

Spin reorientation and in-plane magnetoresistance of lightly doped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ in magnetic fields up to 55 T

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The magnetoresistance (MR) in the in-plane resistivity is measured in magnetic fields up to 55 T in lightly doped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ in the Néel state ($x = 0.01$) and in the spin-glass state ($x = 0.03$) using high-quality untwinned single crystals. In both cases, a large negative MR is observed to appear when the magnetic order is established. For $x = 0.01$, it is found that the MR is indicative of a one-step transition into a high-field weak-ferromagnetic state at around 20 T when the magnetic field is applied from the spin easy axis (b axis), which means that there is no spin-flop transition in the Néel state of this material; this is contrary to a previous report, but is natural in light of the peculiar in-plane magnetic susceptibility anisotropy recently found in this system. In the spin-glass state, we observe that the large (up to $\sim 20\%$) negative MR saturates at around 40 T, and this MR is found to be essentially isotropic when the magnetic field is rotated within the ab plane. Our data show that the large negative MR is inherent to LSCO in a magnetically ordered state, in which the weak-ferromagnetic (WF) moment becomes well-defined; we discuss that the observed MR is essentially due to the reorientation of the WF moments towards the magnetic field direction both in the Néel state and in the spin-glass state.

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I. INTRODUCTION

In the lightly doped regime of the high- T_c cuprates, there is an intriguing dichotomy regarding the coupling between charge carriers and the background spins: On one hand, the long-range Néel order is quickly suppressed with doped holes, and only 2% of hole doping is sufficient to kill the Néel ordering,¹ indicating that the doped holes are strongly coupled to the spin subsystem; on the other hand, a metallic transport with a mobility comparable to that at optimum doping is established with only 1% of hole doping and this metallic transport is completely insensitive to the onset of the Néel ordering,² indicating that the doped holes and the spin subsystem are rather decoupled. Even more intriguingly, the dichotomy is found not only in the hole-doped cuprates but also in the electron-doped cuprates, where the Néel temperature is rather insensitive to the electron doping³ (suggestive of a spin-charge decoupling), and yet a subtle change in the spin arrangement upon the spin-flop transition in magnetic fields causes a large change in the resistivity⁴ (indicative of a strong spin-charge coupling). Therefore, such dichotomy appears to be a ubiquitous feature in the cuprates (though there is an electron-hole asymmetry in its nature) and it is of high importance to clarify the cause of this puzzle to understand the peculiar electronic states in the cuprates. As has been pointed out repeatedly in the literature,^{2,5-10} some form of electron self-organization and a resulting nanoscale phase separation is probably the key to solve this puzzle, but a comprehensive picture to understand the interplay between the charge carriers and the background spins, which is clearly

at the heart of the physics of the cuprates, remains to be developed.

The magnetoresistance (MR) in the lightly doped cuprates has been a useful probe of the peculiar coupling between the charge carriers and the spin subsystem. For example, Thio *et al.* discovered¹¹ a large change in the resistance of a lightly oxygen-doped $\text{La}_2\text{CuO}_{4+y}$ (LCO) sample at the weak-ferromagnetic (WF) transition that occurs at around 4 T when the magnetic field is applied along the c axis. They interpreted this phenomenon in terms of a sort of “spin-valve” effect, which controls the hopping probability depending on the ferromagnetic/antiferromagnetic arrangements of the WF moments between the neighboring CuO_2 planes and thereby changing the c -axis resistivity ρ_c .¹² Remember, the WF moment in LCO [or in antiferromagnetic $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (LSCO)] arises from a slight canting of the Cu spins out of the CuO_2 planes due to the antisymmetric exchange coming from the Dzyaloshinskii-Moriya (DM) interaction^{1,11,13}; when there is a static in-plane antiferromagnetic order, because of the spin canting, each CuO_2 plane develops a weak c -axis ferromagnetic moment, which orders antiferromagnetically along the c axis in the three-dimensional Néel state but can be aligned ferromagnetically upon WF transition when a sufficiently strong c -axis magnetic field is applied. The MR in lightly doped LSCO was recently revisited by Ando, Lavrov, and Komiya (ALK)¹⁴ and they found that not only the ρ_c but also the in-plane resistivity (ρ_{ab}) shows a large change upon the WF transition, which cannot be explained in terms of the spin-valve scenario. ALK therefore proposed an alternative model in which they asserted that

