Dependence of the magnetic properties of $YBa_2Cu_3O_x$ in the superconducting state on the oxygen content

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The temperature and field dependences of the magnetization of polycrystalline $YBa_2Cu_3O_x$ samples were determined in the superconducting state as a function of the oxygen content ($6.24 \leqslant x \leqslant 6.9$). A continuous increase in the volume of superconducting phase as a result of cooling, a scatter of the values of T_c, and a decrease of the upper limit of T_c on lowering of x were observed. In the range $x > 6.55$ the samples contain only the orthorhombic phase, whereas in the $x \leqslant 6.78$ range even at temperatures $T \ll T_c$ the volume of the superconducting phase deduced from the static magnetic susceptibility was considerably less than half the volume of the sample after a correction for porosity. In samples with $6.55 \leqslant x \leqslant 6.78$ the volume of the superconducting phase deduced from the dynamic susceptibility at frequencies from 300 Hz to 8 kHz was considerably greater than that found from static measurements: the difference was independent of the frequency throughout the investigated range, but it decreased on increase in temperature and disappeared at $T = T^*(x) < T_c$. Magnetic hysteresis curves were used to estimate the critical currents and dimensions of Josephson junctions. A qualitative interpretation of all the observed effects was based on the concept of a cluster distribution of oxygen and formation of separate superconducting regions joined by Josephson junctions.

Properties of high-temperature superconductors with the $YBa_2Cu_3O_x$ composition are particularly sensitive to the oxygen content and most of the published investigations have been carried out on samples with the optimal (from the point of view of superconductivity) content. However, it is obvious that detailed investigations of the sample characteristics related to the superconducting state as functions of the oxygen content can throw light on the physical nature of the factors responsible for the actual phenomenon of high-temperature superconductivity.

We investigated how magnetic properties of high-temperature superconductors of this type changed with the oxygen content within limits necessary to convert an orthorhombic sample (superconductor) to the tetragonal nonsuperconducting state. (The oxygen content was varied by quenching samples in liquid nitrogen from various initial annealing temperatures.) The magnetic susceptibilities χ_{dc} and χ_{ac} were determined at temperatures 4.2–100 K in, respectively, constant (Faraday method) and alternating (induction method) fields, and measurements were made also of the magnetization M (in magnetic fields H up to 60 kOe) of compounds of the $YBa_2Cu_3O_x$ type with different oxygen concentrations $6.24 \leqslant x \leqslant 6.9$.

EXPERIMENTAL RESULTS

The influence of random impurities was avoided by making all the measurements on one polycrystalline sample prepared by the cryochemical technology (Ref. 1).[1)] The amount of oxygen was varied by consecutive annealing at various temperatures. We first ensured the maximum oxygen content ($x = 6.9$) after annealing for 12 h in an oxygen stream at 400 °C and slow cooling to room temperature. Then, the oxygen content was reduced to $x = 6.24$ by annealing for 6 h at 900 °C in air and quenching in liquid nitrogen. This was followed by further annealing in air at 700, 600, and 500 °C followed by quenching in liquid nitrogen from each of these temperatures: in this way the oxygen content in a sample was gradually increased to $x = 6.55$, 6.68, and 6.78, respectively.

The phase composition of the samples was monitored by x-ray diffraction using a DRON-2 diffractometer. The parameter x was determined by the method of oxidation–reduction titration to within ± 0.03. The results of an x-ray phase analysis for x ranging from 6.9 to 6.24 revealed structural changes due to disordering of oxygen vacancies and a transition of the structure from the orthorhombic O phase to the tetragonal T phase (Table I).

TABLE I. Characteristics of investigated samples.

