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Effect of low doses of ion irradiation on the superconducting properties of thin NbN films

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Received April 18, 2024
Revised April 18, 2024
Accepted May 8, 2024

For the first time, experiments have been carried out on the effect of low fluences ($\sim 2.7 \cdot 10^{14}\,\text{cm}^{-2}$) of mixed ion irradiation on the superconducting characteristics of an NbN film. The critical transition currents (obtained from static current-voltage characteristics) and the $R(T)$ dependences were measured for the initial film and after irradiation with a low fluence. It was found that at such a fluence, an increase in the critical current for the transition of NbN from the superconducting state to the normal state is observed. It was established that such a small fluence does not influence the critical transition temperature.

Keywords: NbN thin films, mixed ion irradiation, critical currents of superconducting films, effect of irradiation on the superconducting properties of films.

DOI: 10.61011/PSS.2024.06.58693.23HH

1. Introduction

Transistor-based devices are characterized by a number of limitations in clock frequency and power consumption despite the technological development of semiconductor electronics. Alternative computing technologies are developed to overcome these limitations. For example, superconducting computing devices with a classical „Neumann" logical architecture can be used for addressing these limitations.

The SRC „Kurchatov Institute" is developing an element base of logic devices based on switching superconductor sections (nanowires) between normal and superconducting states. Another nanowire with a resistance integrated into it using mixed ion irradiation is used for switching [1]. Niobium nitride NbN was chosen as a superconducting material because of its high critical properties and technological advantages.

Previously, we studied the behavior of NbN exposed to the mixed ion irradiation, which is necessary for the manufacture of elements of superconducting electronics. For instance, a change of the nature of conductivity from superconducting to metallic is observed in case of irradiation of NbN film with a thickness of 5 nm with a mixed proton-oxygen beam in the dose range corresponding to fluences $20-100 \cdot 10^{16}\,\text{cm}^{-2}$. The film becomes dielectric at a temperature of 4.2 K at doses of $100-145 \cdot 10^{16}\,\text{cm}^{-2}$. At the same time, the change of the nature of the conductivity is attributable to a change of the chemical composition of the film [2].

Later, we conducted studies of the effect of mixed ion irradiation with an energy of 1 keV and stabilizing annealing on the superconductivity of 5.5 nm thick NbN films at irradiation fluences of up to $10 \cdot 10^{16}\,\text{cm}^{-2}$. A decrease of the critical properties of the film with an increase of the irradiation fluence is shown in [3]. At the same time, stabilizing annealing in the 200°C, 1 h mode after irradiation with a fluence of more than $40 \cdot 10^{14}\,\text{cm}^{-2}$ increases the critical current of the film. However, annealing can also reduce the critical current in NbN with a fluence of less than $40 \cdot 10^{14}\,\text{cm}^{-2}$ [3].

This decrease of the critical current after annealing is associated with a decrease of the number of defects that are pinning centers as suggested in Ref. [3]. At the same time, there was a strong variation of the values of critical currents due to the differing thicknesses of the initial NbN film for different samples.

The purpose of the proposed work is to study the possibility of the influence of ion irradiation on the critical current of the direct transition of a superconducting thin-film NbN to a normal state because of the creation of defects at low fluences.

2. Experiment

The object of the study is a 5 nm thick NbN film, sputtered onto a sapphire substrate at a temperature of 100°C. The square area of $20 \times 20\,\mu\text{m}^2$ was exposed to irradiation. Contacts for measurements are sputtered on opposite sides of the square. The contact material is platinum with a nickel sublayer.

The following experimental setup was used to prevent the impact of the initial current spread on the result of the experiment. A sample of NbN thin film is placed in a closed-cycle cryostat CFSG-101 manufactured by Cryo Trade Engineering. The current-voltage curve was recorded after cooling in the temperature range of 4−7 K in increments of 0.2 K and 7.1−9.5 K in increments of 0.1 K.

The use of a closed-loop cryostat allows measurements of electrical characteristics without contamination of the sample surface. This makes it possible to irradiate samples after measurements and to perform subsequent measurements in the dose accumulation process.