the in-plane charge transport occurs primarily through a network of antiferromagnetic domain boundaries (that can be viewed as nematic charge stripes) and that a change in the nature of the boundary from antiphase to in-phase (which necessarily happens upon the WF transition) should be the cause of the large MR in ρ_{ab} .¹⁴

Although the nature of the WF transition for $H \parallel c$ is by now well understood,¹⁴ the case for $H \perp c$ is rather controversial. Based on MR measurements of flux-grown LCO crystals for $H \parallel a, b$ up to 23 T, in 1990 Thio *et al.* asserted¹² that when the magnetic field is applied along the spin easy axis (b axis), the spin reorientation takes place in three steps: first, as the WF moment is drawn towards the magnetic-field direction (b axis), the staggered moment is gradually reoriented towards the c axis within the bc plane; second, an ordinary spin-flop transition takes place and the staggered moment flops into the ac plane, perpendicular to the applied field; third, as the WF moment is kept drawn towards the magnetic-field direction, the staggered moment is gradually reoriented towards the c axis within the ac plane. In the final high-field state, the staggered moment is essentially along the c axis and the WF moments are ferromagnetically aligned along the b axis (we call this state a high-field WF state). However, very recently, Gozar *et al.*¹⁵ measured the long-wavelength magnetic excitations in untwinned single crystals of undoped LCO and lightly doped LSCO using Raman spectroscopy and argued that there should be no intermediate “spin-flopped” state where the staggered moments are in the ac plane; in other words, the spin reorientation in this system takes place in just one step, namely, a gradual rotation of the staggered moment towards the c axis within the bc plane until it reaches the c axis to realize the high-field WF state. (It is useful to note that Gozar *et al.* found a new field-induced magnetically ordered state above the Néel temperature T_N for $H \parallel b$, which is essentially the same as the high-field WF state below T_N .¹⁵)

Therefore, it is desirable to conduct careful MR measurements on a state-of-the-art single crystals of lightly doped LSCO up to a high magnetic field and clarify whether the transition into the high-field WF state is achieved in one step or in multiple steps when the magnetic field is applied along the b axis. Also, since the magnetic-field induced spin reorientation in the spin-glass regime¹ of LSCO ($0.02 \leq x \leq 0.05$) has not been studied before, it would be useful to measure the high-field MR in the spin-glass regime as well: for example, recent Raman spectroscopy experiment by Gozar *et al.*¹⁵ mentioned above did not find any feature that can be associated with the field-induced magnetically ordered state for $x = 0.02$ or 0.03 , so it is intriguing to see what is observed in the MR in those samples.

In this work, we measure MR in the in-plane resistivity of lightly doped LSCO at $x = 0.01$ and 0.03 in pulsed magnetic fields up to 55 T. We find that at $x = 0.01$ there is indeed no intermediate spin-flop transition for $H \parallel b$ before the high-field WF state is achieved at the critical

field H_b of ~ 20 T. Intriguingly, while a large negative MR is observed below H_b , the MR above H_b (in the high-field WF state) is found to be positive; this seems to suggest that in cuprates an antiferromagnetic alignment of the background spins gives better charge conduction than a ferromagnetic (spin polarized) alignment, which is contrary to the common wisdom for metals. The MR data for $x = 0.03$ suggest that the charges are strongly coupled to the spin subsystem only at sufficiently low temperature where the spins freeze into a spin-glass state; in this case, a saturation of the negative MR occurs at ~ 40 T, which is roughly twice as much as that for $x = 0.01$. What is unusual for $x = 0.03$ is that both the saturation field and the magnitude of the MR appear to be almost isotropic with respect to the rotation of the magnetic field in the ab plane; we discuss that this unexpected isotropy can be understood to be a result of a short antiferromagnetic correlation length in the spin-glass state, and that the mechanism to produce large negative MR at $x = 0.01$ and 0.03 is essentially the same.

