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Transverse magnetoresistance of $YBa_2Cu_3O_x$ single crystals with different oxygen content

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The normal-state transverse magnetoresistance ($\Delta R/R_0$) has been studied in the *ab* plane of three $YBa_2Cu_3O_x$ single crystals with oxygen indices $x = 6.95$, 6.88, and 6.62. A linear quadratic-field dependence $\Delta R/R_0 = AH^2 + BH$ has been found up to 15 T in the temperature region where superconducting fluctuations are negligible. The linear component dominating in oxygen-deficient samples is probably related to the interaction between charge carriers and dynamic antiferromagnetic spin fluctuations in CuO_2 layers of $YBa_2Cu_3O_x$.

I. INTRODUCTION

In the last few years a lot of interest has focused on the coexistence of both the electron and spin subsystems in the CuO_2 layers of cuprate high-temperature superconductors (HTSC's). Studies of inelastic neutron scattering[1-5] in $La_{1-y}Sr_yCuO_4$ and $YBa_2Cu_3O_x$ single crystals have allowed observation of the transformation from long-range antiferromagnetism to the system of dynamic antiferromagnetically correlated spins (DACS's), as the density of charge carriers (n) in the CuO_2 layers increases. Thus the transition of $YBa_2Cu_3O_x$ at $x = 6.41$ to metallic conductivity (and superconductivity) is accompanied by the decreasing to zero of both the Néel temperature and the mean magnetic moment of the Cu^{2+} ions. With further increase of x (in the metallic phase) the magnetic correlation length ξ_m decreases monotonically and the spin excitation spectrum $\omega(k)$ is shifted to the high-frequency region.[4,5] These facts together with the non-Korringa behavior of ^{63}Cu-NMR spin-lattice relaxation[6] [the normal-state relaxation rate weakly depends on temperature at the planar Cu(2) sites] are interpreted as an effect of strong interaction between the charge-carrier and spin subsystems in the CuO_2 layers. What we are interested in here is a possible manifestation of this interaction in the transport and galvanomagnetic properties of cuprate HTSC's.

In the present study the transverse magnetoresistance $\Delta R/R_0$ of $YBa_2Cu_3O_x$ (YBCO) in the normal state is investigated. The existing studies of magnetoresistance of HTSC single crystals in most cases deal with the strong effects connected with the broadening of the resistive transition and suppression of superconductive fluctuations in a magnetic field (for example, Refs. 7–11). As for the normal-state region ($T > 1.5T_c$), the quantitative analysis of different contributions to $\Delta R/R_0$ is rather complicated because these effects are small (the *ab*-plane magnetoresistance $\Delta R/R_0$ for the yttrium system is 10^{-3}–10^{-4} in this temperature region in the field $H = 10$ T). In fact, the work of Osofsky and co-authors[12] is the only study known to us in which attempts at such analysis have been undertaken for single crystals of $YBa_2Cu_3O_7$ and $GdBa_2Cu_3O_7$. In particular, the transverse magnetoresistance of the $YBa_2Cu_3O_7$ system has been shown to be negative in low fields and to reverse sign in the field ~8 T, the dependence $\Delta R/R_0(H)$ being close to quadratic in high fields. The variation of the field dependence of magnetoresistance as related to change in the oxygen index was not investigated at all to our knowledge.

II. EXPERIMENT

Single crystals of $YBa_2Cu_3O_x$ were grown from solution in a melt of BaO-CuO eutectic in a platinum crucible under slow cooling from 1200 to 910 °C with subsequent decantation of the melt. The resulting plates with the thickness of ~20 μm were used for cutting out samples of $2 \times 0.2 \times 0.02$ mm^3 size. The *c* axis of the samples was oriented along the minimum size of the plate. The optimum oxygen concentration was achieved after annealing the samples in oxygen flow at 450 °C for 48 h. In this treatment the oxygen index x increased up to 6.95 and the oxygen distribution remained homogeneous; this was checked by high-resolution electron microscopy (for more details, see Ref. 13). The samples of compositions $YBa_2Cu_3O_{6.88}$ and $YBa_2Cu_3O_{6.62}$ were obtained by annealing the crystals saturated with oxygen in a flow of gaseous helium at 500 °C for 3 and 10 h, respectively. The characteristics of the superconducting transition for the prepared samples are presented in Table I.

Electric contacts to the samples were made by soldering indium wires to gold pads sputtered onto the surface of the crystals. The electric resistance was measured by the four-probe method, the current passing through the *ab* plane of the crystals.

