Isotropic negative out-of-plane magnetoresistance of $Bi_2Sr_2CuO_{6+\delta}$ single crystals

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In the normal state, the out-of-plane resistivity ρ_c of high-quality $Bi_2Sr_2CuO_{6+\delta}$ single crystals shows an anomalously large negative magnetoresistance in magnetic fields up to 28 T. This phenomenon corresponds to a suppression of the increase in ρ_c with decreasing temperatures as observed in high-T_c superconductors above the critical temperature T_c. In contrast to the magnetoresistance in the superconducting state, this normal-state out-of-plane magnetoresistance is independent of the field orientation (perpendicular and parallel to the CuO planes). This isotropic response points to the importance of spin effects on the pseudogap in the normal state, whereas the highly anisotropic response of the superconducting state is due to orbital effects.

The electrical transport of the layered high-T_c superconductors (HTSC's) shows anomalous properties related to the quasi-two-dimensional (quasi-2D) structure which have been studied very extensively in recent years.[1] One of the very unusual features of the normal-state properties is the coexistence of a metalliclike temperature dependence of the in-plane resistivity ρ_{ab} and a semiconductinglike dependence for the out-of-plane resistivity ρ_c (see, e.g., Refs. 2 and 3). Recently, such an opposite behavior of the resistivities ρ_{ab} and ρ_c was measured by Ando *et al.*[4] in La-doped $Bi_2Sr_2CuO_y$ (superconducting critical temperature $T_c=13$ K) in the normal state down to temperatures as low as $T/T_c\sim0.04$. The latter implies a 2D confinement and is incompatible with a Fermi-liquid behavior.

Many experiments (e.g., NMR,[5] photoemission,[6] tunneling[7]) have provided evidence that in the normal state of underdoped high-T_c superconductors a pseudogap exists in the electronic exitation spectra below a temperature $T^*>T_c$. Photoemission experiments have seen *d*-wave symmetry in the pseudogap structure.[6] In scanning tunneling measurements on $Bi_2Sr_2CaCu_2O_{8+\delta}$, Renner *et al.*[7] have found this pseudogap to be present both in underdoped and overdoped samples, and to scale with the superconducting gap. It has been proposed that the pseudogap in the normal state can be seen as a precursor for the occurrence of superconductivity where the superconducting phase coherence is suppressed by thermal or quantum fluctuations, e.g., Refs. 8–10. In the case of a nonsuperconducting origin, a pseudogap can be formed in the spin part of the excitation spectrum. The response of high-T_c superconductors in the normal state to high magnetic fields can give important information on the question of the nature of the pseudogap.

Because the normal-state properties in the high-T_c superconductors are known to depend strongly on carrier concentration and doping, the reported magnetotransport data in the normal state cannot be easily catagorized in a common picture. Concerning the transport along the *c* axis, the trend can be observed that in the temperature region showing the semiconductinglike *c*-axis resistivity most compounds reveal a negative out-of-plane magnetoresistance: the BiSrCaCuO system,[11–16] the LaSrCuO system,[17,18] the BiSr(La)CuO system[4,19]. In these experiments the relative variation in $\rho(H)$ reaches values up to 10–20 % at the lowest temperatures and highest fields.[4,16] The observed negative out-of-plane magnetoresistance has been discussed in terms of different models, such as *c*-axis hopping with interplanar scattering,[18] reduction of the density of states due to superconducting fluctuations,[11,13,14] and suppression of the pseudogap and/or spin gap.[12,17] The dependence of ρ_c with respect to the magnetic field orientation is of importance to discriminate between these interpretations. From the small anisotropy in the $\rho_c(H)$ transport it was concluded that the spins play an important role in the negative magnetoresistance.[12,17,19]

In this paper, we describe the experimental study of the *c*-axis resistivity ρ_c in the normal state of high-quality nondoped $Bi_2Sr_2CuO_{6+\delta}$ (Bi2201) single crystals under continuous magnetic fields H up to 28 T in the temperature region from 6 to 100 K. The low T_c of these crystals ($T_c\simeq9.5$ K) permits us to investigate the magnetoresistance of a cuprate superconductor in the normal state down to low temperatures. At low temperatures (<28 K) a negative out-of-plane magnetoresistance (MR) up to 60% was observed which is nearly independent of the applied field orientation.