After measuring the current-voltage curve in the initial state, the samples are exposed to mixed ion irradiation on Copra Cube installation manufactured by CCR Technology. A beam consisting of 99% protons and 1% oxygen was used for irradiation. The beam energy was 0.6 keV, the ion current density was $j = 0.849\,\mathrm{A/m^2}$. The fluence rate was $5.3 \cdot 10^{14}\,\mathrm{cm^{-2}}$, the maximum irradiation fluence was $13.25 \cdot 10^{14}\,\mathrm{cm^{-2}}$.

3. Results

The current-voltage curve was recorded in the current stabilization mode by increasing it through the sample to a normal state („forward“ transition), and then by reducing the current to „reverse“ transition to a superconducting state. The critical current of the forward and reverse transitions differ from each other in this case. This difference is caused by the fact that the heat is released according to the Joule-Lenz law in the normal state when a current flows through a superconductor. This heat release results in a local heating of the superconducting phase to a temperature above the critical temperature at a given value of the flowing current. The heat dissipation decreases as the current through the superconductor decreases and the critical temperature corresponding to the flowing current increases. Figure 1 shows an example of the current-voltage curve of the initial and irradiated sample at a temperature of 4.2 K.

It should be noted that the curves shown in Figure 1, plotted for a sample in the form of a microbridge with a width of 20 μm, are characterized by a sharp transition from a superconducting state with low resistance to a normal state with high resistance. This is attributable to the large width of the micro-bridge, at which both boundaries of the sample, on which vortices usually originate at various inhomogeneities, are far from the main volume of the superconducting film. If a nanowire is made from such an NbN film then the features characteristic of this type of superconductors of the second kind begin to appear in case of reduction of the cross section for the flow of a superconducting current. Figure 2 shows a SEM image of a nanowire with a width of 200 nm and a length of 600 nm.

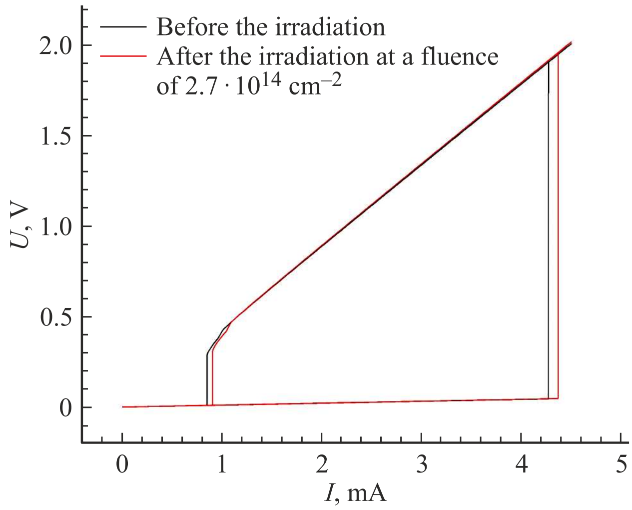

Figure 1. Current-voltage curve of the initial and irradiated sample at a temperature of 4.2 K. The width of the sample is 20 μm.

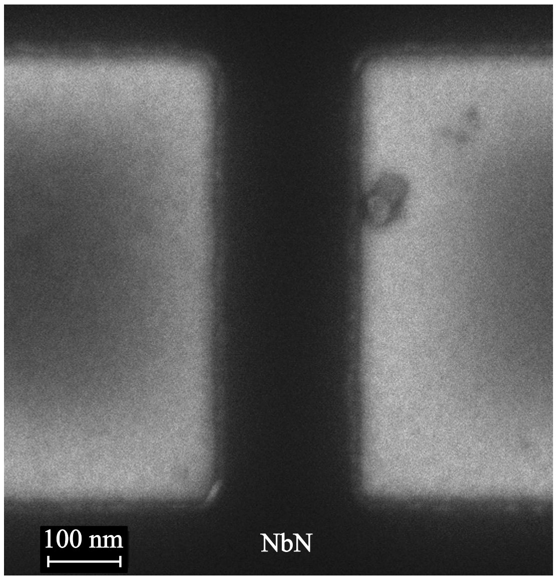

Figure 2. SEM image of a NbN nanowire (dark region) with a thickness of 5 nm, a width of 200 nm and a length of 600 nm on a substrate of single-crystal sapphire.

Figure 3 shows its current-voltage curve (*a* — on a general scale, *b* — with an increase of the low voltage region).