Oxygen content, x	Heat treatment	Crystal struct., from x-ray phase anal	T_c, K	Amount of superconducting phase (%), from χ_{dc}
6,24	annealing at 900°C in air + quenching	T + traces of O	40	0,1
6,55	annealing at 700°C + quenching	O + T	50	–
6,68	annealing at 600°C + quenching	O + traces of T	58	36
6,78	annealing at 400°C + quenching	O	88	45
6,9	annealing at 400°C in oxygen without quenching	O	92	100

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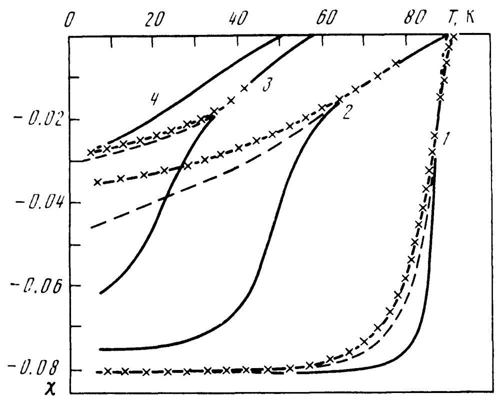

FIG. 1. Temperature dependences of the magnetic susceptibility determined in alternating (χ_{ac}) and constant (χ_{dc}) magnetic fields of $YBa_2Cu_3O_x$ compounds with $x = 6.9$ (1), 6.78 (2), 6.68 (3), and 6.55 (4). Here, the crosses represent χ_{dc} in a field $H = 10$ Oe (cooling in the absence of a field), the continuous curves represent χ_{ac} in the absence of a constant field, and dashed lines give the values of χ_{ac} in the presence of a static field $H \approx 20$ Oe.

The changes in the magnetic properties of a sample containing different amounts of oxygen are presented in Figs. 1–3. It is clear from Fig. 1 that a reduction in the oxygen content x lowered the temperature T_{c0} at which the sample began to exhibit superconducting properties.

Moreover, the results indicated that a reduction in T_{c0} was accompanied by a strong reduction in the relative volume of the superconducting phase in the sample. In fact, the maximum possible value ($-1/4\pi$) of the magnetic susceptibilities $\chi_{dc}(T=0)$ and $\chi_{ac}(T=0)$ was observed only for $x = 6.9$ ($T_c = 91.5$ K). Reduction in the amount of oxygen x from 6.9 to 6.78 lowered T_{c0} from 91.5 to 88 K, gave rise to a smooth temperature dependence of χ_{dc} throughout the interval from T_{c0} to 4.2 K, and reduced the fraction of the superconducting phase at $T=0$ K from 100 to 44% ($\chi_{dc}|_{T=0} \to -0.036$). It should be pointed out that the structure remained orthorhombic: the diffractograms simply manifested broadening of the lines. [The amount of the superconducting phase was found by comparing the $\chi_{dc}(T)$ screening curves obtained during cooling in the absence of a field the Meissner $\chi_{dc}(T)$ dependences (during cooling in a field) allowing for the frozen-in moment.[2] The Meissner curves were not included in Fig. 1 in order not to clutter the figure. The use of an automatic microbalance in the Faraday method ensured that there was practically no displacement of the sample on switching on of the gradient coils when this sample was placed in a homogeneous constant magnetic field in superconducting solenoid. This avoided a possible error in the determination of the susceptibility, which could result from the motion of the sample in the inhomogeneous magnetic field.]

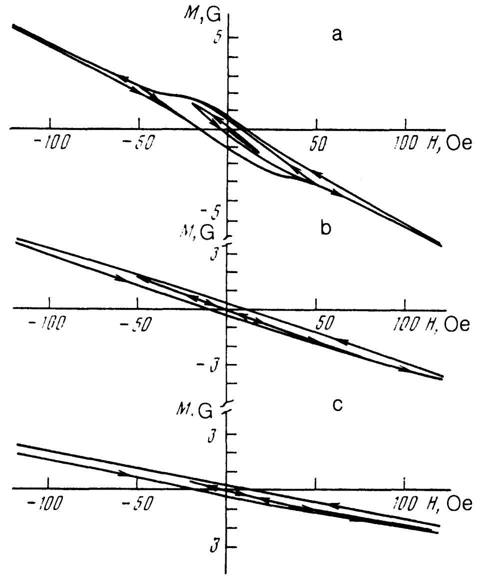

FIG. 2. Dependences of the magnetic moment on the field in the range of weak fields applied to $YBa_2Cu_3O_x$ compounds with $x = 6.9$ (a), 6.78 (b), and 6.68 (c).

