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ORDER, DISORDER, AND PHASE TRANSITION IN CONDENSED SYSTEMS

Mechanism of the Hysteretic Behavior of the Magnetoresistance of Granular HTSCs: The Universal Nature of the Width of the Magnetoresistance Hysteresis Loop

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Abstract—The hysteretic behavior of the magnetoresistance $R(H)$ of granular high-temperature superconductors (HTSCs) of the Y–Ba–Cu–O, Bi–Ca–Sr–Cu–O, and La–Sr–Cu–O classical systems is investigated for transport current densities lower and higher than the critical density (at $H = 0$). All systems exhibit universal behavior of the width of the magnetoresistance hysteresis loop: independence of transport current under identical external conditions. This means that flux trapping in HTSC grains is the main mechanism controlling the hysteretic behavior of the magnetoresistance of granular HTSCs, while pinning of Josephson vortices in the intragranular medium makes no appreciable contribution to the formation of magnetoresistance hysteresis (when transport current flows through the sample). Experimental data on relaxation of residual resistance after the action of a magnetic field also confirm this conclusion.

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1. INTRODUCTION

It is well known that granular high-temperature superconductors (HTSCs) exhibit a hysteretic behavior of magnetoresistance $R(H)$ and critical current $I_c(H)$ in an external magnetic field H [1–7]. Such a behavior was explained on a qualitative level [3, 6, 7]; however, the mechanisms of hysteresis are still being discussed [8–17], and the physical mechanisms of hysteretic behavior of the $R(H)$ and $I_c(H)$ dependences for granular HTSC systems have not been unambiguously established. A granular HTSC can be treated approximately as a two-level system [6], i.e., (1) a HTSC consisting of grains with high critical current densities ($j_{cg} \sim 10^5$–10^7 A/cm^2) and values of upper critical field H_{c2g}, which cannot be attained experimentally (at temperatures not very close to the critical value) and (2) an aggregate of weak Josephson links that form at natural grain boundaries, which determine the critical current density of the granular HTSC ($j_{cJ} \sim 10$–10^3 A/cm^2 at nitrogen temperature, while a lower critical field H_{c1J} may be a few oersteds or less than the magnetic field of the Earth [18]. A drop in voltage (dissipation) during the passage of transport current through a granular HTSC sample indicates that phase coherence is violated in the subsystem of weak links.

For this reason, from possible mechanisms leading to magnetoresistance hysteresis in granular HTSCs, one can mention (1) magnetic flux trapping in grain boundaries (Josephson vortex pinning), (2) the effect of flux trapped by HTSC grains on an intergranular medium, or (3) superposition of the above mechanisms. In [15], we proposed a method for distinguishing the contributions from these mechanisms. This method is based on the fact that the width of magnetoresistance hysteresis loop is proportional to the trapped magnetic flux whose variation is hampered by pinning. In the case when the effect of pinning of Josephson vortices in grain boundaries is significant, an increase in transport current (if it is at least on the same order of magnitude as the critical current) must lead to additional separation of Josephson vortices and ultimately to the dependence of the field width of the hysteresis loop on the current. If the magnetoresistance hysteresis originates only from the flux trapped by grains, the transport current cannot affect the vortices in HTSC grains in view of the obvious condition $I \ll I_{cg}$ (naturally, for temperatures and fields below the melting point of Abrikosov vortices on the H–T diagram).

In [15], the hysteretic dependences $R(H)$ for HTSC-based composites were studied; these are model granular HTSCs with artificially suppressed Josephson links. It was shown that the width of the magnetoresistance hysteresis loop for such systems is independent of transport current, indicating that the hysteresis emerges only due to magnetic flux trapping in HTSC grains. Analysis of the results from investigating the relaxation of the residual resistance of such HTSC composites also confirmed this conclusion [19]: time relaxation of residual resistance is controlled only by processes associated with vortices escaping from grains.

In our opinion, it is expedient to carry out similar experiments with "pure" HTSCs of classical compositions with standard transport properties for such systems. This leads to the conclusion on the mechanism of hysteretic behavior of magnetoresistance in granular HTSCs with natural grain boundaries.

The goal of this study is to establish the mechanism of the hysteretic behavior of granular HTSCs with compositions of $YBa_2Cu_3O_{7-\delta}$, $Bi_{1.8}Pb_{0.3}Sr_{1.9}Ca_2Cu_3O_x$, and $La_{1.85}Sr_{0.15}CuO_4$. Experimental data on hysteretic dependences $R(H)$ and the width of the magnetoresistance hysteresis loop, as well as time relaxation of residual resistance are obtained and analyzed for various transport current densities.