Figure 3, *a* shows that the nanowire is in a superconducting state (S) at low current values (from 0 to $\sim 8\,\mu$A), and it enters into normal state (N) at a current of $\sim 12\,\mu$A. However, a gradual increase of voltage is observed at intermediate current values before a sharp transition to the normal state from 8 to 12 μA (Figure 3, *b*). The increase of voltage before the transition to a normal state is explained by the formation and movement of Abrikosov vortices in the film in this current range. This condition is called resistive

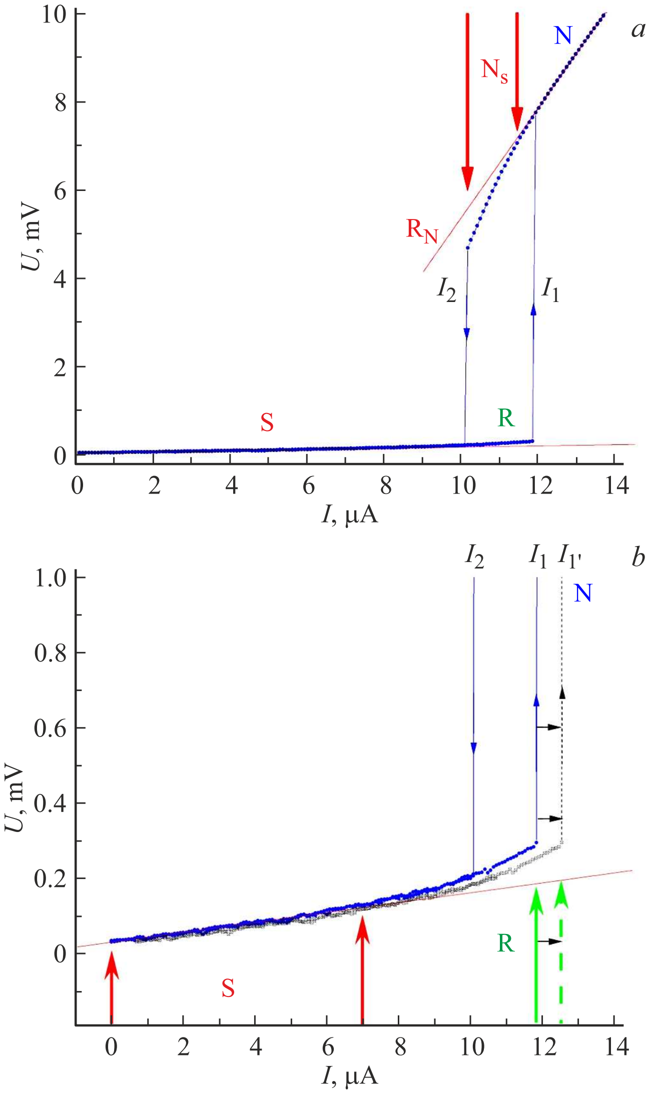

Figure 3. Current-voltage curve ($200 \times 600\,\mathrm{nm}^2$) at a temperature of 4.2 K (a) and an increased current-voltage curve area at low voltage values (b).

(R). The nanowire material is in a superconducting state in the resistive state, and the movement of vortices (the central region of which contains a normal phase) requires work, which causes the occurrence of some (small) voltage at the ends of the nanowire. The process of generating single vortices in the resistive state was initiated in Ref. [4] by creating a sharp concentrator in the narrow part of the nanowire by lithographic methods.

Figure 3, b shows that it is possible to influence the value of the current of direct transition to the normal state in the resistive region (R) immediately preceding the transition of the nanowire to the normal state with an increase of current. For instance, if additional points of attachment of Abrikosov vortices are created in the nanowire (including by introducing defects), this will complicate the movement of vortices and the resistive region may expand into the region of high currents. Such a model case is shown in Figure 3, b by a dotted line. The resistive region increased because of additional defects, and the critical current value I_1 increased to I_1'.