II. EXPERIMENTS

The high-quality LSCO single crystals are grown by the traveling-solvent floating-zone technique.¹⁶ After they are cut and polished into a rectangular platelet shape of $1.2 \times 0.6 \times 0.15$ mm³ suitable for the a -axis resistivity (ρ_a) measurements (the longest edge is parallel to the a axis within an error of less than 1°), the samples are carefully annealed at 500°C in flowing pure Ar to remove the excess oxygen, and then detwinned at 250°C with a uniaxial pressure.¹⁷ The samples are confirmed to be nearly 100% detwinned by the x-ray diffraction (XRD) analysis. The MR is measured in pulsed magnetic fields up to 55 T using a high-frequency (~ 100 kHz) lock-in technique with a four-probe method.^{18,19} The eddy current heating¹⁸ is confirmed to be not adversely affecting the data shown here; however, in the case of the $x = 0.01$ sample, the MR data below 20 K are found to be unreliable, because the resistivity of this sample diverges almost exponentially at low temperature (see Fig. 1), which causes the resistivity value to be extremely sensitive to even a slight change in temperature due to a minor eddy-current heating.

Figure 1 shows the temperature dependences of ρ_a measured on the samples used in the present study. Note the difference in the low-temperature behavior for the two samples: The $x = 0.01$ sample, which is in the Néel state below ~ 230 K, shows a steep resistivity divergence at low temperatures, while the $x = 0.03$ sample shows comparatively modest resistivity divergence.

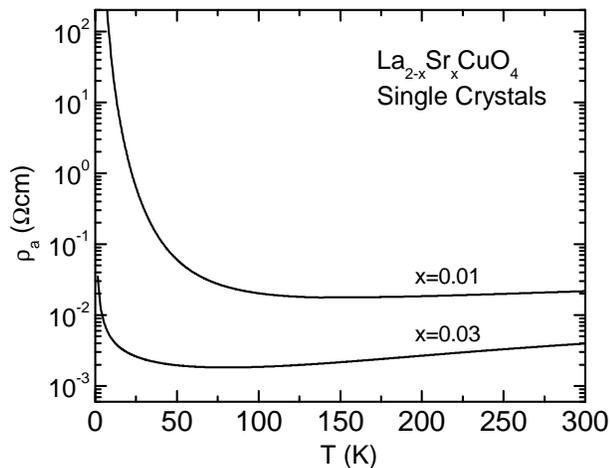


FIG. 1: Temperature dependences of ρ_a for $x = 0.01$ and 0.03 , measured on the untwinned samples used in this study.

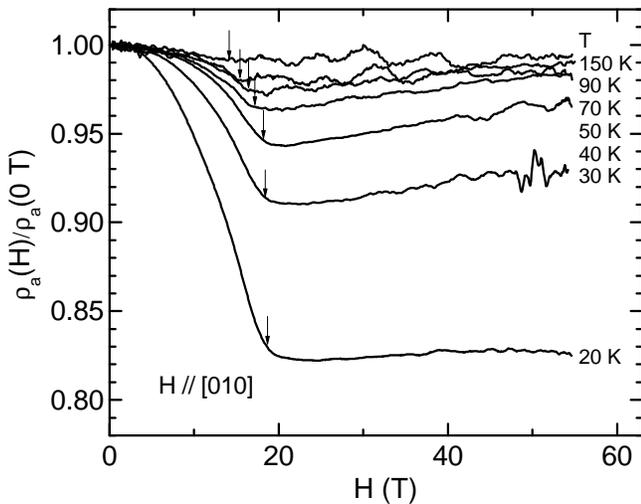


FIG. 2: Magnetoresistance in ρ_a with $H \parallel b$ for the $x = 0.01$ sample at various temperatures. Arrows mark the transition field into the high-field WF state, H_b .

