Negative Magnetoresistance of Bi$_2$Sr$_2$CuO$_x$ Single Crystals in a Strong Magnetic Fields.

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Magnetoresistance (MR) in the out-of-plane resistivity $\rho_c$ for the normal state of the one-layer high-quality Bi$_2$Sr$_2$CuO$_x$ single crystals under various dc magnetic fields up to 28 T over the temperature region 6 – 100 K has been measured. We observed the anomalously large negative longitudinal MR up to 60%. At low temperatures the normal-state MR in contrast to the MR in mixed state is independent of the direction of the current relatively to the field direction suggesting uniquely the spin dominated origin of that. The magnitude of the MR is activated in magnetic field and temperature. We interpret the activated form of $\rho_c$ and the negative MR in terms of 2D stacked alternating metallic and dielectric layers assuming the tunneling between CuO$_2$ planes. If the main fluctuations inside CuO$_2$ planes have magnetic origin, the magnetic field suppresses these fluctuations leading to the uniform spin orientation. In this case the interlayer current will be enhanced well.

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1. The magnetotransport properties of the layered high-$T_c$ superconductors (HTSC) are characterised by anomalous quasi-two-dimensional (2D) states, which are studied very extensively in recent years. One of unusual features of the HTSC normal state properties is the coexistence of the metallic in-plane resistivity $\rho_{ab}$ and the “semiconducting” out-of-plane resistivity $\rho_c$ (e.g. Ref.2,3). Recently, such behaviour of the resistivities $\rho_{ab}$ and $\rho_c$ were measured by Ando et al. in La-doped Bi$_2$Sr$_2$CuO$_y$ ($T_c = 13$ K) down to temperature as low as $T/T_c \sim 0.04$. The latter implies a 2D confinement and is incompatible with a Fermi-liquid behaviour. The small-value (~ 1%) negative out-of-plane magnetoresistance (MR) around $T_c$ for Bi$_2$Sr$_2$CaCu$_2$O$_8$ and YBa$_2$Cu$_3$O$_7$ was explained by a fluctuation conductivity or in terms of a pseudogap in spin system that was reduced by magnetic field. A small negative isotropic MR was observed also in underdoped La$_{2-x}$Sr$_x$CuO$_4$. The normal-state properties essentially depend on a carrier concentration or doping also. In recent experiments the unusual field dependence of the large positive $c$-axis MR of the underdoped cuprate YBa$_2$Cu$_3$O$_8$ has been assigned to the magnetic field decoupled of the chains resulting in a 3D to 2D crossover in the transport behaviour of the whole system. Yoshizaki et al. have observed positive and negative out-of-plane MR up to 2% over the temperature region 35 – 200 K for magnetic fields 0 – 17 T in underdoped Bi$_2$(SrLa)$_2$CuO$_{6+\delta}$. Although magnitude of the negative longitudinal MR was about five times larger than that for the transverse MR they have proposed that data indicated the Lorentz-force independent spin-dominated origin of the MR. These results were discussed from a view point of pseudo-spin-gap formation. Very recently a change of sign from positive near $T_c$ to negative above $T_c$ in the out-of-plane magnetoconductivity of the Tl$_2$Ba$_2$Ca$_2$Cu$_3$O$_{10+\delta}$ compound was again explained by the density-of-states fluctuations. In the same time the one-layer (Hg,Cu)Ba$_2$CuO$_{4+\delta}$ did not show any anomaly in its magnetoconductivity. Present no single theory has been able to account for all the anomalies found in the normal-state properties of HTSC.

In this paper, we describe the experiments to study the $c$-axis resistance in normal state of high-quality nondoped Bi$_2$Sr$_2$CuO$_{6+\delta}$ (Bi2201) single crystals under various dc magnetic fields up to 28 T over the temperature region 6 – 100 K. At low temperatures the almost isotropic negative out-of-plane MR up to 60% was observed.

