Temperature dependence of the upper critical field of $Bi_2Sr_2CuO_x$ single crystals

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The superconducting resistive transition has been investigated in high-quality $Bi_2Sr_2CuO_x$ single crystals with critical temperature T_c (midpoint) $=3.7-9$ K in magnetic fields (continuous up to 28 T and pulsed up to 52 T) applied perpendicular and parallel to the ab plane. The temperature dependence of the resistive upper critical field H^*_{c2} determined as the field at which the in-plane resistivity in the transition region is 90% or more of the normal state resistivity is down to temperatures $T/T_c=0.3$, in close agreement with the Werthamer-Helfand-Hohenberg theoretical curve calculated for conventional type-II superconductors. The following thermodynamic parameters were extracted from the experimental results: $H^*_{c2}(0)_{ab}=16-27$ T, $H^*_{c2}(0)_c=43$ T (for 50% of the normal state resistance), corresponding to superconducting coherence lengths $\xi_{ab}(0)\simeq 35-45$ Å and $\xi_c(0)\simeq 15$ Å. The measured field dependence of the mixed-state Hall effect confirms the temperature dependence of the resistive upper critical field found from the superconducting transitions.
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I. INTRODUCTION

Despite the fact that the upper critical field $H_{c2}(0)$ is one of the fundamental parameters in the problem of high-T_c superconductivity, up to now some ambiguity has existed about the values and the temperature dependence of $H_{c2}(T)$. Due to the structural simplicity and the low critical temperature T_c, the low-T_c phases of the high-T_c superconductors (HTSC) are an excellent choice for studying the superconducting and normal state transport properties. Recently, by measuring the resistive transitions of low-critical-temperature oxides $Tl_2Ba_2CuO_6$ (Tl2201), single crystals (T_c near 20 K),[1] and $Bi_2Sr_2CuO_6$ (Bi2201) films (transition from 19 down to 12 K),[2] in high magnetic fields, an anomalous temperature dependence of $H_{c2}(T)$ was observed. The magnetoresistance (MR) curves were shifted to higher fields with decreasing temperature, but evidence of saturation down to temperatures in the mK range was not found. The value of $H_{c2}(0)$ exceeded the one expected from the slope dH_{c2}/dT at T_c greatly. This anomalous exotic behavior cannot be explained by any well-established model.

The conventional superconductors show a steplike resistive transition with only small broadening for reduced T_c values at increasing field. In this case, $H_{c2}(T)$ values can be directly obtained from the shifts of the resistive transition curves as a function of the applied field. Due to the significant field-induced broadening of the resistive transition in HTSC such a method can lead to great uncertainty.[3] Nevertheless, data for the magnetotransport measurements of $La_{(2-x)}Sr_xCuO_4$ (Ref. 4) suggest that whereas the superconducting transitions of optimally doped (with the maximum T_c) and underdoped samples broaden as the magnetic field is applied, the overdoped samples show a parallel shift. This assumption is supported by the fact that the overdoped samples Tl2201 in the above-mentioned article[1] have relatively sharp magnetic transitions with parallel shifts to higher fields as the temperature is lowered.

It is well known that the preparation of homogeneous, single-phase, and pure single crystals of the HTSC with satisfactory quality is a very difficult task. A significant spatial variation in the concentration of the mobile oxygen can have dramatic effects on the magnetotransport phenomena and on the width of the superconducting transition in a magnetic field. In contrast to the situation for the Tl2201 compound, in the Bi2201 crystals the oxygen ions have a low mobility and a weak sensitivity of T_c to oxygen content in the CuO_2 planes. In this connection we have studied the temperature dependence of the upper critical field $H_{c2}(T)$ by using high-quality Bi2201 single crystals. The investigations of the $H_{c2}(T)$ dependence were performed in magnetic fields up to 52 T.

After a description of the investigated samples and their characteristics, we will discuss the measurements of the magnetic field dependence of the superconducting transition in crystals which reveal conditions close to the flux-flow regime. It is known[5] that in the case of a broadened superconducting transition different criteria can lead to different temperature dependencies for $H_{c2}^{*}(T)$. Therefore, the temperature dependence for the resistive upper critical field H_{c2}^{*} will be determined from the field at which the resistivity ρ in the transition region is 10%, 50%, or 90% of the saturation value of the normal state resistivity ρ_n at a given temperature.

