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Description	

Thermomagnetic hysteretic properties resembling superconductivity in the normal state of $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$

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We report detailed magnetic and thermal hysteresis experiments in the normal-state magnetization of a $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ single crystal. Using a combination of in-field and in-zero-magnetic-field measurements at different stages of thermal history of the sample, we identified subtle effects associated with the presence of magnetic signatures which resemble those below the superconducting transition temperature ($T_c=36$ K) but survive up to 250 K.

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I. INTRODUCTION

The magnetization of type-II anisotropic superconductors displays thermal hysteresis below the irreversibility temperature due to the pinning of superconducting vortices.¹ Contrary to conventional wisdom, recent systematic measurements on $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ single and polycrystalline samples revealed the presence of hysteresis in the temperature dependence of the low-field magnetization up to a doping-dependent characteristic temperature T_s reaching a maximum of 290 K for $x=0.10$.² The temperature, magnetic field, and crystallographic dependences of the onset and strength of the hysteresis were found to resemble fundamental properties of the mixed state. The magnetization results and correspondence with magnetothermal transport experiments^{3,4} opened the exciting possibility of either the presence of superconducting signatures or some form of magnetism encouraging superconductivity above the superconducting transition temperature T_c . However, the nature of the currents responsible for the thermal hysteresis remains to be identified. To this aim we developed an experimental method which can be applied to study subtle magnetic effects in superconducting and magnetic materials in general. As we describe below this method has also the capability of distinguishing the intrinsic thermomagnetic hysteresis from possible contribution arising from extrinsic magnetic impurities. We have studied $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ single crystals and the sample discussed here had a mass of 1.1 mg with onset $T_c=36$ K (transition width of 1.5 K). The samples were prepared and characterized as in Ref. 5. Our results indicate that the onset of the thermal hysteresis at T_s is due to a transition from a fluctuating (at $T>T_s$) to a pinned ordered state ($T\leq T_s$), whose magnetic moment may be associated with superconductivity related long-lived persistent currents.

The paper is organized as follows. In Sec. II we briefly describe the experimental technique, in Sec. III we present the results on the thermal hysteresis effects observed by setting the applied magnetic field below and above T_c (Secs. III A and III B, respectively). In Sec. III C we show that T_s is a transition temperature and in Sec. III D we report experi-

ments showing the presence of magnetic flux conservation when crossing T_c (with both decreasing and increasing temperature) in zero applied magnetic field. Section IV summarizes the present work.

II. EXPERIMENT

Measurements were carried out over a course of 12 months using a Quantum Design (MPMS-XL) superconducting quantum interference device (SQUID) magnetometer at 3 cm scan length after we first applied a zero-magnetic-field procedure to suppress the remnant field of its superconducting magnet down to $\leq \pm 1.5$ G. No extra material was added to hold the sample, and a sample holder with no discontinuity was used so there was no background due to the sample holder—at least to the resolution of the XL-SQUID magnetometer. (The sample was held mechanically by two plastic straws placed inside an almost equal length outer straw supplied by Quantum Design.) It is the discontinuity which is responsible for the SQUID signal and, therefore, in the experimental configuration with no discontinuity in the sample holder the measured signal reflects purely the sample's magnetization. The thermomagnetic hystereses have been checked separately in Cambridge and Tohoku. The results reported here were confirmed several times over the 12-month course of the present experiments and with different scan lengths (4 and 6 cm) and heating and cooling times (0.5, 1, 2, and 4 K/min). Note that the results shown here have been obtained by stabilizing the sample temperature at every temperature the data was collected. We have been particularly careful to eliminate the possibility of thermal lags and external noise such as that coming from the mains. The raw data and SQUID response function were monitored for each data point.