III. RESULTS AND DISCUSSIONS

A. Néel State ($x = 0.01$)

The MR in ρ_a with $H \parallel b$ for $x = 0.01$ is shown in Fig. 2. Although the data are noisy at high temperatures, one can see that the low-field negative MR at 150 K tends to saturate at around 13 T, which is consistent with our previous data measured in dc magnetic field (see the 160 K data in Fig. 2 of Ref. 14), giving confidence in the present pulsed magnetic field measurements. It should be noted that, according to the previous study,¹⁴ only the b -axis component of the in-plane magnetic field is responsible for the low-field negative MR. As the temperature is lowered, the absolute value of the low-field negative MR grows [Fig. 3(a)] and the critical field moves

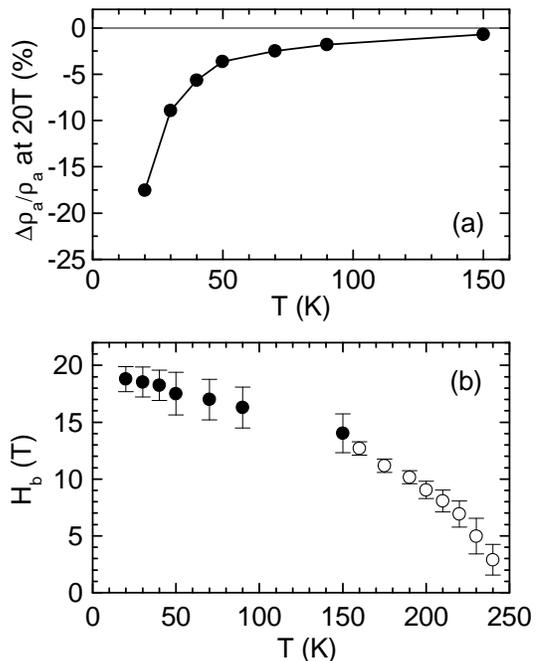


FIG. 3: (a) Temperature dependence of the magnetoresistance in ρ_a with $H \parallel b$ for the $x = 0.01$ at 20 T. (b) Temperature dependence of the transition field into the high-field WF state, H_b , extracted from the present pulsed magnetic field data (solid circles) and the previous dc magnetic field data¹⁴ (open circles).

to higher field [Fig. 3(b)], both of which are consistent with the previous reports.^{12,14} However, there are two features that are different from the old data of Thio *et al.*¹²: (i) the MR above H_b is positive, and (ii) there is no kink in the MR data below H_b . The latter indicates that there is no intermediate spin-flop transition proposed by Thio *et al.* The absence of the spin-flop transition for $H \parallel b$ is actually natural in light of the recent magnetic susceptibility data¹⁷ measured on an untwinned single crystal of LSCO at $x = 0.01$, which showed that χ_a is always smaller than χ_b at temperatures below 300 K, even though the b axis is the spin easy axis; remember, in ordinary antiferromagnets in the Néel state, the perpendicular susceptibility χ_{\perp} is larger than the longitudinal susceptibility χ_{\parallel} , which provides the source of the energy gain in the spin-flop transition. Therefore, the unusual anisotropy in the magnetic susceptibility (which is probably due to the peculiar role of the DM interaction¹⁷) makes the occurrence of the ordinary spin-flop transition unlikely in this system.