As for the reverse current I_2, the transition of a nanowire from a normal state to a superconducting state is accompanied by the following processes [5]. The voltage linearly depends on the current when the current decreases through the nanowire, which is in a normal state, as long as the current is greater (or slightly less) than the direct junction current I1 (see curve R_N in Figure 3, a). The energy release of the normal nanowire material exceeds the cooling system's ability to remove it from the system in this current range. A normal nanowire maintains its temperature above T_c due to the Joule heat generated in it. However, the thermal power released in the normal region decreases as the current decreases, which results in a decrease of the length of the thermal domain (compression of the thermal domain). This is manifested in the deviation of the voltage from the linear dependence towards lower voltages (region N_s in Figure 3, a). The heat output in the normal domain can no longer compete with the existing heat dissipation to maintain its temperature above the critical one when the current value I_2 is reached. The thermal domain reverses to a superconducting state. Therefore, the reverse transition current is determined by the minimum thermal power capable of competing with the existing heat dissipation to maintain the domain temperature above the critical transition temperature [5]. It is known that the value of the reverse transition current can be realized at the level of ten percent of the value of the forward transition current when special conditions are created to minimize heat removal, for example, when forming „weighted“ nanowires without contact with the substrate in a dry cryostat. The normal state of the nanowire is stabilized by resistive heating up to the minimum current values [6].

Diagrams of changes in critical currents depending on the irradiation fluence (Figure 4) at different temperatures are constructed on the basis of the removed current-voltage curve.

Figure 4, a shows that an increase of the direct critical current is observed in the temperature range of 4–7 K after the first irradiation with fluence $2.65 \cdot 10^{14}\,\mathrm{cm}^{-2}$. The forward critical current does not significantly change in the temperature range 7–9 K. The maximum increase of the direct critical current was recorded at a temperature of 6.2 K and reached 9%. The direct critical current gradually decreases relative to the level of the first irradiation in case of subsequent irradiations.

Figure 4, b shows that an increase of the reverse transition current from the normal state to the superconducting state is also observed after the first irradiation. Since the reverse transition, as mentioned above, is associated with the thermal equilibrium of the minimum thermal domain, let us consider the factors affecting heat generation and heat dissipation in this state. The generally accepted approach to considering the heat balance equation includes taking into

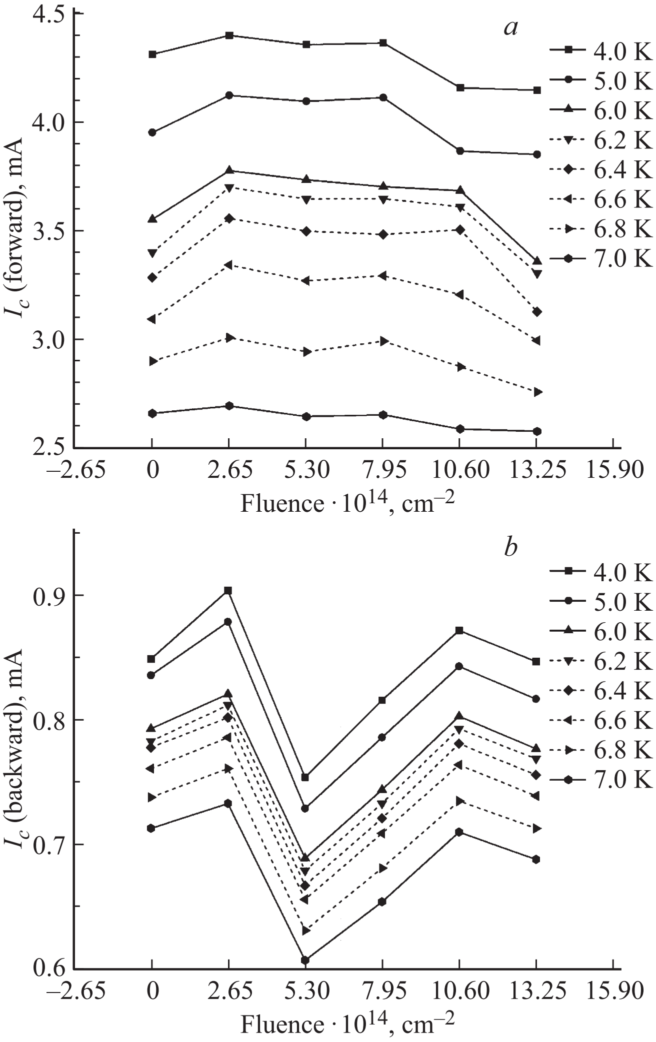

Figure 4. Change in critical currents depending on the irradiation fluence for forward (*a*) and reverse (*b*) transitions.

