Magnetoresistance in Heavily Underdoped YBa$_2$Cu$_3$O$_{6+x}$: Antiferromagnetic Correlations and Normal-State Transport

A. N. Lavrov, Yoichi Ando, Kouji Segawa and J. Takeya
Central Research Institute of Electric Power Industry, 2-11-1 Iwato-kita, Komae, Tokyo 201-8511, Japan
(March 24, 2022)

We report on a contrasting behavior of the in-plane and out-of-plane magnetoresistance (MR) in heavily underdoped antiferromagnetic (AF) YBa$_2$Cu$_3$O$_{6+x}$ ($x\leq 0.37$). The out-of-plane MR ($I\parallel c$) is positive over most of the temperature range and shows a sharp increase, by about two orders of magnitude, upon cooling through the Néel temperature $T_N$. A contribution associated with the AF correlations is found to dominate the out-of-plane MR behavior for $H\parallel c$ from far above $T_N$, pointing to the key role of spin fluctuations in the out-of-plane transport. In contrast, the transverse in-plane MR ($I\parallel a(b); H\parallel c$) appears to be small and smooth through $T_N$, implying that the development of the AF order has little effect on the in-plane resistivity.

74.25.Fy, 74.20.Mn, 74.72.Bk

High-$T_c$ superconductivity (SC) in cuprates occurs as a crossover phenomenon in the doping range between an antiferromagnetic (AF) insulator and a Fermi-liquid metal state. While the hole (electron) doping destroys the long-range AF order in the CuO$_2$ planes, short-range AF correlations persist well into the superconducting compositions \[\text{[1]},\] and thus it is likely that the interplay of the doped carriers with the AF correlations underlies the physics of cuprates in a wide range of carrier concentrations \[\text{[2–4]}.\]

In order to clarify the role of the magnetic interactions in cuprates one may study the temperature and doping regions which are peculiar for the spin subsystem. So far, a crossover at a temperature $T^*$ corresponding to the formation of a pseudogap in the spin and charge excitation spectra has attracted most attention. An additional decrease in the in-plane resistivity $\rho_{ab}$ below $T^*$ observed in underdoped YBa$_2$Cu$_3$O$_{6+x}$ (Y-123) and in YBa$_2$Cu$_4$O$_8$ (Y-124) suggests the possibility that the in-plane transport is determined to a large extent by the spin scattering \[\text{[5,6]}.\] The pseudogap (or spin gap) was also employed to explain both the activated behavior of the out-of-plane resistivity $\rho_c(T)$ and the negative out-of-plane magnetoresistance (MR) in Y-123 and Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ \[\text{[7,8]}.\]

The vicinity of the Néel transition is another peculiar region for the spin subsystem besides the crossover at $T^*$. The dynamic AF correlations developed in the CuO$_2$ planes evolve into the long-range AF order upon crossing the Néel temperature $T_N$ \[\text{[9]},\] and one can expect some singular behavior to show up in the properties governed by the magnetic interactions. A magnetotransport study in the vicinity of $T_N$ is therefore an attractive possibility to clarify the role of spin degrees of freedom in the peculiar electron transport.

In this Letter, we present a study of the in-plane and out-of-plane MR of heavily underdoped antiferromagnetic Y-123 crystals, supplemented by measurements of the Lu-123 crystal used earlier for the study of the phase diagram \[\text{[10]}.\] We find that the out-of-plane MR undergoes a drastic change in the vicinity of the Néel temperature, increasing by about two orders of magnitude with a transition into the AF state. At the same time, quite unexpectedly, no feature associated with the AF ordering was observed in the transverse in-plane MR $I\parallel a(b); H\parallel c$. Therefore, the development of the AF correlations and the formation of the long-range Néel order have a profound influence only on the charge transport between the CuO$_2$ planes, leaving the in-plane transport unchanged. Moreover, we find that the longitudinal out-of-plane MR ($H\parallel c$) is apparently governed by the AF fluctuations even in the temperature range above $T_N$, indicating that the spin fluctuations are playing a major role in the out-of-plane transport regardless of the presence of the Néel order.

