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Ion-irradiation effect on electron transport in YBCO thin films

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The disorder effect on superconducting properties of thin-film YBCO nanostructures in external magnetic fields is experimentally studied. The disorder was produced by irradiation with xenon ions. The research included transport measurements of narrow bridges based on HTSC YBCO films (thickness 50 nm) in strong magnetic fields (up to 12 T). Thus, for samples with different degrees of disorder, critical dependences have been studied, i.e. the $H_{c2}(T)$ phase-transition line, the $H_{irr}(T)$ irreversibility line, etc. The dependences of the mean-free path and critical temperature on the concentration of defects created by ion irradiation have been experimentally studied. The experimental data are described using formulas obtained within the framework of well-known models, such as the Ginzburg-Landau theory, the Drude theory, and the Gorkov equations.

Keywords: B HTSP, thin films, ion irradiation, defects, resistive measurements, upper critical field, line of irreversibility, vortices.

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1. Introduction

In the last decade, there has been a bright outbreak in the research of superconductors with an unusual type of pairing (in particular, high-temperature superconductors [HTSC]) with varying degree of disorder (see, for example, papers [1–6]). Interest in them is due to a number of reasons. In particular, for high degrees of disorder, when the sample is near the „superconductor–normal metal" (or „superconductor–insulator") transition, it is possible to change the type of superconducting pairing [2,3,6], as well as the occurrence of an unusual temperature dependence of the upper critical field $H_{c2}(T)$ — it turns out to be monotone increasing, which leads to the possibility of stimulating superconductivity by applying an external magnetic field H [4], while usually the external magnetic field always suppresses the critical temperature T_c [7]. A very compelling experimental result found for the non-superconducting state of YBCO films containing a lattice of nanoholes is the possibility of electric current flow, the elementary carrier of which is the Cooper pairs, but not individual electrons, as is usually in the case of the normal state of conventional superconductors [1]. For low degrees of disorder in HTSCs, it is also possible to observe rather unusual effects, which may be of interest from both theoretical and applied points of view. For example, in such systems anomalous features of phase diagrams on the external magnetic field H — temperature T plane are possible [5,6]. Namely, an unusual decrease in the slope of the upper critical field near $T_{c0} = T_c(H = 0)$ was observed, while usually the defects concentration increasing leads to the local slope increasing of the phase-transition line $H_{c2}(T)$. It was also found that the temperature dependence of the upper critical field has a positive curvature near T_{c0}. In some cases, disorder in HTSC samples is created by radiation exposure or irradiation (see, for example, works [5,6,8–18]). It is notable that at moderate doses of irradiation the radiation exposure can be used to increase the critical parameters of HTSC structures [17]. In particular, irradiation can lead to the critical temperature increasing in HTSCs (see, for example, [7,14]), as well as providing the possibility of creating nanostructures with an increased critical current (see, for example, [18]). By selecting the parameters of ions as sources of radiation damage, one can create a lattice of narrow quasi-cylindrical defect regions in films, leading to pinning enhancement and, consequently, to higher values of the critical current density j_c compared to unirradiated samples. Thus, the controlled introduction of defects in HTSC samples (including due to radiation exposure) can be used to create superconducting structures with high critical parameters and new unusual properties. In the present paper, we carried the study necessary to achieve this fundamental goal. For the samples based on YBCO with varying degrees of disorder, the critical dependences, i.e., the phase-transition line $H_{c2}(T)$, the irreversibility line $H_{irr}(T)$, etc. were studied, as well as the features of the vortex states of irradiated samples, which are extremely important, since it is vortices that determine the thermodynamic and transport characteristics of superconductors.