To measure the transverse magnetoresistance, the samples were identically mounted on a rotary platform in the center of a superconducting solenoid generating a mag-

TABLE I. Temperatures T_c and widths ΔT_c (10%/90%) of superconducting transition of the samples studied.

Sample number	Oxygen content	T_c (K)	ΔT_c (K)
1	6.95	92.8	0.3
2	6.88	91.5	1.9
3	6.62	34.0	22.0

netic field up to 15 T. The angle φ between the magnetic field and the *ab* plane of the samples could be changed by rotation of the platform, whereas the current parallel to the axis of rotation always made the angle of 90° with the magnetic field. The values of both the magnetic field and the angle φ (which was varied from 0 to 90°) were determined by means of two Hall detectors.

The cell temperature was controlled by an automated system with a copper-Constantan thermocouple as a sensor. Temperature was measured with a resolution of 1 mK with a carbon resistance thermometer manufactured and calibrated in a magnetic field at the Central Institute of Committee of Standards (Moscow, Russia). The temperature gradient between the thermometer and the copper container with the samples was checked by a differential thermocouple. The field dependence of the sample resistivity $R(H)$ was taken under a slow (~ 0.005 T/S) sweep of the solenoid current. Improved precision of the measurements was achieved by means of a computerized data acquisition and processing system. The system ensured the evenness of parameter increments and made corresponding corrections. The inevitable temperature drift $\Delta T(H)$ due to increasing magnetic field (0.1–0.2 K for the maximum field) was measured at each point. The field dependence of magnetoresistance of the samples was reduced to the initial (without magnetic field) temperature by the expression

$$\Delta R/R_0 = [R(H) - \Delta T(H) dR/dT - R(0)]/R(0) .$$

III. RESULTS

The experimental dependence of magnetoresistance $\Delta R/R_0(H) = [R(H) - R(0)]/R(0)$ on the magnetic field for the angle $\varphi = 90°$ (magnetic field H perpendicular to the *ab* plane) at different temperatures is presented in Figs. 1(a)–1(c). For sample 1 (with the maximum oxygen content) the form of the $\Delta R/R_0(H)$ dependence is in qualitative agreement with the data from Ref. 12. For samples 2 and 3 the character of the curves changes noticeably. The results of approximation of the obtained data by the expression

$$\Delta R/R_0 = AH^2 + B|H| \quad (1)$$

are represented in Fig. 1 as solid lines. The A and B parameters determined by the rms method are presented in Table II.

It can be seen from Fig. 1 that the field dependences of the magnetoresistance are adequately described by expression (1). According to Table II, the relation between the linear and quadratic contributions to (1) depends sub-

TABLE II. Approximation parameters A, B, and R_0 of the magnetoresistance field dependencies in the expression $\Delta R/R_0 = AH^2 + BH$. Here $\Delta R/R_0(H) = [R(H) - R(0)]/R(0)$ is the experimental magnetoresistance (for magnetic field perpendicular to the *ab* plane of the crystals).

Sample number	T (K)	R_0 ($10^{-3}\Omega$)	A (10^{-6} T^{-2})	B (10^{-5} T^{-1})	A/B (T^{-1})
1	140.8	138.789	3.23	2.87	0.112
	120.2	116.387	9.47	3.94	0.240
2	140.8	128.874	2.60	38.62	0.0067
	120.2	114.942	8.04	55.88	0.0144
3	140.8	930.180	0.616	41.08	0.00150
	120.2	826.751	3.660	50.71	0.00722
	81.5	652.897	5.14	85.14	0.00604