As shown in Ref. 2 the thermal hysteresis is stronger when the applied magnetic field $H\parallel ab$ than when $H\parallel c$. It is also for $H\parallel c$ that the magnetic field dependence of the thermal hysteresis is weaker. Also the onset temperature of the hysteresis, T_s , decreases with increasing field, is smaller for $H\parallel c$, and becomes negligibly small at $H>1$ kG. For these

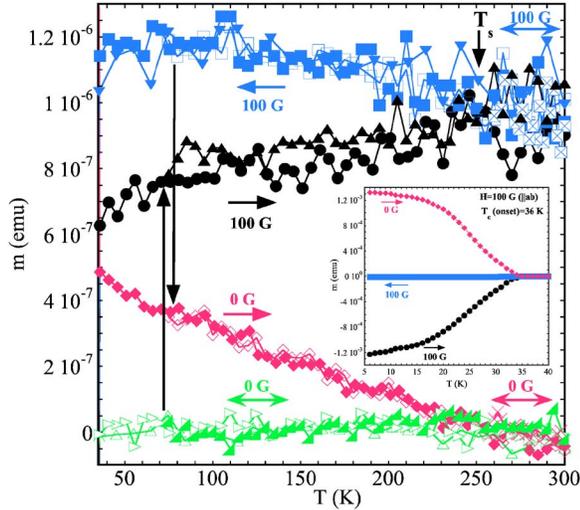


FIG. 1. (Color online) Magnetic moment m versus temperature T for a $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ single crystal in applied magnetic field $H \parallel ab = 100$ G. Experimental run [sequence (1)]: (●) Field applied at 6 K and sample warmed to 300 K with the field on. (■) Sample cooled from 300 K to 6 K in the presence of applied field. The main panel and inset depict the high-temperature ($T > T_c$) and low-temperature regions, respectively. (◆) Field decreased to zero at 6 K and sample warmed in zero field up to 300 K. (△) Sample cooled in zero field from 300 K to 6 K. Experimental run [sequence (2)]: (□) Field applied at 300 K and sample cooled in field to 75 K. (◇) Field decreased to zero at 75 K (along the big arrow pointing downward) and sample warmed in zero field up to 300 K. (▲) Sample cooled in zero field from 300 K to 75 K. (▲) Field applied at 75 K (along the large arrow pointing upward) and sample warmed in field up to 300 K. (▼) Sample cooled in field from 300 K to 6 K. Experimental run [sequence (3)]: (⊠) Field applied at 300 K and sample cooled in field to 260 K. (×) Field decreased to zero at 260 K and sample warmed in zero field (ZFW) up to 300 K. (▷) Sample cooled in zero field from 300 K to 6 K.

reasons in the present paper we performed all the experiments with $H \parallel ab = 100$ G.

III. RESULTS AND DISCUSSION

A. Hysteresis effects by setting the applied magnetic field on at $T < T_c$

The experimental procedure employed is as follows. First, we cooled the sample from 300 K to 6 K in zero-field. Once the sample was at 6 K, the applied magnetic field was set to 100 G and the magnetic moment m was measured first with increasing temperature up to 300 K [field warming (FW)] (Fig. 1, black solid circles) and then with decreasing temperature back to 6 K [field cooling (FC)] (Fig. 1, blue solid squares). While at 6 K the magnetic field was set to zero and the sample was warmed to 300 K [zero field warming (ZFW)] (Fig. 1, red solid diamonds). The sample was then cooled back to 6 K in zero field [zero field cooling (ZFC)] (Fig. 1, main panel, green open triangles). The procedure is schematically shown in sequence (1):

$$\begin{aligned}
 &300 - 6 \text{ K in } 0 \text{ G (ZFC), } 6 - 300 \text{ K in } 100 \text{ G (FW),} \\
 &300 - 6 \text{ K in } 100 \text{ G (FC), } 6 - 300 \text{ K in } 0 \text{ G (ZFW),} \quad (1) \\
 &300 - 6 \text{ K in } 0 \text{ G (ZFC).}
 \end{aligned}$$

Cooling the sample from above T_c but in the presence of 100 G, is known to result in homogeneously distributed trapped vortices at $T < T_c$ (Fig. 1, inset, blue solid squares).¹ By setting the applied field to zero at 6 K, we observed the effect of induced currents which keep the field (vortices) trapped inside the sample and give rise to a paramagnetic moment below T_c (Fig. 1, inset, red solid diamonds). As expected, these currents decrease with temperature. However, instead of vanishing at T_c the moment survives at $T > T_c$ (Fig. 1, main panel, red solid diamonds). The presence of the positive moment (established at $T < T_c$ in order to keep the vortices trapped) up to T_s is suggestive for the presence of trapped vortices above T_c .² Next we cooled the sample from 300 K to 6 K in zero field, and the data followed the “zero line” (Fig. 1, main panel, green upper open triangles), indicating the absence of any magnetization in the sample, as expected in an equilibrium paramagnetic state. We note that the existence of a straight “zero line” (represented by the green upper open triangles in Fig. 1, main panel), as well as other extensive tests we performed on our samples,⁶ indicates that the above hysteretic effect cannot be due to traces of possibly undetected magnetic impurities.

