Anisotropy of the hopping magnetoresistance of antiferromagnetic $La_2CuO_{4+\delta}$ single crystals

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The anisotropy of the hopping conductivity of antiferromagnetic $La_2CuO_{4+\delta}$ single crystals with $T_N \approx 188$ K is investigated in the temperature range 5–295 K and the anisotropy of their magnetoresistance, in the temperature range 5–55 K. The resistance is measured by the Montgomery method for different combinations of directions of the transport current and magnetic field relative to the crystallographic axes. For the case when the field and transport current are directed parallel to the CuO_2 layers, a transition from negative to positive magnetoresistance is observed when the temperature is raised to $T \approx 20$ K. For fields perpendicular to the CuO_2 layers, only negative magnetoresistance is observed. The nature of the positive magnetoresistance is discussed. It is shown that the effect is most likely due not to the interaction of the spin of the charge carriers with the surrounding magnetic medium but to the orbital motion of these carriers. The corresponding values of the positive magnetoresistance and its behavior as a function of magnetic field and temperature are found using the well-known model of Shklovskii and Efros, which is based on taking into account the compression of the impurity wave functions of the charge carriers in the magnetic field. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1542472]

1. INTRODUCTION

The unique magnetic and conducting properties of $La_2CuO_{4+\delta}$ oxides with an excess of oxygen have been attracting considerable attention in the last 15 years, since the discovery of high-temperature superconductivity in perovskite copper oxides. The stoichiometric oxide La_2CuO_4 ($\delta = 0$) is an antiferromagnetic (AF) insulator with a Néel temperature T_N of around 300–320 K.[1–3] Saturation with excess oxygen ($\delta > 0$) gives rise to charge carriers (oxygen holes) and leads to suppression of the AF order (lowering of T_N). At a sufficiently high value of δ (>0.05) the system becomes a metal and, below 35–40 K, a superconductor. The crystal lattice of $La_2CuO_{4+\delta}$ belongs to the family of layered perovskite lattices. For $T<530$ K the lattice is orthorhombic. In the symmetry group $Bmab$ the **c** axis is perpendicular to the CuO_2 layers, while the **a** and **b** axes are parallel to them.[3] The excess oxygen is found in the interstices between CuO_2 layers. The conductivity of the system is governed by holes in the CuO_2 layers, and the magnetic state is determined by the spin of the copper ions Cu^{2+} ($S=1/2$). At a sufficiently low value of δ (i.e., in the insulating state) antiferromagnetic ordering is observed within the CuO_2 layers. The magnetic interaction between layers is extremely weak. It has been established that all of the spins in the CuO_2 layers are rotated by a small angle from the plane of the layer ($\approx 0.17°$).[1] The CuO_2 layers therefore have small ferromagnetic moments, which undergo AF ordering in the direction perpendicular to the layers for $T<T_N$ (Ref. 1).

For layered perovskite compounds one might expect: 1) quasi-two-dimensional behavior of the conductivity (including the hopping conductivity); 2) substantial anisotropy of the conductivity for directions perpendicular to and parallel to the CuO_2 planes. The first of these expectations is not confirmed by experiment. It has been found[4,5] that for insulating $La_2CuO_{4+\delta}$ crystals at sufficiently low temperatures the resistance has a temperature dependence of the form

$$R(T) \propto \exp[(T_0/T)^{1/4}], \qquad (1)$$

corresponding to the case of three-dimensional hopping conductivity with a variable hopping length (VHLC). This means that the charge carriers execute hops not only within the CuO_2 layers but also between them. In addition, a significant anisotropy of the hopping conductivity has been observed in high-quality samples:[2,6] the conductivity σ_{ab} in the direction parallel to the CuO_2 layers is one to two orders of magnitude higher than the conductivity σ_c in the direction perpendicular to the layers.