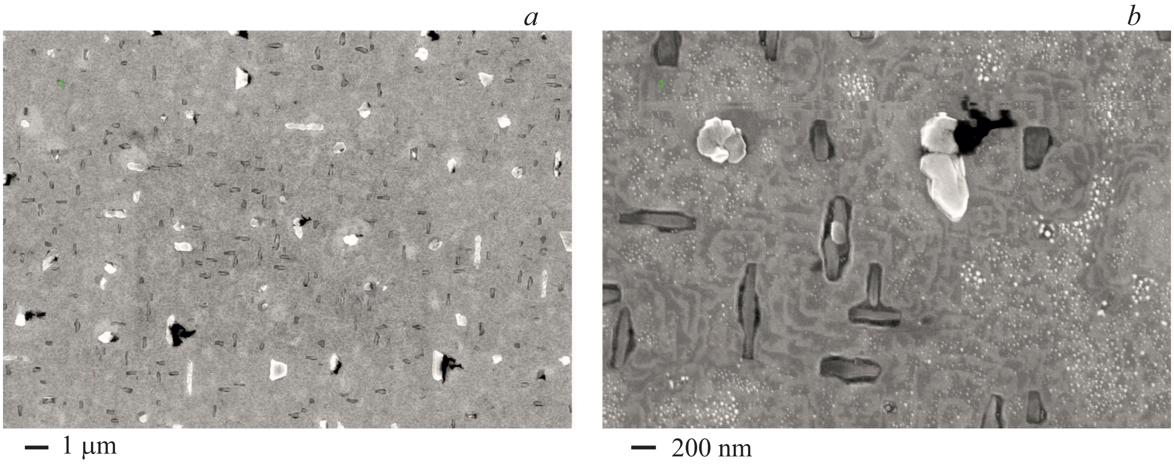

Figure 1. Scanning electron microscope (SEM) image of the YBCO film under study.

It is well known that the external magnetic field H can penetrate II type superconductors in the form of vortices [19]. In the idealized case of a bulk superconductor without defects, the lattice of vortices is strictly ordered and has hexagonal symmetry. The lattice period depends on H. When the field H exceeds the critical value H_{c2}, the vortex density becomes so large that the lattice period is of the order of the vortex size. The vortices come into contact with their normal cores, and a second-order phase transition to the normal state occurs. The critical value H_{c2}, called the second critical field, depends on the temperature. Near the critical temperature T_{c0}, this dependence is linear

$$H_{c2} = \Phi_0(1 - T/T_{c0})/2\pi\xi_0^2, \quad (1)$$

where Φ_0 is the magnetic flux quantum, ξ_0 is the superconducting coherence length at $T = 0$. The dependence $H_{c2}(T)$ determines the line of the phase transition to the normal state of a bulk superconductor without defects. Typical samples have finite dimensions; therefore, the normal state in the superconductor is achieved at a higher value of the external field associated with the formation of nuclei near the boundaries. Moreover, the samples are always heterogeneous. Thus, when the field H increases, defects can lead to competition between different types of superconductivity nucleation. As a result, phase transition line turns out to be nonlinear near T_{c0} (see, for example, [5]). Since in the experiment the superconducting transition has a finite width, which increases with H increasing, the question arises of choosing the $R = \text{const}$ level for which one or another phase transition line should be determined. Obviously, this level may depend on the degree of disorder. In the present paper we carried out the experimental study of this issue as applied to thin YBCO HTSC films irradiated with different doses of xenon ions. Thus, in the work we found: (a) the mean-free path and the critical temperature T_{c0} for different radiation doses, (b) temperature dependences of critical magnetic fields for different radiation doses, and (c) the criterion $R = \text{const}$ for determining the boundary of the vortex state of irradiated samples. The data found in the measurements are described using formulas derived from well-known models such as the Ginzburg–Landau theory, the Drude theory, and the Gorkov equations.