2. EXPERIMENT

Yttrium-, bismuth-, and lanthanum-based ceramics were synthesized using a standard solid-phase synthesis technique from the corresponding especially pure oxides and carbonates Y_2O_3, $BaCO_3$, CuO, Bi_2O_3, PbO, $SrCO_3$, $CaCO_3$, and La_2O_3. The Debye powder diagram for $YBa_2Cu_3O_{7-\delta}$ contains only reflections from the 1–2–3 structure. For $Bi_{1.8}Pb_{0.3}Sr_{1.9}Ca_2Cu_3O_x$ HTSC, X-ray diffraction analysis shows that the 2223 phase is dominating; analysis of reflections proved that the fraction of the 2212 phase is less than 5%. The Debye powder diagram for $La_{1.85}Sr_{0.15}CuO_4$ also contains reflections from this structure; no reflections from the initial ingredients are observed. We will henceforth denote the samples of yttrium, bismuth, and lanthanum ceramics as YBCO, BSCCO, and LSCO.

The results of scanning electron microscopy show that the samples have a clearly manifested granular structure. The average grain size was approximately 6 μm for YBCO, 8 μm for BSCCO, and 5 μm for LSCO. Superconducting transition temperature T_c determined from magnetic measurements was approximately 92 K for YBCO, 108 K for BSCCO, and 35 K for LSCO.

The temperature dependences of resistance in magnetic fields and magnetoresistance isotherms were measured using the standard four-point probe method. The parameters of the samples varied (the cross-sectional area was (0.5–1.0) × (0.5–1.0) mm^2 and the distance between the potential contacts was 1–2 mm). Magnetoresistance will be given below in milliohms, and the $R(H)$ dependences can be compared to the value of contribution R_{nJ} to the sample resistance from grain boundaries at T_c; the value of $R_{nJ}/R(T_c)$ is independent of the sample parameters and is a characteristic of the material. The field dependences of magnetoresistance $R(H) = U(H)/I$ were measured at a constant transport current I under zero field cooling ($\mathbf{H} \perp \mathbf{I}$). Measurements for YBCO and BSCCO samples were taken at 77.4 K; the samples were immersed in liquid nitrogen, which ensured effective removal of heat released in a contact for a transport current of 700 mA. The $R(H)$ dependences were measured at 4.2 K at a current of up to 50 mA (for $I > 50$ mA, sample heating was observed). After each measurement of $R(H)$, the sample was heated to above T_c. We will denote the external field by $H_\uparrow$ if it increases and by $H_\downarrow$ if it decreases.

Relaxation of remanent resistance $R_{\text{rem}}(t)$ (i.e., $R(H_\downarrow = 0, t)$ of the YBCO and BSCCO samples was measured after application of magnetic field in accordance with the following scheme: the sample was cooled in zero external field, the $R(H)$ dependence was measured during smooth variation in the external field to $H_\uparrow = H_{\max}$, then to $H_\downarrow = 0$, after which the $R_{\text{rem}}(t)$ dependence was recorded.

Magnetic measurements were taken on an automated vibration magnetometer [20]. Field dependences of magnetization and time relaxation of remanent moment $M_{\text{rem}}(t)$ were measured analogously to the $R(H)$ and $R_{\text{rem}}(t)$ dependences.

3. RESULTS AND DISCUSSION

3.1. Superconducting Transition of Granular HTSCs in an External Field as a Characteristic of Natural Josephson Junctions

The effect of Josephson junctions that form at grain boundaries of polycrystalline HTSCs is manifested in the structure of the resistive transition in a magnetic field. Figure 1 shows temperature dependences of resistivity $\rho(T)$ of the samples in various external magnetic fields. The beginning of the resistive transition coincides with temperature T_c determined from magnetic measurements (see Section 2). The sharp resistivity jump corresponds to the superconducting transition in HTSC crystallites. The smooth part of the $\rho(T)$ dependence is controlled by the transition of weak Josephson links and broadens in a magnetic field up to the temperature at which the resistance is instrumental zero. The curves in Fig. 1 can be used to determine "normal resistance" R_{nJ} of the Josephson network in a HTSC polycrystal. This is in fact the maximal possible magnetoresistance[1] associated with grain boundaries at $T < T_c$. The values of R_{nJ} for each specific sample are indicated below in the magnetoresistance curves.