The known studies of the magnetoresistance of $La_2CuO_{4+\delta}$ in the hopping conductivity regime cannot be considered exhaustive.[1,7,8] A positive magnetoresistance should be expected for VHLC.[9] However, only negative magnetoresistance has been observed for $La_2CuO_{4+\delta}$. The case when the magnetic field **H** and transport current **J** are perpendicular to the CuO_2 layers (i.e., $\mathbf{H}\|\mathbf{c}$ and $\mathbf{J}\|\mathbf{c}$) has been studied in detail. Under such conditions one observes a rather sharp decrease of the resistance in the vicinity of certain characteristic fields H_c, which depend on the temperature.[1,7,8] For fields far from the value H_c the magnetoresistance is negligible (for $H \ll H_c$) or has a quite large constant value (for $H \gg H_c$). The value of H_c is maximum (around 50 kOe) for $T \to 0$. With increasing temperature the value of H_c decreases, and the decrease is especially rapid near T_N. This effect is absent above T_N. All of this indicates that the magnetoresistive effect in this case is due to changes of the magnetic state of $La_2CuO_{4+\delta}$ under the influence of the external field. Indeed, it has been shown[1,10] that a sufficiently high magnetic field leads to a transition of $La_2CuO_{4+\delta}$ from the AF to a weak ferromagnetic (WF) state, in which the ferromagnetic moments of the CuO_2 layers are parallel. Possible magnetoresistance models involving this transition are considered in Refs. 7 and 10.

The information available on the behavior of the magnetoresistance for other combinations of magnetic-field and transport-current directions is extremely spotty and often contradictory. It was found in Ref. 1 that the magnetoresistance is close to zero for fields parallel to the CuO_2 layers (i.e., for $\mathbf{H} \perp \mathbf{c}$) for $T > 20$ K (for transport currents both parallel and perpendicular to the CuO_2 layers). At the same time, in Ref. 7 a rather significant negative magnetoresistance was observed for $\mathbf{H} \perp \mathbf{c}$ (for currents perpendicular to the CuO_2 layers), which can be attributed to a spin-flop (SF) transition at sufficiently high fields ($H > 100$ kOe).[7,10] Since the spin-flop transition that occurs with increasing field parallel to the CuO_2 layers is continuous (with no jump in magnetoresistance at the transition), it can have a noticeable influence on the magnetoresistance even in fields below 50 kOe.[7] We note that in the published studies known to us only a negative magnetoresistance is observed, and only spin mechanisms of hopping magnetoresistance have been invoked to explain it.[10] On the whole it can be stated that the features and mechanisms of the magnetoresistance in $La_2CuO_{4+\delta}$ crystals in the hopping conductivity region have not been studied in sufficient detail, and it therefore seems that further studies are urgently needed.

In this paper we present the results of a study of the anisotropy of the hopping conductivity and magnetoresistance of $La_2CuO_{4+\delta}$ single crystals. The samples were antiferromagnetic, with $T_N \approx 188$ K. The conductivity in zero magnetic field was measured in the interval 5–295 K, and the magnetoresistance was measured in the interval 5–55 K. The magnetoresistance was measured for different combinations of directions of the magnetic field and transport current relative to the principal crystallographic axes. For the case when both the field and the transport current are directed parallel to the CuO_2 layers, one observes a transition from negative to positive magnetoresistance with increasing temperature. For fields perpendicular to the CuO_2 layers one observes only negative magnetoresistance. We know of no previous published reports of positive magnetoresistance of $La_2CuO_{4+\delta}$ crystals in the AF state. These results suggest that this effect is due not to spin but to orbital processes (compression of the wave functions of the charge carriers in a magnetic field, in accordance with the model of Ref. 9).

2. SAMPLES AND EXPERIMENTAL TECHNIQUES

The initial material for preparing the samples was a $La_2CuO_{4+\delta}$ single crystal (with $T_N \approx 230$ K), the conducting

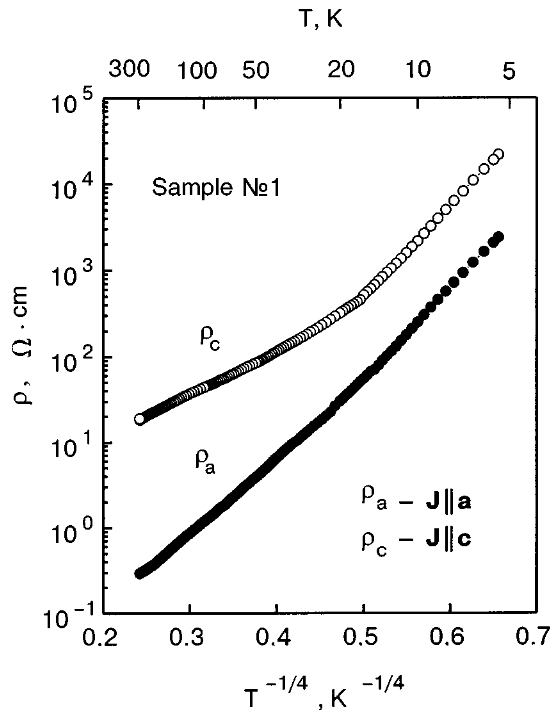

FIG. 1. Anisotropy of the hopping conductivity of the $La_2CuO_{4+\delta}$ crystal. The measurements of the resistivities ρ_a and ρ_c were done for directions of the measuring current (100 μA) along the crystallographic axes **a** and **c**.