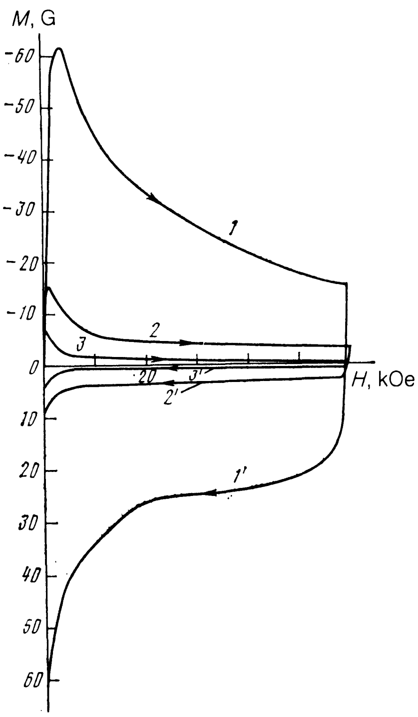

FIG. 3. Dependences of the magnetic moment on the field in the range of strong fields applied to $YBa_2Cu_3O_x$ compounds with $x = 6.9$ (1), 6.78 (2), and 6.68 (3).

We thus found that the susceptibilities of "poor" high-temperature superconductors with lower values of x and T_{c0} behaved as if the sample contained a considerable fraction of a nonsuperconducting phase with an orthorhombic structure.

Additional information was provided by the $\chi_{ac}(T)$ dependences in Fig. 1. (Measurements in the frequency range from 300 Hz to 8 kHz failed to reveal any frequency dependence of this susceptibility.) It's clear from Fig. 1 that an alternating field (of ≈ 0.1Oe amplitude) induced currents which flowed at low temperatures in a volume considerably greater than that occupied by the superconducting phase and the nonsuperconducting component seemed to be screened. Increase in temperature ($T > 60$ K for $x = 6.78$) destroyed the difference between the values of χ_{ac} and χ_{dc}, i.e., the dc and ac induction currents flowed in the same effective volume, which was however considerably less than the total volume of the sample. It can be seen from Fig. 1 that a similar effect could occur also on application of a weak constant external field of ≈ 20 Oe intensity against the background of the alternating field.

Lowering of the oxygen content reduced the effective volume screened by alternating currents at low temperatures and at $x = 6.55$, when—according to the x-ray phase analysis data—there was a considerable proportion of the tetragonal phase, the $\chi_{\rm ac}(T)$ and $\chi_{\rm dc}(T)$ curves practically coincided throughout the investigated range of temperatures.

Determination of the $M(H)$ dependences (Fig. 2) at $T = 4.2$ K showed that the slopes of the initial parts of the curves were in practice correlated with the amount of matter in the superconducting state in the investigated samples. Then, at $x = 6.9$ (Fig. 2a) a "weak" hysteresis of the $M(H)$ curve was observed in fields -50 Oe $< H < 50$ Oe, with the curve transformed into a reversible $M(H)$ dependence in fields in the range ~ 100–300 Oe (at $T = 4.2$ K), whereas a reduction in the oxygen content maintained the reversibility even in the lowest fields -50 Oe $< H < 50$ Oe, i.e., in this case no "weak" hysteresis was observed at all.

The hysteretic $M(H)$ dependences plotted in Figs. 2b and 2c for a sample with a reduced oxygen content, and observed as a result of cyclic variation of the magnetic field within a wide range ($H \gtrsim 100$ Oe), represented the initial parts of loops due to a "strong" hysteresis. Curves 2 and 3 in Fig. 3 represented $M(H)$ curves obtained in high magnetic fields. These curves demonstrated that the first critical fields associated with the penetration of the magnetic field into the crystallites, were considerably less for samples with a reduced oxygen content than for a sample with $x = 6.9$. The significant reduction in the peak-to-peak amplitude of the "strong" hysteresis indicated that a reduction in the oxygen content altered greatly the superconducting characteristics of the material.