II. EXPERIMENT

The Sr-deficient $Bi_{(2+x)}Sr_{2-(x+y)}Cu_{(1+y)}O_z$ single crystals were grown in a gaseous phase in the large closed cavities of a KCl solution melt.[6] In this case, the crystals do not have contact with the solidified melt in the crucible, thereby avoiding thermal stresses during cooldown. The number of the crystals in one cavity reached several tens. The crystals were grown in the temperature range $830-850\,°C$ with the ratio Bi/Sr $=1.4-1.5$ provided the Cu content was increased insignificantly. Due to the long growth time our single crystals have high cation ordering. The sizes of the crystals were around $(0.5-2)$ mm$\times(0.4-0.7)$ mm$\times(1.5-5)$ μm. Properties of the crystals grown inside the same cavity were very similar. The high quality of the crystals was verified by measurements of the dc resistance, ac susceptibility, x-ray diffraction, and scanning electron microscopy.[6] Composition of the crystals was studied using a Philips CM-30 electron microscope with Link analytical AN-95S energy dispersion x-ray spectrometer. It is believed that the low-T_c Bi compound crystals are either overdoped when the Bi/Sr ratio is larger than 1 (Bi excess is localized at the Sr position) or overdoped at a significant excess of Cu with substitution of part of the Bi atoms by Cu. In the present measurements only crystals with Bi excess were investigated. It is also known[6,7] that even in nondoped pure Bi2201 compound, both superconducting and ''semiconducting'' crystals can be obtained by changing the Bi/Sr ratio. The Bi2201 samples with the ratio Bi/Sr$=1$ contain many other phases and a single-phase solid solution can be formed by changing the ratio of Bi to Sr only. It was also established[7] that with increasing Bi content the lattice parameters increase without formation of any other phases. The T_c value of the crystals formed by our free growth method ranges up to 13 K. But we have found that the more perfect superconducting Bi2201 single crystals have a very narrow region of values of the lattice parameters $a=5.360-5.385$ Å, $c=24.60-24.63$ Å. In this case the T_c (midpoint) values of the crystals lie in the region 3.5 -9 K. The half-width of the main reflections in the x-ray rocking curves for the single crystals consisting of two or three blocks did not exceed 0.3°, whereas for the crystals comprised of one block only (with dimensions of 0.3 mm $\times 0.3$ mm) it was less than 0.1°. This value is close to the resolution limit of a diffractometer and for the analysis of the crystal perfection we used the supercell parameters which are more sensitive to structural imperfections. In our single crystals there is a linear relationship between the T_c value and the monoclinic superlattice angle value[6] which is directly related to the concentration of the carriers.

Low-Ohmic contacts were made to the samples by using evaporated and fired-on gold films. A four-probe contact configuration with symmetrical position of the contacts on both ab surfaces of the sample was used for the measurements of the in-plane and out-of-plane resistances. In order to convert the measured resistances to specific resistivities, the dimensions of the samples were measured with a high resolution optical microscope. In the employed dc method for the resistance measurements, the current was about 50 -100 μA for in-plane and $100-1000$ μA for out-of-plane resistance measurements. The crystals were studied with the magnetic field **H** applied either parallel or perpendicular to the **c** axis. In the latter case a configuration with **J** perpendicular to **H** for the in-plane transport current **J** was used. High magnetic field measurements were carried up to 28 T in continuous fields and up to 52 T in pulsed magnetic fields.

The magnetotransport measurements have been performed on crystals with T_c (midpoint) $=3.7-9$ K. The transition width defined by the 10% and 90% points of the superconducting transition of our samples ranged from 0.5 to 1.7 K. We note that the onset temperatures of the superconducting transition from dc resistance and ac susceptibility measurements were close and the transition widths almost the same.

The in-plane resistivity of the crystals with $T_c=4-6$ K showed a linear temperature dependence at high temperatures which saturates below $20-40$ K to a residual resistivity. Crystals with $T_c=8-9.5$ K had a nearly linear temperature dependence $\rho_{ab}(T)$ down to T_c. The slope $\Delta\rho_{ab}/\Delta T$ of the in-plane resistivity at high temperatures ranged from 0.5 to 1.5 $\mu\Omega$ cm/K. The residual resistivity $\rho_{ab}(0)$ lies between 60 and 180 $\mu\Omega$ cm. In the samples with higher T_c, smaller $\rho_{ab}(0)$ and larger $\Delta\rho_{ab}/\Delta T$ were observed. These $\rho_{ab}(0)$ and $\Delta\rho_{ab}/\Delta T$ values were obtained using the Montgomery method. However, as was shown[8] they can be twice as large as the values obtained by means of the Van der Pauw four-probe methods. It is known that this discrepancy is due to the strong anisotropy of the layered cuprates. It has been shown[9] that in $YBa_2Cu_3O_7$ with $\rho_c \gg \rho_{ab}$ the absolute value of ρ_{ab} measured by the contacts on one side of the single crystal is roughly a factor of 2 larger than that obtained with the current and voltage contacts on opposite sides of the sample. We have also observed a similar phenomenon in our specimens. The out-of-plane resistivity $\rho_c(T)$ of Bi2201 single crystals varies as a power law $T^{-\alpha}$ over the temperature region $T=3-300$ K with $\alpha=0.7-1.6$. The largest anisotropy ratio ρ_c/ρ_{ab} is 5.3×10^4 at $T=0.4$ K.

In a two-dimensional model from the residual resistivity $\rho_{ab}(0)$ and the lattice parameter c we determined the disorder parameter values[10] $(k_F l)_{ab}=39-23$ where k_F is the Fermi wave vector and l the elastic scattering length in the ab plane. We measured a normal-state Hall coefficient R_H in our crystals at 4.2 K and found that the carrier density equals $(4.8-6.3)\times10^{21}$ cm^{-3}. The carrier density in the samples with a lower T_c was larger than that in samples with T_c $=9$ K. If we take the effective mass in the ab plane for Bi2201 $m^*=6.5m_0$ (m_0 represents the free-electron mass), calculated by Hou *et al.*[11] based on the assumption of a cylindrically shaped Fermi surface, we obtain $k_F\approx0.7$ Å^{-1} and $l=60-35$ Å at T less than 4 K. According to the optical data obtained by Tsvetkov *et al.*[12] on our Bi2201 single