properties of which are described in Ref. 5. Two samples in the form of parallelepipeds with dimensions of $1.3 \times 0.3 \times 0.39$ mm (sample No. 1) and $0.75 \times 0.3 \times 0.29$ mm (sample No. 2) were cut from it. The crystallographic orientation of the samples was determined by an x-ray method. The samples were subjected to a prolonged homogenizing anneal in an oxygen atmosphere (400 °C, 7 days). As a result of the annealing the oxygen content increased somewhat, so that the resistivity ρ of the samples decreased by approximately two orders of magnitude, and the Néel temperature T_N (determined from measurements of the temperature dependence of the magnetic susceptibility) decreased to 188 K. Here the samples remained in an insulating state and possessed hopping conductivity. Measurements showed that the two samples had essentially the same resistive and magnetoresistive characteristics. The resistivity was measured by the Montgomery method,[11] which permits a more reliable study of the anisotropy of the conductivity than does to usual four-contact method. Contacts between the measuring wires and samples were made using a conducting silver paste with a subsequent high-temperature annealing of the samples with the contacts. The following directions of the measuring current were used: $\mathbf{J} \| \mathbf{a}$ and $\mathbf{J} \| \mathbf{c}$ (sample No. 1) and $\mathbf{J} \| \mathbf{a}$ and $\mathbf{J} \| \mathbf{b}$ (sample No. 2). The magnetic fields were directed parallel to the **b** or **c** axis and always perpendicular to the current.

3. RESULTS AND DISCUSSION

The temperature dependence $\rho(T)$ for sample No. 1 (under conditions such that Ohm's law holds) is shown in Fig. 1 for different directions of the measuring currents relative to the crystallographic axes. It is seen that Mott's law for VHLC, i.e., Eq. (1), holds for $T \geq 20$ K. No noticeable change in the behavior of $\rho(T)$ near the Néel temperature

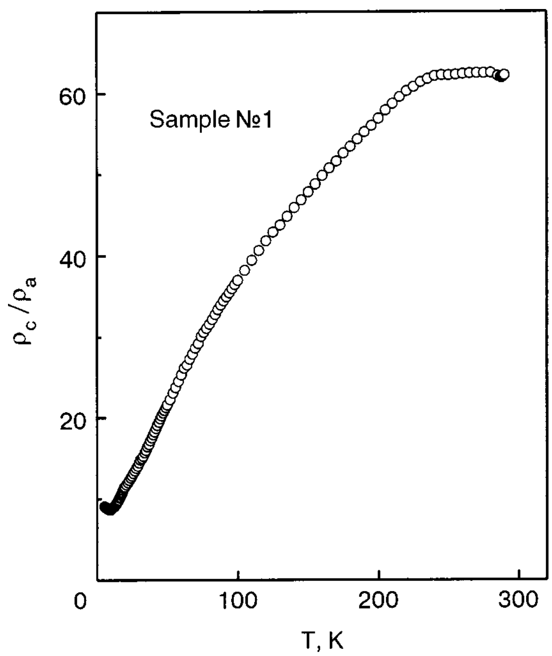

FIG. 2. Temperature dependence of the ratio of the resistivities ρ_a and ρ_c obtained for measuring currents directed along the crystallographic axes **a** and **c**.

