

# Analysis of experiments on the ion irradiation of $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ films: $d$ pairing or anisotropic $s$ pairing?

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The influence of ion irradiation on the critical temperature  $T_c$  and the resistivity  $\rho_{ab}(T)$  of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  films with different oxygen contents ( $T_{c0} \approx 90$  K and 60 K) is investigated. Plots of the dependence of  $T_c/T_{c0}$  on the residual resistivity  $\rho_0$  are obtained over the very broad ranges  $0.2 < T_c/T_{c0} < 1$  and  $0 < \rho_0 < 800 \mu\Omega \cdot \text{cm}$ . It is established that the critical values of  $\rho_0$ , at which superconductivity vanishes, are an order of magnitude greater than those predicted by the theory of  $d$  pairing. For  $0.5 - 0.6 < T_c/T_{c0} < 1$  the experimental data are consistent with the theoretical plots of  $T_c(\rho_0)$  obtained for a superconductor with anisotropic  $s$  pairing within the BCS model, while for  $T_c/T_{c0} < 0.5$  they are consistent with the model based on the localization of a Bose condensate having anisotropic  $s$  symmetry. © 1996 American Institute of Physics. [S1063-7761(96)02708-4]

## 1. INTRODUCTION

Under the conditions in which experiments performed to determine the symmetry of the superconducting order parameter  $\Delta$  in the  $a$ - $b$  crystallographic plane of a high- $T_c$  superconductor give conflicting results (see, for example Refs. 1 and 2), great importance is attached to investigations of the physical characteristics of high- $T_c$  superconductors, which, according to theory, should differ quantitatively for superconductors with  $s$  and  $d$  pairing and should, therefore, make it possible to indirectly evaluate the symmetry of  $\Delta$ . They include, in particular, the dependence of the critical temperature  $T_c$  on the concentration  $n_{\text{im}}$  of nonmagnetic defects, which may be nonmagnetic substitutional impurities or defects resulting from irradiation.

Several hundred experimental studies of the influence of ion, neutron, and electron irradiation on the properties of high- $T_c$  superconductors have been reported (see, for example, Refs. 3–5 and the references therein). It was established that  $T_c$  and the critical current density  $J_c$  decrease with increasing  $n_{\text{im}}$  and vanish at a certain critical value  $n_{\text{im}}^c$ . An analysis of the data reveals that the dependence of the reduced critical temperature  $T_c/T_{c0}$  (here and in the following  $T_{c0}$  is the value of  $T_c$  for a sample without defects, i.e., before irradiation) is a universal function of the reduced defect concentration  $n_{\text{im}}/n_{\text{im}}^c$ .<sup>5</sup> Nonmagnetic impurities have a similar influence on the critical properties of high- $T_c$  superconductors (see, for example, Ref. 6). Thus, it can be concluded that a phase transition from the superconducting state to the normal state can occur under the influence of defects. In high- $T_c$  superconductors this transition takes place when the inverse momentum relaxation time  $1/\tau$  of the carriers reaches a value on the order of the Fermi energy  $E_F$  (Refs. 5 and 6), rather than a value of order  $\Delta$ , as might have been expected for  $d$ -wave pairing (here and in the following we use a system of units in which  $\hbar = k_B = 1$ ).

Within the BCS theory, under the assumption of a weak dependence of the electron density of states  $N$  on the energy of the quasiparticles  $\varepsilon$  in the vicinity of  $\varepsilon = E_F$ , the depen-

dence of  $T_c$  on the momentum relaxation time  $\tau$  for  $1/\tau < E_F$  is determined by the equation<sup>7</sup>

$$\ln\left(\frac{T_{c0}}{T_c}\right) = \chi \left[ \psi\left(\frac{1}{2} + \frac{1}{4\pi\tau T_c}\right) - \psi\left(\frac{1}{2}\right) \right]. \quad (1)$$

Here  $\psi(z)$  is the digamma function, and  $\chi = 1 - \langle \Delta \rangle^2 / \langle \Delta^2 \rangle$  is the anisotropy parameter, which is determined by averaging  $\Delta(\mathbf{k})$  and  $\Delta^2(\mathbf{k})$  over the Fermi surface with the weight  $v^{-1}(\mathbf{k})$ , where  $v(\mathbf{k}) = \partial \varepsilon(\mathbf{k}) / \partial \mathbf{k}$  (Ref. 7), and which is a quantitative expression of the degree of anisotropy  $\Delta(\mathbf{k})$  (here and in the following we are dealing with the “two-dimensional anisotropy”  $\Delta$  in the  $a$ - $b$  plane, and we do not specify the mechanism of the electron-boson interaction). The quantity  $1/\tau$ , for its part, is related by the expression<sup>8</sup>

$$\frac{1}{\tau} = \frac{\omega_{\text{pl}}^2}{4\pi\rho_0} \quad (2)$$

to the residual resistivity  $\rho_0$ , which is found directly from experimental data (here  $\omega_{\text{pl}}$  is the plasma frequency) and is proportional to  $n_{\text{im}}$  in a first approximation.