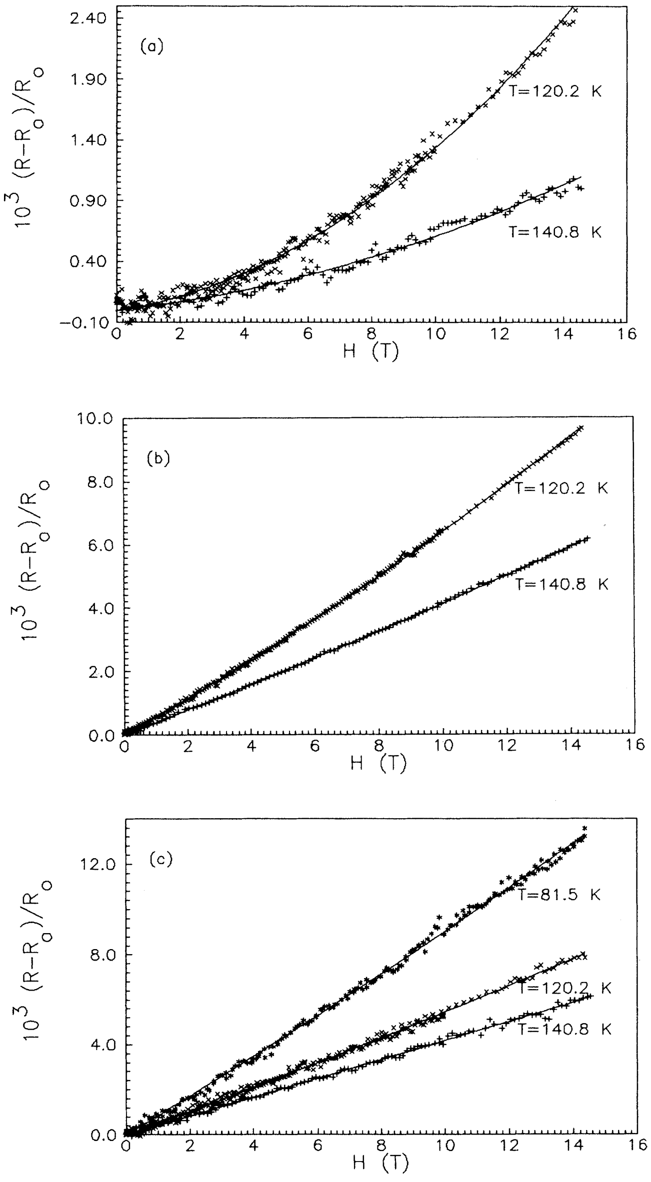

FIG. 1. Magnetoresistance vs magnetic field curves at $\varphi = 90°$ for samples (a) 1, (b) 2, and (c) 3.

stantially on the oxygen content of the samples. It should be noted that the sharp (by more than an order of magnitude) increase of the linear contribution when x decreases takes place within the narrow interval of the oxygen index $6.88 < x < 6.95$.

Results of measurements of the angle dependence $\Delta R/R_0(\varphi)$ for different fixed magnetic fields at $T = 140.8$ K are presented in Figs. 2(a)–2(c). The set of approximating curves is described by the expression

$$\Delta R/R_0 = (A_H + A_\varphi \sin^2\varphi)H^2 + (B_H + B_\varphi \sin\varphi)|H| \ . \qquad (2)$$

Results of the approximation are shown in Fig. 2 by solid lines, and the coefficients A_H, A_φ, B_H, and B_φ are presented in Table III.