THEORY

High-temperature superconductors are widely regarded as assemblies of superconducting regions joined by Josephson junctions (see, for example, Ref. 3). Application of the Bean theory[4] within the framework of this general picture makes it possible to explain qualitatively the observed magnetic effects, as demonstrated below.

We shall begin by stating a number of general formulas taken from Bean's theory and some of their modifications necessary to estimate the parameters of the system described above.[2)] According to Refs. 4 and 5, the distribution of the critical bulk transport current J_c in a sample can be described by the equilibrium condition for a vortex lattice

$$[\mathbf{J}_c\mathbf{B}] = \mathbf{P}(\mathbf{B}) \tag{1}$$

($\mathbf{P}$ is the bulk vortex-pinning force and $\mathbf{B}$ is the magnetic induction), which together with the equation

$$[\nabla \mathbf{H}] = \frac{4\pi}{c}\mathbf{J}_c \tag{2}$$

provides a complete formulation of the problem of the critical state if we know the $\mathbf{H}(\mathbf{B})$ dependence. We shall assume that the range of fields $H > H_{c1}$ and the condition $H \approx B$ can be satisfied with precision sufficient for our estimates. The solution of Eqs. (1) and (2) for a cylindrical sample gives a simple expression for the static magnetization in the range of fields $H_{c1} \ll H \ll H_{c2}$:

$$M_0(B) = J_c R/3c, \quad J_c(B) = P(B)B^{-1}, \tag{3}$$

where R is the radius of the investigated sample.

The theory of Ref. 5 also predicts the peak-to-peak amplitude of a hysteresis loop

$$M_+(B) - M_-(B) = 2M_0(B). \tag{4}$$

The quantities M_+ and M_- in Eq. (4) represent the magnetizations measured during increase and reduction in the field. The sum

$$4\pi(M_+ + M_-) = 2(4\pi M_e) \tag{5}$$

gives the reversible part of the magnetization M_e, which is due to surface currents in the superconductor.

Equations (3)–(5) apply to a homogeneous hard type-II superconductor, whereas in our case it is applied to the volume inside the superconducting region. We shall now analyze the pinning mechanisms for weak links, i.e., in the Josephson junction between grains (Fig. 4).

A magnetic field penetrates a Josephson junction (along the y axis in Fig. 4) to a depth

$$\lambda_J = (\Phi_0 c/4\pi^2 J_m d)^{1/2} \ll \lambda_{1,2}, \tag{6}$$

where $\lambda_{1,2}$ are the depths of penetration of the field (along the x axis) into grains in contact; $d = \lambda_1 + \lambda_2$ is the thickness of the junction; J_m is the maximum density of the current through the Josephson junction; $\Phi_0 = hc/2e$ is the flux quantum; c is the velocity of light; e is the electron charge.

The critical field for the penetration of vortices into a Josephson junction is given by[6]

$$H_{c1}' = \frac{1}{\pi^2}\frac{\Phi_0}{d\lambda_J} \ll H_{c1} = \frac{1}{4\pi}\frac{\Phi_0}{\lambda^2}\ln\left(\frac{\lambda}{\xi}\right). \tag{7}$$

The quantity ξ in Eq. (7) is the coherence length.

Vortices form a one-dimensional chain (Fig. 4a), and in fields $H \gg H_{c1}'$ the current density in a junction is

$$J(y) = J_m \sin[2\pi(y - y_0)/a] \tag{8}$$

so that the corresponding magnetic field in the junction

$$H(y) = H - (4\pi/c)J_m \cos[2\pi(y - y_0)/a] \tag{9}$$

oscillates as a function of the coordinate y along the junction (Fig. 4a). The oscillation frequency is

$$a = \Phi_0/dH. \tag{10}$$

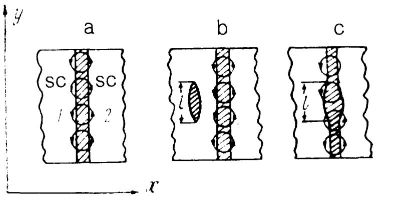

FIG. 4. a) Josephson junction between superconductors SC (1 and 2) free of defects, showing a chain of vortices which have penetrated the junction (the magnetic field of the vortex is normal to the xy plane). b) Josephson junction with a defect of the cavity type near the contact. c) Josephson junction with an intrinsic defect representing a thicker region of the nonsuperconducting layer. The quantity l represents the size of the defect.