For isotropic  $s$  pairing we have  $\langle \Delta \rangle^2 = \langle \Delta^2 \rangle$ ; therefore,  $\chi = 0$ , and when there is relatively weak disorder ( $1/\tau < E_F$ ), according to (1), the value of  $T_c$  is independent of  $\tau$  and, therefore,  $\rho_0$  (Anderson's theorem<sup>9</sup>), as is observed experimentally in ordinary superconductors with a low  $T_c$ . If  $N(\varepsilon)$  has a peak near the Fermi level,  $T_c$  varies significantly as  $n_{\text{im}}$  increases (i.e., as  $\tau$  decreases).<sup>10</sup> Then, depending on the relative positions of the Fermi level and the maximum of  $N(\varepsilon)$ ,  $T_c$  can either decrease (for example, in  $\text{Nb}_3\text{Sn}$ ) or increase (for example, in  $\text{Mo}_3\text{Si}$ ).<sup>11</sup>

In superconductors with  $d$  pairing we have  $\chi = 1$  (since  $\langle \Delta \rangle = 0$ ); therefore, according to (1),  $T_c$  should decrease rapidly with increasing  $1/\tau$  and vanish at  $1/\tau \approx T_{c0}$ , which for the values typical of high- $T_c$  superconductors  $T_{c0} \approx 100$  K and  $\omega_{\text{pl}} \approx 1$  eV corresponds to  $\rho_0 \approx 50 \mu\Omega \cdot \text{cm}$ . This conclusion remains valid<sup>8</sup> when the strong electron-boson coupling effects are taken into account in the Éliashberg theory.

The range  $0 < \chi < 1$  corresponds to anisotropic  $s$  pairing,<sup>7</sup> which includes the special case of so-called  $s^*$  pairing, in which  $\Delta(\mathbf{k}) = \Delta_0[\cos(k_x a) + \cos(k_y a)]$ . In this case, according to (1),  $T_c$  tends asymptotically to zero as  $1/\tau$  increases,  $T_c$  decreasing more rapidly or  $\chi$  becomes larger. In particular, the equation<sup>7</sup>

$$\frac{T_c}{T_{c0}} = 1 - \frac{\pi}{8} \frac{\chi}{\tau T_{c0}} \quad (3)$$

holds in the limiting case of weak disorder ( $1/\tau \ll 4\pi T_{c0}$ ). However,  $T_c$  does not vanish at a finite value of  $1/\tau$ , attesting to the inadequacy of the standard approximations of the BCS model for describing the dependence of  $T_c$  on  $1/\tau$  when  $1/\tau \sim E_F$  holds.

As shown by the cluster calculations in Ref. 12, which were performed within the Emery model by exact diagonalization, the pairing correlations in the  $d$  channel are suppressed in the presence of atomic disorder considerably more strongly than those in the  $s^*$  channel. Thus, different theoretical approaches<sup>7,8,12</sup> lead to the same result: the value of  $T_c$  of a superconductor with  $d$ -wave pairing is far more sensitive to defects than is the value of  $T_c$  of a superconductor with anisotropic  $s$  pairing.

Various aspects of the relationship between  $T_c$  and  $\rho_0$  were recently discussed in the literature. For example, a theory, which states that  $T_c$  is governed by the value of  $\rho(T_c)$  and vanishes when  $\rho(0) \sim \hbar/e^2$ , was proposed in Ref. 13. This theory was developed for "bad" metals, in which the mean-free path of the carriers is less than their de Broglie wavelength at sufficiently high temperatures even though the temperature dependence  $\rho(T)$  has a metallic character.

