

Magnetoresistance Anomalies in Antiferromagnetic $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$: Fingerprints of Charged Stripes

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We report novel features in the in-plane magnetoresistance (MR) of heavily underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$, which unveil a developed “charged stripe” structure in this system. One of the striking features is an anisotropy of the MR with a “*d*-wave”-like symmetry upon rotating the magnetic field H within the ab plane, which is caused by the rotation of the stripes with the external field. With decreasing temperature, a hysteresis shows up below ~ 20 K in the MR curve as a function of H and finally below 10 K the magnetic-field application produces a persistent change in the resistivity. This “memory effect” is caused by the freezing of the directionally ordered stripes.

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High- T_c cuprates can be viewed as doped antiferromagnetic (AF) insulators with the charge carriers transferred into perfect CuO_2 planes from the charge reservoir layers. In general, there is a tendency for AF insulators to expel doped holes and to show a phase separation into regions with and without the doped holes [1]. When the ions are not mobile and the Coulomb interaction between doped holes is effective, an intriguing microscopic state with holes gathered within an array of quasi-1D “stripes” may be realized [1–3]. An ordered striped structure has actually been observed by the neutron diffraction in a doped antiferromagnet, $\text{La}_2\text{NiO}_{4.125}$ [4]. Inelastic neutron scattering studies of superconducting (SC) cuprates found incommensurate magnetic fluctuations [5], which can be considered as dynamical stripe correlations [1]; it has been discussed that such dynamical stripes are pinned in $\text{La}_{1.6-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$, which shows a static striped structure similar to that of the nickelate except for the direction of the stripes [6].

Whether the microscopic charge inhomogeneities in the form of dynamic or static stripes, if present at all, are relevant to the unusual normal-state and SC properties in cuprates remains unclear. For SC compositions, the fast dynamics of the stripe correlations [5] make it implausible to study them by methods other than spectroscopy. In fact, very little is known about the electron dynamics in the stripes. The existence of the static stripes in cuprates has been documented for at least two cases: (a) in $\text{La}_{1.6-x}\text{Nd}_{0.4}\text{Sr}_x\text{CuO}_4$ [6], where the stripes are pinned by lattice deformations, and (b) in heavily underdoped, nonsuperconducting cuprates [2], in which the dynamical stripe correlations presumably cross over into a static stripe array as AF domains grow in the CuO_2 planes. To study the detailed properties of the stripes, it is natural to turn to either of these systems.

In this Letter, we present novel magnetoresistance (MR) features observed in heavily underdoped, antiferromagnetic $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$, which can hardly be explained without assuming a hole segregation into some flexible stripes.

The most intriguing observation is the possibility to induce a persistent in-plane anisotropy just by exposing a crystal to the magnetic field, upon which the higher conductivity axis is adjusted to the field direction. This phenomenon can be understood to originate from a field-induced topological ordering of the stripes, whose direction remains unchanged after removing the magnetic field at low temperatures.

The high-quality $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ single crystals were grown by the flux method in Y_2O_3 crucibles, and a high-temperature annealing was used to reduce their oxygen content. The exact oxygen content was determined by iodometry [7]. The magnetoresistance was measured using a standard ac four-probe technique by sweeping the magnetic field at fixed temperatures stabilized by a capacitance sensor with an accuracy of about 1 mK. The angular dependence of the MR was determined by rotating the sample within a 100° range under constant magnetic fields up to 16 T (Cernox sensor was used for the temperature control during the rotation).

Figure 1 shows the in-plane resistivity ρ_{ab} of the heavily underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ crystals studied in this work. One can see here that the evolution of the in-plane transport upon reducing the carrier concentration is much more gradual than is often considered; in Fig. 1, apart from the low- T region below ~ 50 K, $\rho_{ab}(T)$ retains a metal-like behavior even in these samples with $x \approx 0.30$ or 0.32 , which are located deep in the AF region of the phase diagram (see inset of Fig. 1). The growth of ρ_{ab} at low temperatures is notably slower than $A \exp(B/T)^k$ with $k = 1/4 - 1$, which also implies that the system we are dealing with is not a simple band-gap insulator nor an Anderson insulator.