The Bi2201 investigated single crystals were grown by a KCl-solution-melt method.[20] A temperature gradient along the crucible results in the formation of a big closed cavity inside the solution melt. The several tens of crystals grown in such a cavity share common properties. The quality of the crystals was verified by measurements of the dc resistance, ac susceptibility, x-ray diffraction, and energy-dispersive x-ray microprobe analysis. Our crystals showed x-ray rocking curves with a width of about 0.1°–0.3°. Two crystals with $T_c=9.5$ K (midpoint) and $\Delta T_c\simeq1$ K were investigated. Composition measurements of our crystals with $T_c\simeq9.5$ K have shown that they are slightly underdoped by oxygen depletion.[21] The dimensions of the crystals were 0.5 mm $\times1$mm$\times3$ μm (crystal No. 1) and 0.5 mm$\times1$ mm$\times10$ μm (crystal No. 2).

A four-probe contact configuration with symmetrical positions of the low-resistance contacts ($<1\Omega$) on both *ab* surfaces of the sample was used for the measurements of the

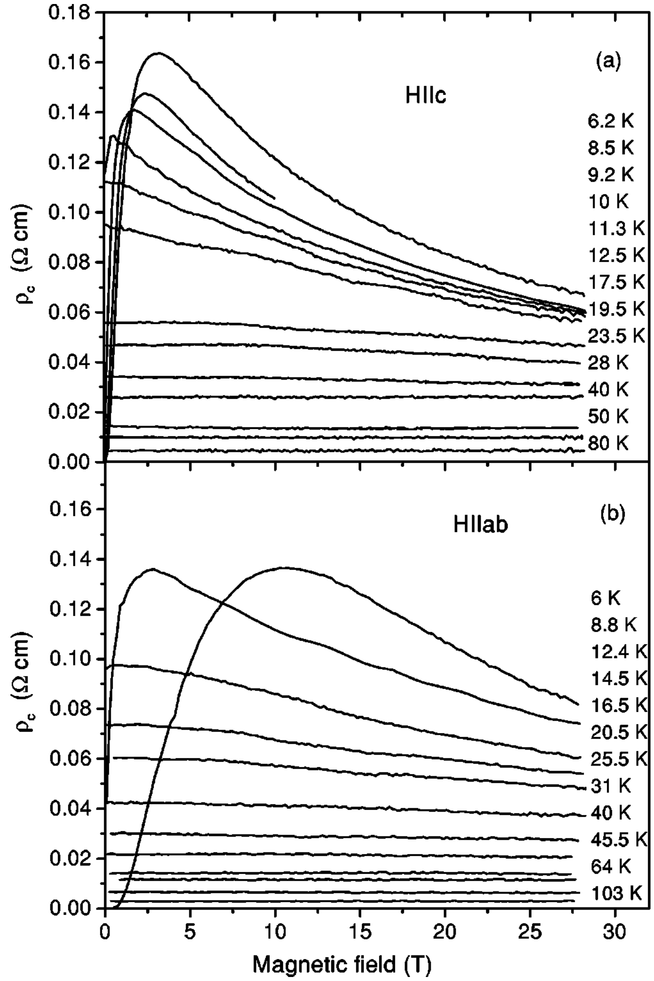

FIG. 1. The out-of-plane resistivity ρ_c as a function of the magnetic field H in the longitudinal $\mathbf{H}\|\mathbf{c}$ (a) and transverse $\mathbf{H}\|\mathbf{ab}$ (b) configurations for the crystal No. 1 at different temperatures just below and above $T_c=9.5$ K.