2. The Bi2201 single crystals were grown by KCl-solution-melt method. A temperature gradient along a crucible results in formation a big closed cavity inside the solution-melt. Number of the crystals which share the common properties reached several tens in the cavity. The quality of the crystals was verified by the measurements of the $dc$ resistance, $ac$ susceptibility, X-ray diffraction and energy dispersive X-ray microprobe analysis. Our crystals showed the X-ray rocking curves width about 0.1° – 0.3°. The two crystals with $T_c = 9.5$ K (midpoint) and $\Delta T_c \simeq 1$ K were investigated. Dimensions of the crystals were 0.5 mm × 1 mm × 3 $\mu$m (#1) and 0.5 mm × 1 mm × 10 $\mu$m (#2). A four-probe contact configuration with symmetrical position of the low-resistance contacts (< 1Ω) on both $ab$-surfaces of the sample was used for the measurements of the in-plane and out-of-plane resistances. The measured resistances was transformed to $\rho_c$ and $\rho_{ab}$ using the crystal dimensions. In zero magnetic field the samples showed nearly linear temperature dependence $\rho_{ab}(T)$ which saturates below 20 K to a residual resistivity 50 and 80 $\mu$Ω·cm. The out-of-plane resistivity of the single crystals $\rho_c(T)$ over the temperature region $T = 10 – 300$ K showed the conventional “semiconducting” normal-state behaviour of $\rho_c$.
in layered Bi-family and was in reasonable good agreement with a power law $T^{-a}$ with $a = 1.65$ (crystal #1) and 1.3 (crystal #2) without a linear-$T$ term. The $\rho_c$ values at $T = 100$ K of the thin and thick samples are equal 2.7 and 13.5 m$\Omega$-cm, respectively. The anisotropy ratio $\rho_c/\rho_{ab}$ was nearly $5 \times 10^3$ at low temperatures. By measuring the Hall coefficient $R_H$ in the crystals we determined the carrier density $n = 4.8 \times 10^{21}$ cm$^{-3}$. The crystals were studied with the magnetic field $H$ in longitudinal ($H \parallel c \parallel J$) and transverse ($H \parallel ab \perp J$) configurations.

3. Fig.1 displays the field dependence of the longitudinal $\rho_c$ (a) and MR, $\Delta \rho_c/\rho_{c0}$, (b) as $H$ is parallel to $c$-axis for the sample #1 at different temperatures just below $T_c$ and above $T_c$. (At $T < T_c$ the peak value of $\rho_c(H)$ was selected as $\rho_{c0}$). The out-of-plane resistivity is decreasing with increasing magnetic field, which is consistent with the preceding papers.

![FIG. 1. The field dependence of the out-of-plane resistivity $\rho_c$ (a) and longitudinal MR, $\Delta \rho_c/\rho_{c0}$, (b) as $H$ is parallel to $c$-axis for the sample #1 at different temperatures just below $T_c$ and above $T_c$. (At $T < T_c$ the peak value of $\rho_c(H)$ was selected as $\rho_{c0}$).](image1)

However there are two distinguishing features to note from this figure. First, at low temperatures the $\rho_c$ decreases too rapidly as compared with other works. For example, at $T \approx 6$ K the $\rho_c$ decreases its value more than by a factor of 2.5. With increasing temperature the MR decreases and becomes positive above $\approx 26$ K. In the very high fields $\rho_c$ shows tendency to the saturation. We have observed the clearly defined saturation of the out-of-plane MR after its twofold decreasing in our crystals at $T = 1.9 - 4.2$ K in the pulsed magnetic fields the longitudinal geometry at $40 - 50$ T$^1$. However, the precise measurement of $\rho_c$ in the pulsed magnet was found a difficult task due to very low out-of-plane resistance of our crystals and higher noise level. It should be noted that Ando et al.$^4$ have observed the 10% negative out-of-plane MR in La-doped Bi$_2$Sr$_2$CuO$_y$ at $T = 0.8$ K by suppressing superconductivity with pulsed magnetic fields. They have found that the magnetic-field dependence of the negative MR is approximately linear and there is no sign of saturation up to 60 T. The second peculiarity is the extremely large negative MR which was observed in the parallel configuration where the macroscopic Lorentz force should be absent.