The presence of a trapped field at $T < T_s$ implies the presence of a thermal hysteresis. Indeed, this leads to the second observation of our experiments—i.e., the thermal hysteresis at $T > T_c$. The sensitivity of the experiment allows us to see that the FW curve saturates at $T > T_s \approx 250$ K, at which point it also starts to be significantly noisier (Fig. 1, main panel, black solid circles). Notably, the FC curve, from 300 K to T_s (Fig. 1, main panel, blue solid squares), remained noisy too but basically reversible, and with decreasing temperature at $T < T_s$, m increased slightly and was always above the equilibrium background ($\approx 10^{-6}$ emu). This indicates that T_s is not just a crossover temperature. Note that along the FW curve (black solid circles) at $T_c < T < T_s$ the moments are lower than the equilibrium background (defined at $T_s < T \leq 300$ K) indicating that persistent diamagnetic currents may have survived as the sample crossed T_c .

B. Hysteresis effects by setting the applied magnetic field on at $T > T_c$

To investigate whether setting the applied magnetic field at $T < T_c$ is essential for observing a hysteresis above T_c , we performed the following experiments.

(i) We first warmed the sample to 300 K in zero field, then applied a magnetic field of 100 G at 300 K, and finally cooled the sample again but only down to 75 K. The data from the sequence (Fig. 1, main panel, blue open squares) traced the curve obtained previously for the original FC run (Fig. 1, main panel, blue solid squares).

(ii) While the sample was at 75 K, we decreased the applied magnetic field to zero (as indicated by the big arrow

pointing down in the main panel of Fig. 1) and subsequently warmed the sample in zero field to 300 K. The data from this sequence (Fig. 1, main panel, red open diamonds) traced the curve obtained for the ZFW run (Fig. 1, main panel, red solid diamonds).

(iii) Next we cooled the sample from 300 K, in zero field, along the zero line (Fig. 1, main panel, green solid right angle triangles), increased the applied magnetic field to 100 G when the sample reached 75 K (along the big arrow pointing up in the main panel of Fig. 1), and warmed the sample in field up to 300 K. In this sequence the data (Fig. 1, main panel, black solid triangles) traced the FW curve obtained previously by increasing the field while the sample was at $T < T_c$ (Fig. 1, black solid circles). The sample was then cooled in field down to 6 K (Fig. 1, main panel, blue solid triangles). The process (i)–(iii) is schematically shown in sequence (2):

$$\begin{aligned} &6 - 300 \text{ K in } 0 \text{ G (ZFW)}, \quad 300 - 75 \text{ K in } 100 \text{ G (FC)}, \\ &75 - 300 \text{ K in } 0 \text{ G (ZFW)}, \quad 300 - 75 \text{ K in } 0 \text{ G (ZFC)}, \quad (2) \\ &75 - 300 \text{ K in } 100 \text{ G (FW)}, \quad 300 - 6 \text{ K in } 100 \text{ G (FC)}. \end{aligned}$$

(iv) Moreover, increasing the applied field to 100 G, or decreasing it to zero and cooling or warming the sample in field or in zero field in the region $260 \text{ K} < T \leq 300 \text{ K}$ resulted in a zero line (for 0 G) (Fig. 1, main panel, red crosses) and in noisy data for 100 G—on a paramagnetic background of $\approx 10^{-6}$ emu (Fig. 1, main panel, blue crossed squares). This experimental run is described in sequence (3):

$$\begin{aligned} &300 - 260 \text{ K in } 100 \text{ G (FC)}, \quad 260 - 300 \text{ K in } 0 \text{ G (ZFW)}, \\ &300 - 6 \text{ K in } 0 \text{ G (ZFC)}. \quad (3) \end{aligned}$$

Furthermore, the data obtained at 100 G in this temperature region (Fig. 1, main panel, blue open and solid squares) were always noisier than the data obtained in the temperature region $T < T_s$.