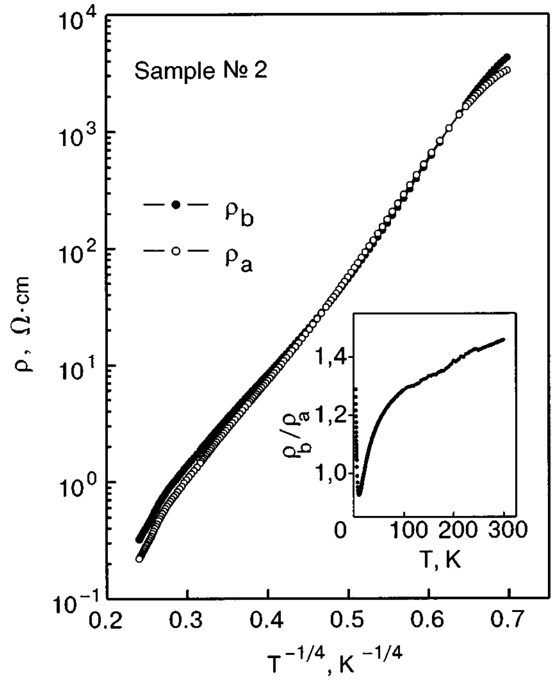

FIG. 3. Temperature dependence of the resistivities ρ_a and ρ_b measured along the crystallographic axes **a** and **b**, which lie in the CuO_2 planes. The inset shows the temperature dependence of the ratio of these resistivities.

(188 K) was observed. The $\rho(T)$ curves, as expected, are highly anisotropic, i.e., the values of ρ_c in the direction perpendicular to the CuO_2 layers is much greater than the value ρ_a in a direction parallel to these layers (Fig. 1). In the investigated temperature interval (5–295 K) the anisotropy of the resistivity is minimum in the region of helium temperatures, where $\rho_c/\rho_a \approx 10$ (Fig. 2). With increasing temperature the ratio ρ_c/ρ_a increases, reaching a value of ≈ 62 at $T \approx 20$ K. Further warming all the way up to room temperature did not lead to any substantial change in ρ_c/ρ_a (Fig. 2). The large values of ρ_c/ρ_a attest to the high crystalline perfection of the samples. In our studies we did not observe strong anisotropy of ρ in the crystal planes parallel to the CuO_2 layers (Fig. 3), primarily because of the unavoidable presence of twins in $La_2CuO_{4+\delta}$ crystals.

For $T < 20$ K we observed a deviation from Mott's law. This effect was investigated earlier[5] and has been attributed to the phenomenon of phase separation in $La_2CuO_{4+\delta}$ crystals.[12] At sufficiently low temperatures, phase separation leads to the presence of small superconducting inclusions in the insulating matrix. This causes a deviation from Mott's law and leads to a negative magnetoresistance which increases with decreasing temperature (see Ref. 5).

Measurements showed that not only the magnitude but even the sign of the magnetoresistance depend on the direction of the transport current relative to the CuO_2 layers. Figures 4–6 show data on the behavior of the magnetoresistance in fields parallel to the CuO_2 layers. In the case when the current is parallel to the CuO_2 layers one observes a transition from negative to positive magnetoresistance with increasing temperature. The transition occurs near $T \approx 18$ K (Fig. 5). At the same time, when the current is perpendicular to the CuO_2 layers one observes only negative magnetoresistance, the value of which increases monotonically with decreasing temperature (Fig. 6). In the case when both the current and field are perpendicular to the CuO_2 layers, one observes the expected behavior of the magnetoresistance, due to the AF–SF transition.[1,7,8]

The $R(H,T)$ curves shown in Figs. 4–6 demonstrate the dependence of the absolute value and sign of the magnetoresistance on the direction of the transport current for the same direction of the magnetic field (along the CuO_2 layers). These results are new. In particular, the positive magnetoresistance of $La_2CuO_{4+\delta}$ crystals in the VHLC regime has

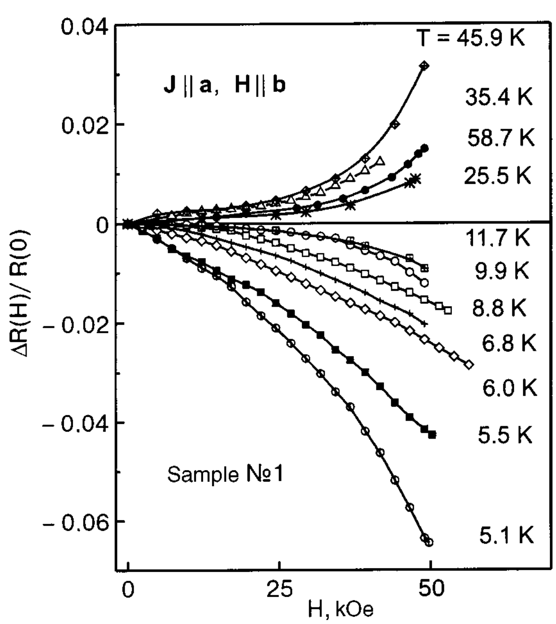

FIG. 4. Field curves of the magnetoresistance for different temperatures in the case when the measuring current is parallel to the CuO_2 layers. The magnetic field is directed parallel to these layers, along the crystallographic axis **b**.