We also mention the model based on the localization of a Bose condensate,<sup>14</sup> which predicts a phase transition from a superfluid state to a localized state and a linear dependence of  $T_c$  on  $q/\tau$  (i.e., on  $\rho_0$ ):

$$\frac{T_c}{T_{c0}} = 1 - \frac{\delta}{\tau E_F} = 1 - \frac{\rho_0}{\rho_0^c}, \quad (4)$$

where we have written  $\rho_0^c = \beta(\hbar/e^2)$ , and  $\delta$  and  $\beta$  are numerical coefficients of order unity. We note, in passing, that Eq. (4) was derived in Ref. 14 from the Ginzburg–Landau equation, which is valid only for  $(T_{c0} - T_c)/T_{c0} \ll 1$ , i.e., if  $\rho_0 \ll \rho_0^c$ . However, there is some basis to assume that Eq. (4) is applicable over a broader range of  $\rho_0$ . The physical meaning of Eq. (4) is as follows.<sup>14</sup> An increase in  $n_{im}$  causes localization of a portion of the bosons in the condensate with resultant decreases in the density of the superfluid component  $n_s$  and, accordingly, in  $T_c$ :

$$T_c \propto n_s = n_0 \left( 1 - \frac{n_{im}}{n_0} \eta \right), \quad (5)$$

where  $n_0$  is the density of the Bose condensate,  $n_s$  is the density of the superfluid component, and  $\eta$  is a numerical coefficient that depends on the matrix element of the interaction of electrons with the defects.

The experimental dependence of  $T_c$  on  $\rho_0$  can, in principle, be found by replacing any atoms in the crystal lattice (for example, the copper atoms) with atoms of other chemi-

cal elements. This generally leads to a decrease in  $T_c$  and an increase in  $\rho_0$ . For example, the plots of  $\rho(T)$  for samples of high- $T_c$  superconductors of different types (ceramics, single crystals, and thin films) and with different concentrations of substitutional impurities were obtained for  $\text{YBa}_2(\text{Cu}_{1-x}\text{M}_x)_3\text{O}_y$  ( $\text{M} = \text{Zn}, \text{Co}, \text{Ni}$ ) in Refs. 15–23, for  $\text{Y}_{1-x}\text{Pr}_x\text{Ba}_2\text{Cu}_3\text{O}_y$  in Refs. 6 and 24, for  $\text{YBa}_2(\text{Cu}_{1-x}\text{M}_x)_4\text{O}_8$  ( $\text{M} = \text{Fe}, \text{Ni}$ ) in Ref. 25, for  $\text{Bi}_2\text{Sr}_2\text{Ca}(\text{Cu}_{1-x}\text{M}_x)_2\text{O}_{8+y}$  ( $\text{M} = \text{Fe}, \text{Ni}, \text{Zn}$ ) in Ref. 28, for  $\text{Bi}_2\text{Sr}_2(\text{Ca}_{1-x}\text{R}_x)\text{Cu}_2\text{O}_{8+y}$  ( $\text{R} = \text{Pr}, \text{Gd}, \text{Er}$ ) in Ref. 29, etc. However, the interpretation of such experiments is complicated by the fact that the impurities (even the nonmagnetic ones) can induce local magnetic moments<sup>23,28,30–33</sup> and (or) alter the concentration of charge carriers,<sup>18,19,29</sup> thus causing the appearance of additional (apart from elastic scattering) channels for suppressing superconductivity.

As was noted above, one alternative to substitutional impurities is controlling the variation of the defect concentration by irradiating high- $T_c$  superconductors with ions, electrons, or other particles.<sup>3–6,34–37</sup> First, radiation defects apparently do not have magnetic moments, since they are host atoms that have been displaced from their equilibrium positions (there are, incidentally, different opinions on this issue; see Ref. 3 and the references therein). Second, the atomic disorder resulting from irradiation is not accompanied by any significant changes in carrier concentration.<sup>34,38</sup> Thus, the experimental plots of the dependence of  $T_c$  on  $\rho_0$  obtained by irradiating high- $T_c$  superconductors can be compared directly with the theoretical predictions for superconductors with different symmetry and different degrees of anisotropy of the order parameter [Eqs. (1)–(3)].

The following circumstance must be noted here. While at small irradiation doses the dependence of  $\rho_{ab}$  on  $T$  at temperatures  $T > T_c$  remains linear, making it possible to find  $\rho_0$  by extrapolating  $\rho_{ab}(T)$  to  $T=0$ , at large doses (at which  $T_c$  is significantly smaller than  $T_{c0}$ ) the derivative  $d\rho_{ab}/dT$  generally changes sign from plus to minus as the temperature is lowered, although a superconducting transition can take place.<sup>5,34</sup> Consequently, it is not possible to determine  $\rho_0$  precisely when  $T_c$  is low. Therefore, this approach is often restricted to very small doses, under which the change in  $T_c$  under irradiation ( $T_{c0} - T_c$ ) amounts to only a few degrees and is insufficient for comparing theory with experiment, since it does not enable us to determine the functional form of the dependence of  $T_c$  on  $\rho_0$  and to find (or at least to estimate) the critical value  $\rho_0^c$ , at which the superconductivity vanishes.