Another point to be elaborated on is the positive MR observed above H_b in Fig. 2. This MR is not likely to be the ordinary positive (orbital) MR of a metal, because (i) the system is in the strongly localized (hopping conduction) regime where the electron motion is not coherent²⁰ and (ii) the magnetic-field (H) dependence of the MR (particularly at 30 – 90 K) is not $\sim H^2$ but is rather linear in H . Thus, the MR above H_b is

most likely caused by a change in the spin subsystem. If so, the positive MR is rather unexpected, because above H_b the main moment is gradually polarized at the expense of the staggered component of the moment [all the spins are expected to be ferromagnetically aligned (completely polarized) when the Zeeman energy exceeds the exchange energy (order of 100 meV in cuprates) and the thermal energy]. Remember, it is a common sense that the ferromagnetic arrangement of the spins promotes the charge motion while the antiferromagnetic arrangement tends to localize the charges; therefore, according to the common wisdom, the positive MR above H_b in LSCO is unusual in that the charge mobility tends to become worse when the system is approaching a spin-polarized state. However, the argument of whether the ferromagnetic/antiferromagnetic arrangement promotes/hinders the charge motion is based on a picture that the charges are essentially uniformly distributed and the physics is determined by the motion of a single charge in the magnetic background. If, on the other hand, the charges are phase segregated from the magnetic subsystem and form a self-organized network of “rivers of charges”, the above consideration does not apply. In fact, in our series of works on the lightly doped cuprates, we have shown that various peculiar properties in the lightly doped regime can be best understood in terms of the electron self-organization picture.^{2,10,14,17,21,22} Therefore, the positive MR observed above H_b is another indirect support to the notion that charges and spins are spatially decoupled in lightly doped LSCO, but the exact reason why the MR is positive in the high-field WF state (where the “rivers of charges” must constitute in-phase magnetic domain boundaries¹⁴) needs to be clarified by future studies.

B. Magnetic Shape Memory Effect

We note that one should always be careful about the “magnetic shape memory effect”²³ in lightly doped LSCO crystals when making measurements under high magnetic fields. This effect causes a swapping of the orthorhombic a and b axes in a *fixed* sample, where the b axis tends to become parallel to the applied in-plane magnetic field. Therefore, no axes swapping takes place in untwinned crystals when the magnetic field is within $\pm 45^\circ$ of the b axis direction. However, when a high magnetic field (usually in excess of 10 T) is applied within $\pm 45^\circ$ of the a axis, new twin boundaries are created and initially untwinned crystals become twinned. In our previous work using dc magnetic fields up to 14 T,²³ we observed that this phenomenon takes place for $x = 0.01$ only near room temperature where the thermal fluctuations help the axes swapping, but in the present 55 T experiment we found that this phenomenon can happen even at low temperatures when the applied magnetic field is sufficiently strong. In fact, after the $H \parallel b$ measurements are finished, we tried to measure the MR for $H \parallel a$ in our $x = 0.01$ sample at low temperatures (below 70 K),