These (i)–(iv) tests show that the same results are obtained at $T > T_c$ as long as the field is increased or decreased below T_s . They also show a degree of reproducibility of the data between individual experimental runs. Although there is significant noise, the reproducibility of the data is within an error of about $\pm 10\%$ (see Fig. 1, main panel, blue solid and open rectangles and blue solid triangles).

The main panel in Fig. 1 also indicates that the in-field and in-zero-field data have different backgrounds on which the hysteresis is superimposed: one for $H=0$ (Fig. 1, green triangles) and one for $H=100$ G (Fig. 1, blue crossed squares). In order to compare the levels of the hystereses we corrected the in-field data on their paramagnetic background at $T_s < T \leq 300$ K (10^{-6} emu) and replotted them in Fig. 2. The result we obtain is a nearly symmetric picture at $T_c < T < T_s$ (Fig. 2, black solid circles and red solid diamonds). Interestingly, this picture resembles the behavior observed at $T < T_c$ (Fig. 1, inset), indicating a common origin in the moments giving rise to the thermal hysteresis below and above T_c . Furthermore, the in-field data are noisier than the zero-field data, and the noise is higher at $T_s < T \leq 300$ K. Decreasing

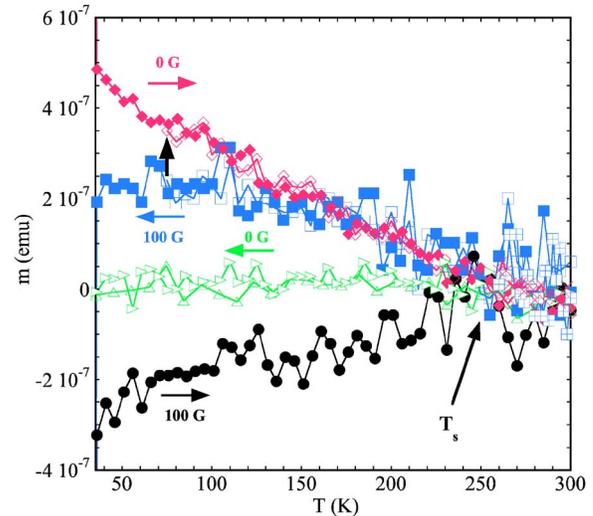


FIG. 2. (Color online) Magnetic moment m versus temperature T for a $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ single crystal in applied magnetic field $H \parallel ab = 100$ G. Data from field warming and field cooling runs are corrected on their equilibrium background (the value of the background is 10^{-6} emu and is defined as the reversible part which exists between 250 K and 300 K—see Fig. 1). The meanings of the symbols are the same as in Fig. 1.

ing the field to zero from the FC data at 75 K (represented by a large arrow pointing down in Fig. 1, main panel) in Fig. 2 we observe an increase in m along the short arrow pointing up (blue open squares \rightarrow red open diamonds), suggesting the presence of persistent paramagnetic currents induced by the change in the field (similarly as in Fig. 1, inset, red solid diamonds).

C. Tests showing that T_s is a transition temperature

The presence of a thermal hysteresis when H is applied at $T_c < T < T_s$ (Fig. 1, main panel, black and blue solid triangles) and the increased noise above T_s (Fig. 1, main panel, blue solid, open, and crossed rectangles and blue solid triangles) indicates that the latter is not a mere crossover. To examine whether an actual transition occurs at T_s , we studied the behavior of the hysteresis when we return from $T_c < T < T_s$. Figure 3 depicts properties of such partial magnetic and thermal hysteresis loops:

(i) We warmed the sample to 150 K after applying a magnetic field of 100 G at 6 K. The sample was then FC down to 75 K (Fig. 3, blue upper solid triangles). By decreasing the applied field to zero when the sample was at 75 K, the measured moment m dropped along the big arrow—starting from the blue upper solid triangles and pointing down to red lower solid triangles in Fig. 3.