In addition, the decrease in  $T_c$  with irradiation is usually accompanied by a considerable increase in the width of the superconducting transition  $\Delta T_c$  (which is defined as the difference between the temperatures for the onset and completion of the transition), which can be comparable to the value of  $T_{c0} - T_c$  or even exceed it. The transition width  $\Delta T_c$  can be regarded as the error in the determination of  $T_c$  (the latter is usually determined as the temperature at the middle of the transition). The possible causes of large values of  $\Delta T_c$  include, first, poor quality of the original samples and, second, the excessively high temperature  $T_{irr}$  at which the irradiation is performed. It is known<sup>5</sup> that  $T_{irr} \approx 300$  K is already suffi-

cient to thermally anneal the radiation defects responsible for the changes in  $T_c$  and  $\rho_0$  in high- $T_c$  superconductors; therefore, the results obtained in this way are a consequence of the superposition of two processes, viz., radiation-induced defect formation and annealing of the defects formed.

Thus, for a detailed comparison with theory, first, we must have an experimental plot of the dependence of  $T_c$  on  $\rho_0$  over as broad a range of  $T_c/T_{c0}$  and  $\rho_0$  as is possible, and, second, the condition  $\Delta T_c \ll T_{c0} - T_c$  must be satisfied over that entire range. The purpose of the present work is to experimentally investigate the influence of ion irradiation at  $T_{\text{irr}} = 300$  K and  $T_{\text{irr}} = 12$  K on the values of  $T_c$  and  $\rho_0$  for high-quality thin films of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  with  $x \approx 0$  ( $T_c \approx 90$  K) and  $x \approx 0.4$  ( $T_c \approx 60$  K) and to analyze the function  $T_c(\rho_0)$  theoretically. As will be shown below, the experimental data are consistent with anisotropic  $s$  pairing and contradict the  $d$  symmetry of the order parameter.

## 2. EXPERIMENT

The technique used to fabricate the films was thoroughly described in Ref. 39. The quality of the films is evidenced by the high value of  $J_c$  (for films with  $x \approx 0$  the value of  $J_c$  exceeds  $10^6$  A/cm<sup>2</sup> at  $T = 77$  K and  $10^7$  A/cm<sup>2</sup> at  $T = 4.2$  K). The films were irradiated by helium ions with an energy of 1.2 MeV both at room temperature ( $T_{\text{irr}} = 300$  K) and at a low temperature ( $T_{\text{irr}} = 12$  K) to avoid annealing the defects and their migration to the grain boundaries. For this purpose, a measurement of  $\rho_{ab}(T)$  in the crystallographic  $a$ - $b$  plane was performed after each stage of the low-temperature ( $T_{\text{irr}} = 12$  K) irradiation by heating the films to a temperature above 100 K. This made it possible to achieve a uniform distribution of the radiation defects throughout the sample and, as a consequence, a comparatively small value of  $\Delta T_c$  even when  $T_c$  is very low.

At  $T_{\text{irr}} = 300$  K, the plots of the temperature dependence of  $\rho_{ab}$  are linear over the entire range  $T_c < T < 300$  K for all values of the fluence  $\Phi$  for which the transition to the superconducting state still takes place ( $0.2 < T_c/T_{c0} < 1$ ; we were unable to adjust  $\Phi$  to ensure  $0 < T_c/T_{c0} < 0.2$ ). This makes it possible to determine  $\rho_0$  by extrapolating  $\rho_{ab}(T)$  to  $T = 0$ . An increase in  $\Phi$  results in a decrease in  $T_c$ , an increase in  $\rho_{ab}$  for  $T > T_c$ , and an increase in  $\rho_0$ . The derivative  $d\rho_{ab}(T > T_c)/dT$  then varies in the ranges 1.09–1.45 and 3.20–3.59  $\mu\Omega \cdot \text{cm}/\text{K}$  for the films with  $x \approx 0$  and  $x \approx 0.4$ , respectively [during irradiation the value of  $d\rho_{ab}(T > T_c)/dT$  first increases and then decreases approximately back to its original value].