![FIG. 2. The out-of-plane resistivity $\rho_c$ (a) and $\Delta \rho_c/\rho_{c0}$ (b) in the transverse configuration ($H \parallel ab, J \parallel c$) as a function of $H$ for the same sample #1 at selected temperatures.](image2)

The $\rho_c$ (a) and $\Delta \rho_c/\rho_{c0}$ (b) in the transverse configuration ($H \parallel ab, J \parallel c$) are plotted in Fig.2 as a function of $H$ for the same sample #1 at selected temperatures. It is apparent that a very large negative out-of-plane MR is present in the normal-state once again. The difference in the normal-state magnitudes of $\Delta \rho_c/\rho_{c0}$ in Fig.2 and Fig.1 is no more than several percent. One can also see...
that the normal-state MR in contrast to the MR in mixed state is almost independent of the magnetic field direction with respect to the current direction. The larger resistive onset field at $T < T_c$ is a direct consequence of the large anisotropy of upper critical field in Bi2201. The critical field in this direction is much larger than that when the field is perpendicular to the $ab$-plane. While Lorentz-force-independent MR in normal state uniquely indicates the spin dominated origin of that.

Similar behaviour of the $\rho_c$ and $\Delta \rho_c / \rho_{ab}$ from second crystal #2 was obtained except that at high magnetic fields the magnitude of $\Delta \rho_c / \rho_{ab}$ was $\approx 2$ times smaller previous one. The crystals #1 and #2 have the same nominal composition, the same $T_c$, but were grown in different crucibles. The fact that $\rho_c$ and MR of the crystals significantly differ presumably reflects varying disorder along $c$-axis, which is believed to be due to additional insulating layers in the thick sample. Our studies shown that those layers existed generally in the crystals thicker than $3 - 5 \mu$m.

The temperature dependencies of the out-of-plane resistivity $\rho_c(T)$ in sample #1 over the temperature region $6 - 80$ K at zero-field curve (solid line) together with values of $\rho_c(T)$ at 10, 20, and 28 T extracted from Fig.1(a). The inset shows similar $\rho_c(T)$ at zero-field (solid line), at 10, and 20 T data for sample #2.

The temperature dependencies of the out-of-plane resistivity in sample #1 over the temperature region $6 - 80$ K is represented in Fig.3, where we show the zero-field $\rho_c(T)$ curve (solid line) together with values of $\rho_c(T)$ at 10, 20, and 28 T extracted from Fig.1(a). The $\rho_c(T)$ curves intersect the zero-field $\rho_c(T)$ curve at $T \approx 26$ K where the MR shows a change of sign as the temperature is increased. The inset shows similar $\rho_c(T)$ at zero-field (solid line), at 10, and 20 T data for sample #2.

In the quasi-classical model the longitudinal MR arises from a spin-dependent scattering which is independent of the field direction. This MR in conventional metals is positive, very small ($\sim 10^{-3}$) and increases quadratically with $H$. In cited above works$^{7,10}$ the negative out-of-plane MR is quadratic of the magnetic field in the wide range of the field up to 14 T to a good approximation or that is linear up to 60 T$^4$. Our $\rho_c$ data can be well fitted to the functional form $\rho_c(H) = A_0 + A_1 \exp(-H/bT)$, where $H$ and $T$ are in T. The constant $b$ equals $\approx 2.8$, with the Zeeman field taken as $H_Z = k_B T / g \mu_B \approx 0.74$ T at 1 K ($g = 2$ and $\mu_B$ are $g$ factor and Bohr magneton). The anomalously large negative longitudinal MR has been observed more early in the transition metal dichalcogenides$^{14}$ which have a layer type structure. Fukuyama and Yosida$^{15,16}$ have explained this phenomenon on basis of a variable-range hopping mechanism in Anderson localized states$^{17}$. The application of the magnetic field introduces Zeeman shifts of each eigenstate dependent on the spin directions and causes the repopulation among the localized states. The energy difference decrease with increasing field and the interplane conduction increases exponentially$^{16}$. If we assume that this scenario is correct for Bi2201 compound also then it may explain the negative longitudinal MR. However, the magnitude of the observed MR is too large to be considered for this model and the field value at which the MR should fall off sharply is more less than we observe.