These crystals demonstrate an unusual behavior of the in-plane MR, $\Delta\rho_{ab}/\rho_{ab}$, when the magnetic field H is applied along the CuO_2 planes. Figure 2 shows the in-plane MR measured in the longitudinal geometry [$H \parallel I \parallel a(b)$]. At low fields, this longitudinal in-plane MR is negative and follows roughly a T -independent parabolic curve ζH^2 , but

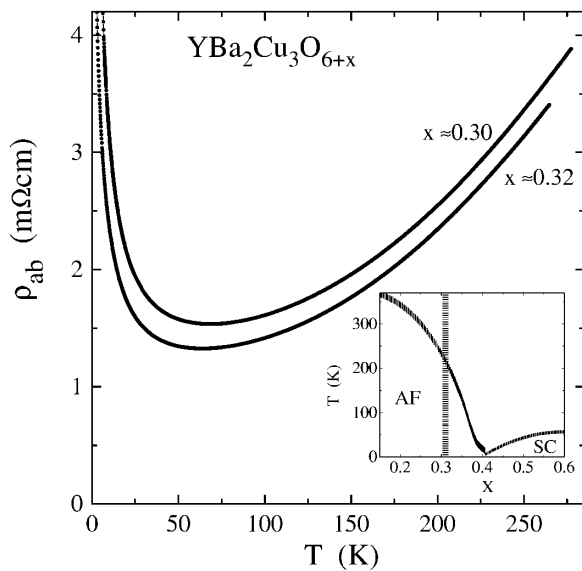


FIG. 1. $\rho_{ab}(T)$ of the two $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ single crystals. The inset illustrates the part of the phase diagram being studied.

abruptly saturates above some threshold field. The threshold field H_{th} and the magnitude of the saturated MR gradually increase with decreasing temperature. Above H_{th} , the MR can be fitted with the usual γH^2 dependence; namely, we can write the H dependence of the MR above H_{th} as $\Delta\rho_{ab}/\rho_{ab} = (\Delta\rho_{ab}/\rho_{ab})_0 + \gamma H^2$. Therefore, the behavior of the MR in Fig. 2 can be viewed as a superposition of the low-field feature [whose size can be measured by $(\Delta\rho_{ab}/\rho_{ab})_0$] onto a weak background MR of γH^2 . Note that γ is positive at high T and changes its sign to negative at about 50 K, where $\rho_{ab}(T)$ acquires the localizing behavior (see Fig. 1).

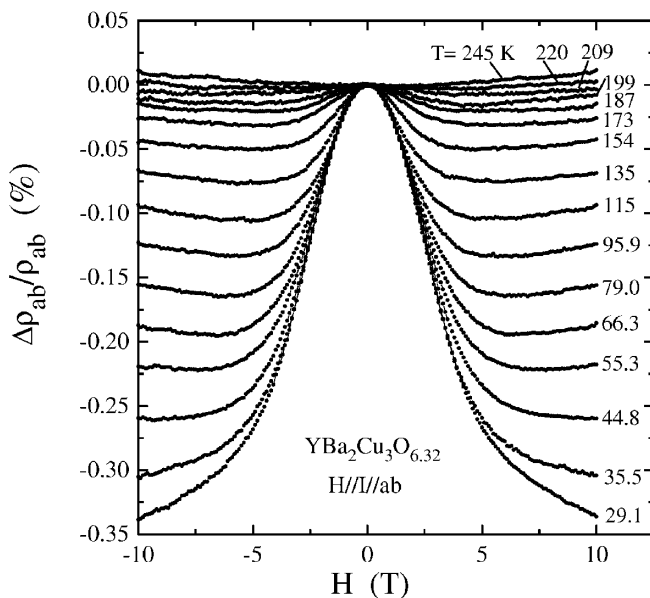


FIG. 2. In-plane longitudinal MR of $\text{YBa}_2\text{Cu}_3\text{O}_{6.32}$. The data are averaged over several field sweeps.

To our surprise, when the magnetic field is turned in the plane and becomes perpendicular to the current [$H \parallel a(b); H \perp I$], we find that the low-field MR term just switches its sign, retaining its magnitude and the threshold-field value [Fig. 3(a)]. We thus performed detailed MR measurements upon rotating H within the ab plane, which revealed a striking anisotropy with a “ d -wave” like symmetry [Fig. 3(b)]; namely, $\Delta\rho_{ab}/\rho_{ab}$ changes from negative at $\alpha = 0^\circ$ (α is the angle between H and I) to positive at $\alpha = 90^\circ$, passing through zero at about 45° . Note that the MR diagram in Fig. 3(b) is fairly symmetric. Some T -dependent difference between the longitudinal and transverse segments is mainly caused by the background MR; at 68 K, for example, the positive γH^2 term extends the “+” arm at $\alpha = 90^\circ$ and reduces the “-” arm at $\alpha = 0^\circ$.