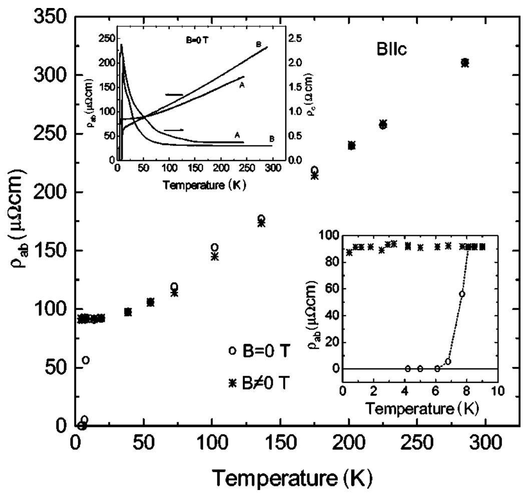

FIG. 1. Upper inset: the temperature dependencies of the in-plane ρ_{ab} and out-of-plane ρ_c resistivities for two single crystals with $T_c = 4.6$ K (A) and 8.5 K (B) at $H = 0$ T. Main panel: the zero field resistivity vs temperature for one of the crystals (open circles). The stars show the resistivity data at 20 T taken from the saturation region of the resistivity. The lower inset shows an expanded scale of the low-temperature data.

crystals, the carrier density $n = 5.1\times10^{21}$ cm^{-3} at 10 K and the effective mass in the ab plane $m^* = 3m_o$. With this value of m^* we calculated $k_F = 0.44$ Å^{-1}, Fermi velocity $v_F = 1.7\times10^7$ cm/s and $l = 90-50$ Å. It seems likely that m^* extracted from optical data is more suited to the determination of the transport parameters of crystals.

III. RESULTS AND DISCUSSION

The zero field temperature dependence of the in-plane resistivity of our low-T_c Bi2201 single crystals was described in considerable detail earlier.[13] As an example we display in the upper inset of Fig. 1 the typical temperature dependences of the in-plane ρ_{ab} and out-of-plane ρ_c resistivities for two single crystals with $T_c = 4.6$ K (A) and 8.5 K (B) under zero magnetic field. In the study of the superconducting resistive transition of the crystals in magnetic fields we observed that all transition curves in the magnetic field saturated at temperatures down to 0.4 K. In Fig. 1 (main panel) the zero field resistivity is plotted as a function of temperature (open circles) for one of the studied crystals. The stars represent the data points for $\rho_{ab}(T)$ measured in a magnetic field $H = 20$ T. At temperatures $T < T_c$ the resistivity data were defined from the resistive transition as a function of magnetic field in the saturated portion of the $\rho_{ab}(H)$ curves. The lower inset in Fig. 1 shows an expanded scale of the low-temperature data. The high magnetic-field measurements testify that the ab plane resistivity in the normal state shows the ordinary behavior for a metal down to 0.4 K. Analogous results were obtained on the low-T_c phases of Tl2201,[1] $YBa_2(Cu_{0.97}Zn_{0.03})_3O_{7-\delta}$,[5] and very recently on La-doped Bi2201 ($T_c \simeq 13$ K).[14] Since the finding is in close agreement with data published earlier, we will not discuss these results here.

The resistive superconducting transitions have shown a significant field-induced broadening even at low measuring

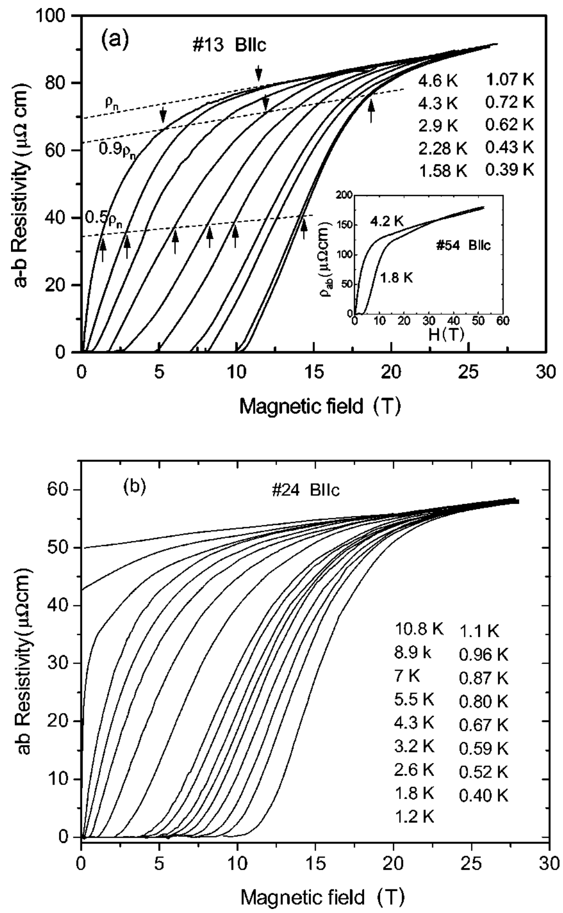

FIG. 2. Resistive transitions of crystal #13 (a) and #24 (b) in a magnetic field **H**∥**c** at different temperatures. The dashed lines and the arrows show the method used to define H_{c2}^* from the measured $\rho_{ab}(H)$ curve. Inset in (a) shows the magnetic field dependence of the in-plane resistivity for single crystal #54, at 4.2 K and 1.8 K in a 52-T pulsed magnet. The magnetic field is applied parallel to the **c** axis.

dc currents (30 μA). In Fig. 2 we report the magnetoresistance curves for two samples #13 (a) and #24 (b) at various temperatures with the field direction perpendicular to the ab plane of the crystal. The zero-field temperature region of the transition defined by the 10% and 90% points of the transition equals 5.1−6.8 K for sample #13 and 8.7−9.5 K for sample #24. The dashed lines in Fig. 2(a) show the method used to define H_{c2}^* from the measured $\rho_{ab}(H)$ curve. Three other studied samples #10, #5, and #14 had the transition between 5.8−6.5 K, 4.3−4.8 K, and 4.5−5 K, respectively. The field-induced resistive transitions for all three samples were similar to those shown in Fig. 2. In spite of the strong broadening of the magnetic transitions one can see in Fig. 2 that the resistive transitions in the normal state are completed at 25 T, even at the lowest experimental temperature (a weak increase of the normal-state resistance is due to a MR contribution in high magnetic fields).