Equations (8)–(10) allow us to estimate the dependence of the pinning force P on a or on H in the limit $a \to 0$. The pinning forces may be due to various factors.

We shall discuss two very different pinning mechanisms which however result in a universal asymptotic $P(H)$ dependence.

We shall assume that pinning is due to a magnetic interaction, such as that caused by reflection of a vortex at a defect boundary of the superconductor–vacuum type (Fig. 4b). In the case of this mechanism the interaction force is proportional either to the oscillatory part of $H(y)$ or to $J(y)$. In both cases we have

$$P \propto J_m \int dy\, u(y) \cos[2\pi(y-y_0)/a], \tag{11}$$

where $u(y)$ is the dependence of the force of interaction between vortices on the position relative to a defect.

A similar expression for P is obtained also if we postulate the other pinning mechanism, i.e., pinning at junction inhomogeneities. Such inhomogeneities (Fig. 4c) result in a local change in J_m and, consequently, in λ_J. The energy U for binding a vortex lattice to such a localized defect can be estimated by perturbation theory using an expression for the functional of the free energy of a Josephson junction[6]

$$U \propto \int dy' [\lambda_J^{-2}(y') - \lambda_{J_0}^{-2}] \cos\left(\frac{2\pi y'}{a}\right) \cos\left(\frac{2\pi y}{a}\right), \tag{12}$$

where λ_{J_0} is the value of $\lambda_J(y)$ far from a defect and y is the coordinate of the vortex lattice relative to the defect. Bearing in mind that $P \sim dU/dy$ and assuming that $\lambda_J^{-2}(y) - \lambda_{J_0}^{-2} = J_m u(y)$, we again obtain Eq. (11). It therefore follows that Eq. (11) is a universal expression for the pinning force. The only parameter in Eq. (11) which depends on the field H is the quantity a [see Eq. (10)]. The pinning force depends strongly on a if $u(y)$ is a sufficiently smooth function. In the limit when $u(y)$ is an analytic function, which is the most realistic case, we find that

$$P \propto J_m \exp(-\alpha l/a) = J_m \exp(-\alpha H dl/\Phi_0), \tag{13}$$

where l is the characteristic size of the defect and $\alpha \approx 1$ is a numerical coefficient. It therefore follows that the pinning force decreases exponentially on increase in the field.

The expression (13) for the pinning force makes it possible to use the experimental $P(H)$ dependence, i.e., the magnitude of the hysteresis [Eqs. (1)–(5)], to estimate the characteristic size of the defect. It is interesting to note that apart from the dependence on the pinning mechanism, Eq. (13) predicts also a linear relationship between P and J_m. In other words, Eq. (13) is a quantitative expression of the obvious fact that the pinning force decreases on deterioration of the contacts (junctions).

DISCUSSION AND INTERPRETATION OF THE RESULTS

The most interesting among the experimental data is the disagreement between the actual volume of a sample and the volume occupied by the superconducting phase (even in the limit $T \to 0$), observed for all samples with $x < 6.9$; an increase in temperature reduces smoothly to zero the volume of the superconducting phase in these samples. It follows from the x-ray phase analysis data, however, that samples with $x = 6.9$, 6.78, and 6.68 are orthorhombic and the sample with $x = 6.55$ is a mixture with tetragonal (nonsuperconducting) and orthorhombic structures. Therefore, the superconducting state in samples with $x = 6.55$ is not realized over the whole volume of the sample even at very low temperatures and this is true in spite of the homogeneity of the crystal structure of the investigated material.