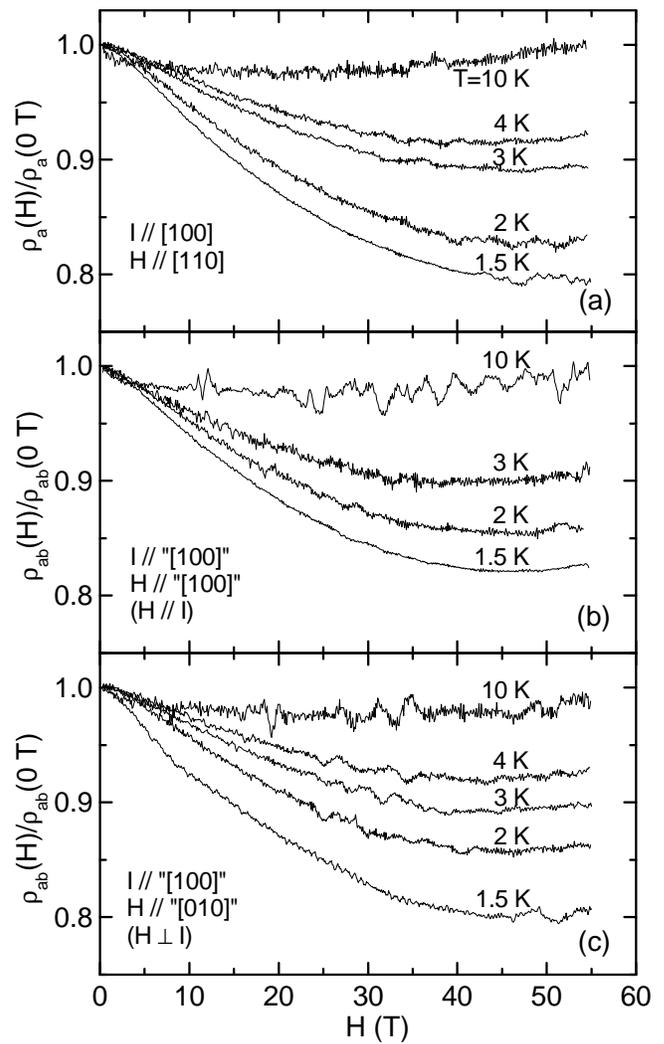


FIG. 4: Magnetoresistance of the $x = 0.03$ sample for three different configurations of the in-plane magnetic field.

but the sample was re-twinned during this experiment, making the data to be not very meaningful (this is why only the data for $H \parallel b$ are shown here).

C. Spin-Glass State ($x = 0.03$)

Figure 4 shows the MR for $x = 0.03$, where the magnetic field is applied from three different in-plane directions, $H \parallel [110]$ (diagonal to the orthorhombic axes and thus is parallel to the Cu-O-Cu bond direction), $H \parallel [100]$ (orthorhombic a axis), and $H \parallel [010]$ (orthorhombic b axis); we note that the measurements are done in this order. This $x = 0.03$ sample was initially 100% detwinned and was prepared so that the current flows along the a axis ($I \parallel [100]$), and we did not expect the magnetic shape memory effect to occur at this composition; however, to our surprise, we found that the sample was retwinned after the measurements, with about 45% of

misoriented domains. Since the axes swapping never occurs for $H \parallel [110]$ (first set of the data), it must have occurred during the measurement for $H \parallel [100]$ (second set of the data). Therefore, we should consider that the data for $H \parallel [100]$ and $H \parallel [010]$ [Figs. 4(b) and 4(c)] are actually on a twinned sample; this is why the current directions and the magnetic field directions are denoted using double-quotation marks in Figs. 4(b) and 4(c). Nevertheless, the data shown in Fig. 4 are useful, because they show that the MR is essentially isotropic at $x = 0.03$: If the MR were intrinsically anisotropic and the nature of the anisotropy were similar to that for $x = 0.01$ (*i.e.*, only the b -axis component of the in-plane magnetic field is effective in causing the negative MR), the characteristic magnetic field scale for the MR would be $\sqrt{2}$ times larger for $H \parallel [110]$ compared to that for $H \parallel [010]$ even in a twinned sample. In Figs. 4(a)-4(c), the minimum in MR at each temperature occurs at essentially the same magnetic field for the three configurations, indicating that both the a - and b -axis components of the magnetic field are equally effective in causing the MR. Note that not only the characteristic magnetic field scale but also the magnitude of the MR is essentially isotropic, growing up to $\sim 20\%$ at 1.5 K for all three configurations.