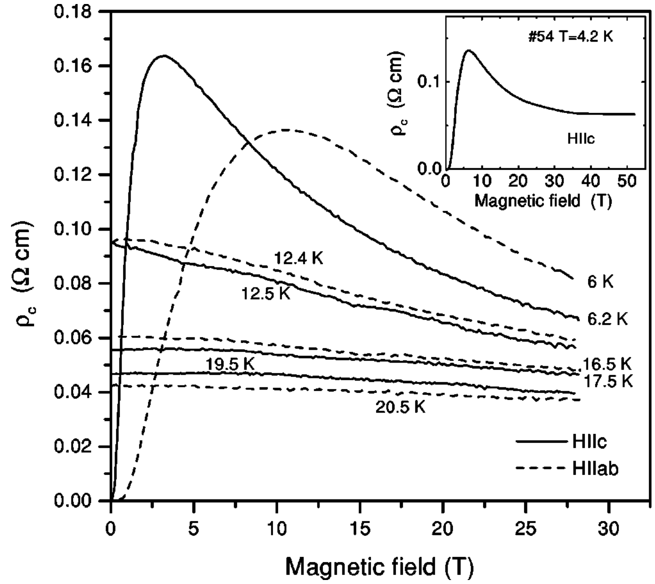

FIG. 2. $\rho_c(H)$ curves for the longitudinal and transverse configurations at $T\simeq 6$, 12.5, 17, and 20 K to show the isotropic field dependence in the normal state. The inset show the saturating magnetoresistance for another sample ($T_c=5.5$ K) in a pulsed-field experiment (Ref. 21).

in-plane and out-of-plane resistances. The measured resistances were transformed to the respective resistivities ρ_c and ρ_{ab} using the crystal dimensions. In zero magnetic field, the samples showed a nearly linear temperature dependence of the in-plane resistivity $\rho_{ab}(T)$ which saturates below 20 K to a residual resistivity of 50 and 80 $\mu\Omega$ cm, respectively. The out-of-plane resistivity $\rho_c(T)$ of the single crystals showed semiconductinglike normal-state behavior over the temperature region $T=10$–300 K. The temperature dependence down to T_c could be reasonably described by a power-law dependence $T^{-\alpha}$ with $\alpha=1.65$ (crystal No. 1) and 1.3 (crystal No. 2). The ρ_c values at $T=100$ K of the thin and thick samples are equal to 2.7 and 13.5 mΩcm, respectively. The anisotropy ratio ρ_c/ρ_{ab} was nearly 5×10^4 at low temperatures. By measuring the Hall coefficient R_H in the crystals we determined the carrier density $n=4.8\times10^{21}\,\mathrm{cm}^{-3}$. For the applied current J parallel to the c axis, the magnetic field H was applied both parallel to the c axis and parallel to the ab plane in the longitudinal ($\mathbf{H}\|\mathbf{c}\|\mathbf{J}$) and transverse ($\mathbf{H}\perp\mathbf{c}\|\mathbf{J}$) configuration.

Figure 1 displays the field dependence of the out-of-plane resistivity ρ_c in the longitudinal (a) and transverse (b) configuration for the thin sample No. 1 at different temperatures just below and above T_c. After an increase of ρ_c at low fields due to the suppression of superconductivity, ρ_c decreases with increasing magnetic field in the normal state. The in-plane resistivity ρ_{ab} shows a metallic temperature dependence in the magnetic-field-induced normal state down to low temperatures.[21] The negative out-of-plane magnetoresistance is consistent with other experiments on high-T_c superconductors showing a semiconductinglike ρ_c as reported in the above cited publications. However, two new features can be distinguished in our data.

First, at low temperatures ρ_c shows a much stronger negative magnetoresistance compared to reported experimental results. For example, at $T\approx 6$ K, ρ_c decreases by more than a factor of 2.5 in the highest applied fields. With increasing temperature the magnetoresistance decreases and becomes positive above ≈ 28 K. In very high fields ρ_c shows a tendency to saturation. The inset of Fig. 2 shows the $\rho_c(H)$ data for a Bi2201 crystal with $T_c=5.5$ K (overdoped) measured up to 52 T (Ref. 21) with a clear saturation in high fields after a twofold decrease. For the La-doped Bi2201 system [BiSr(La)CuO] studied previously,[4] only a 10%-negative out-of-plane magnetoresistance has been reported in pulsed magnetic fields up to 60 T at $T=0.8$ K. We note that the high-temperature c-axis resistivity of our crystals is about two orders of magnitude smaller compared to the BiSr-(La)CuO crystals investigated by Ando *et al.*[4] and Yoshizaki and Ikeda.[19] For our crystals we also observe a much larger increase of ρ_c at low temperatures. The larger negative magnetoresistance observed in our samples could be connected with this characteristic in $\rho_c(T)$.