It is natural to assume that there are regions with such configurations of the oxygen atoms which favor the superconducting state (or which prevent the appearance of this state). The assumptions made can be used to explain quite readily the smooth reduction in the volume of the superconducting phase on increase in temperature: obviously, each configuration (and they form a continuous set) corresponds to its own superconducting transition temperature. Lowering of the value of T_{c0} on reduction in x is explained equally simply: when x is reduced, there is a drop in the statistical weight of those configurations which correspond to high oxygen concentrations, i.e., to high values of T_c [the highest T_c corresponds to $x \approx 6.9$, i.e., to the highest oxygen concentration; for $x = 6.9$ the weight of "unfavorable" configurations is so small that already several degrees below T_{c0} practically the whole of the sample become superconducting (lowest curve in Fig. 1)].

As pointed out already, a quantitative determination of the volume of the superconducting phase was made by measuring the value of χ_{dc}. The temperature dependence of the dynamic susceptibility obtained for samples $x = 6.78$ and $x = 6.68$ in the range $T < 60$ K yielded a much greater volume of the superconducting phase than that obtained from an analysis of χ_{dc}, irrespective of the frequency ω in the range from 300 Hz to 8 kHz; in the range of higher temperatures the two susceptibilities agreed. These results can be explained on the basis of a pattern of superconducting regions separated by Josephson junctions and bearing in mind the phenomenon of creep in a Josephson junction. If we assume that the characteristic frequencies of vortex creep in a Josephson junction are considerably less than the frequency of the dynamic experiments (but higher than the reciprocal of the time in static measurements, $\approx 10^{-2}$ Hz), then in the presence of an alternating magnetic field the Josephson junction becomes a "harder" superconductor than in a constant field. Therefore, in an alternating magnetic field a network of superconducting regions joined by Josephson junctions extends over a large part of the volume. Heating causes deterioration of the quality of the Josephson junctions and, consequently, it reduces the number of bonds between the superconducting regions. At some temperature ($T^* \approx 60$ K, see Fig. 1) a percolation limit is reached and at higher temperatures the separate superconducting regions behave independently of one another. A further increase in temperature reduces the total volume of the superconducting phase and this is now solely due to the scatter of the critical temperatures T_c of the individual regions. It is clear, that above the percolation temperature T^* the values of χ_{dc} and χ_{ac} coincide. For a sample with $x = 6.9$ the superconducting phase occupies practically the whole volume so that we have $T_{c0} \approx T_c$ and the effects discussed above are insignificant. In the case of a sample with $x = 6.55$ the concentration of the superconducting regions is insufficient for the percolation process at any temperature, as indicated by a considerable proportion of the nonsuperconducting tetragonal phase (Table I).

The results of our dynamic experiments carried out using a weak (20 Oe) static magnetic field H can be explained simply by allowing for the fact that an increase in H reduces strongly [exponentially, see Eq. (13)] the critical current through a Josephson junction. Consequently, as in the case of an increase in temperature, these junctions deteriorate and we are again dealing with a pattern of isolated superconducting regions. Clearly, we then have $\chi_{dc} = \chi_{ac}$.

We shall now discuss the dependences of the magnetic moments of samples on a constant magnetic field H. In the range of weak fields ($H \lesssim 100$ Oe) only the sample with $x = 6.9$ exhibited a magnetic hysteresis loop. The absence of a small loop for samples with $x < 6.9$ could be explained either by a weak pinning force in the Josephson junctions (and, consequently, a reduction in the peak-to-peak amplitude of the hysteresis to a value no longer detectable) or by a high rate of creep of vortices inside the junctions. In a sample with $x = 6.9$ the pinning forces were clearly quite strong (and the rate of creep was low), so that in fields up to ≈ 100 Oe the current through the Josephson junctions screened a large part of the volume. Equation (13) and the experimental values of $M_0(H)$ [see Eq. (4)] can be used to estimate the characteristic size of the defect responsible for pinning. One should then bear in mind that $M_0(H) \propto J_c(H) \propto P(H)$. Substitution of $\alpha = 1$ and $d = 2\lambda = 4\times 10^3$ Å gives $l = 1.5\times 10^4$ Å. This estimate is quite reasonable because it is of the same order of magnitude as the linear dimensions of grains composing our samples. In other words, it is quite likely that the Josephson junctions and the pinning centers in samples with $x = 6.9$ appear at the boundaries between the individual grains, i.e., the superconducting phase in a sample with $x = 6.9$ occupies practically the whole of the inner volume of a grain.