We also measured the resistive transitions for one of our Bi2201 crystals (#54, $T_c = 5.5$ K) in a 52-T pulsed magnetic field with a 10-kHz excitation current of 10 μA using a lock-in amplifier. Similar results were obtained. The inset in Fig. 2(a) shows the magnetic field dependence of the in-plane resistivity for the Bi2201 single crystal at 4.2 K and

1.8 K. The magnetic field is applied parallel to the **c** axis.

We have studied the resistive transitions with the magnetic field parallel to the *ab* plane and obtained similar results. Because of the very large values of the upper critical fields in the **H**∥**ab** geometry we defined the H_{c2}^* values in this case for 50% and 10% of the normal-state resistivity ρ_n only. The transport current was in the *ab* plane of the crystals and orthogonal to the field in all cases.

It is well known that in layered HTSC the magnetoresistance near T_c is strongly influenced by phenomena related to thermal fluctuations and quasi-two-dimensional vortex states.[15,16] Because of the low pinning energies and large fluctuations in the case of the high-T_c phases of HTSC, transport measurements determine an irreversibility line rather than the thermodynamic critical field H_{c2}. However, in spite of the low T_c values of our samples and the low measuring temperatures where thermal fluctuations must play an insignificant role, the MR curves are broadened as before. A similar behavior of the magnetic transitions in the **H**∥**c** geometry in Bi2201 at temperatures $T=2.2-5.4$ K was briefly reported previously.[10] If we assume that because of the significant excess of Bi our crystals are heavily doped, the data in Fig. 2 are in strong contrast to the data for overdoped Tl2201 single crystals[1] and Bi2201 films.[2] In cited papers,[1,2] relatively sharp magnetic transitions with parallel shifts to higher fields are observed even at temperatures near T_c. However, in present measurements as may be seen from Fig. 2 the transition widths decrease and the curves shift parallel to higher fields at lower temperatures only. Most probably such a sharpening of the superconducting transitions is caused by reduced flux-flow effects at the lowest temperatures. In the optimally La-doped Bi2201 polycrystals samples which have the maximum value $T_c \simeq 25$ K for Bi2201 a carrier density near $n=3\times10^{21}$ cm^{-3} was obtained.[7] Then our single crystals with $T_c=3.5-4$ K need to be assigned to be heavily doped. On the other hand, as is reported in Ref. 17, the carrier concentration in the underdoped $Bi_2Sr_{2-x}La_xCuO_{6+\delta}$ single crystals with $T_c=13$ K are similar. Because the largest value of T_c in our pure Bi2201 single crystals approximately equals 13 K, the samples studied here with $T_c=9-10$ K are most likely to be near optimal doping. Taking into account the relatively large value of the elastic scattering length, this assumption seems plausible.[18] This is consistent with the observed wide superconducting transition of the La-doped Bi2201 samples in pulsed magnetic fields.[14] La doping reduces the hole concentration[7] and the width of the transition curves increases. Unfortunately, such doping deteriorates the crystal quality also and the superconducting transition width in magnetic fields is as large as 20 T even at $T=0.8$ K. The sharpening of the superconducting transitions upon lowering the temperature was recently observed in the underdoped $La_{2-x}Sr_xCuO_4$ system.[19]

Another prominent feature of the MR curves for the low-T_c samples (Fig. 2) is that all curves in the **H**∥**c** geometry saturate at high enough fields and to the same value. This is consistent with the temperature independence of the normal state resistivity of our samples at low temperatures.[13]

Figure 3 shows the temperature dependencies of the resistive upper critical field H_{c2}^* in the **H**∥**c** geometry for samples #13 (a) and #24 (b) extracted from the MR curves. The H_{c2}^* values are obtained from the fields at which the resistivity of the samples has reached 10%, 50%, 90%, and 100% of its normal-state values ρ_n. The temperature dependence of H_{c2}^* was also determined for the field oriented perpendicular to the **c** axis in the **H**∥**ab** geometry using the 10% and 50% criteria. This dependence is close to the one found in the **H**∥**c** geometry. The broadening of the superconducting transition and the rounding of the top of the transition make it impossible to define the value of the upper critical field exactly. The temperature dependencies of H_{c2}^* in Fig. 3 obtained from the 10% and 50% criteria exhibit a strong upward curvature as observed in overdoped Tl2201,[1] Bi2201,[2] and $YBa_2(Cu_{0.97}Zn_{0.03})_3O_{7-\delta}$.[5] By using criteria for the definition of H_{c2}^* more closely to ρ_n we obtain almost a linear $H_{c2}^*(T)$ dependence. For the **H**∥**ab** geometry the linear $H_{c2}^*(T)$ dependence was even observed for 10% and 50% criteria. The latter fact seem to give evidence for the strong influence of the character of the vortex motion on the extracted $H_{c2}^*(T)$ dependence from the resistive measurements.