It is useful to note that the MR for $x = 0.03$ shown in Fig. 4 remains small down to 10 K, but suddenly grows at lower temperatures. Thus, the growth of the MR is clearly related to the spin-glass order, which is established below 6–10 K for $x = 0.03$.^{17,24} Intriguingly, in the magnetic susceptibility measurement of an untwinned single crystal with $x = 0.03$ by Lavrov *et al.*,¹⁷ it was found that the spin-glass ordering temperature T_g is anisotropic, meaning that the spin-glass order parameter vanishes upon heating at different temperatures depending on the direction of the applied field to measure the magnetic susceptibility. Obviously, this anisotropy in T_g is related to the anisotropy in the magnetic susceptibility itself that is indicative of the staggered moment to be confined in the bc plane, as is the case with $x = 0.01$; this suggests that the direction of the spins are not random in the “spin-glass” phase of LSCO, a departure from the conventional picture of the spin glass or the cluster spin glass. Since the local spin structure as suggested by the magnetic susceptibility anisotropy at $x = 0.03$ appears to be similar to that at $x = 0.01$, it is natural to expect that the negative MR observed for $x = 0.03$ is caused by essentially the same mechanism as that for $x = 0.01$, namely, the reorientation of the WF moments towards the magnetic-field direction. The fact that the characteristic magnetic field for the saturation of the MR for $x = 0.03$ (~ 40 T) is larger than that for $x = 0.01$ (~ 20 T) is also consistent with this interpretation, because in the spin-glass phase the spin correlation length is rather short (4–10 nm depending on the in-plane direction)²⁵ and thus the magnitude of the WF moment (which is given by an integration of the canted moments over the antiferromagnetically correlated area in the CuO_2 planes) becomes accordingly small.

One might think that the isotropic nature of the MR for $x = 0.03$ evident in Fig. 4 is not very consistent with the interpretation that the WF moment (that is confined in the bc plane) is responsible for the MR. However, one should keep in mind that the antiferromagnetically correlated region in the spin-glass regime is rather small, which makes it easier for the applied magnetic field, with the help of the thermal energy, to overcome the spin-anisotropy energy and to rotate the moments associated with antiferromagnetically correlated regions out of the bc plane. To make a crude estimate, the correlation area of $4 \times 10 \text{ nm}^2$ (Ref. 25) contains roughly 300 Cu ions, for which the integrated anisotropy energy for the WF moments, $\sum z J_{bc} S M_F / g \mu_B$ (where $z = 4$ is the number of nearest neighbors, $J_{bc} \simeq 0.7 \text{ meV}$ is the in-plane anisotropic exchange, $S = 1/2$ is the Cu spin, and $M_F \simeq 0.002 \mu_B$ is the WF moment per Cu),¹² is only about 0.4 meV — this energy is actually smaller than the Zeeman energy for the integrated WF moment in 40 T ($> 1 \text{ meV}$) and is comparable to the thermal energy at the spin-glass ordering temperature (6–10 K), giving confidence in this estimate. Therefore, it is likely that the spins are “deconfined” from the bc plane in high magnetic fields before the high-field WF state is established, and this can explain the observed isotropy in the MR that is expected to be associated with the reorientation of the WF moment.

Whatever the cause of the isotropy in the MR for $x = 0.03$, it is intriguing that the magnitude of the MR can become as large as 20% at low temperature, indicating that a large MR is a common feature of insulating LSCO that shows some kind of magnetic order, with which the WF moment can become well-defined. Thus, it is most reasonable to conclude that the large MR in the insulating LSCO is essentially associated with the reorientation of the WF moment and the resulting change in the antiferromagnetic domain boundaries, into which the doped-holes are presumably segregated.¹⁴

IV. CONCLUSIONS

We measure the magnetoresistance (MR) in the in-plane resistivity of high-quality LSCO single crystals with $x = 0.01$ and 0.03 in pulsed magnetic fields up to 55 T. Contrary to the previous report by Thio *et al.* in 1990,¹² we observe that there is no spin-flop transition in the Néel state when the magnetic field is applied from the spin easy axis (b axis), and the high-field weak-ferromagnetic (WF) state (where the WF moments are polarized along the b axis) is achieved in just one step. For $x = 0.03$, a large (up to $\sim 20\%$) negative MR is observed in the spin-glass state, where the saturation of this MR occurs at around 40 T. Intriguingly, the large MR in the spin-glass state is essentially isotropic, which is probably related to the short antiferromagnetic correlation length in the spin-glass state that allows a deconfinement of spins out of the bc planes in high-magnetic

fields.

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