The "strength" of Josephson links in a polycrystal can be estimated from the effect of the magnetic field on the smooth part of the resistive transition (depen-

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[1] Normal resistance of grain boundaries may depend on temperature and this dependence is determined by the type (metal or insulator) of boundaries; however, this does not change the results of this study.

dence $T_{c0}(H)$). The data presented in Fig. 1 and the critical current densities for investigated materials (j_c(77.4 K) ≈ 100 A/cm^2 for YBCO, j_c(77.4 K) ≈ 50 A/cm^2 for BSCCO, and j_c(4.2 K) ≈ 80 A/cm^2 for LSCO) and compared to the available data from the literature for similar systems [21–23] make it possible to characterize our samples as standard. It should be noted that a stronger effect of the external field on the resistive transition in the network of weak links is observed for yttrium-based HTSC composites [24, 25] and bismuth-containing composites [26], as well as in polycrystals annealed in an inert atmosphere (with oxygen deficiency) [10, 27], in which the subsystem of Josephson junctions is artificially suppressed.

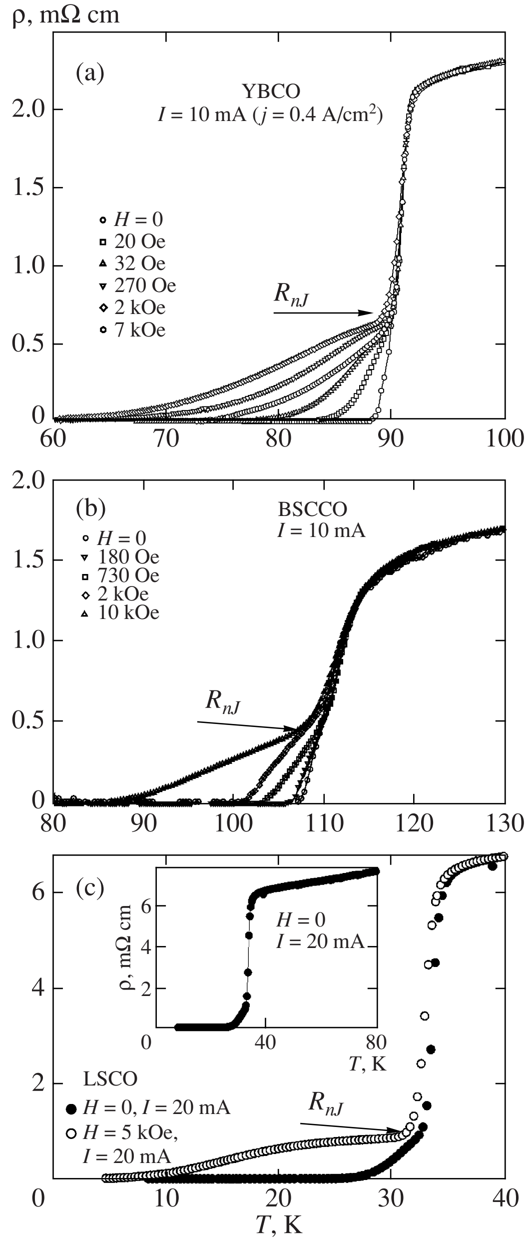

Fig. 1. Temperature dependences of resistivity of (a) yttrium, (b) bismuth, and (c) lanthanum ceramic samples for various values of external magnetic field H; the values of "normal resistance R_{nJ} of the Josephson network" (i.e., contribution from grain boundaries to resistivity; see Section 3.1) and transport current I are also indicated. The inset shows the behavior of the dependence for LSCO at $T > T_c$.

3.2. Hysteretic Dependences R(H) and Width of the Magnetoresistance Hysteresis Loop

Figure 2 shows the field dependences of magnetoresistance measured for various values of transport current I. All these dependences are characterized by a clearly manifested hysteresis. Let us briefly consider the premises for analysis of the dependence of the width of the magnetoresistance hysteresis loop on transport current [15].