From the linear part of the $H_{c2}^*(T)$ dependencies near T_c

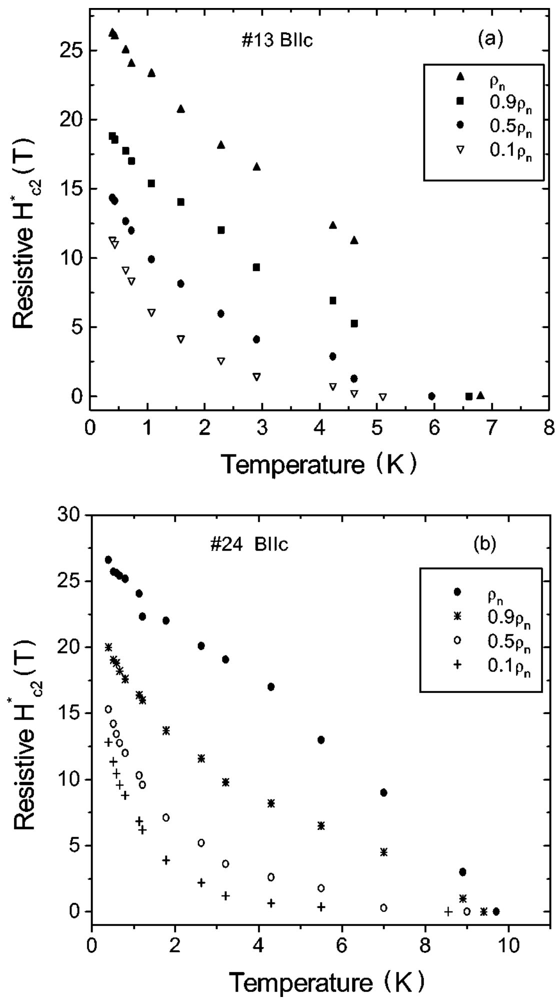

FIG. 3. Temperature dependencies of the resistive upper critical field H_{c2}^* in the **H**∥**c** geometry for samples #13 (a) and #24 (b) extracted from the resistive transitions of the crystals. The H_{c2}^* values are the fields at which the resistivity of the samples has reached 10%, 50%, 90%, and 100% of its normal-state value ρ_n.

as determined from the $\rho=\rho_n$ criterion we obtained the slopes dH^*_{c2}/dT_c ranging from -2.9 T/K to -4.7 T/K for the five samples in the **H**∥**c** geometry. For the **H**∥**ab** geometry the values $dH^*_{c2}/dT_c=-12$ T/K (sample #7, $T_c=3-4.5$ K) and -5 T/K (sample #26, $T_c=6-7.7$ K) for the 50% and 10% criteria, respectively, were obtained. Using a linear extrapolation of $dH^*_{c2}(T)$ from T_c to zero temperature we found $H^*_{c2}(0)\simeq 27$ T, 22 T, 20 T, 21 T, and 16 T (**H**∥**c** axis) for samples #24, #13, #10, #5 and #14, respectively. In the case of the **H**∥**ab** plane geometry $H^*_{c2}(0)=40-43$ T for sample #7 and #26 (50% criterion) was obtained. Using a Werthamer-Helfand-Hohenberg (WHH)-type extrapolation to lower temperature[20] with $H^*_{c2}(0)=0.693(-dH^*_{c2}/dT)T'_c$ we obtained from the $\rho=\rho_n$ points $H^*_{c2}(0)=22$ T, 15 T, 14 T, 14.5 T, and 11 T (samples #24, #13, #10, #5, and #14, respectively) for **H**∥**c** axis. Here, the dH^*_{c2}/dT is taken to be the tangent of $H^*_{c2}(T)$ at temperatures close to T_c and T'_c the intersection of this tangent with the temperature axis. Our results differ strongly from those found in Ref. 2 for Bi2201 films which have higher values of T_c (the midpoint transition at $\simeq 15$ K and the transition width $\simeq 7$ K). Extrapolating the data near T_c, the slope $dH^*_{c2}/dT=-0.29$ T/K and $H_{c2}(0)=3.8$ T were found for these films.[2] Since the superconducting transitions of our Bi2201 samples are broadened in a magnetic field and different criteria lead to different temperature dependencies for $H^*_{c2}(T)$, we will consider that the data obtained from $\rho\simeq\rho_n$ (90% and 100% criteria) show the most accurate reflection of the $H^*_{c2}(T)$ dependence. For comparison we determined $H^*_{c2}(T)$ also from crossing between the extrapolated normal state resistivity and the tangent of the transition. As a result, smaller values for $H^*_{c2}(T)$ and $H^*_{c2}(0)$ were obtained but the general view of the $H^*_{c2}(T)$ curves remained the same.

Returning to Fig. 3, we observe that the temperature dependence of H^*_{c2} obtained from the 10% and 50% criteria demonstrate a strong upward curvature as observed earlier in overdoped materials.[1,2,5] In these cited articles[1,2] the positive curvature of the $H^*_{c2}(T)$ dependence was independent of the chosen criterion. It can be supposed that our data deduced from $0.1\rho_n$ and $0.5\rho_n$ criteria determine the irreversibility line which lies well below $H^*_{c2}(T)$ and has an upward curvature.[21] The irreversibility line, as is known, does not occupy a well defined position in the H-T diagram insofar as that it depends on the sample, sample size, and other experimental conditions.

For our single crystals with a low T_c subsequently involving low measuring temperatures, the fluctuation effects should be quite small except for temperatures very close to T_c. However, as can be seen from the braodened transitions close to H_{c2} in Fig. 2, the short coherence length and the large anisotropy probably enhance the contribution of thermal fluctuations. The resistive transition widths decrease with increasing magnetic field for temperatures lower than $T\simeq 3$ K, where the transition curves are shifted showing a clear flux-flow regime with a resistivity linear in field. For the 2D superconductors a crossover between free flux flow and thermally activated motion should take place at a melting temperature T_m of the vortex lattice[22] given by $T_m=(1/8\pi\sqrt{3})\Phi_0^2 s/16k_B\pi^2\lambda_{ab}^2\simeq 5.5$ K using the London penetration depth $\lambda_{ab}\simeq 3100$ Å (Ref. 23), the CuO_2 interlayer spacing $s=12.3$ Å, and the flux quantum Φ_0. The estimated T_m value is close to that found in our experiment.