The second striking result is that the strong negative magnetoresistance is observed for both geometries ($\mathbf{H}\|\mathbf{c}\|\mathbf{J}$ and $\mathbf{H}\perp\mathbf{c}\|\mathbf{J}$). In contrast to the magnetoresistance in the superconducting state, the normal-state magnetoresistance of ρ_c is independent of the field orientation with respect to the current direction. To illustrate this, we show in Fig. 2 $\rho_c(H)$ curves at different temperatures for both configurations. For

$T<T_c(H=0)$, $\rho_c(H)$ differs in absolute value between the two field orientations. However, the relative variation in magnetic field is comparable. The considerable difference in the field position of the maxima of $\rho_c(H)$ between the two field orientations (see Figs. 1 and 2) is a direct consequence of the anisotropy of the upper critical field in Bi2201 due to a difference in the orbital effect of the magnetic field. The similarity in the normal-state data for the two field orientations excludes probably an explanation of the negative out-of-plane magnetoresistance in terms of superconductivity.

Similar magnetoresistance behavior was obtained for ρ_c of the second crystal No. 2 except that at high magnetic fields the relative change of $\rho_c(H)$ was ≈ 2 times smaller compared to crystal No. 1. Crystals No. 1 and No. 2 have the same nominal composition and the same T_c, but were grown in different crucibles. The fact that the magnitude of the magnetoresistance significantly differs for the two crystals reflects presumably the disorder along the c axis, which is believed to be related to the presence of additional insulating layers in the thick sample (cf. the difference in ρ_c at zero field for the two crystals). Our x-ray studies have shown a better crystalline quality for the thinner samples.

The temperature dependence of the out-of-plane resistivity in sample No. 1 is presented in Fig. 3 for magnetic fields parallel [Fig. 3(a)] and perpendicular [Fig. 3(b)] to the c axis. All $\rho_c(T,H)$ curves intersect the zero-field $\rho_c(T)$ curve at $T\approx 28$ K where the magnetoresistance changes sign. The inset of Fig. 3(a) shows similar dependences for the $\rho_c(T,H)$ data of sample NO. 2. From the temperature-dependent data in Fig. 3 it can be concluded that the observed negative magnetoresistance corresponds to a suppression of the semiconductinglike behavior in $\rho_c(T)$. A comparison of the data in Figs. 3(a) and 3(b) confirms the previous mentioned isotropy of the magnetoresistance. The inset of Fig. 3(b) displays the relative variation $\Delta\rho_c/\rho_{c0}=[\rho_c(H,T)-\rho_c(0,T)]/\rho_c(0,T)$ in the normal state at different temperatures for both configurations at a magnetic field $H=28$ T.

In quasiclassical models of conventional metals spin-dependent scattering leads to a magnetoresistance which is independent of the field orientation. This magnetoresistance is positive, usually very small ($\sim 10^{-3}$), and has a quadratic field dependence. The observed out-of-plane magnetorsistance in our Bi2201 single crystals is negative and has a much stronger variation with field. The strong negative magnetoresistance observed in our experiments shows an exponential decrease with magnetic field. The slope in the ln ρ_c vs H plot of Fig. 4 decreases with increasing temperature. From the inset of Fig. 4 (plot of $\ln\rho_c$ vs H/T) it follows that our data can be described like $\rho_c(H,T)=\rho_{c0}+A\exp(-g\mu_B H/k_B T)$ with g factor $g=2$ and Bohr magneton μ_B where the scaling factor A depends on temperature. For a more accurate determination of this exponential decrease with magnetic field, experiments in the saturating region at higher fields have to be done. The $\rho_c(H)$ data obtained from pulsed-field experiments shown in the inset of Fig. 2 can be well described with a similar exponential decrease.