Equations (3) and (4) make it possible to estimate the critical current J_c through a Josephson junction from the peak-to-peak amplitude M_0 of a hysteresis loop (Fig. 2). Assuming that $R \approx 1$ mm (which was the size of the sample) we find that $J_c \approx 10^3$ A/cm^2, which is in good agreement with the published values of J_c obtained for ceramic samples.[7]

In the case of the reversible part of the magnetization $M_e(H)$ (representing the sum $M_+ + M_-$) we find that the magnetization is governed only by the total magnetizations of the individual grains when vanishingly low transport currents flow through the Josephson junctions and when the individual grains can be regarded as isolated. The experimental results confirmed this prediction, because the slope of the reversible part of the experimental curve $M(H)$ (Fig. 2a) obtained in the range 1000–300 Oe was very nearly equal to $-1/4\pi$ if we introduce a correction for the porosity (see Ref. 2).

In strong magnetic fields there is again a hysteresis of the magnetization. An investigation of its dependence on the magnetic field H and determination in this way of $J_c(H)$ have been the subject of many papers (see, for example, Ref. 8). We shall consider briefly the question of the dependence of J_c on the oxygen content x.

The hysteresis loop observed in high fields is due to the pinning of those vortices which penetrate into the superconducting regions. Since the peak-to-peak amplitude of the hysteresis loop is proportional to J_c, it follows from the results in Fig. 3 that lowering of x rapidly reduces the critical current density in grains. An estimate of $J_c(H)$ obtained on the basis of Eqs. (3) and (4) gives, for example, $J_c(x = 6.9) = 2.4\times 10^7$ A/cm^2, $J_c(x = 6.78) = 3\times 10^6$ A/cm^2, $J_c(x = 6.68) = 7.5\times 10^5$ A/cm^2 in a field $H = 10$ kOe; the corresponding values for $H = 30$ kOe are $J_c(x = 6.9) = 1.5\times 10^7$ A/cm^2, $J_c(x = 6.78) = 2.1\times 10^6$ A/cm^2 and $J_c(x = 6.68) = 4.5\times 10^5$ A/cm^2. (In these estimates the value of R was assumed to be constant and equal to 10^4 Å, because a reduction in the amount of the superconducting phase by a factor of 2 changed R by just 30%.)

It is clear from all the above predictions that a reduction in the amount of oxygen (by quenching the samples in liquid nitrogen from various annealing temperatures) not only lowers the quality of the junctions, but decreases the dimensions of the superconducting regions and results in deterioration of the current-carrying capability of these regions, whereas a reduction in x reduces the vortex-pinning force in the same regions.

The proposed scheme thus makes it possible to explain largely the formation of superconducting properties of the investigated materials as a function of the oxygen content. The disagreement between the volumes of the superconducting phase and the total volume of the orthorhombic samples with compositions in the range $x < 6.9$ and the scatter of the values of T_c for different superconducting regions are very interesting results. A similar scatter of T_c observed for samples with $x < 7$ was discovered recently[9] in experiments on single crystals, so that one could assume that it is not related directly to the ceramic nature of our samples. The disagreement between the volumes and the scatter of T_c can be explained in a natural manner by postulating a cluster distribution of oxygen in a crystal. Such an inhomogeneity of the oxygen concentration can be in the form of random large-scale (in any case larger than the coherence length) fluctuations or it may be the result of the presence of twinning planes. The actual physical mechanism of the appearance of superconducting regions will require further studies.

[1)]The authors are grateful to A. R. Kaul' and I. É. Graboĭ for supplying the investigated sample.

[2)]After the present paper was sent to press, we became acquainted with a preprint reporting the work of S. Senoussi, M. Oussena, J. Bieri, J. Arabski, and R. Reich, where this was done to explain static magnetic hysteresis loops of a sample with $x \approx 6.9$.

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