences (a) and displacement damage in the same memristive structure irradiated with 150 keV ($Si^+ + O^+$) ions reproducing the effect of 1 MeV neutrons (b). The profile of ionization losses of 150 keV ($Si^+ + O^+$) ions with the fluences corresponding to the fluence of fission neutrons of 10^{15} cm^{-2} is plotted in Fig. 1a (curve 3) for the comparison purposes.

150 keV produces ionizing effect in SiO_2 film that is equivalent to the effect of high-energy protons at the fluence of 10^{15} cm^{-2}. The same proportion is reasonable for any other proton fluences. The calculated displacement damage at the irradiation with both high-energy and medium-energy protons is found to be negligible and is not considered further.

The depth profiles of displacements at various fluences of ($Si^+ + O^+$) co-implantation are shown in Fig. 1b. The fluences of ions were chosen in such a way that the total concentrations of displacements in the central region of the SiO_2 layer would be equal to the concentrations of displacements generated by fast neutrons. The calculated profile of ionization losses for the fluences of ($Si^+ + O^+$) ions corresponding to the neutron fluence of 10^{15} cm^{-2} is plotted in Fig. 1a to compare them with the ionization energy losses of protons at the fluence of 10^{15} cm^{-2}. It is seen that the ionizing effect under such ($Si^+ + O^+$) ion irradiation is several orders of magnitude smaller than the effect of protons, so, in the first approximation, it can be neglected in further analysis.

The calculated values of equivalent fluences of 150 keV H^+ and ($Si^+ + O^+$) ions required for the simulation of 15 MeV protons and 1 MeV neutrons, respectively, are summarized in Table 1.

4. Experimental results and discussion

For the experimental realization of ion-beam simulation, the Au/Zr/SiO_2/TiN/Ti capacitor devices with the same geometrical parameters as used in the calculation were fabricated by