account the processes of heat generation attributable to the flow of current through the normal domain, as well as heat dissipation into the substrate and heat dissipation along the nanowire [7]:

$$J^2\rho + \kappa \frac{\partial^2 T}{\partial x^2} - \frac{\alpha}{d}(T - T_{sub}) = \frac{\partial cT}{\partial t}, \qquad (1)$$

where J — current density through the nanowire, ρ — electrical resistance of the normal section of the nanowire, κ — thermal conductivity of NbN, thermal conductivity of the film interface with the substrate, d — film thickness, T_{sub} — substrate temperature, c — specific heat capacity of the film. Stationary conditions are realized in case of slow recording of the current-voltage curve in the current stabilization mode, therefore equation (1) can be equated to zero. Additionally, it can be considered based on the data of Ref. [6], that the heat removal through a single-crystal sapphire substrate is the most significant heat removal channel from the resistive region, in connection with which we pay attention only to the first term responsible for heat generation and the third term responsible for heat removal in the substrate.

Let us consider which of these two parameters of the equation (1) can change under the impact of irradiation with small fluence, when only point defects are formed and there is still no significant change in the atomic composition because of the replacement of nitrogen atoms with oxygen atoms, characteristic of large fluences.

Figure 1 shows that low-fluence irradiation increases resistance in the normal state by several percent. A slight increase of the resistance of the normal phase is attributable to the formation of defects in the NbN. If an increase of resistance in the normal state (ρ) would be the only consequence of exposure to a small irradiation fluence, this would result in a decrease of the magnitude of the current of transition to the superconducting state I_2. This is attributable to the fact that the value of the minimum power generated by the normal domain is achieved at a lower current, taking into account the increase of resistance in the normal state. This is what happens at high irradiation fluences, when the resistance in the normal state begins to increase due to radiation-induced changes in the atomic composition of the film and the magnitude of the reverse transition current significantly decreases [3].

Since the NbN film has a thickness of 5.5 nm and the energy of the mixed ion beam is 0.6 keV, the defect generation zone, according to the simulation results, extends deep into the target, capturing both the substrate and the interface between the NbN film and the sapphire single crystal. According to our assumption, the radiation-induced processes at the interface of the film with the substrate, for instance, ion mixing of the interface, increases the thermal conductivity α. This improves the heat dissipation into the substrate, and therefore the value of the minimum power generated by the normal domain during its transition to the superconducting state is achieved at a higher current I_2. It is clear that such a mechanism can be implemented only in a limited range of fluences, since the subsequent strong increase of resistance in the normal state causes an inevitable drop of the reverse transition current [3].

The most important thing for us is the demonstrated increase of the critical current of transition from the superconducting state to the normal state in the range of minimum temperatures available for measurement from 4 to 7 K. This effect does not significantly manifest itself at higher temperatures. Presumably, this is attributable to the approximation to T_c and the expanded temperature transition characteristic of type II superconductors.

Also, the dependences of the electrical resistance of 20 μm wide samples of micro bridges on temperature are plotted on the basis of the recorded current-voltage curve (Figure 5). The value of the measuring current corresponded to 100 μA.

A comparison of the dependences $R(T)$ plotted for different fluence values in Figure 5 shows that the critical temperature and width of the superconducting junction

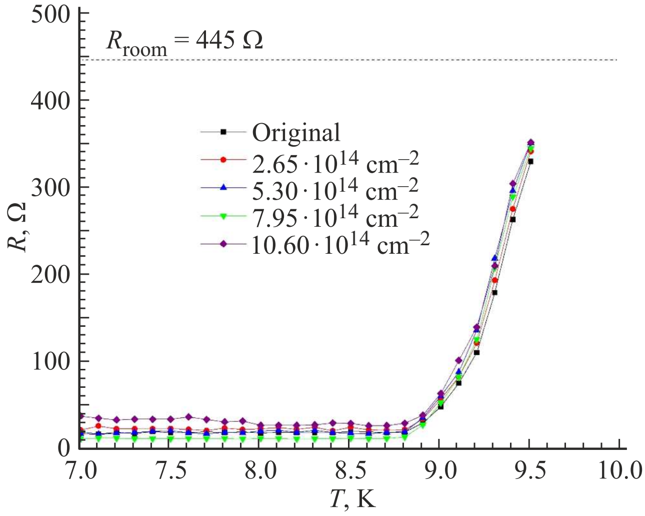

Figure 5. The dependences of the electrical resistance of 20 μm wide samples on the temperature at various fluences of ion irradiation, based on the current-voltage curve.

do not significantly change in the range from zero to $13.25 \cdot 10^{14}\,\mathrm{cm}^{-2}$.