An anomalously large negative longitudinal MR (almost 90% at 0.05 K and 8 T) has been observed previously in the transition metal dichalcogenides[22] which also have a layered structure. These compounds show the typical temperature dependence due to variable-range hopping between localized

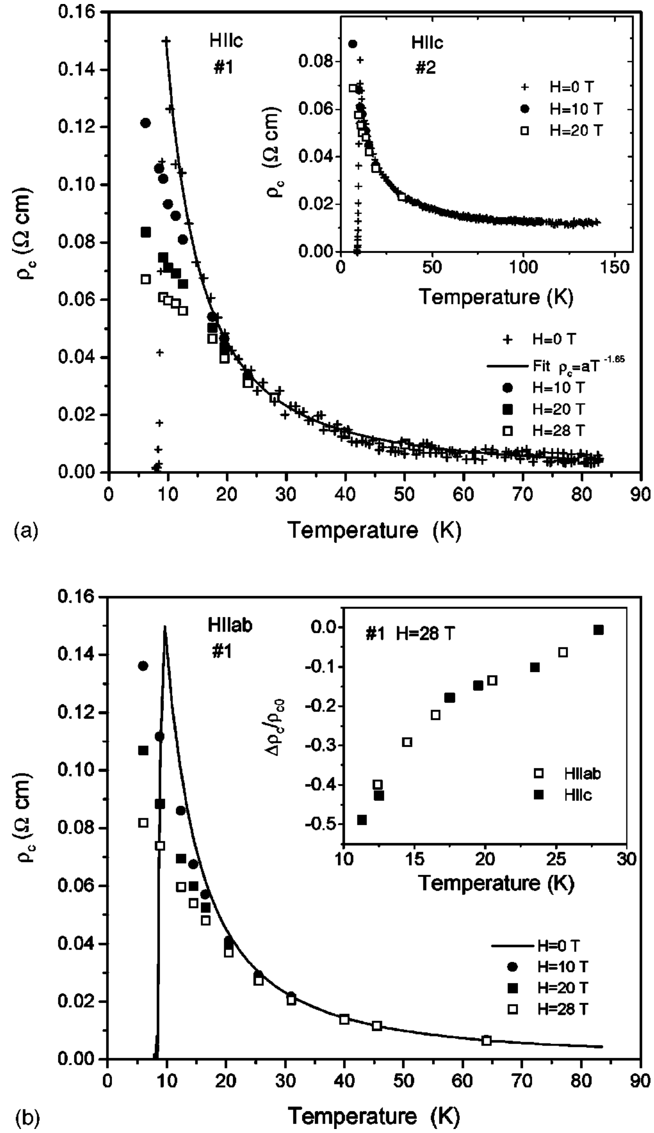

FIG. 3. The temperature dependencies of the out-of-plane resistivity $\rho_c(T)$ for sample No. 1 at 10, 20, and 28 T (data points) in the **H**∥**c** configuration (a) and in the **H**∥**ab** configuration together with the zero-field data (solid line). The inset in (a) shows the **H**∥**c** data for sample No. 2. The inset of (b) shows the relative variation $\Delta\rho_c/\rho_{c0}=[\rho_c(H,T)-\rho_c(0,T)]/\rho_c(0,T)$ at different temperatures for both configurations at a magnetic field $H=28$ T.

states. Fukuyama and Yosida[23] have explained this phenomenon by introducing Zeeman shifts for the Anderson localized states leading to enhanced conductivity (exponential in $g\mu_B H/k_B T$) with the energy levels of one spin component closer to the mobility edge. However, because the in-plane resistivity in our Bi2201 crystals is metallic, the carrier localization is highly improbable to explain the semiconductinglike $\rho_c(T)$ data.