(ii) The sample was then ZFW to 300 K, with m ending on the curve which is below that obtained by the ZFW process—starting either at $T < T_c$ ($T = 6$ K) (red solid diamonds) or at 75 K (Fig. 1, main panel, red open diamonds). This experimental run is represented in sequence (4):

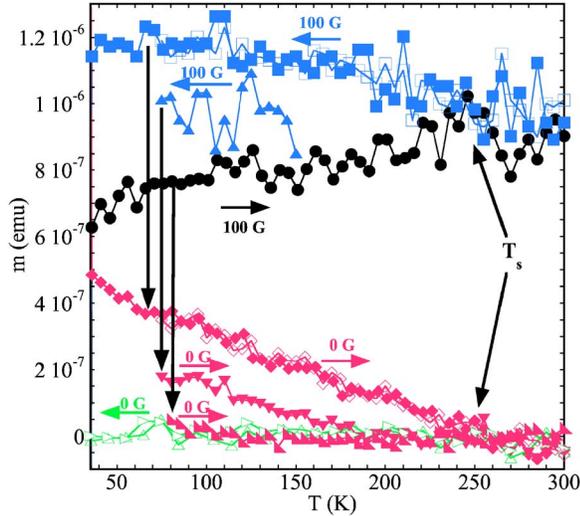


FIG. 3. (Color online) Magnetic moment m versus temperature T for a $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ single crystal in applied magnetic field $H \parallel ab = 100$ G. \bullet , \blacksquare , \blacklozenge , \blacktriangle , \square , \triangleright , \diamond Their meanings are the same as in Fig. 1. Experimental run [sequence (4)]: (\blacktriangle) Field applied at 6 K, sample warmed in field up to 150 K (these points are not shown in the figure) and then cooled in field to 75 K. (\blacktriangledown) Field decreased to zero at 75 K (along the large arrow starting from upper solid triangles and pointing down to lower solid triangles) and sample warmed in zero field to 300 K. Experimental run [sequence (5)]: (red \blacktriangle) Field applied at 6 K, sample warmed in field up to 75 K (along the solid circles), then field set to zero (along the large arrow pointing from solid circles down to solid right-angle triangles), and sample zero field warmed up to 300 K.

$$\begin{aligned} &300 - 6 \text{ K in } 0 \text{ G (ZFC)}, \quad 6 - 150 \text{ K in } 100 \text{ G (FW)}, \\ &150 - 75 \text{ K in } 100 \text{ G (FC)}, \quad 75 - 300 \text{ K in } 0 \text{ G (ZFW)}. \end{aligned} \quad (4)$$

Moreover, in Fig. 3 we show that by lowering H to zero at 75 K on the FW curve (black solid circles), m dropped along the big arrow pointing from black solid circles down to red solid right-angle triangles, and it is slightly above the zero line. [This run is described below as sequence (5).] It also has weak, but distinct, temperature dependence. Therefore, it is only when we decrease the applied field to zero while the sample is at $T > T_s$ that the true zero moment is observed:

$$\begin{aligned} &300 - 6 \text{ K in } 0 \text{ G (ZFC)}, \quad 6 - 75 \text{ K in } 100 \text{ G (FW)}, \\ &75 - 300 \text{ K in } 0 \text{ G (ZFW)}. \end{aligned} \quad (5)$$

Hence, T_s represents a transition from a fluctuating to a pinned ordered state in which long-lived weak persistent currents may exist—depending of course on the magnetic history of the sample. Let us note that the drop of the magnetic moment at 75 K to nearly zero as the field was decreased (Fig. 3, large arrow pointing from black solid circles down to red solid right-angle triangles) differs from observations in conventional spin glasses, where the moments decay very slowly.⁷

D. “Flooding” experiments

The question arising next is how to collectively understand these and earlier² observations. Although there is no

theoretical interpretation of our results, given the absence of bulk superconductivity above T_c and based on the striking resemblance of the hystereses at $T < T_c$ and $T_c < T < T_s$, one suggestion may be the presence of superconducting “vortices” at $T > T_c$.² In fact signatures for their possible presence in the normal state have been observed in magnetothermal transport and local magnetic imaging experiments in LSCO.^{3,4,8} As discussed previously² such a presence would explain many of our observations, in particular the similarities in the hystereses below and above T_c and the increased moment in Fig. 2 (short arrow pointing up). Overall, the proposed picture here would be similar to the critical state model below T_c . Alternatively, the data may be governed by a mechanism incorporating magnetic domains—for example, in the form of droplets or rivers.⁹