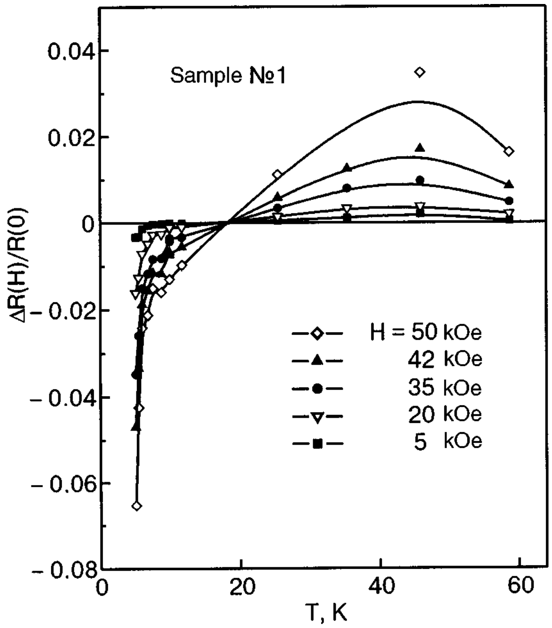

FIG. 5. Change in sign of the magnetoresistance upon an increase in temperature for the case of a measuring current parallel to the CuO_2 layers. The magnetic field is directed parallel to these layers, along the crystallographic axis **b**.

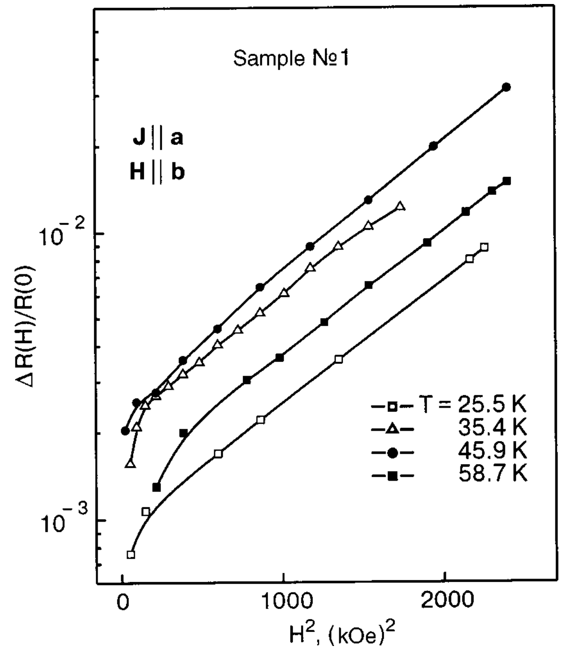

FIG. 7. Plot of ln[$R(H)/R(0)$] versus H^2 at different temperatures. The measuring current and magnetic field are parallel to the CuO_2 layers.

been observed for the first time. The curves in Fig. 4 reflect the competition of at least two mechanisms for the magnetoresistances of different sign. Below $T \approx 20$ K the predominant contribution is the negative magnetoresistance due to the phase separation effect inherent to $La_2CuO_{4+\delta}$, i.e., the separation of the system into regions enriched with and depleted of charge carriers (holes).[12] At a small value of δ the hole-enriched regions are of the nature of isolated metallic inclusions in an insulating matrix. At high temperatures the presence of these inclusions has a weak effect on the hopping conductivity of the system. The transition of these inclusions to the superconducting state leads to a deviation of the $\rho(T)$ curve from Mott's law (1) and to the appearance of a rather strong negative magnetoresistance (see the detailed discussion in Ref. 5).