The high-quality YBa$_2$Cu$_3$O$_{6+x}$ single crystals are grown by the flux method in Y$_2$O$_3$ crucibles to avoid incorporation of impurities, and their oxygen content is reduced by subsequent high-temperature annealing. While the c-axis resistivity is easily measured in these samples owing to the high anisotropy, special care is paid to measure $\rho_{ab}$ reliably; samples with a length/thickness ratio $\geq 100$-150 are used and the current contacts are carefully placed to cover the crystal side surfaces. The MR measurements are performed either by sweeping temperature (controlled by a Cernox resistance sensor) in constant magnetic fields up to 16 T, or by sweeping the field at a fixed temperature stabilized by a capacitance sensor to an accuracy of about 1 mK. The latter method allows measurements of $\Delta \rho/\rho$ as small as $10^{-5}$ at 10 T.

It was recently reported that heavily underdoped RBa$_2$Cu$_3$O$_{6+x}$ ($R=$Tm, Lu) exhibits a maximum in $\rho_c(T)$, which originates from a competition between two distinct mechanisms contributing to the interplane transport \[\text{[11]}.\] The long-range AF ordering brings about an additional peculiarity; an abrupt increase in $\rho_c$ occurs upon cooling through $T_N$, which is followed by a resistivity di-
FIG. 1. $\rho_c(T)$ of an Y-123 single crystal at three different oxygen contents. Curves for $H=0$ and 16 T [$H\|[a(b)]$ are shown. The kink in the resistivity curves marks the AF transition. Inset: The derivative $d\rho_c/dT$ near $T_N$ for $x=0.32$ ($H=0$).

vergence at lower $T$. We confirm essentially the same behavior in YBa$_2$Cu$_3$O$_{6+x}$, and in Fig. 1 we present a set of $\rho_c(T)$ curves obtained for the same Y-123 single crystal at slightly different oxygen contents in the AF region. The rise in $\rho_c$ induced by the AF transition becomes more and more evident as $T_N$ is lowered; for high $T_N$, a derivative plot helps to highlight the anomaly and to evaluate $T_N$ (see inset to Fig. 1). It is worth noting that $T_N$ is extremely sensitive to the hole doping, as can be seen in Fig. 1, and the width of the AF transition observed here (10-15 K) is almost the smallest achievable value, indicating the high quality of our crystals.

Figure 2 demonstrates the unusual behavior of the out-of-plane MR in the heavily underdoped Y-123, where a step-like increase in $\Delta \rho_c/\rho_c$ is observed upon cooling through $T_N$. Except for a small difference in the MR step width, which is obviously related to the width of the AF transition, this striking feature is very reproducible within a set of Y-123 crystals and in the Lu-123 crystal. One would expect that the Néel transition in Y-123, like other phase transitions associated with the magnetic subsystem, is considerably affected by the application of magnetic fields. If a strong magnetic field suppresses AF order and lowers $T_N$, the out-of-plane MR should then become negative, because $\rho_c$ is enhanced below $T_N$. What we have found is opposite to these naive expectations; $T_N$ appears to be quite insensitive to the magnetic field and the out-of-plane MR, surprisingly, is positive.

Upon tilting the magnetic field (inset to Fig. 3), we observe a considerable anisotropy in the out-of-plane MR, $\Delta \rho_c/\rho_c$, which becomes largest for the transverse geometry at lower $T$. Besides the difference in the magnitude, the $T$ dependence of the out-of-plane MR is remarkably different between the $H\|[a(b)]$ and the $H\perp c$ geometries; while $\Delta \rho_c/\rho_c$ for $H\|[ab]$ keeps growing below $T_N$ (Fig. 2) after it shows a small peak, $\Delta \rho_c/\rho_c$ for $H\perp c$ gradually diminishes below $T_N$ after it shows a pronounced peak (Fig. 3). Therefore, the magnetoresistance becomes more anisotropic as the temperature is lowered below $T_N$.

To obtain an idea about the mechanisms which couple mobile carriers with the AF order, it is helpful to look also at the in-plane transport. For the oxygen contents under study the in-plane resistivity demonstrates a crossover behavior, passing through a minimum at $T = 50-60$ K [Fig. 4(a)]. To our surprise, the in-plane MR $\Delta \rho_{ab}/\rho_{ab}$, as well as $\rho_{ab}$ itself, is always smooth in the vicinity of $T_N$ and we do not find any anomaly which can be associated with the Néel transition. For the transverse in-plane MR $[H\|[a(b)]$, a small magnitude of the MR and a possible admixture of the asymmetric Hall component ($\propto H$) in raw data required more precise measurements with field sweeping to be performed; the resulting field dependences of $\rho_{ab}$ are presented in Fig. 4(b). The $T$ dependence of $\Delta \rho_{ab}/\rho_{ab}$ is shown in Fig. 4(c), where the in-plane MR reveals no correlation with the AF transition even at this sensitivity level ($\sim 10^{-5}$). Instead it is rather small and remains almost constant down to the temperature region where $\rho_{ab}$ acquires localizing behavior [Fig. 4(a)] and $\Delta \rho_{ab}/\rho_{ab}$ changes its sign. The Néel transition and the corresponding changes in the spin-excitation spectrum have therefore no effect on the charge transport within the CuO$_2$ planes.