Irrespective of the nature of the domain structure (vortices or not) causing the thermal hysteresis, the fundamental question which needs to be experimentally addressed is whether this form of magnetism cooperates or competes with superconductivity. Already, our earlier work has shown that $T_s(x) \sim T_c(x)$, suggesting a cooperative relation (x is the carrier concentration level).²

To address this question in some more detail here we “flooded by superfluid” this “magnetism.” We first FW the sample from 6 K to 300 K in 100 G and then FC to 6 K. While the sample was at 6 K, we decreased the applied magnetic field to zero and ZFW the sample to 75 K (Fig. 4, red solid diamonds). From that point, but still in zero field, we cooled the sample to 6 K (Fig. 4, red crosses) and warmed it again to 300 K (Fig. 4, red crossed squares). This experimental run is schematically shown in sequence (6):

$$\begin{aligned} &300 - 6 \text{ K in } 0 \text{ G (ZFC)}, \quad 6 - 300 \text{ K in } 100 \text{ G (FW)}, \\ &300 - 6 \text{ K in } 100 \text{ G (FC)}, \quad 6 - 75 \text{ K in } 0 \text{ G (ZFW)}, \quad (6) \\ &75 - 6 \text{ K in } 0 \text{ G (ZFC)}, \quad 6 - 300 \text{ K in } 0 \text{ G (ZFW)}. \end{aligned}$$

The data obtained by cooling and warming the sample in the range of 75-6-75 K was found to be (a) reversible, (b) the magnetic moment did not depend on temperature, and (c) the magnetic moment crossed smoothly from the normal to the superconducting state and vice versa. A similar behavior was observed also in another experimental run [sequence (7)] when the sample was ZFW to 150 K and then cooled to 6 K and warmed to 300 K (Fig. 4, grey open squares and grey solid triangles):

$$\begin{aligned} &300 - 6 \text{ K in } 0 \text{ G (ZFC)}, \quad 6 - 300 \text{ K in } 100 \text{ G (FW)}, \\ &300 - 6 \text{ K in } 100 \text{ G (FC)}, \quad 6 - 150 \text{ K in } 0 \text{ G (ZFW)}, \quad (7) \\ &150 - 6 \text{ K in } 0 \text{ G (ZFC)}, \quad 6 - 300 \text{ K in } 0 \text{ G (ZFW)}. \end{aligned}$$

This reversibility shows clearly a return-point-memory effect, indicating the crossing through T_c evolves smoothly

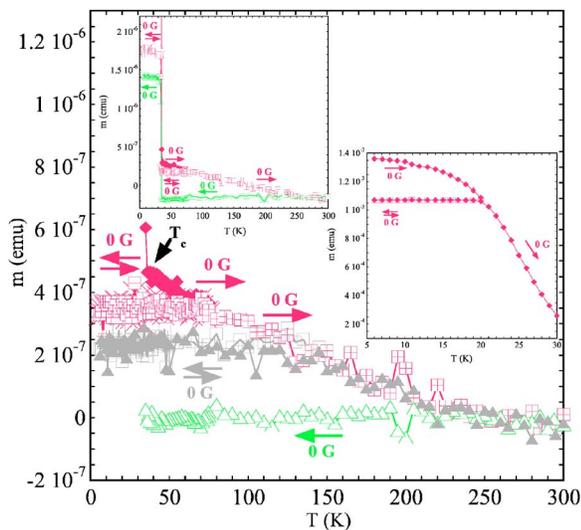


FIG. 4. (Color online) Magnetic moment m versus temperature T for a $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ single crystal in applied magnetic field $H_{\parallel ab}=100$ G. (\blacklozenge) (\blacktriangle) Their meanings are the same as in Fig. 1. To arrive to the crosses (\times) the following procedure was performed [sequence (6)]: A 100-G field was applied at 6 K; then the sample was warmed in field up to 300 K and consequently cooled in field to 6 K. The field was then decreased to zero at 6 K and the sample warmed in zero field up to 75 K. Then the sample was cooled down to 6 K in zero field. The crosses shown in the figure capture the latter part of the procedure. The data shown as crossed squares (\boxtimes) capture the part where the sample was warmed again from 6 K and in zero field up to 300 K. To arrive to the open squares (\square) (main panel) the following procedure was performed [sequence (7)]: A 100-G field was applied at 6 K; then the sample was warmed in field up to 300 K and consequently cooled in field to 6 K. The field was then decreased to zero at 6 K and the sample warmed in zero field up to 150 K. Then the sample was cooled down to 6 K in zero field. The open squares shown in the figure capture the latter part of the procedure. The data shown as solid triangles (\blacktriangle) capture the part when the sample was warmed again from 6 K but in zero field up to 300 K. In the upper inset all the symbols have the same meaning as in the main figure, but the data are not corrected for the background. In the lower inset (\blacklozenge) captures the part described in Fig. 1 for the same symbols but at $T < T_c$, whereas (\square) and (\blacklozenge) at $6 \text{ K} \leq T \leq 12 \text{ K}$ represent a thermal cycle [sequence (8)] performed to test the memory of the moment below T_c .