Since the temperature was not raised above 100 K during the low-temperature irradiation ( $T_{\text{irr}} = 12$  K) in order to avoid annealing defects, the temperature range  $T_c < T < 100$  K was insufficiently wide for determining  $\rho_0$  by extrapolating  $\rho_{ab}(T)$  to  $T = 0$ . We note, however, that the weak dependence of the derivative  $d\rho_{ab}(T > T_c)/dT$  on  $\Phi$  is evidence that radiation disordering results in variation of the temperature-independent component  $\rho_{ab}(T)$ , i.e., variation of  $\rho_0$  itself, which is governed by elastic scattering. Therefore, for the films irradiated at  $T_{\text{irr}} = 12$  K, we set  $\rho_0 = \rho_{ab}^{\text{irr}}(100 \text{ K}) - \rho_{ab}^{\text{unirr}}(100 \text{ K})$ , where  $\rho_{ab}^{\text{irr}}$  and  $\rho_{ab}^{\text{unirr}}$  are

the values of  $\rho_{ab}$  in the irradiated and original samples, respectively [ $\rho_{ab}^{\text{unirr}}(100 \text{ K}) = 95 \mu\Omega \cdot \text{cm}$  for  $x \approx 0$  and  $224 \mu\Omega \cdot \text{cm}$  for  $x \approx 0.4$ ]. Such a determination of  $\rho_0$  can produce an appreciable error only when  $\Phi$  is very small [when  $\rho_0$  is comparable to the value  $\rho_{ab}^{\text{unirr}}(T \rightarrow 0) = 5\text{--}30 \mu\Omega \cdot \text{cm}$  estimated by extrapolation to  $T = 0$ ], but it is valid for  $\rho_0 > \rho_{ab}^{\text{unirr}}(T \rightarrow 0)$ , i.e., over practically the entire range  $\rho_0 < 800 \mu\Omega \cdot \text{cm}$ . (We note that a similar method for determining  $\rho_0$  was used in Ref. 35.)

We stress that a transition from metallic to semiconductor conductivity takes place at  $\rho_0 \approx 1000 \mu\Omega \cdot \text{cm}$ . This value of  $\rho_0$  coincides in order of magnitude with the maximum resistivity of a layered (quasi-two-dimensional) metal  $\rho_{\text{max}} \approx l/\sigma_{\text{min}}$ , where  $l$  is the distance between layers (for high- $T_c$  superconductors it is the lattice constant  $c \approx 1$  nm), and  $\sigma_{\text{min}} \approx e^2/\hbar$  is the minimum metallic conductivity in two dimensions.<sup>40</sup> The fact that  $\rho_{\text{max}}$  is achieved in our experiments only at large values of  $\Phi$ , at which  $T_c/T_{c0} < 0.2$  holds, is a consequence of the small value of  $\rho_0$  before irradiation and attests to the high quality of the samples.

Plots of  $T_c/T_{c0}$  versus  $\rho_0$  for  $T_{\text{irr}} = 300$  K and  $T_{\text{irr}} = 12$  K are presented in Figs. 1 and 2, respectively. It is seen that these plots are nearly linear over the entire range  $0.2 < T_c/T_{c0} < 1$  for both  $x \approx 0$  ( $T_{c0} \approx 90$  K) and  $x \approx 0.4$  ( $T_{c0} \approx 60$  K), regardless of  $T_{\text{irr}}$ . This makes it possible to evaluate the critical value  $\rho_0^c$ , at which  $T_c$  vanishes:  $\rho_0^c \approx 1100 \mu\Omega \cdot \text{cm}$  ( $x \approx 0$ ,  $T_{\text{irr}} = 300$  K);  $\rho_0^c \approx 1350 \mu\Omega \cdot \text{cm}$  ( $x \approx 0.4$ ,  $T_{\text{irr}} = 300$  K);  $\rho_0^c \approx 750 \mu\Omega \cdot \text{cm}$  ( $x \approx 0$ ,  $T_{\text{irr}} = 12$  K);  $\rho_0^c \approx 750 \mu\Omega \cdot \text{cm}$  ( $x \approx 0.4$ ,  $T_{\text{irr}} = 12$  K). Our results for  $x = 0$  and  $T_{\text{irr}} = 300$  K are in qualitative agreement with the data in Refs. 6 and 34, where the value  $\rho_0^c \approx 2000 \mu\Omega \cdot \text{cm}$  was obtained. For  $x = \text{const}$  a decrease in  $T_{\text{irr}}$  from 300 to 12 K results in a decrease in  $\rho_0^c$  by 30–40%. This apparently happens because  $T_{\text{irr}} = 300$  K radiation defects diffuse to the grain boundaries, with the resultant formation of narrow (with a thickness of the order of the coherence length) intervening layers with an increased concentration of defects near the grain boundaries; hence,  $\rho_0$  increases with increasing dose more rapidly than when  $T_{\text{irr}} = 12$  K, and  $T_c$  decreases at the same rate. We note that in the case of low-temperature irradiation the values of  $\rho_0^c$  for the films with  $x \approx 0$  ( $T_{c0} \approx 90$  K) and  $x \approx 0.4$  ( $T_{c0} \approx 60$  K) are identical.