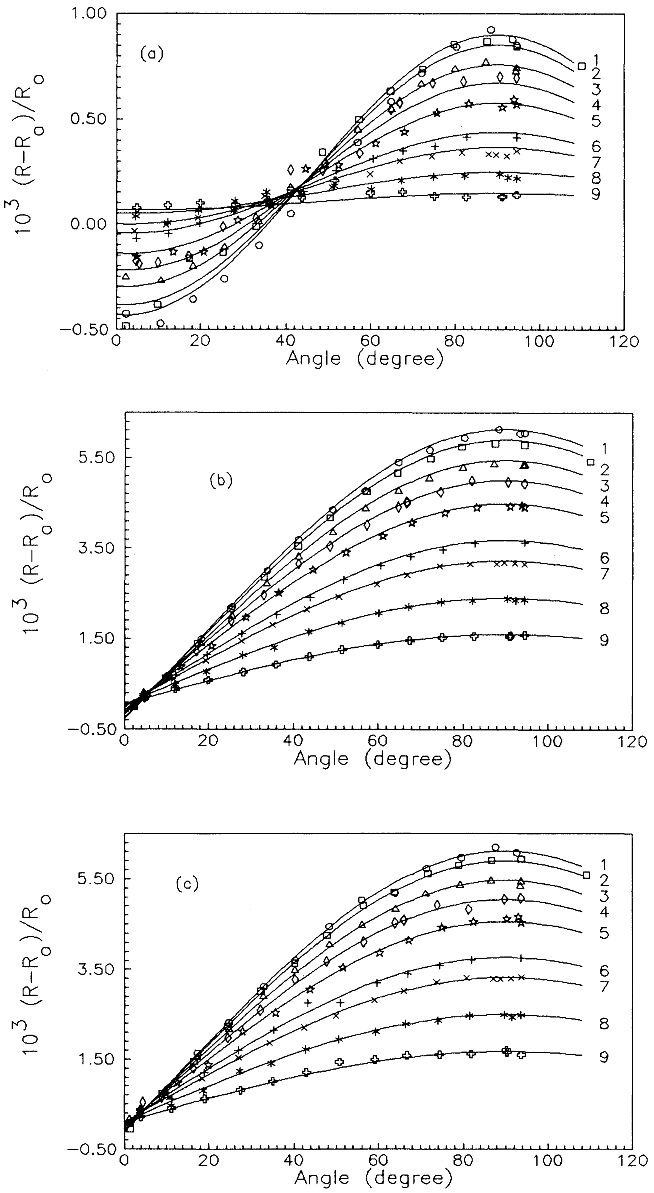

FIG. 2. The family of angular dependencies of magnetoresistance in magnetic fields at the temperature 140.8 K for samples (a) 1, (b) 2, and (c) 3. Intensity of magnetic fields (T): curve 1, 14.5; curve 2, 14.0; curve 3, 13.0; curve 4, 12.0; curve 5, 10.8; curve 6, 8.9; curve 7, 7.9; curve 8, 5.9; and curve 9, 3.9.

TABLE III. Approximation parameters A_H, A_φ, B_H, and B_φ of the family of magnetoresistance angular dependencies at $T = 140.8$ K in the expression $\Delta R/R_0 = (A_H + A_\varphi \sin^2\varphi)H^2 + (B_H + B_\varphi \sin\varphi)|H|$. Here $\Delta R/R_0 = [R(H) - R(0)]/R_0$ is the experimental magnetoresistance.

Sample number	A_H (10^{-6} T^{-2})	A_φ (10^{-6} T^{-2})	B_H (10^{-5} T^{-1})	B_φ (10^{-5} T^{-1})
1	−4.44	6.76	3.52	−0.694
2	−3.06	4.69	2.66	36.70
3	−3.02	2.70	3.59	38.80

The analysis of the data obtained shows that the difference in the angular dependencies of the magnetoresistance for different samples is connected mainly with the contribution linear in $\sin\varphi$. Indeed, according to Table III, the coefficient B_φ changes its sign and increases in size almost by two orders of magnitude when going from sample 1 to samples 2 and 3, while the other coefficients change considerably less. Thus the next changes in importance have been found in coefficient A_φ which differs for samples 1 and 3 by no more than the factor of 2.5. As to the isotropic contributions into (2) (A_H and B_H), they may be considered independent of x within the experimental error.

IV. DISCUSSION

The data obtained for the normal-state YBCO HTSC indicate the presence of an anisotropic component of magnetoresistance linear in magnetic field. This component is predominant for oxygen-deficient samples. Since the linear run of $\Delta R/R_0(H)$ is seldom found we have focused our interest on this contribution. Let us consider possible causes of its appearance. Our investigation was carried out in the low effective magnetic field range, where the Larmor radius of the charge carrier (r_h) is more than its mean free path (l). In such a case the one-zone approximation gives the dependence $\Delta R/R_0 \sim H^2$ for the magnetoresistance of metals without long-range magnetic order. The two-zone model gives a similar result ($\Delta R/R_0 \sim H^2$) provided the charge-carrier mobilities differ in different zones.[14] In the intermediate magnetic field ($r_h \sim l$), where peculiarities of the Fermi surface come into play, the field dependence of the magnetoresistance could look linear on a finite interval near the change from square dependence in the low field to saturation in the intermediate one. In our case a quasilinear interval of this type could be expected to appear first in the magnetoresistance of sample 1 with the greatest l and last for sample 3. However, that is not the case here. Besides, the linear components in $\Delta R/R_0$ of both the first and the second samples must be close to each other because their specific resistances (and consequently l) differ by no more than 10% in the temperature region under consideration. However, as can be seen from Table II, the coefficients B of the samples 1 and 2 differ by more than one order of magnitude.

For high-temperature superconductors, suppression of fluctuations of the superconductive order parameter by the external field can contribute markedly to the magnetoresistance. This contribution is most substantial near T_c, but it could also be notable at temperatures comparatively far from the transition point. Samples 1 and 2 have T_c values close to each other (92.8 and 91.5 K), so the contribution in question is expected to be approximately the same for both of them. The data obtained, however, do not show it to be the case. Besides, sample 3 with the transition temperature of only 34 K demonstrates the largest linear component of $\Delta R/R_0$, which is rather difficult to explain.