The small negative out-of-plane MR in Bi2212 (1% at 14 T and 100 K) (Ref. 12) and in BiSr(La)CuO (2% at 17 T and 35 K) (Ref. 19) have been interpreted in terms of a slight reduction of the pseudogap by the magnetic field. Following this approach, the large decrease of ρ_c with magnetic field at low temperatures can be interpreted as a gradual closing of the pseudogap. From the isotropic field dependence, we can conclude that superconductivity is probably not at the origin of the pseudogap.

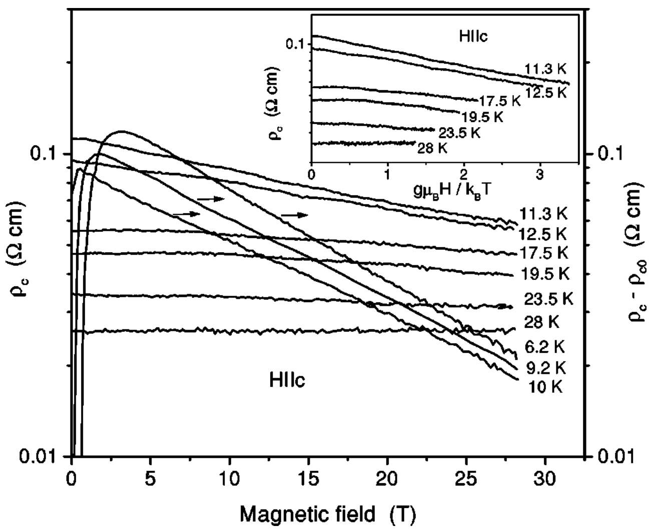

FIG. 4. The ρ_c data for sample No. 1 on a logarithmic scale as a function of magnetic field ($\mathbf{H}\|\mathbf{c}$ configuration) for different temperatures. For the lowest temperatures $\rho_c - \rho_{c0}$ has been plotted with $\rho_{c0} = 0.043\ \Omega$ cm. The inset shows the same plot as a function of the ratio $g\mu_B H/k_B T$.

According to a widely believed view,[8–10] the pseudogap (a suppression of the electron density of states) in the normal state above T_c can be of superconducting nature, i.e., a superconducting gap without the existence of phase coherence. In angle-resolved photoemission spectroscopy (ARPES) measurements,[6] the pseudogap appears below T^* at the points $(\pi,0)$ on the Fermi surface and the superconducting gap appears at lower temperatures below T_c at the points (π,π) (both are *d*-wave type). The transitions leading to the pseudogap and the superconducting gap are separated in temperature. In the framework of this scenario it is difficult to explain the isotropy of MR because the response of a superconductor to the orbital effect of a magnetic field should be highly anisotropic. This is clearly visible in Figs. 1 and 2 on comparison of the $\rho_c(H)$ curves in the $\mathbf{H}\|\mathbf{c}$ and $\mathbf{H}\|\mathbf{ab}$ configurations at temperatures below T_c. The isotropic behavior of the normal-state MR with respect to the orientation of the magnetic field shows that actually only the effect of the magnetic field on the spins (Zeeman effect) is important in the normal state. The pseudogap may have nonsuperconducting origin, for example, a spin-density wave or another more complicated spin excitation. For this situation, there is no problem to reconcile the isotropic magnetoresistance in the normal state with the anisotropic one in the superconducting state. Although we can conclude on the different nature of the pseudogap with respect to superconductivity, our spin-dominated effect on the magnetotransport along the *c* axis gives no further information on the possible spin structure to explain the pseudogap.

To summarize, an anomalously large negative longitudinal MR up to 60% has been observed in the out-of-plane MR in single crystals of the single-CuO-layer compound $Bi_2Sr_2CuO_{6+\delta}$ in magnetic fields up to 28 T in the normal state up to 28 K. In contrast to the MR in the superconducting state, the normal-state MR is independent of the applied field direction with respect to the current, suggesting a uniquely spin-dominated origin of MR. Interpreting the magnetic-field-induced suppression of the low-temperature upturn in $\rho_c(T)$ by a suppression of the pseudogap, our results would put serious doubts on the superconducting nature of the pseudogap.

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