For $H \geq H_{c1J}$, the external field penetrates the subsystem of grain boundaries in the form of hypervortices, which are transformed into Josephson vortices with increasing external field [8, 18]. As in the classical analysis of dissipation processes in type II superconductors with transport current, the Lorentz force modifies the profile of the coordinate function of pinning potential for Josephson vortices. This leads to creep (in this case, the pinning energy for Josephson vortices is much higher than thermal energy kT) or to flow of vortices (when the pinning energy is on the order of kT), which ultimately results in dissipation [21, 23, 28]. The magnetoresistance of a granular superconductor is controlled by these processes and depends on the transport current, as well as on effective field $\mathbf{B}_{\text{eff}}$ in the intergranular medium, which is a superposition of the external field and magnetic moments of adjacent grains. In turn, the magnetic moments of superconducting grains are controlled by the superposition of fields from Meissner currents and Abrikosov vortices (for $H > H_{c1g}$, where H_{c1g} is the field of the first penetration of vortices into grains). In the intergranular medium, we can expect that induced field will be enhanced due to the mutual effect of magnetic moments of neighboring grains, as well as demagnetizing factors associated with the geometry of grains [7]. Consequently, we can introduce proportionality factor α between the mean magnetic moments of grains and the field in the intergranular space induced by these moments. This quantity characterizes magnetic flux being compressed in the intergranular medium [10] and the averaged demagnetizing factor determined by the shape of grains [7]. The magnetic induction at the grain boundary averaged over all boundaries through which charge carriers tunnel for $\mathbf{H} \perp \mathbf{I}$, is defined as

$$\mathbf{B}_{\text{eff}}(\mathbf{H}) = \mathbf{H} + 4\pi\mathbf{M}(\mathbf{H})\alpha, \quad (1)$$

where $\mathbf{M}$ is the magnetization of the HTSC (in gausses). Strictly speaking, $\alpha = \alpha(H)$, where $\alpha(H_{\uparrow}) \neq \alpha(H_{\downarrow})$. If the magnetization of an HTSC (and, accordingly, of grains) is negative, the field induced in grain