2. Fabrication of samples and experimental details

Narrow bridges formed on the basis of the thin epitaxial YBCO film (50 nm thick) grown by magnetron sputtering on a lanthanum aluminate substrate $LaAlO_3$ were studied in this work. Fig. 1 shows image of the YBCO film surface obtained with a scanning electron microscope (SEM). Fig. 2 shows optical image of the bridge we studied. The bridges were obtained using the method of ion irradiation through a photoresist mask, which was then removed in acetone. This method implementation was similar to how it was done earlier in paper [20], which also presents the results of structural studies of similar YBCO films. The original bridges had the following geometric dimensions: width 50 μm, length 250 μm. The contact pads were fabricated by thermal spraying of silver through a metal mask. The silver layer was 100 nm thick. Critical temperature $T_{c0} = 89$ K,

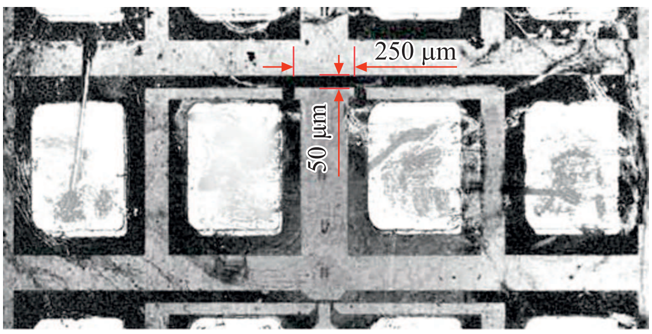

Figure 2. An optical microscope image of the bridge under study.

critical current density $j_c = 4 \cdot 10^6$ A/cm^2 at $T = 77$ K. The specific resistance of each of the bridges at a temperature of $T = 100$ K was $100\,\mu\Omega$cm, which corresponds to the optimum oxygen doping of the film, i.e., $x = 0.1$ in the $YBa_2Cu_3O_{7-x}$ film. Note that the film is imperfect and contains many structural defects with characteristic sizes ranging from a few nm to hundreds of nm (see Fig. 1). The largest of them are usually called precipitates (see light and dark inclusions). However, the average distance between them substantially exceeds their characteristic size, and the superconducting transition in a zero magnetic field turns out to be rather narrow — about 1 K. This means that in terms of theoretical concepts, the sample can be approximately considered as homogeneous with a given critical temperature T_c, but with some spatial modulation of the coherence length. Such model is the basis of the theoretical description of the temperature dependences of critical magnetic fields, which will be discussed below.

During the study the bridges were irradiated with Xe^{2+} ions with the energy of 150 keV and different doses. The maximum accumulated dose n_D was $7.3 \cdot 10^{12}$ cm^{-2}. Irradiation was carried out at room temperature.

The sample resistance was measured by the standard four-probe method. The transport current was $10\,\mu$A. Resistive measurements were carried out with a magnetic field sweep up to 12 T in the temperature range from helium values to 100 K. To create the magnetic field a closed-cycle cryogenic system of two cryostats was used, one of which contained a superconducting solenoid with a hole 52 mm in diameter (Oxford Cryofree SC magnet), and the second cryostat (Oxford Optistat PT) with controlled temperature (from 1.6 K and above) was inserted into this hole, in it the test sample was located. The temperature T was determined by a special calibrated thermometer with a resolution of 50 mK. The magnetic field H was determined with a resolution 12 G.

3. Experimental results

In this paper, we experimentally study the dependences of the normal resistance of sample $R_N = R(100\,\mathrm{K})$ and the critical temperature T_{c0} on the concentration of defects created by ion irradiation. Using the Drude theory, as well as the Gorkov equations, the measured data made it possible to study the dependence of the mean-free path ℓ on the ion dose n_D. Thus, in a wide range of n_D values up to $5 \cdot 10^{12}$ cm^{-2} the dependences $R_N(n_D)$ and $T_{c0}(n_D)$ turn out to be quasilinear (see Fig. 3). This allows us to approximate the experimental data $T_{c0}(n_D)$ by a simplified expression for the critical temperature

$$T_{c0} \cong T_{co}^{(virg)} - \pi\hbar/8k_B\tau, \tag{2}$$

where $T_{c0}^{(virg)}$ is the critical temperature of the sample without irradiation, τ is the mean-free time of electrons. Estimating the Fermi velocity $v_F \cong 5 \cdot 10^5$ m/s (see, for example, [5]), we obtain the mean-free path $\ell \cong 50$ nm for