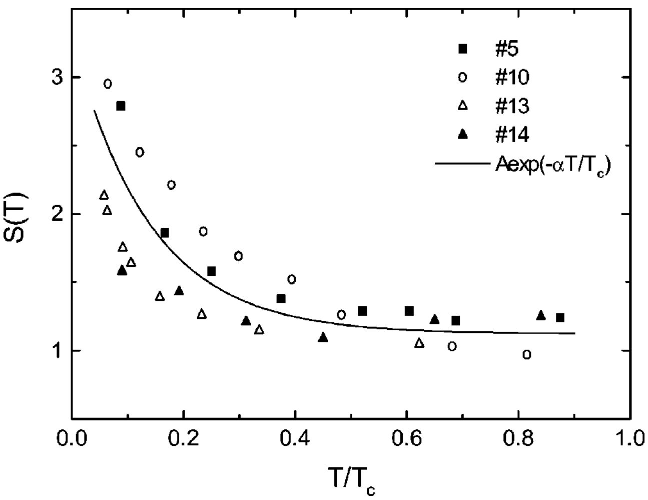

FIG. 4. The normalized slope $S(T)$ of the transition curves vs reduced temperature T/T_c for the **H**∥**c** geometry extracted from the resistive transitions in four crystals. The solid curve corresponds to the formula $A\times\exp(-\alpha T/T_c)$ with $A=2.17$ and $\alpha=7.14$.

Earlier measurements of the flux-flow resistivity ρ_f in conventional type-II superconductors have shown that ρ_f is proportional to the magnetic field at low temperatures, i.e., $\rho_f/\rho_n=H/H_{c2}(0)$ (Ref. 24), where ρ_n is the normal-state resistivity. For the comparison of experimental data on different samples it is convenient to use the dimensionless ratio $S(T)=[Hd(\rho_f/\rho_n)/dH]_{H_{c2}}$. This parameter extracted from our MR data for the **H**∥**c** geometry as a measure for the slope of the transition curves is plotted in Fig. 4 as a function of the reduced temperature. As is seen from Fig. 4, the range of $S(T)$ values for Bi2201 is almost comparable to that for the conventional dirty Nb-Ta, In-Bi, and Pb-Tl alloys,[25] but their parameters S depend inversely on temperature. In the present case the behavior of $S(T)$ reflects the fact that the depinning field (the onset of resistance) of Bi2201 increases faster than H^*_{c2} with decreasing temperature. This is also evident from Fig. 2. Since a depinning line due to thermally activated flux creep is the irreversibility line[26,27] and the irreversibility field[21] follows an exponential law $H_{ir}\simeq\exp(-T/T_c)$ or a power law $H_{ir}\simeq T^{-2}$, the observed temperature dependence of S seems plausible. The solid curve in Fig. 4 corresponds to the formula $A\times\exp(-\alpha T/T_c)$ with $A=2.17$ and $\alpha=7.14$.

It is known that resistance measurements provide the lower limit for ρ_f because pinning may be present. Then using the expression for flux-flow viscosity $\eta=\Phi_0 H/c^2\rho_f$ we find the upper limit for $\eta\leqslant 1.8\times 10^{-7}$ g/cms at $T=4.3$ K and $H=1$ T (Fig. 2). This value is close to the theoretical viscosity $\eta\simeq 3\times 10^{-7}$ g/cm s, estimated by use of the formula $\eta=\Phi_0 H_{c2}(0)_{ab}/c^2\rho_n$ with the averaged value of the normal-state resistivity in the ab plane $\rho_n=120$ $\mu\Omega$ cm. Based on the data presented in Fig. 4 and the calculated values of T_m and η, we believe that for the measurements of the MR of our crystals in the mixed state the conditions close to the flux-flow regime were realized. For this reason at the significant broadening of the superconducting transition in a magnetic field, the influence of the flux-

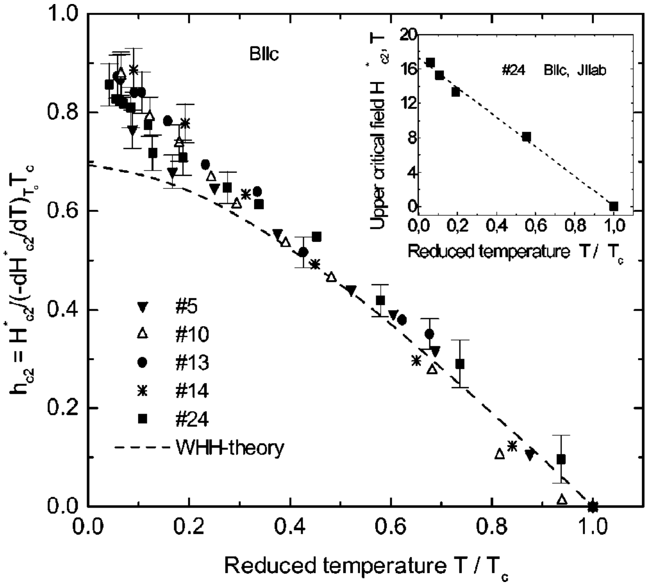

FIG. 5. Reduced critical field $h_{c2}=H^*_{c2}/(-dH^*_{c2}/dT)_{T_c}T_c$ as a function of the reduced temperature T/T_c for five samples together with the theoretical WHH curve (dashed line).[20] The inset shows the temperature dependence of H^*_{c2} at points where the Hall resistivity $\rho_{xy}(H)=0$.