## 3. DISCUSSION OF RESULTS AND CONCLUSIONS

We stress that the resistive transition  $\Delta T_c$  for all  $\Phi$  is less than  $T_{c0} - T_c$  (see Figs. 1 and 2). This makes it possible to compare the experimental data with theory over the broad range  $0.2 < T_c/T_{c0} < 1$ . The theoretical curves calculated from Eqs. (1) and (2) for several values of the anisotropy parameter  $\chi$  are shown in these figures. Different authors have presented slightly different values of  $\omega_{\text{pl}} = 1.1\text{--}1.4$  eV for  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  (see the references in Ref. 8). We set  $\omega_{\text{pl}} = 1.25$  eV [note that an increase or decrease in  $\omega_{\text{pl}}$  results in contraction or extension, respectively, of the  $\rho_0$  axis, according to Eq. (2)].

It is seen from Figs. 1 and 2 that the best agreement between theory and experiment is achieved for  $\chi \approx 0.2$ , but only in the range  $0.5\text{--}0.6 < T_c/T_{c0} < 1$ . It is not possible to

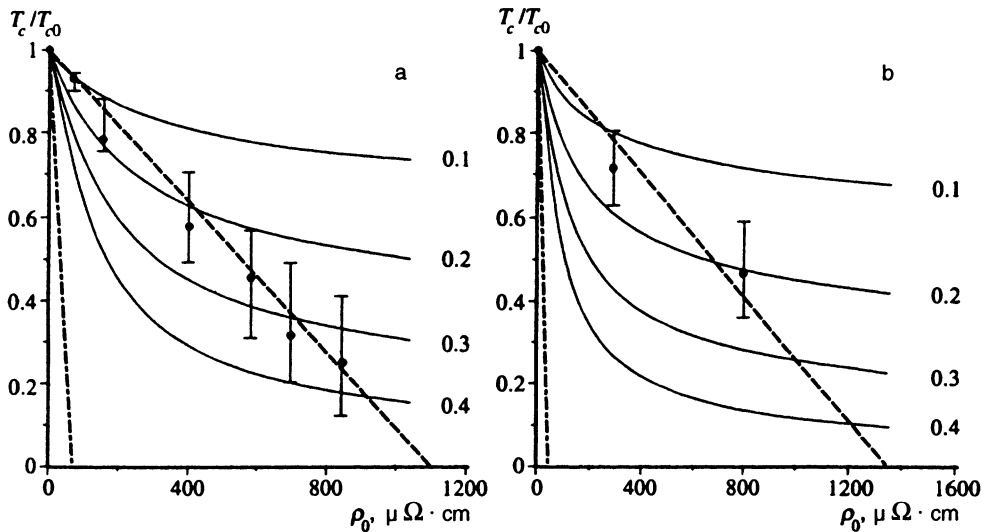


FIG. 1. Dependence of  $T_c$  on the residual resistivity  $\rho_0$  for  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  films obtained by irradiation at  $T_{\text{irr}}=300$  K. Points – experimental data corresponding to the middle of the resistive superconducting transition (the errors are due to the finite width of the transition, and they were determined in the standard manner at the 0.9 and 0.1 levels from the value of  $\rho_0$  at the onset of the transition). Solid lines – theoretical curves calculated from Eqs. (1) and (2) with  $\omega_{\text{pl}}=1.25$  eV for anisotropic  $s$  pairing ( $0 < \chi < 1$ ); the numbers next to the curves are the values of the anisotropy parameter  $\chi$ . Dot-dashed lines – theoretical curves calculated for  $d$  pairing ( $\chi=1$ ). a)  $x \approx 0$  ( $T_{c0} \approx 90$  K); b)  $x \approx 0.4$  ( $T_{c0} \approx 60$  K).

achieve agreement at all values of  $T_c/T_{c0}$  by adjusting  $\chi$ . In addition, it is clear that the variation of  $\omega_{\text{pl}}$  likewise does not contribute anything in this area, since the functional form of the theoretical and experimental dependences of  $T_c$  on  $\rho_0$  differ strongly (the latter are practically linear over the entire range  $0.2 < T_c/T_{c0} < 1$ , see Figs. 1 and 2).