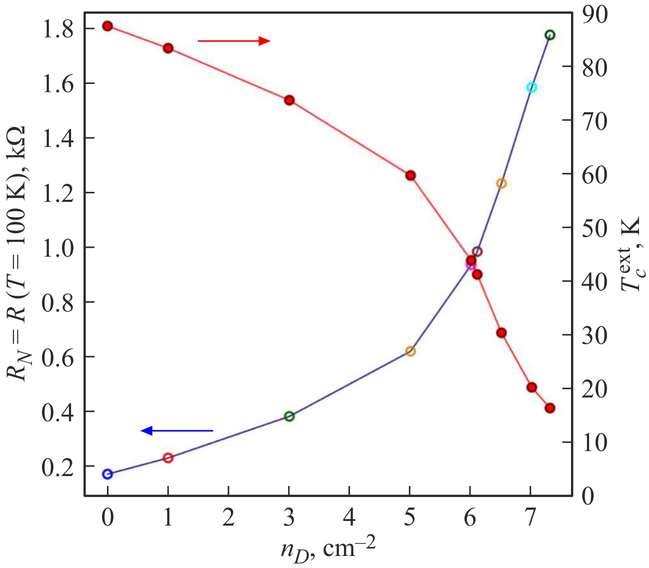

Figure 3. Normal resistance of sample $R_N = R(100\,\mathrm{K})$ and critical temperature T_{c0} vs. irradiation dose n_D.

$n_D = 5 \cdot 10^{12}$ cm^{-2}. Obviously, this is an overestimation by ℓ, since $T_{c0}^{(virg)} = T_{c0}(\tau \to \infty)$, which exceeds the value for the critical temperature of the original sample ($T_{c0} = 89$ K, see Section 2). Alternatively, the Drude model gives the following expression for τ:

$$\tau \cong \frac{m}{e^2 n \rho_{100}}, \tag{3}$$

where ρ_{100} is resistivity corresponding to $R_N = R(100\,\mathrm{K})$, $n \cong 2 \cdot 10^{21}$ cm^{-3} is carrier concentration (see data for $YBa_2Cu_3O_{7-x}$ in [21]). Thus, for $n_D = 5 \cdot 10^{12}$ cm^{-2} we find: $\ell \cong 5$ nm. However, this assessment is underestimated due to the fact that the actual residual resistance is lower than ρ_{100}, which was used in formula (3). Thus, we find limits for the mean-free path for each of the considered doses. In particular, for the maximum value $n_D = 7.3 \cdot 10^{12}$ cm^{-2}, we found that ℓ turns out to be about several nanometers, and, therefore, it is fair to consider the sample within the clean limit when ℓ exceeds ξ_0, which is 1.5 nm in this material. This means that after each act of ion irradiation the average coherence length in the sample remained unchanged

$$\xi_0(n_D) = inv. \tag{4}$$

The nonlinearity of $R_N(n_D)$ and $T_{c0}(n_D)$ identified at $n_D > 5 \cdot 10^{12}$ cm^{-2} can be related to the overlap of defective clusters that appear during irradiation (see Fig. 3), since at such doses the lateral sizes of individual clusters L_c turn out to be comparable with the characteristic distance between them ΔL_c. Indeed, $L_c \sim 10$ nm (see calculations in SRIM [22] for Xe^{2+} with the chosen parameters specified in Section 2 of this paper), and characteristic distance $\Delta L_c \propto n_D^{-1/2}$. In particular, for $n_D = 10^{12}$ cm^{-2}:

$$\Delta L_c \cong 10\,\mathrm{nm}.$$

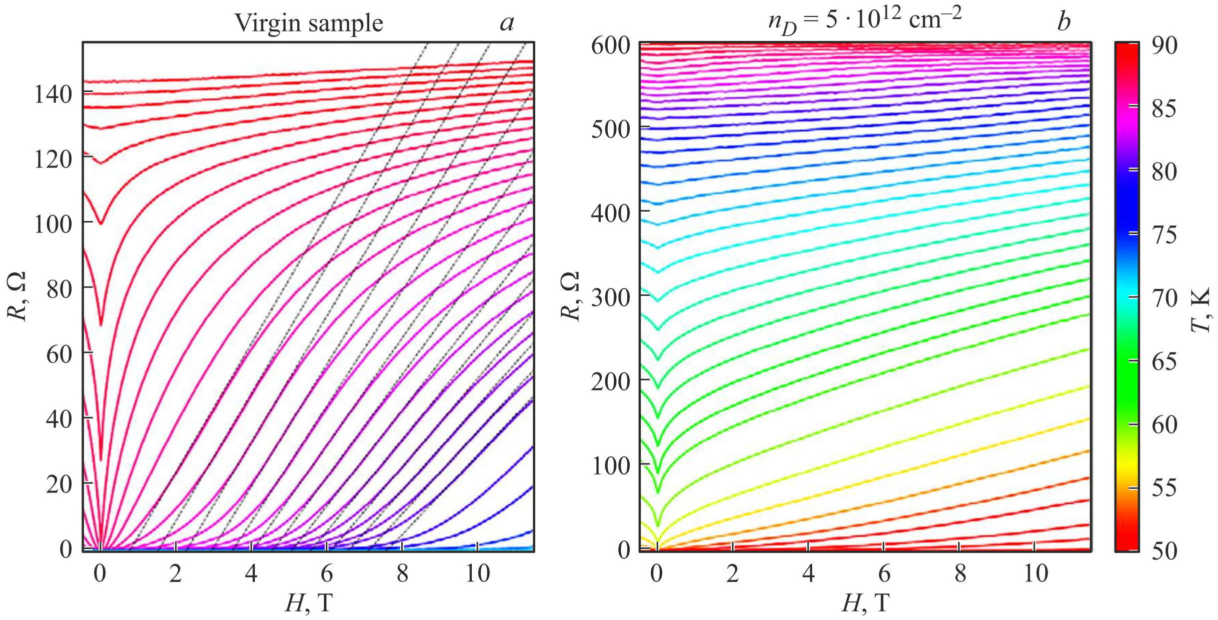

Figure 4. Dependences $R(H)$ for different temperatures (the scale is shown on the right) for the original sample (a) ($n_D = 0$) and for the irradiated sample (b) ($n_D = 5\cdot10^{12}\,\mathrm{cm}^{-2}$).

Next, we present the results of transport measurements of the samples under study in external magnetic fields. Resistive measurements were carried out with a magnetic field sweep up to 12 T in the temperature range from helium values to 100 K. The magnetic field orientation was chosen in the direction of the axis c of the film. Thus, the dependences $R(H)$ were taken for the given temperature T. Choosing the value $R = \mathrm{const}$, we obtain a series of points on the external magnetic field — temperature plane, which represents the dependence $H(T)$. According to the predictions of paper [23], for high levels $R \to R_N$ we expected the appearance of nonlinear distortions in the $H(T)$ dependences near T_{c0} with a gradual increasing of the ion irradiation dose n_D. The description of these lines for different levels was made in the framework of the modified Ginzburg–Landau theory, similarly to how it was described in [5]. It turned out that the distortions manifest themselves most clearly at $R \to 0$, when the $H(T)$ line corresponds to the $H_{irr}(T)$ irreversibility line associated with the entry/exit of vortices. For high levels, the expected effect turned out to be weakly expressed. However, for such resistances we found some other unusual features on $H(T)$ lines. It turned