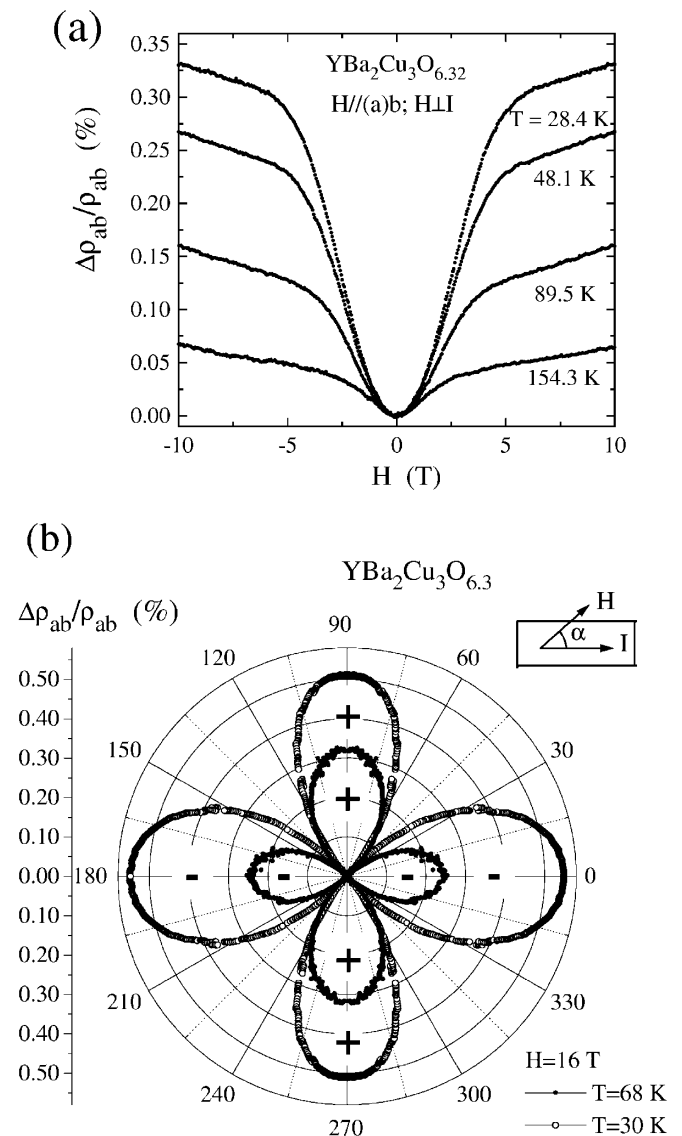


FIG. 3. (a) In-plane MR for the same sample as in Fig. 2, but with the field turned transverse to the current [$H \parallel a(b), H \perp I$]. (b) Angular dependences of the MR [$H \parallel (ab); H = 16 \text{ T}$] for $\text{YBa}_2\text{Cu}_3\text{O}_{6.3}$; the sign of MR is indicated. Actual data were taken for two quadrants at 68 K and for one quadrant at 30 K, and then extended to all angles.

It is worth noting that this low-field MR feature is not observed at all when the magnetic field is applied along the c axis; the in-plane MR for $H \parallel c$ is weak and approximately similar to the γH^2 term in the longitudinal MR (see also Ref. [8]). This means that the orbital part of the MR and interference effects are irrelevant to the anomalous low-field behavior observed here.

Since the samples being studied here are antiferromagnets, it is natural to look for a connection between the long-range AF order and the observed behavior. The Néel temperature T_N for these two samples with $x = 0.30$ and 0.32 has been evaluated to be about 230 and 200 K, respectively, from the measurements of c -axis resistivity ρ_c [8] [the slope of $\rho_c(T)$ shows a distinct anomaly at T_N [9]]. The sharpness of the anomaly in ρ_c tells us there is a very high homogeneity of the oxygen in our samples [8]. The magnitude of the low-field MR term, $(\Delta\rho_{ab}/\rho_{ab})_0$, plotted in Fig. 4 as a function of temperature, actually becomes noticeable near T_N and gradually grows with decreasing temperature. However, it is not clear whether the long-range Néel order itself plays a dominant role in the low-field MR anomalies, because $(\Delta\rho_{ab}/\rho_{ab})_0$ does not show an abrupt increase at T_N (which at least tells us that the feature is not related to the spin-flop transition). It is more likely that the anomaly is governed by the AF correlation within the CuO_2 planes, rather than the long-range order. It is worth noting that even at high T (near 200 K) the low-field MR feature remains sharp with a well-defined threshold field (inset of Fig. 4). This suggests that the anomaly is a co-

operative phenomenon; if the anomaly is just an integration of the behaviors of independent spins, the feature would be smeared by the thermal fluctuations at such high temperatures. We also note that, since Y has no magnetic moment, the anomaly observed here cannot be associated with magnetic ordering of the rare-earth sublattice.