According to the observed anisotropy of the magnetoresistance one can conclude that the process under discussion takes place in a quasi-two-dimensional entity (this pertains first of all to the component of $\Delta R/R_0$ linear in field; see Table III). We assume the resistivity as well as the galvanomagnetic properties of the normal phase to be governed by the scattering processes of charge carriers in the CuO_2 layer. In such a case, the interaction of charge carriers with the system of magnetic moments formed by the *d* electrons of the Cu^{2+} ions can contribute markedly to the magnetoresistance of the YBCO HTSC. In accordance with the ideas developed in Ref. 15, the structure of *d*-electron spins can be described as a system of localized magnetic moments with intensive antiferromagnetic correlations; however, no long-range order is observed at a relatively high oxygen index ($x > 6.4$). Such a situation is not typical for three-dimensional magnetics in which the correlation region is limited by the vicinity of the ordering temperature. However, behavior of this kind could be realized for two-dimensional magnetic systems, since, according to the Mermin, Wagner, and Hohenberg theorem,[16] fluctuations prevent long-range order.

There are both theoretical (Turov and Shavrov[17]) and experimental[18] investigations of transverse magnetoresistance where the contribution to $\Delta R/R_0$ linear in field has been shown to be possible in metallic antiferromagnetics whose symmetry allows for the existence of weak ferromagnetism. In a recent work[19] such a contribution has been discovered in the magnetoresistance of the heavy-fermion system UPt_3 in which superconductivity and antiferromagnetism coexist. It has been found in Ref. 19 that the coefficient *B* of the linear component of $\Delta R(H)$ remains nonzero also above the Néel point, but it falls steeply in the short range of temperature where ξ decreases. This fact may show the Turov-Shavrov result to be true not only for the region of magnetic order but also (under specific conditions) for the region of fluctuations. In all likelihood, the same model could be used to explain the results of our study. However, as opposed to Ref. 19, in YBCO the magnetic correlation length ξ depends only weakly on temperature, but considerably on *x*, as demonstrated in Refs. 4 and 5. Indeed, the ratio ξ/a_0 falls from 2.4 to 0.8 as *x* increases from 6.5 to 6.9, whereas an increase of temperature up to 250 K does not result in a marked variation of ξ. Such behavior might be caused by the quasi-two-dimensionality of the magnetic system as well as by strong electron-spin interactions in the CuO_2 layer.[20]

Finally, we would like to offer a possible explanation of the sharp decrease of the linear contribution in $\Delta R/R_0$ as the oxygen index approaches 7. As *x* increases, there occur a simultaneous decrease of the correlation radius and an increase of the characteristic frequency of spin fluctuations ω. According to Ref. 21,

$$\omega = \Gamma/\beta^{1/2}\pi(\xi/a_0)^2 , \tag{3}$$

where $\beta = \pi^2$, $\Gamma \sim 0.4$ eV, and $a_0 \sim 3.8$ Å is the lattice parameter. Based on (3) as well as the data on the variation of ξ versus the oxygen index,[4,5] we can estimate that in the interval $6.5 < x < 6.9$ ω changes in the range of $10^{13}-10^{14}$ Hz. On the other hand, the characteristic frequencies of charge carriers for $YBa_2Cu_3O_x$ are $\Omega \sim k_F v_F \sim 10^{14}-10^{15}$ Hz (v_F and k_F being the Fermi velocity and quasimomentum). For the case of $\omega \ll \Omega$ and $(\xi/a_0) > 1$, the conduction electrons respond to such a dynamic magnetic system as to a steady long-range-ordered antiferromagnetic state. As *x* approaches 7 and (ξ/a_0) approaches 1, ω approaches Ω, enhancing "resonantly" the interaction of charge carriers with DACS's. As a consequence, extra degrees of freedom of the spin system are excited and the Turov-Shavrov effect disappears.

V. CONCLUSIONS

The field dependences of transverse magnetoresistance in the *ab* plane of single crystals of $YBa_2Cu_3O_x$ with different oxygen content have been shown to contain contributions both square and linear in magnetic field. The linear contribution dominates in oxygen-deficient samples and it drops sharply as the oxygen content approaches 7. Different possible models of the discovered effect were considered. The interaction of charge carriers with antiferromagnetically correlated magnetic moments of Cu^{2+} ions in the CuO_2 layers was suggested as the most probable cause of this effect.

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