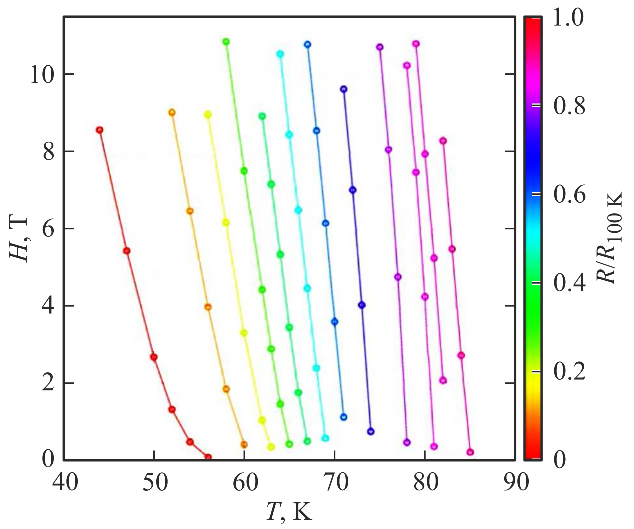

Figure 5. Dependences $H(T)$ for $n_D = 5\cdot10^{12}\,\mathrm{cm}^{-2}$ and different levels of constant resistance $R = \mathrm{const}$ in fractions of $R_N = R(100\,\mathrm{K})$.

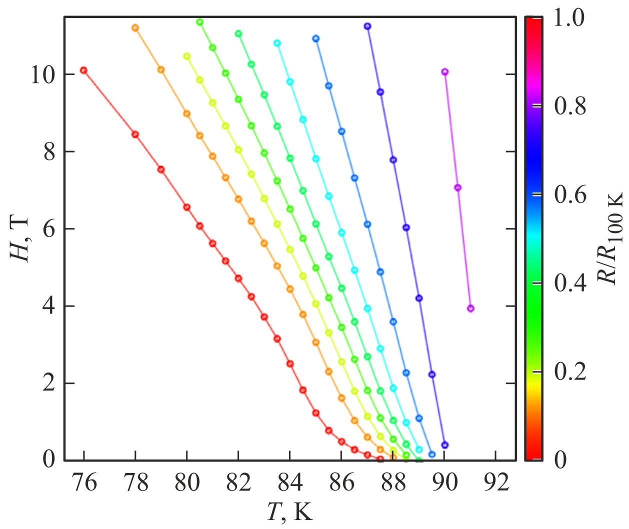

Figure 6. Dependences $H(T)$ for a non-irradiated sample ($n_D = 0$) and different levels of constant resistance $R = \mathrm{const}$ in fractions of $R_N = R(100\,\mathrm{K})$.

out that for the irradiated sample, the $R(H)$ dependences become quasilinear and, moreover, quasiparallel for different temperatures (see Fig. 4). This leads to the fact that for different R the lines $H(T)$ almost do not change their slope, as can be seen in Fig. 5, which differs significantly from the case when the sample was not irradiated (see Fig. 6).

4. Discussion

For each temperature T the $R(H)$ transition can be divided into two regions — the region of low resistances, when vortices enter the sample, they participate in dissipation during the flow of the transport current, and the region of high resistances, when the sample is predominantly in the normal state, and only local regions can participate in the non-dissipative current flow. The boundary $R = \text{const}$ between these regions determines the field H_{c2} at a given temperature T. This limit may change when the sample is irradiated. Therefore, a selection criterion R is needed, which allows one to determine the $H_{c2}(T)$ dependence for a superconductor with different irradiation doses. For the virgin sample (without irradiation), the criterion usually corresponds to the choice of R as the maximum resistance value in the region where $R \sim H$ (the flux-flow regime). In our experiments, this corresponds to the level $R = 0.4R_N$. Such a criterion should be chosen to determine $H_{c2}(T)$ for $n_D = 0$. Indeed, with such definition we obtain $\xi_0 = 1.5\,\text{nm}$, which agrees with known literature data (see, for example, [24]). Let us now define the criterion for each of the chosen doses n_D. Fig. 7 shows the dependence of the slope dH/dT on the dimensionless resistive level for different n_D. Note that for $R < 0.4R_N$ the dH/dT dependence from the level becomes quasiconstant for any n_D. It follows from formula (1) that ξ_0 can be experimentally determined from the dependence $H_{c2}(T)$ as a value inversely proportional to the root of the product of dH/dT and T_{c0}. The critical temperature T_{c0} decreases as the dose increases. This means that in order to fulfill formula (4) with a gradual increasing of n_D, the slope dH/dT must increase. Therefore, for the correct extraction of H_{c2} from the experimental curves $R(H)$, it is necessary to increase R when n_D increases. So, for example, for maximum accumulated dose $n_D = 7.3 \cdot 10^{12}\,\text{cm}^{-2}$, this level should be chosen near the onset (the beginning of the superconducting transition), i.e., at $R \to R_N$. Thus, despite the fact that the sample approaches the dirty limit during irradiation, the vortex region (low resistance region) expands up to $R \to R_N$.