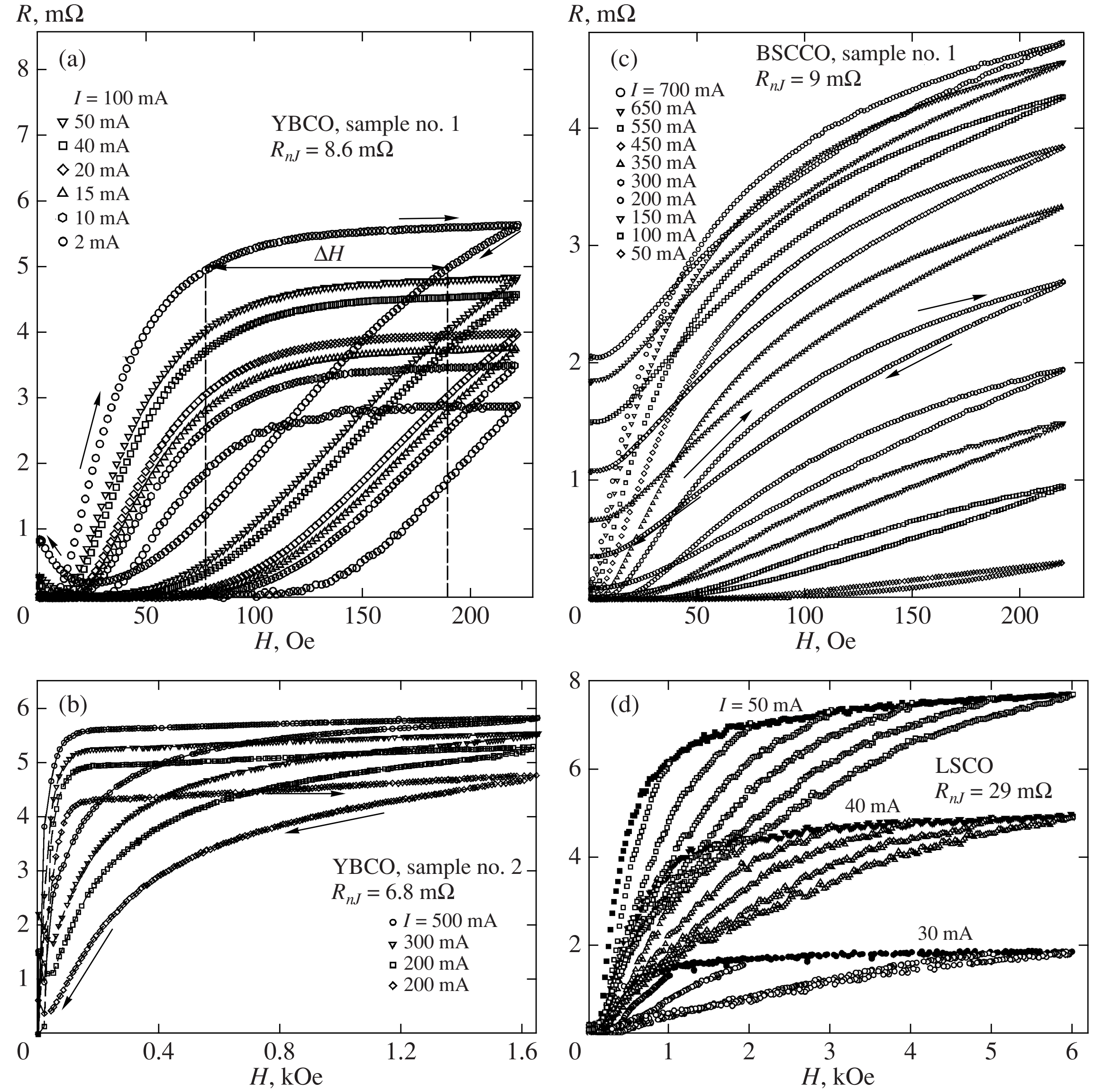

Fig. 2. Hysteretic dependences of the magnetoresistance of grains of HTSCs studied here for various values of transport current I: (a, b) various YBCO samples, $T = 77.4$ K; (c) BSCCO, $T = 77.4$ K; (d) LSCO, $T = 4.2$ K; dark and light symbols correspond to an increase and decrease in field. The value of R_{nJ} is determined from the $R(T, H)$ dependences in Fig. 1. Arrows indicate the direction of variation of the external field. An example of determining the field width $\Delta H = H_{\downarrow} - H_{\uparrow}$ of the magnetoresistance hysteresis loop (for $H_{\downarrow} = 180$ Oe) for $R = \text{const}$ is given.

boundaries, in which $\mathbf{H} \perp \mathbf{I}$, is parallel to the external field; if, however, $M > 0$, the induced field is antiparallel to the external field [15]. For $R(H_{\downarrow}) = R(H_{\uparrow})$, the effective fields at points $H_{\uparrow}$ and $H_{\downarrow}$ are identical and $B_{\text{eff}}(H_{\downarrow}) = B_{\text{eff}}(H_{\uparrow})$. Then width $\Delta H(H_{\downarrow}) = H_{\downarrow} - H_{\uparrow}$ of the magnetoresistance hysteresis loop can be determined from expression (1):

$$\begin{aligned}\Delta H(H_{\downarrow}) &= H_{\downarrow} - H_{\uparrow} \\ &= 4\pi M(H_{\downarrow})\alpha(H_{\downarrow}) - 4\pi M(H_{\uparrow})\alpha(H_{\uparrow}).\end{aligned} \tag{2}$$

If the value of $B_{\text{eff}}(H)$ is determined only by the external field and magnetization of grains as proposed above, the right-hand side of relation (2) must be independent of transport current I (the value of I is several orders of magnitude smaller than I_{cg}). If $\Delta H = \text{const}$ in experiments with various values of I, the effect of magnetic flux trapping in the intergranular medium is negligibly small. If, however, the total magnetic induction in the intergranular medium is the sum of three comparable contributions (of the external field and the fields induced by the magnetic moments of grains and pinned Josephson vortices), the contribution of the latter vortices to B_{eff} will change upon a variation of transport current. Such a behavior of the system of vortices in a magnetic field with transport current is expected since, in accordance with prevailing concepts [29], vortices will be detached from pinning centers upon an increase in transport current for $I \sim I_c$, and the contribution of Josephson vortices will decrease. In this case, $\Delta H(H_\downarrow)$ is a function of transport current.

Figure 3 shows the $\Delta H(H_\downarrow)$ dependences for the results presented in Fig. 2. To within the error in experimental data (which is determined by the error in the fixation of the maximal applied field $H_{\max}$ as well as by the error of measurement of H and R), we did not observe any effect of variation of transport current on the field width of the hysteresis loop $R(H)$. For YBCO, measurements were taken for currents of I = 10–150 mA (in a field of up to 220 Oe) and I = 100–500 mA (in a field of up to 1600 Oe), i.e., practically to I_c (I_c(77.4 K) ≈ 600 mA). Such a behavior is typical of relatively weak fields (200 Oe), as well as for fields of up to 1600 Oe. For BSCCO, the field width of the hysteresis loop is independent of current for $I < I_c$ as well as for $I > I_c$ (I = 50–700 mA, I_c(77.4 K) ≈ 400 mA) in the range of weak fields (up to 220 Oe) as well as strong fields of up to 2 kOe (the range of strong fields is not depicted in the figure since the magnetoresistance hysteresis loop is fairly narrow). As regards the LSCO sample, we managed to take measurements at a temperature of 4.2 K in a narrow range of transport currents I = 30–50 mA (I_c(4.2 K) ≈ 300 mA); in this case, the width of the magnetoresistance hysteresis loop is also independent of I (for this sample, the $R(H)$ dependences were measured up to various values of $H_{\max}$).