Since NbN is a superconductor with s-wave pairing type, Anderson's is applicable for it [8]. We compare the calculated values of the average distance between ions with the average size of the defect formed under the action of irradiation from one ion for the studied case of introduction of defects into a thin film under the action of mixed irradiation. The chosen value of fluence for the calculation is $F = 2.65 \cdot 10^{14}\,\mathrm{cm}^{-2}$, since it maximizes the increase of critical current.

The average distance between ions at fluence $F = 2.65 \cdot 10^{14}\,\mathrm{cm}^{-2}$ can be estimated by the formula

$$\Delta L \cong \frac{1}{\sqrt{F}} = 0.6\,\mathrm{nm}. \qquad (2)$$

The average size of the defect formed by a single ion can be estimated using the SRIM solid ion propagation simulation program [9]. The program takes into account all the processes of interaction of charged particles with atoms of a solid body, and also records not only the acts of primary dislocation of lattice atoms as a result of interaction with beam ions, but also the complete history of all events, including the formation of secondary defects, as a result of scattering of primary dislodged atoms and recoil atoms.

Only discharged cascades of atomic collisions are formed for the used ions (primarily protons) at energies 0.6 keV consisting of point defects spaced apart [10]. Therefore, the average distance between several point defects within one cascade of atomic damage initiated by one ion can be taken as the size of defect L from one ion. The estimation of L for the irradiation parameters used, the thickness of the NbN film, its density, and the presence of a massive sapphire substrate was performed using the SRIM code [9] in the mode of tracking and recording to disk of the full cascade of atomic defects for the number of ion injections (Monte Carlo method) equal to ~ 1 million of particles. Vacancies were identified in the NbN subsystem of nitrogen (N) atoms after the simulation and the average distance between these vacancies was calculated within the framework of a single cascade of atomic collisions. The obtained average distance between vacancies for a large number of single cascades was $L \sim 6$ nm, which is about an order of magnitude higher than the average distance between ΔL ions for the studied fluence

$$\Delta L < L. \qquad (3)$$

The ratio (3) together with the immutability of T_c at a given irradiation fluence (Figure 5) provides the basis for the manifestation of Anderson's theorem.

Similar estimates were performed for YBCO films irradiated with xenon ions [11–13]. A noticeable decrease of the critical temperature T_c was observed in cited works when the condition was met (3), since Anderson's theorem does not hold for the superconducting compound YBCO.

4. Conclusion

It was shown that irradiation of the NbN film with a mixed ion beam of energy of 0.6 keV and fluence of $2.65 \cdot 10^{14}\,\mathrm{cm}^{-2}$ increased the forward and reverse critical currents of the superconducting junction without increasing the critical temperature. Thus, for the first time, results were obtained in which an increase of the critical currents of thin-film NbN is observed at low fluences ($\sim 2.7 \cdot 10^{14}\,\mathrm{cm}^{-2}$) of mixed ion irradiation.

The increase of the direct critical transition current is explained by the pinning of Abrikosov vortices on the created defects in NbN, while this fluence of mixed irradiation is too small for a significant change of the atomic composition as a result of the process of selective substitution of nitrogen atoms by oxygen atoms. The absence of the effect of such irradiation on the temperature of the superconducting transition, despite the formation of defects under the action of irradiation, which corresponds to Anderson's theorem [8], valid for superconductors with s-wave type of pairing, to which NbN belongs.

The results obtained in this work regarding the increase of critical currents at low fluences of mixed ion irradiation set the radiation-induced method of directional transformation of the properties of thin-film materials that we used among other radiation methods for changing the properties of superconducting materials aimed at creating defects, for instance, irradiation with neutrons or heavy ions with high energy.

Acknowledgments

The authors express their deep gratitude to D.A.Savinov for his invaluable assistance and advice in the process of

working on the manuscript, which resulted in a qualitatively new understanding of the experimental results obtained and significantly improved the presentation of the material.

Funding

This study was carried out under the state assignment of SRC „Kurchatov Institute“.

Conflict of interest

The authors declare that they have no conflict of interest.

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Translated by A.Akhtyamov