An analysis of the published data<sup>6,15–24,34,35</sup> reveals that  $T_c$  varies linearly with  $\rho_0$   $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  both in the cases of the chemical replacement of specific host atoms<sup>6,15–24</sup> and in the case of irradiation.<sup>6,34,35</sup> For the purpose of comparing our results with the data of other investigators, we evaluated  $\chi$  using Eqs. (2) and (3) and experimental plots of the dependence of  $T_c$  on  $\rho_0$  taken from Refs. 6, 15–24, 34, and 35 [to analyze the data obtained on polycrystalline samples, we used an approximate method to convert  $\rho$  into  $\rho_{ab}$  (Ref. 25)]. The spread in the values of  $\chi$  is found to be fairly great,  $\chi=0.05$ – $0.5$ . Also, the value of  $\chi$  is generally greater in the chemical substitution experiments<sup>16,22,23</sup> than in the irradiation experiments.<sup>6,34,25</sup> As noted above, this can be attributed to the action of secondary factors, such as the variation of the

carrier concentration and (or) scattering on the magnetic moments.

We note that a deviation from the linear dependence of  $T_c(\rho_0)$  and a decrease in the derivative  $|dT_c/d\rho_0|$  or  $\rho_0$  increases (which corresponds to a decrease in  $\chi$ ) were observed in several studies (for example, Refs. 6 and 21). We note in this context that all four plots of the dependence of  $T_c$  on  $\rho_0$  that we obtained (see Figs. 1 and 2) have a small positive curvature (although this effect scarcely exceeds the range of error, i.e.,  $\Delta T_c$ ).

Thus, most of the experiments on the influence of non-magnetic impurities and radiation defects on  $T_c$  and  $\rho_0$  in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  are consistent with the estimate  $\chi=0.1$ – $0.3$ . The smallness of  $\chi \ll 1$  attests to the anisotropic  $s$  pairing of the charge carriers in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ , since  $\chi=1$  holds in the case of  $d$  pairing. We note, in passing, that the calculations performed within the model of  $(d+s)$ -wave symmetry for  $\Delta$  (Ref. 41), according to which  $\Delta(\mathbf{k}) = \Delta_d + \alpha \Delta_s$ , where  $\Delta_d = \Delta_0 \cos 2\varphi$  and  $\Delta_s = \Delta_0$ , also lead to an equation like (1). Here the role of  $\chi$  is played by the parameter  $1/(1+2\alpha^2)$ ,

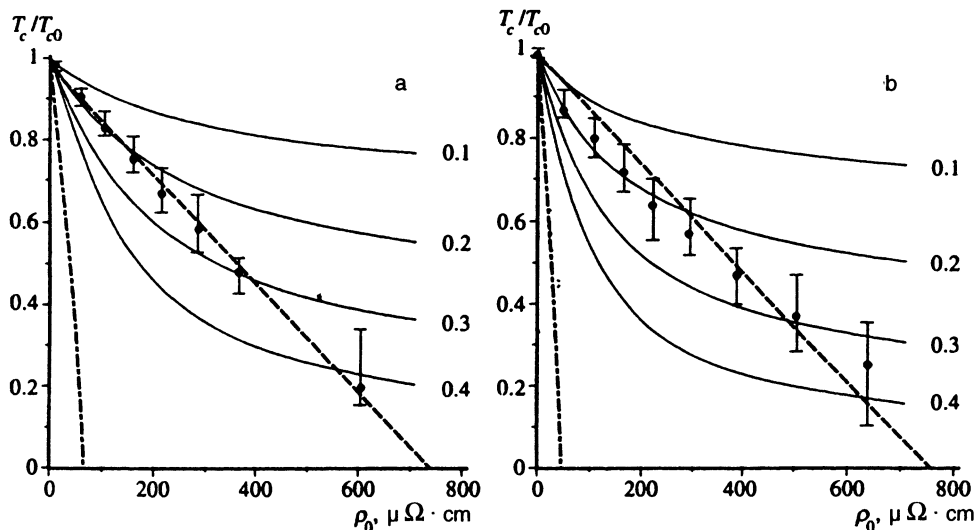


FIG. 2. Same as in Fig. 1 for  $T_{\text{irr}}=12$  K.

i.e., an increase in the contribution of the isotropic  $s$  channel to  $\Delta(\mathbf{k})$  leads to a decrease in  $\chi$ . However, the estimate  $\chi=0.1-0.3$  corresponds to  $\alpha^2=1-4$ , implying that of the  $s$ -wave component dominates in  $\Delta(\mathbf{k})$ . At such values of  $\alpha$  the order parameter  $\Delta(\mathbf{k})=\Delta_d+\alpha\Delta_s$  actually corresponds to anisotropic  $s$ -wave pairing.