Thus, parameter $\Delta H(H_\downarrow)$ is independent of transport current not only for HTSC composites with suppressed Josephson links [15], but also in granular HTSCs with Josephson links formed by natural grain boundaries. The observed behavior can be treated as sound proof that the hysteretic behavior of the magnetoresistance of granular HTSCs is mainly controlled by the flux trapped in superconducting grains. Parameter $\Delta H(H_\downarrow)$ (field width of the magnetoresistance hysteresis loop) can be regarded as universal to a certain extent. It is controlled not only by external conditions for the sample (temperature and maximal applied field), but also intragranular pinning since, in accordance with formula (2), the value of $\Delta H(H_\downarrow)$ is proportional to the difference between the forward and backward magnetizations (and, hence, to intergranular pinning).[2]

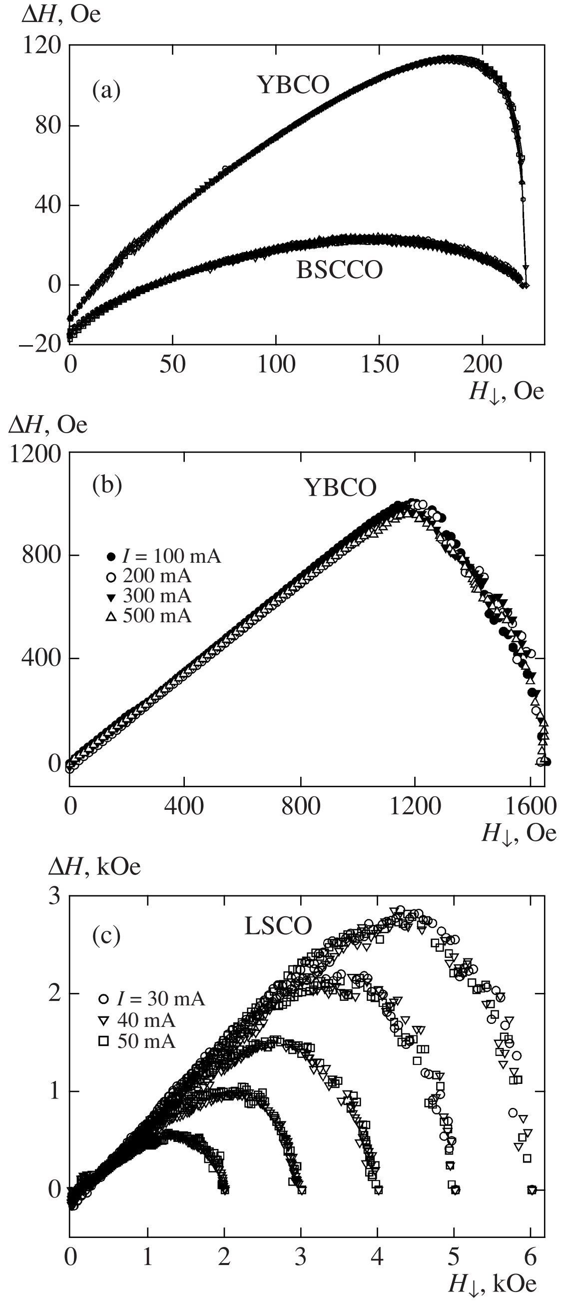

Fig. 3. Field width $\Delta H = H_\downarrow - H_\uparrow$ of the hysteresis loop, determined from the data in Fig. 2 in accordance with condition R = const for various values of transport current I: (a) YBCO and BSCCO, $H_{\max}$ = 220 Oe, T = 77.4 K (values of I are the same as in Figs. 2a and 2b); (b) YBCO, $H_{\max}$ = 1640 Oe, T = 77.4 K; (c) LSCO, T = 4.2 K, $H_{\max}$ = 2, 3, 4, 5, and 6 kOe.