An intriguing issue is whether the out-of-plane transport is sensitive exclusively to the long-range order arising below $T_N$. If the short-range AF correlations above $T_N$ also contribute to $\rho_c$ and its MR, we may expect that
the AF fluctuations play an essential role not only in the AF compositions but also in the SC compositions. Apparently, the more precise field-sweeping technique should be employed to investigate the behavior above \( T_N \), where the MR becomes very small. Besides, below \( T_N \) such measurements allow us to single out the main \( \gamma H^2 \) term from possible additional contributions to the MR [10]. The \( T \) dependences of \( \gamma_H H^2 \) and \( \gamma_c H^2 \) components of \( \Delta \rho_c/\rho_c \) (for \( H \perp c \) and \( H \parallel c \), respectively) presented in Fig. 5 depict the qualitative difference in the MR behavior for the two directions of the magnetic field. For \( H \parallel ab \) [Fig. 5(a)], the out-of-plane MR changes at \( T_N \) in a step-like manner by up to two orders of magnitude, where the width of the step is the same as the width of the AF transition itself. The step separates regions below and above \( T_N \) with relatively weak dependence of the MR on temperature. This behavior implies that the sensitivity to the magnetic field appears abruptly with the onset of the long-range AF order. On the other hand, for \( H \parallel c \) [Fig. 5(b)] we observe a MR peak at \( T_N \), which is accompanied by a tail spreading to far above \( T_N \). The MR as a function of temperature has no discontinuity at \( T_N \) and one can infer from Fig. 5(b) that \( \Delta \rho_c/\rho_c \) grows as \( T^{-k} \) with lowering \( T \) until the Néel transition interrupts this tendency. However, the right-hand side of the MR peak for \( H \parallel c \) (the \( T^{-k} \) behavior) apparently shifts with \( T_N \) when the \( x \) is changed, which indicates its relation to the AF ordering. Therefore, we can conclude that a mechanism associated with the AF fluctuations dominates the out-of-plane MR in a wide temperature range above \( T_N \) as well. This observation clearly demonstrates that the short-range AF correlations play an essential role in the interplane transport. At high \( T \), the longitudinal MR turns out to be weakly \(<(1.5-2.5) \times 10^{-5} \) at 10 T\) negative [Fig. 5(b)], which is reminiscent of the large negative MR observed in moderately underdoped Y-123 [8]. We note that this weak negative background has a negligible effect on the MR behavior in the temperature range up to \( 2T_N \) [Fig. 5(b)] shows how its subtraction modifies the data and is not important for the present discussion.

The contrasting behavior of the in-plane and out-of-plane MR indicates that changes which occur in the spin subsystem at \( T_N \) are influential only on the electron transport between the CuO\(_2\) planes and apparently not on the in-plane one. It is known that the heavily underdoped Y-123 above \( T_N \) possesses well-developed dynamic AF correlations in the CuO\(_2\) planes [1] and the Néel temperature actually corresponds to the establishment of AF order along the c-axis. The symmetry change accompanying the long-range order and a change in the spin dynamics are the only two mechanisms for the Néel transition to influence the electron transport. In spite of the sharp increase in \( \rho_c \) upon cooling through \( T_N \), opening of a gap in the quasiparticle energy spectrum due to the magnetic superstructure is unlikely, since one can hardly imagine a gap formation to have no impact on the in-plane transport. On the other hand, it is possible that the freezing of the spin degrees of freedom below \( T_N \) causes an increase in \( \rho_c \), if the spin fluctuations assist the electron hopping between the CuO\(_2\) planes. Since an

FIG. 3. \( T \)-dependences of the longitudinal out-of-plane MR (\( H \parallel c \)) at 16 T. Inset: Angular dependence of the out-of-plane MR measured at 30 K for a Y-123 crystal with \( T_N = 108 \) K (measurements performed within the 100\(^\circ\) range and extended for other angles).