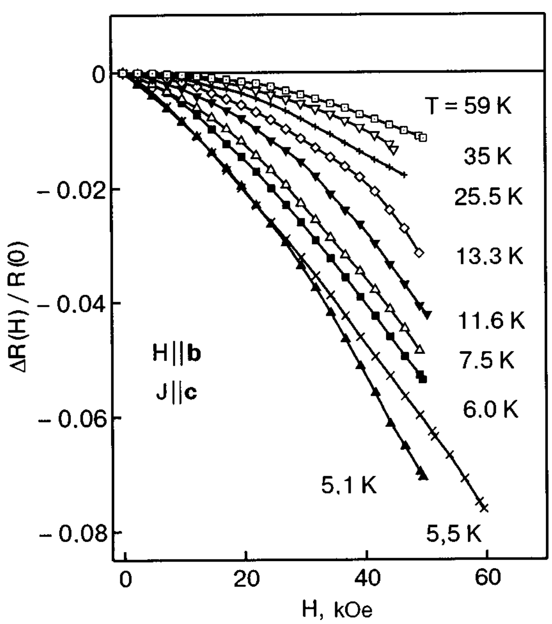

FIG. 6. Field dependence of the magnetoresistance at different temperatures in the case of a measuring current perpendicular to the CuO_2 layers. The magnetic field is directed parallel to these layers, along the crystallographic axis **b**.

An extremely probable cause of the positive magnetoresistance in the case of VHLC is the mechanism[9] based on compression of the impurity wave functions in a magnetic field. For the case of a weak magnetic field ($L_H \gg L_c$, where $L_H = (\hbar/2H)^{1/2}$ is the magnetic length and L_c is the localization length) the value of the magnetoresistance according to this mechanism is given by the expression[9]

$$\ln\frac{R(H)}{R(0)} = t_1\left(\frac{L_c}{L_H}\right)^4\left(\frac{T_0}{T}\right)^{3/4}, \tag{2}$$

where $t_1 = 5/2016$, and T_0 is the quantity that appears in Eq. (1) for $R(T)$ in the case of VHLC. Analysis of the data has shown that this mechanism is in qualitative (and even semi-quantitative) agreement with the experimental results. Indeed, we have found that the field dependence of the positive magnetoresistance is quadratic (ln[$R(H)/R(0)$]$\propto H^2$; see Fig. 7), in accordance with with formula (2). The decrease of the magnetoresistance at high temperatures (Fig. 5) also agrees with this mechanism. Moreover, the measured values of $\Delta R(H)/R(0)$ correspond to formula (2) for $L_c \approx 2$ nm. Estimates of L_c in previous papers[4,5] gave a value of around 1 nm. It should be noted, however, that the samples investigated in the present study were much less resistive than the samples in Ref. 5, for example. Therefore, the localization length in them may be somewhat larger. It is apparently this circumstance (i.e., the rather large localization length in the samples studied here) that has made it possible for us to observe the positive magnetoresistance of $La_2CuO_{4+\delta}$ crystals in the insulating state.

Thus the positive magnetoresistance observed in the present study corresponds in a number of respects to the mechanism proposed in Ref. 9. On the other hand, the competition between the two magnetoresistance mechanisms of different sign in the samples studied here precludes a completely reliable comparison of the results with the theory of the positive magnetoresistance in the case of VHLC.[9] It also cannot be ruled out that the positive magnetoresistance may be due to features of the magnetic structure of $La_2CuO_{4+\delta}$, e.g., to AF domains and stripes.[13] These possibilities are considered in Ref. 14 for the AF crystal $YBa_2Cu_3O_{6.32}$, where for fields and currents parallel to the CuO_2 layers a positive magnetoresistance is observed for $\mathbf{H}\perp\mathbf{J}$ and a negative magnetoresistance for $\mathbf{H}\|\mathbf{J}$. Behavior of this sort can be explained if it is assumed that the stripes are aligned along the applied magnetic field. Such an orientation of the stripes in the field is possible if they have a local ferromagnetic moment.[14] Of course, this explanation cannot be regarded as conclusive, and it must be verified by further experimental checks. It follows from Ref. 14 that the AF cuprates can have anisotropy of the magnetoresistance related to the mutual orientation of the current and magnetic field. For the CuO_2 layers this form of anisotropy can depend weakly on the crystallographic anisotropy within the layers. In the studies described in the present paper, for $La_2CuO_{4+\delta}$ single crystals, the magnetic field was always directed perpendicular to the current. We plan further investigations of $La_2CuO_{4+\delta}$ crystals in which we intend to use other mutual orientations of the current and magnetic-field directions to elucidate the nature of the positive magnetoresistance in fields parallel to the CuO_2 layers.

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