We also note that the value  $\rho_0^c \approx 1000 \mu\Omega \cdot \text{cm}$  is more than an order of magnitude greater than the value  $\rho_0^c \approx 50 \mu\Omega \cdot \text{cm}$  calculated from Eqs. (2) and (3) for a superconductor with  $T_{c0}=60-90$  K,  $\omega_{pl}=1.25$  eV, and  $\chi=1$  (i.e., with  $d$  symmetry of the order parameter); the corresponding curves are shown in Figs. 1 and 2. The proposed  $d$  symmetry can be brought into agreement with experiment only by postulating the unrealistically small values  $\omega_{pl}=0.3-0.4$  eV, which are three to four times smaller than the smallest published value of  $\omega_{pl}$  for  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ .

As for other high- $T_c$  superconductor systems, the values of  $\chi$  that we determined for them from experimental plots of the dependence of  $T_c$  on  $\rho_0$  (Refs. 25–29) in the same manner as for  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  are:  $\chi \approx 0.15$  in  $\text{YBa}_2\text{Cu}_4\text{O}_8$  (Ref. 25),  $\chi \approx 0.1$  in  $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$  (Refs. 26 and 27), and  $\chi \approx 0.1$  in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  (Refs. 28 and 29). [We set  $\omega_{pl} \approx 1$  eV for all these compounds. If the true value of  $\omega_{pl}$  for a particular compound is greater (or smaller) than 1 eV, the value of  $\chi$  is smaller (greater) than our evaluation.] We note that the calculations that we previously performed<sup>42,43</sup> on the basis of angle-resolved photoemission spectroscopy (ARPES) also showed that the value of  $\chi$  for the high- $T_c$  superconductor  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  is approximately equal to 0.1, despite the strong anisotropy of  $\Delta$  in  $\mathbf{k}$  space.<sup>44</sup>

As was noted above, Eq. (1) is incapable of explaining the experimental plots of the dependence of  $T_c$  on  $\rho_0$  for  $T_c/T_{c0} < 0.5$ . At the same time, these plots are described well by Eq. (4), which is based on the model of the localization of a condensate of Cooper pairs.<sup>14</sup> It is important to stress that the critical value  $\rho_0^c$  in (4), at which  $T_c$  vanishes, is consistent in order of magnitude with the experimental value  $\rho_0^c \sim 1000 \mu\Omega \cdot \text{cm}$  (which corresponds to  $1/\tau \sim E_F$ ). We also note that in practically all the experimental studies known to us the transition from the linear metallic dependence of  $\rho_{ab}(T)$  to a semiconductor dependence with  $d\rho_{ab}/dT < 0$  was observed at  $\rho_0 \sim 1000 \mu\Omega \cdot \text{cm}$ , attesting to the appearance of charge-carrier localization effects. Since localization takes place at  $1/\tau \sim E_F$  and since the destruction of superconductivity in the  $d$ -wave channel occurs at  $1/\tau \sim T_{c0} \ll E_F$ , localization can appear only when the order parameter does not have  $d$ -wave symmetry. This also favors anisotropic  $s$  pairing.

We note that the disparity between theory and experiment at small values of  $T_c/T_{c0}$  can also be attributed to such factors as 1) the inadequacy of the BCS approximation (i.e., the need to take into account retardation effects) and 2) the possible presence of magnetic moments in the radiation defects (which would require consideration of the scattering on the magnetic impurities).

Thus, an analysis of the data obtained in the present work and the published experimental data on the influence of impurities and radiation defects on the critical temperature and the residual resistivity of high- $T_c$  superconductors pro-

vide evidence in support of the anisotropic  $s$  symmetry of the order parameter and against the  $d$ -wave picture of high- $T_c$  superconductivity. The small (an order of magnitude smaller than for  $d$  pairing) value of the anisotropy parameter  $\chi$  is an argument indicating that anisotropic  $s$  pairing occurs in different high- $T_c$  superconductor systems. The smallness of  $\chi$  accounts for the relatively weak (at least, far weaker than for  $d$  pairing) sensitivity of  $T_c$  to atomic disorder.

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