We have attempted to describe qualitatively our data in terms of 2D stacked alternating metallic and dielectric layers assuming the tunneling of electrons between the adjacent CuO$_2$ planes. Based on the resistance anisotropy ($\rho_c / \rho_{ab} \sim 10^4 - 10^5$) it is believed that a transition amplitude of the charge carriers between the planes is two order of magnitude less than that within the CuO$_2$ layer over the same distance. As an added argument of the 2D charge transport in the Bi2201 crystals is a slight negative in-plane MR $\Delta \rho_{ab} / \rho_{ab0}$, which we observed only when the field is applied perpendicular to the CuO$_2$ layers. In this case the magnetic field dependence of in-plane MR is partially described by weak localization theory. Observed here the extremely large negative out-of-plane MR is independent of the magnetic field direction, suggesting the spin-dominated origin of MR.

We will proceed from the assumptions that: (i) - the current $J$ along $c$-axis determines by the electron tunneling between the neighbouring metallic weakly linked layers with the lower-order transition amplitude equals $T$ $(J \sim |T^2|)$; (ii) - the crystal is perfected, then, the charge carriers transport between the layers should be elastic with conserving of the quasiparticle momentum along the layers and spin; (iii) - the planar density of states in the vicinity of a Fermi level is free from the singularities. From the last assumption it follows that in the absence of an interaction between electrons in each layer the $c$-axis current has not to depend on the magnetic field.

Really, though a spin-splitting of the 2D-bands leads to an electron density redistribution among the bands, the
total current is given by the sum of both spin components remains constant. It follows that the negative MR can be associated with the electron gas unideality only which should depend on the magnetic field. In terms of an one-particle Green functions it is inferred that a renormalization constant $G = Z / (\omega - \varepsilon_k - U)$ is less than 1, where $\varepsilon_k$ is a 2D electron spectr in the layer and $U$ is a layer potential. (For the ideal gas $Z = 1$).

According to our assumptions (i) and (ii), the tunnel current is determined by a convolution of the Green function in the adjacent CuO$_2$ layers. Vertices of the convolution are transition amplitudes among the layers which equal $T$. Hence one can expect that the interlayer current $J$ is proportional to $Z^2$. If the magnitude of $Z$ increases with increasing field, the current $J$ will be enhanced well. This is possible when main fluctuations determining $Z$ value in the layer have a magnetic origin. In this case the magnetic field suppresses these fluctuations leading to the uniform spin orientation. It is evident that in an intense fields saturation of the magnetic field dependence of the tunnel conductance is expected when all spins are oriented in the same direction and the temperature dependence of MR has activated character.

Quantitative calculation of $Z(H,T)$ behaviour essentially depends on choosing of a model of the magnetic correlations in CuO$_2$ layer. These correlations may be caused by a band magnetism, interaction of the copper magnetic moments with the band electrons, various paramagnons and so on. The different theoretical models should be compared with the MR experimental data in the further in order to make clear a nature and role of the magnetic interactions in the superconductivity of high-$T_c$ cuprates.

To summarize, we measured the MR in the out-of-plane resistivity for the normal state of the one-layer high-quality Bi$_2$Sr$_2$CuO$_{6+\delta}$ single crystals under various dc magnetic fields up to 28 T over the temperature region 6 – 100 K. We observed the anomalously large negative longitudinal MR up to 60%. At low temperatures the normal-state MR in contrast to the MR in mixed state is independent of the direction of the current relatively to the field direction suggesting uniquely the spin dominated origin of that. The magnitude of the MR is activated in magnetic field and temperature. With increasing temperature the MR decreases and becomes positive above $\approx 26$ K. In the very high fields MR shows tendency to the saturation. We interpret the activated form of $\rho_c$ and the negative MR in terms of 2D stacked alternating metallic and dielectric layers assuming the tunneling between CuO$_2$ planes. If the main fluctuations inside CuO$_2$ planes have magnetic origin, the magnetic field suppresses these fluctuations leading to the uniform spin orientation. In this case the interlayer current will be enhanced well.

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