motion dissipation becomes less noticeable when H^*_{c2} is determined at a higher fraction of ρ_n. Hence, we suppose that the obtained data for $H^*_{c2}(T)$ in the $\mathbf{H}\|\mathbf{c}$ geometry using the $\rho=0.9\rho_n$ and $\rho=\rho_n$ criteria are rather close to the true values. Then the upper critical field for our Bi2201 single crystals is $H^*_{c2}(0)_{ab}=16-27$ T. Because of the very high upper critical fields in the $\mathbf{H}\|\mathbf{ab}$ geometry, we could determine $H^*_{c2}(0)_c\simeq 43$ T for the $\rho=0.5\rho_n$ criterion only. If we take $H^*_{c2}(0)_{ab}$ at $\rho=0.5\rho_n$ equal to 16 T (Fig. 3), we find that the anisotropy $H^*_{c2}(0)\|_{ab}/H^*_{c2}(0)\|_c$ equals 2.7. We may also estimate the ab plane and $\mathbf{c}$ axis coherence lengths at zero temperature from the anisotropic Ginsburg-Landau relations[28] $H^*_{c2}(0)\|_c=\Phi_0/2\pi\xi^2_{ab}$ and $H^*_{c2}(0)\|_{ab}=\Phi_0/2\pi\xi_{ab}\xi_c$. Using the data from the $\rho=\rho_n$ criterion we get for four samples $\xi_{ab}=35-45$ Å in the ab plane and $\xi_c=14$ Å in the $\mathbf{c}$ direction. The ξ_{ab}/ξ_c anisotropy value is found to be 2.7. Comparing these values of ξ_{ab} with the averaged mean-free path $\overline{l}=70$ Å our crystals appear to be neither in the clean limit nor in the dirty limit with $\xi_{ab}/\overline{l}\approx 0.6$. The values of $H^*_{c2}(0)_{ab}$ and $\xi_{ab}(0)$ are close to the ones obtained earlier for optimally doped Bi2201 single crystal.[10] Using the energy gap value[29] $\Delta_0=3.5$ meV, we can estimate the Clogston paramagnetic limit $H_p(0)=\Delta_0/\mu_B\sqrt{2}\simeq 24$ T (μ_B is Bohr magneton). That is close to the measured values of $H^*_{c2}(0)_{ab}$ but lower than the measured $H^*_{c2}(0)_c$. A similar suppression of the paramagnetic effect was also observed in the conventional layered superconductors $NbSe_2$ and TaS_2.

In Fig. 5 we display the reduced critical field $h_{c2}=H^*_{c2}/(-dH^*_{c2}/dT)_{T_c}T_c$ as a function of the reduced temperature T/T_c for five investigated samples at field orientation $\mathbf{H}\|\mathbf{c}$ together with the theoretical WHH curve[20] which describes the behavior of the upper critical field in conventional type-II superconductors without spin paramagnetic and spin-orbit effects. Except for the small region at lower temperatures $T/T_c<0.3$ the experimental data can be described by the conventional WHH theory. We did not observe any unusual behavior of the resistive upper critical field H^*_{c2} in any Bi2201 single crystals that were studied. Notice that the agreement between WHH theory and $H_{c2}(T)$ data has previously been observed also by Chen[30] in studies of a single-crystalline Bi2201 sample in magnetic field up to 8 T at temperatures 4.3−7 K.

The result in Fig. 5 clearly shows that the data for the temperature dependence of the reduced critical field h_{c2} can be described by a linear dependence which is very close to the WHH curve. One can suppose that the deviation of the experimental points at $T/T_c<0.3$ is indicative of a positive curvature down to the lowest temperatures. However, one must note that at these temperatures it is rather impossible to determine the superconducting transition exactly due to the presence of superconducting fluctuations. In addition, at $T\ll T_c$ the coherence length is very short and inhomogeneities on the scale of $\xi(T)$ can be resolved, leading to deviation from the averaged properties.[31] Coffe, Muttalib, and Levin[32] calculated the temperature dependence of H_{c2} for highly disordered superconductors and predicted that because the magnetic field diminishes the localization effect, the low-temperature values of H_{c2} can show an enhancement over the WHH curve for dirty superconductors. However, this phenomenon plays probably no role here because in this case one would also expect a negative normal-state in-plane MR. At low temperatures we always observed a positive MR.

Hall effect measurements are a useful method in determining the transport behavior in metals and semimetals. Previous experiments on $YBa_2Cu_3O_7$ revealed a sign-reversal anomaly of the Hall coefficient R_H near T_c.[33] Kopnin, Ivlev, and Kalatsky[34] describe the flux-flow Hall conductivity as a sum of the quasiparticle and vortices parts $\sigma_H=\sigma^n_H+\sigma^f_H$. σ^f_H is only determined by the imaginary part of the relaxation time and has the same sign for any vortex direction.[35] The Hall conductivity is independent of pinning. The change of sign in Hall resistivity $\rho_{xy}(H)$ relates to exceeding a positive σ^n_H relative to a negative σ^f_H. Roughly approximated, the applied field with $\rho_{xy}(H)=0$ may be thought of as an approximate estimation of $H^*_{c2}/2$ at given temperature. We have measured the field dependence of the mixed-state Hall effect on one of our Bi2201 single crystals #24 at different temperatures down to 0.4 K in magnetic fields up to 23 T perpendicular to the ab plane. We find that $\rho_{xy}(H)$ is negative with a broad minimum at low fields and holelike at high fields in the normal state. By plotting the magnetic fields at points where $\rho_{xy}(H)=0$ against reduced temperature, we find in the inset of Fig. 5 that H^*_{c2} varies linearly with T. The $H^*_{c2}(T)$ data determined by this means are in close agreement with those extracted from the ρ_{ab} resistivity for the same sample (Fig. 2).