The most intriguing peculiarity of the weak-field MR becomes evident at low temperatures. As the temperature decreases below 20–25 K, the H dependence of ρ_{ab} becomes irreversible. Initially this irreversibility appears as a small hysteresis on the MR curve [Figs. 5(a) and 5(b)]. However, as can be seen in Fig. 5(c), the irreversibility becomes much more pronounced upon cooling to 10 K. In Fig. 5(c), the $\rho_{ab}(H)$ curve similar to that at higher temperatures can be observed only in the first sweep which starts from $\Delta\rho_{ab}/\rho_{ab} = 0$; the data from the subsequent field sweeps significantly differ from the first one, with a strongly reduced low-field feature [the peak height is suppressed and the position of the peak is further shifted compared to the peaks in Fig. 5(b)]. The important point here is that the resistivity does not return to its initial value after removing the magnetic field; namely, the system acquires a memory. The application of the magnetic field at low temperatures therefore results in a persistent change in the resistivity; it can be easily seen that the sign and the size of this persistent change depends on the field direction (with respect to the current) according to the diagram in Fig. 3(b). This indicates that the magnetic field induces a persistent anisotropy of the in-plane resistivity, which has the “ d -wave”-like symmetry of Fig. 3(b).

It is very difficult to understand the novel MR features found here, especially the memory effect, without considering an inhomogeneous state or a superstructure in the CuO_2 planes instead of a uniform AF state. The observed hysteresis tells us that the structure is frozen in below ~ 20 K, while it is mobile at high and moderate temperatures. There are only two candidates which can produce a nonuniform structure: the ion rearrangement and the hole segregation. Since one can hardly imagine some rearrangement of ions to occur at such low temperatures and to be driven by the magnetic field, the holes can be the only possible factor responsible for the inhomogeneous structure.

The picture of charge inhomogeneities in the CuO_2 planes with holes being confined to the stripes [1] actually allows one to account for all the observed MR peculiarities. Since the AF interactions are essential for the formation of the stripes, it is expected that the development of the stripes is closely tied to the growth of AF domains in the CuO_2 planes. Indeed, for the heavily underdoped region with the long-range AF order, the measurements of the staggered magnetization and the Néel temperature [2] have provided a substantial support for the presence of the striped phase [3]. It is naturally expected that a strong local anisotropy is introduced by the stripes with confined carriers moving along; however, such an anisotropy will not be observed at long length scales if the orientational order of the stripes is

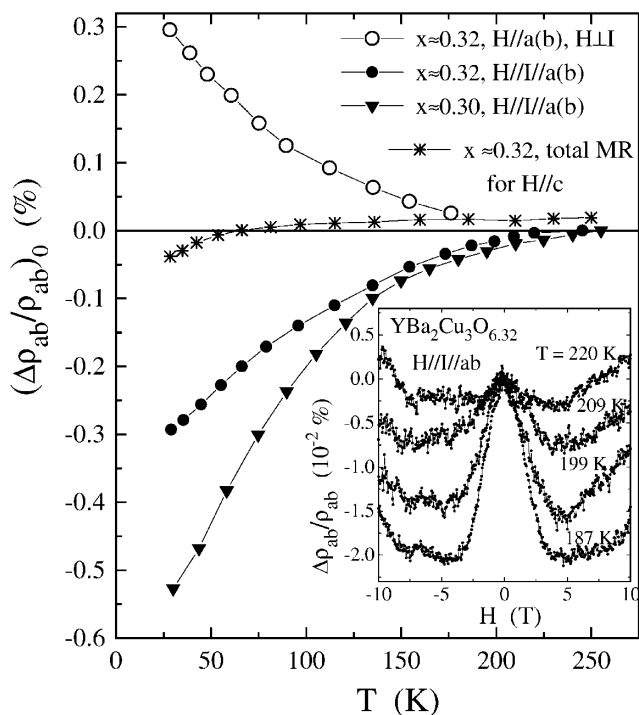


FIG. 4. Magnitude of the low-field MR for $H \parallel I$ and $H \perp I$. For comparison, total magnitude of the MR at 10 T is shown for $H \parallel c$, where there is no low-field anomaly. Inset: H dependences of the longitudinal MR at high T .