FIG. 4. (a) \( \rho_{ab}(T) \) of an Y-123 single crystal. (b) \( H \)-dependences of the transverse in-plane MR. (c) \( T \)-dependences of the MR at 10 T for two \( x \) values.
FIG. 5. (a) T-dependences of the $\gamma_H H^2$ term in the transverse out-of-plane MR for Y-123 (open circles) and Lu-123 (solid circles). The arrows indicate $T_N$. (b) T-dependence of the $\gamma_H H^2$ term ($H \parallel c$) for Lu-123 (solid circles). The open triangles illustrate the data for Lu-123 after subtracting the weak negative background. Inset: The Lu-123 data on a linear scale. In both (a) and (b), the MR data for Y-123 for sweeping of the temperature are shown (dots) for comparison.

increase in $\rho_c$ also takes place when the magnetic field is applied, one can infer that the field suppression of the spin fluctuations is likely to be the main source of the positive out-of-plane MR in our heavily underdoped Y-123. Also, the dramatic changes in the out-of-plane transport associated with the evolution of the magnetic state might suggest that it is the spin subsystem that is responsible for the charge confinement within the CuO$_2$ planes.

Now let us discuss the actual mechanism which gives rise to the peculiar transport properties observed here. A possible picture to account for the observed features is the segregation of the doped holes into “stripes” which separate AF domains. In this picture, the confinement of the charges into the CuO$_2$ planes is substituted in a sense by the confinement to the quasi-1D stripes. Since the formation of the stripes themselves is governed by the magnetic interactions, it is not surprising that the spin degrees of freedom are playing a dominant role in the hole confinement and hence in the out-of-plane charge transport. Also, we can expect the in-plane MR to be very weak in this picture; the orbital MR term is irrelevant for the carriers moving along quasi-1D stripes, since the magnetic field cannot bend their trajectories. The spin-charge separation (naturally expected for 1D stripes) and the spin gap formed in both the stripes and their AF environment suggest that the spin-dependent scattering for the charge transport along stripes should not be large.

In summary, the out-of-plane transport in heavily underdoped YBa$_2$Cu$_3$O$_{6+x}$ is found to show anomalous magnetoresistance associated both with the Néel ordering and with the AF correlations above $T_N$. The MR behavior gives evidence that the spin fluctuations play an essential role in the interplane transport and hence suggest that the charge confinement within the CuO$_2$ planes is also fundamentally related to the spin degrees of freedom.

We are grateful to L. P. Kozeeva for providing the Lu-123 crystals. A.N.L. acknowledges the support from JIS-TEC through an STA fellowship.

* On leave from the Institute of Inorganic Chemistry, Lavrentyeva-3, 630090 Novosibirsk, Russia.

[1] A. P. Kampf, Phys. Rep. 249, 219 (1994); G. Aeppli et al., Science 278, 1432 (1997).
[2] B. P. Stojevik and D. Pines, Phys. Rev. B 55, 8576 (1997).
[3] V. J. Emery, S. A. Kivelson, and O. Zachar, Phys. Rev. B 56, 6120 (1997).
[4] S. C. Zhang, Science 275, 1089 (1997); N. Nagaosa, ibid. 275, 1078 (1997).
[5] T. Ito, K. Takenaka, and S. Uchida, Phys. Rev. Lett. 70, 3995 (1993).
[6] B. Bucher et al., Phys. Rev. Lett. 70, 2012 (1993).
[7] K. Takenaka, K. Mizuhashi, H. Takagi, and S. Uchida, Phys. Rev. B 50, 6534 (1994).
[8] Y. F. Yan, P. Matl, J. M. Harris, and N. P. Ong, Phys. Rev. B 52, R751 (1995).
[9] A. N. Lavrov, M. Yu. Kameneva, and L. P. Kozeeva, Phys. Rev. Lett. 81, 5636 (1998).
[10] The field dependence of $\rho_c$ at $T < T_N$ is not always trivial; for samples with $T_N \leq 50-60$ K, $\Delta \rho_c/\rho_c \propto H^2$ up to 16 T, but an additional negative low-field MR term appears to be superimposed on the $\gamma H^2$ behavior as the doping level of the sample is further reduced. This additional low-field MR feature can be explained by the rearrangement of the AF domain boundaries (stripes).
[11] Y. Ando, A. N. Lavrov, and K. Segawa, cond-mat/9905074.