Figure 6 shows the resistive transitions of sample #13 in a magnetic field $\mathbf{H}\|\mathbf{c}\|\mathbf{J}$ at different temperatures. The maximum value of the out-of-plane resistivity ρ_c increases with decreasing temperature in the magnetic field just as in studies of $Bi_2Sr_2CaCu_2O_{8+\delta}$.[36,37] This reflects the common "semiconducting" normal-state behavior of ρ_c in the layered Bi family. The question of the coexistence of the metallic in-

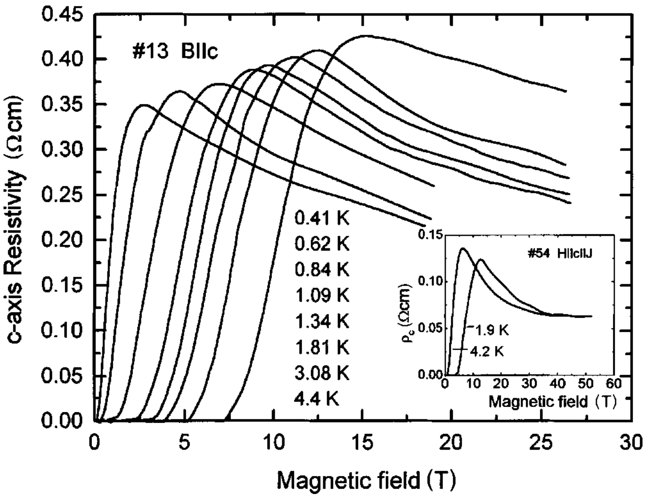

FIG. 6. Magnetic-field dependences of the out-of-plane resistivity ρ_c of sample #13 at different temperatures. Inset shows the magnetic field dependence of ρ_c for crystal #54 at $T=4.2$ K and 1.9 K in the 52-T pulsed magnet.

plane resistivity with the ''semiconducting'' out-of-plane resistivity down to low temperatures has attracted much attention in last few years and several models have been proposed to explain it. Here, we do not want discuss this topic. However, we would like to notice that the out-of-plane resistivity ρ_c at $T \ll T_c$ shows a tendency to saturation in very high magnetic fields for the samples with $T_c \leqslant 5$ K. We observed this saturation of the out-of-plane MR in pulsed magnetic fields with the $\mathbf{H}\|\mathbf{c}\|\mathbf{J}$ geometry at $40-50$ T. The inset in Fig. 6 shows the magnetic field dependence of ρ_c for crystal #54 at $T=4.2$ K and 1.9 K. The lock-in-amplifier operates at 10 kHz with an excitation current 500 μ A. Unfortunately, precise measurements of ρ_c are a difficult task in the pulsed magnet, due to the very low resistance of our crystals along the **c** axis yielding a higher noise level.

Considering that Bi2201 belongs to the family of the highly anisotropic layered superconductors BiSrCaCuO, it could be expected that the out-of-plane resistivity ρ_c cannot provide information on H_{c2}^* because in this case the behavior of ρ_c is related to Josephson effects and not with the formation of a superconducting order parameter inside the CuO_2 layers. The resistivity anisotropy $\rho_c/\rho_{ab} \sim 10^3-10^4$ shows that Bi2201 is a highly anisotropic superconductor. However, in contrast to an anisotropy parameter $\gamma=\xi_{ab}/\xi_c =200-1000$ for $Bi_2Sr_2CaCu_2O_8$,[22] for Bi2201 the value of $\xi_{ab}/\xi_c \simeq 2.7$ was obtained, which could lead to the opposite of the above given conclusion. Indeed, the $H_{c2}^*(T)$ dependence determined by the maximum in the $\rho_c(H)$ curves exhibits a significant positive curvature. The $H_c^*(T)$ data lie close to the points in Fig. 3(a) obtained for the same sample using the 50% criteria. This is consistent with the experimental data because the maxima of $\rho_c(H)$ coincide with the field positions of $\rho_{ab}(H)=0.4\rho_n$. Hence, the peak effect in $\rho_c(H)$ is a property of the mixed state and results from a competition between the ''semiconducting'' behavior of ρ_c and the superconducting transition.

All $\rho_c(H)$ curves (Fig. 6) have a pronounced break point in the derivative well above the $\rho_c(H)$ peak, which shifts to higher fields with decreasing temperature. At $T \simeq T_c$, this break point disappears. The field position of these break points in the derivative coincide with the H_{c2}^* values determined by the $\rho_{ab}=\rho_n$ criterion with a similar temperature dependence. Since the mechanism of a negative out-of-plane longitudinal magnetoresistance is still unclear, we can only give a qualitative discussion of this phenomenon.

In conclusion, we have studied the superconducting resistive transition in several high-quality $Bi_2Sr_2CuO_x$ single crystals with critical temperature T_c (midpoint)$=3.7-9$ K in magnetic fields up to 28 T for continuous fields and up to 52 T for pulsed fields, both perpendicular and parallel to the ab plane. We found that the temperature dependence of the resistive upper critical field H_{c2}^*, determined as the field at which the in-plane resistivity in the transition region is 90% or more of the normal-state resistivity, is down to the temperatures $T/T_c=0.3$, in close agreement with the Werthamer-Helfand-Hohenberg theory for conventional type-II superconductors. The measured field dependence of the mixed-state Hall effect at different temperatures down to 0.4 K in the magnetic field up to 28 T confirms the temperature dependence of the resistive upper critical field extracted from the superconducting transitions.

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