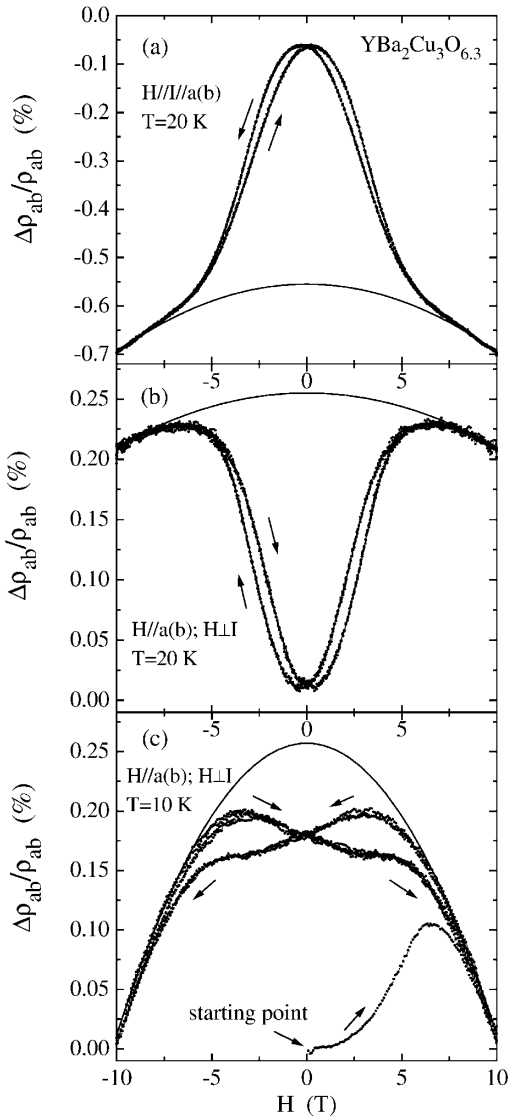


FIG. 5. Hysteresis in the low- T MR. Each curve contains data of four field sweeps (no averaging was made) performed at a rate of 1 T/min. In panel (c), the first run is different from subsequent ones and the resistivity does not return to its zero-field value. The solid lines depicts the background MR.

not established or the stripe direction alters from one CuO_2 plane to another. Within this picture, one can imagine that the magnetic field would give rise to a topological ordering of the stripes, aligning them along the field direction and changing the array of the current paths. The existence of the threshold field for the low-field feature is presumably coming from the establishment of the directional order of the stripes. Also, the “ d -wave”-like anisotropy of the in-plane MR can be understood as a consequence of the rotation of the stripe direction with respect to the current direction.

The observation that the stripes can be directed with external magnetic field implies that the stripes should

have local ferromagnetic moment. (Otherwise the stripes cannot couple to the magnetic field.) This is possible if the stripes are “in-phase boundaries,” at which the spin directions across the boundary are the same. We note that the MR behavior observed here surprisingly resembles that of a ferromagnetic metal with a mesoscopic domain structure [10].

As the temperature is lowered, it is expected that the stripe dynamics slows down and the magnetic domain structure in the CuO_2 planes is frozen, forming a cluster spin glass [11]. The spin-glass transition temperature has been reported for heavily underdoped $\text{Y}_{1-y}\text{Ca}_y\text{Ba}_2\text{Cu}_3\text{O}_6$ to be about 20–25 K for the AF compositions [11], which is in good agreement with the temperature where the hysteretic MR behavior is found to show up in our experiment.

In summary, the in-plane MR in heavily underdoped $\text{YBa}_2\text{Cu}_3\text{O}_{6+x}$ is found to demonstrate a variety of unusual features, including a striking “ d -wave” shaped angular dependence, anomalous low-field MR and its saturation above a well-defined threshold field, and a hysteretic behavior at low temperatures. The overall features can be consistently explained by assuming a developed array of charged stripes and the field-induced topological ordering of the stripes. At temperatures below ~ 10 K, the external magnetic field of the order of a few T can produce a persistent directional ordering of the stripes, giving rise to a “memory effect” in the resistivity. These findings give a strong evidence that the charge inhomogeneities in the CuO_2 planes in the form of stripes exist and actually have a considerable impact on the electron transport. Also, our data show that the magnetic field can be used as a tool to manipulate the striped structure.

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