We can specify the experimental conditions in which the magnetoresistance hysteresis loop is presumably not independent of the transport current: (1) at temperatures close to the superconductor transition temperature and in fields for which the transport current cannot affect the vortices inside grains (above the melt-

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[2] A few remarks concerning coefficient α in formula (1) are appropriate here. The value of effective field $B_{\text{eff}}(H)$ in the intergranular medium can be estimated by substituting into expression (2) the experimental values of magnetization $M(H\uparrow)$ and $M(H\downarrow)$ (under the same experimental conditions (T, $H_{\max}$)) for the same sample and comparing the right-hand side of formula (2) with the value of ΔH given in Fig. 3. It was found that to satisfy condition (2), the value of a must noticeably differ from unity. This is indirect experimental proof that the magnetic flux is concentrated in the intergranular medium since diamagnetism in grains is much stronger and magnetic flux compression indeed takes place in the intergranular layers [10].

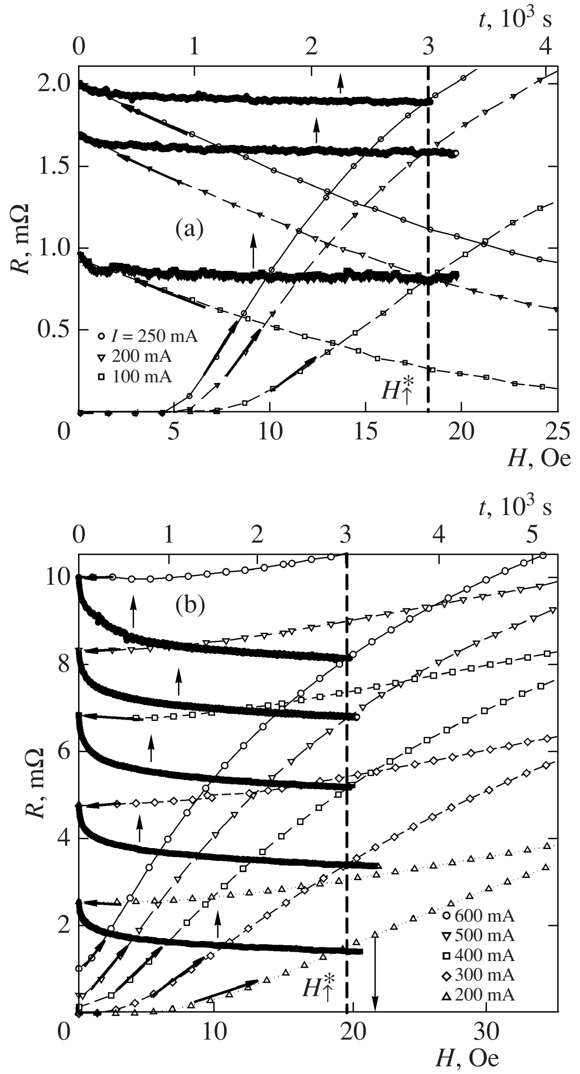

Fig. 4. Initial segments of the hysteretic dependences of magnetoresistance for (a) YBCO sample no. 3 and (b) BSCCO sample no. 2 at $T = 77.4$ K, $H_{\max} = 250$ Oe and for various values of transport current I. Arrows along the curves indicate the direction of variation of the external field. Results of time relaxation of remanent resistance $R_{\text{rem}}(t)$ are given on the same scale. The scales for t and H are chosen so that the $R_{\text{rem}}(t)$ and $R(H_{\uparrow})$ dependences for the same transport current intersect at $t = 3000$ s. Vertical dashed lines indicate that the intersection of the dependences $R_{\text{rem}}(t = 3000\text{ s})$ with $R(H_{\uparrow})$ takes place for $H_{\uparrow}^{*} = 18.2$ Oe for YBCO and $H_{\uparrow}^{*} = 19.5$ Oe for BSCCO, which are independent of transport current (see Section 3.3 for details).

ing point of the lattice of Abrikosov vortices); (2) for high values of transport current density ($j \sim 10^3$ A/cm^2, $I \approx 1$–3 A for the standard size of the sample), when the field produced by the current itself makes a significant contribution to the effective field [4]; and (3) if the critical parameters (j_c and pinning energy) of the Josephson medium are on the same order of magnitude with those for grains.

3.3. Relaxation of Remanent Resistance with Time

The independence of the width of the magnetoresistance hysteresis loop of the transport current also indicates that for $H_{\downarrow} = 0$, the effective field in the intergranular medium is controlled by the field induced by the magnetic moments of grains. The magnetization of a superconductor (including $M_{\text{rem}} = M(H_{\downarrow} = 0)$ relaxes with time. The effective field in the intergranular medium also decreases, and remanent resistance R_{rem} also relaxes with time, which was observed in a number of experiments [5, 6, 8, 13, 19, 27].