It is important to note that for $R > 0.4R_N$, the dependences $dH/dT(R)$ turn out to be monotone increasing, reaching the maximum at $R \to R_N$ (see Fig. 7). However, when the dose n_D gradually increases, the change in $dH/dT(R)$ becomes less and less significant. Thus, after reaching the dose $n_D = 7.3 \cdot 10^{12}\,\text{cm}^{-2}$, it can be seen that the dependence $dH/dT(R)$ becomes quasiconstant. According to our estimates, it is fair to describe the sample within the framework of the pure limit, which means that $\xi_0(n_D) = inv$. Therefore, the question arises of how the $dH/dT(R)$ dependence will look for large doses, when condition (4) is still valid, and the critical temperature T_{c0} should probably decrease. It can be assumed that, on the contrary, T_{c0} may stop changing as n_D increases, while the dependence $dH/dT(R)$ remains quasiconstant. This could confirm the transition from the d-wave phase of superconducting pairing to the s-wave phase (see, for example, [25]), which was partially studied by us experimentally in [6]. However, this assumption requires direct transport measurements and will be fulfilled by us in the future.

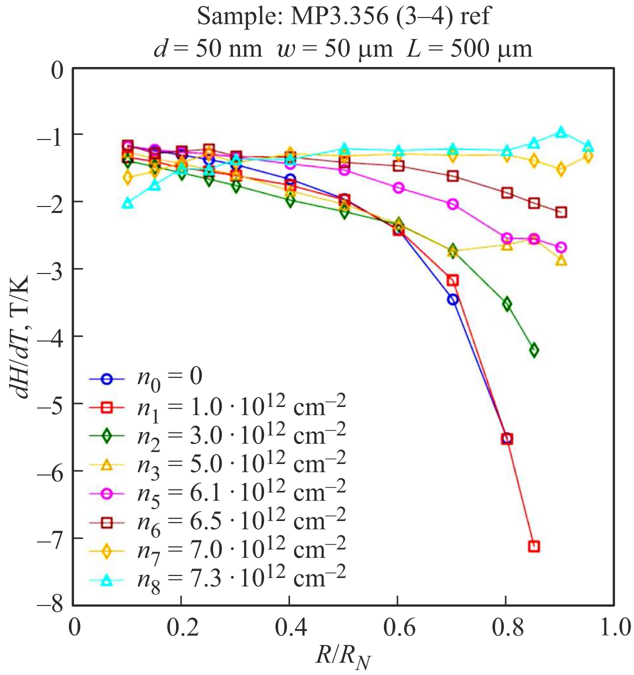

Figure 7. Slope dH/dT vs. normalized resistance for different irradiation doses n_D.

In order to detect the nonlinear features of $H(T)$ near T_{c0}, which were predicted in paper [21], we assume that one should choose more moderate radiation doses $n_D \sim 10^{10}\,\text{cm}^{-2}$, when the characteristic distance between defective clusters ΔL_c is much larger than their lateral size L_c. These studies will be carried out on new similar samples irradiated with appropriate doses of xenon ions.

5. Conclusion

In the present paper, we carried out an experimental study of YBCO HTSC thin films irradiated with different doses of xenon ions and studied the criterion that makes it possible to determine the dependence $H_{c2}(T)$ from the experimental data $R(H, T)$. Besides, the mean-free path and critical temperature T_{c0}, as well as the temperature dependences of critical magnetic fields for different irradiation doses, are found in the paper. The phase-transition lines may be of interest for studying the effect of disorder on the type of superconducting pairing in HTSC materials. Besides, the studied effect of ion irradiation on YBCO films is also important in superconducting electronics — in the creation of defect-induced weak bonds and for studying their physical nature.

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Conflict of interest

The authors declare that they have no conflict of interest.

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