The value of R_{rem} for a certain value of t^* can be compared to the $R(H_{\uparrow})$ dependence. Condition $R_{\text{rem}}(t = t^*) = R(H_{\uparrow} = H_{\uparrow}^{*})$ indicates that effective fields in the intergranular medium are the same for $H_{\downarrow} = 0$, $t = t^*$ and $H = H_{\uparrow}$: $B_{\text{eff}}(H_{\downarrow} = 0, t^*) = B_{\text{eff}}(H_{\uparrow})$. In this case, the value of $\Delta H(H_{\downarrow} = 0, t^*)$ determined from expression (2) is equal to $-H_{\uparrow}^{*}$, where $H_{\uparrow}^{*}$ is the field in which $R(H_{\uparrow}) = R_{\text{rem}}(t = t^*)$. If the independence of quantity $\Delta H(H_{\downarrow} = 0, t)$ of the transport current does not change with time, this additionally confirms that pinning of Josephson vortices does not affect the hysteretic behavior of magnetoresistance and relaxation of the remanent resistance.

Figure 4 shows the $R(H)$ dependences for YBCO and BSCCO samples in the range of weak fields; the value of $H_{\max}$ is 250 Oe for both samples. In addition, this figure shows the field dependences of time relaxation for these samples, measured after the application or removal of the external field. It turns out that condition $R_{\text{rem}}(t = t^*) = R(\text{H}_{\uparrow} = H_{\uparrow}^{*})$ holds for the same values of field $H_{\uparrow}^{*}$, which is independent of transport current. Figure 4 illustrates this situation for $t^* = 3000$ s; the H and t scales are adjusted so that the value of $t = 3000$ s coincides with field $H_{\uparrow}^{*}$, for which dependences $R(H_{\uparrow})$ and $R_{\text{rem}}(t = 3000\text{ s})$ intersect (i.e., the above condition $R_{\text{rem}}(t = t^*) = R(H_{\uparrow} = H_{\uparrow}^{*})$ holds). Such a behavior also takes place for other values of t^*. Comparison of dependences $R_{\text{rem}}(t)$ and $R(H_{\downarrow})$ leads to an analogous result.

In accordance with the results presented in Fig. 4 for $t = 3000$ s ($H_{\max} = 250$ Oe), the value of $H_{\uparrow}^{*}$ amounts to 18.2 Oe for the YBCO sample and 19.5 Oe for BSCCO. The independence of this quantity of transport current indicates that the hysteretic dependence of magnetoresistance, as well as time relaxation of remanent resistance, is controlled only by the field induced by magnetic moments of HTSC grains, while pinning of Josephson vortices does not affect these processes.

Magnetization relaxation of YBCO and BSCCO samples measured under analogous conditions amounts approximately to 5.5 and 18.0%, respectively, which is typical of these systems [30]. The $R_{\mathrm{rem}}(\ln(t))$ and $M_{\mathrm{rem}}(\ln(t))$ dependences are linear in these coordinates, which indicates the existence of thermoactivation processes of the emergence of Abrikosov vortices from grains [30, 31].

Thus, the observed relaxation of remanent resistance of the granular systems with the classical composition after the action of magnetic field is associated with the emergence of Abrikosov vortices from HTSC grains and, as a consequence, with a decrease in the effective field in the intergranular medium.

4. CONCLUSIONS

Analysis of experimental data on magnetoresistance $R(H)$ and relaxation of remanent resistance $R_{\mathrm{rem}}(t)$ after the action of a magnetic field, which were obtained in this study on granular HTSCs with classical compositions, leads to the conclusion that the dominant mechanisms controlling hysteretic dependence $R(H)$ and the behavior of $R_{\mathrm{rem}}(t)$ are magnetic flux trapping in grains and the effect of the magnetic moments of grains on the field in the intergranular medium. Pinning of Josephson vortices does not appreciably affect the hysteretic behavior of $R(H)$ and evolution of $R_{\mathrm{rem}}(t)$, while dissipation in granular HTSCs is controlled by the flow of Josephson vortices in the intergranular medium. Obviously, the effect of pinning of Josephson vortices is manifested in the case when the transport current is so small that the sample resistance itself in an external field is close to instrumental zero or when the sample does not carry a macroscopic transport current (e.g., in susceptibility measurements) [8]. The width of the hysteresis loop for granular HTSCs is a universal parameter independent of transport current, which characterizes intergranular pinning and magnetic flux compression in the intragranular medium.

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