from one state ($T < T_c$) to another ($T > T_c$), and there a common origin for the magnetic moment responsible for the hystereses above and below T_c . This behavior indicates a magnetic flux conservation effect (Faraday effect) rather than a Meissner effect. To see it more clearly, we show the raw data (Fig. 4, upper inset) of the sample ZFW to 75 K, then cooled to 6 K, and finally warmed to 300 K [sequence (6)]. The magnetic moment corresponding to the zero line (green open triangles) is negative at $T > T_c$, and there is a jump to positive values at T_c as the temperature drops below T_c . This effect is similar to the decrease of the applied magnetic field to zero when the sample is in the superconducting state (e.g., at $T=6$ K)—after it was FC from 300 K in 100 G (Fig. 1, inset, blue solid rectangles, red solid diamonds).

We note that the nonzero magnetic moment of the zero

line at $T > T_c$ is a consequence of a trapped magnetic field in the superconducting magnet of the SQUID magnetometer used to perform the measurements. Because the magnetic moment $m = [\mu_0^{-1} V / (1 - N)] (B - \mu_0 H_a)$ (in SI units) (V , sample volume; $0 < N < 1$, demagnetizing factor; B , magnetic flux density; H_a , applied magnetic field) is negative, it means that $B < \mu_0 H_a$. A jump in the magnetic moment to positive values at $T < T_c$ can in principle be caused by the Meissner effect, if the trapped magnetic field of the superconducting magnet H_a is negative. When the field $B=0$, $m = [\mu_0^{-1} V / (1 - N)] (-\mu_0 H_a) > 0$. Now, because the trapped field H_a of the magnet does not change, cooling the sample from 75 K, when B in the sample is higher than that on the zero line, should give at least the same positive magnetic moment as for the zero line, or lower (because of a possible partial screening), but not higher as observed in the present experiment (red crosses and red crossed squares in the upper inset of Fig. 4).

It is difficult to identify the exact effects of a trapped field on the “zero line” because we measure only m , which is a difference of two unknown quantities B and H_a . Furthermore, the value of the trapped field is random. More importantly, however, the data corrected on the background due to the trapped field (by its subtraction) (Fig. 4, the main panel) show no jump when crossing T_c —in both directions. This means that the flux in the sample is conserved, and no Meissner effect is observed. The magnetic flux conservation points towards the presence of a Faraday effect. This means that when crossing T_c , at $T < T_c$ superconducting screening currents are induced which keep the field inside the sample unchanged. Because the trapped field H_a of the superconducting magnet does not change with the temperature of the sample (these are two independent systems), the only possibility is that the Faraday effect is triggered by a change in the magnetic flux density B inside the sample at $T=T_c$. In other words, the paramagnetism of the sample at $T > T_c$ must start to diminish (its increase is less probable because the magnetic moment does not change with temperature as we start to cool down the sample from, e.g., 75 K or warm it up, Fig. 4, red crosses and red crossed squares), giving rise to the induced surface superconducting currents, which then keep the paramagnetic moment in the bulk unchanged. By warming the sample above T_c these macroscopic induced superconducting currents disappear at T_c because no bulk superconductivity is observed in the normal state of the sample. This suggests a cooperative behavior of the magnetism and superconductivity—in the sense that the paramagnetism of the sample at $T > T_c$ tends to disappear in the superconducting state as the temperature drops below T_c ; i.e., the paramagnetic moments tend to change their orientation. This normal-state paramagnetism may be caused either by pinned vortices, stripes, or by some other form of magnetic domains. However, there must be some form of interaction between the supercurrent and these domains at T_c , leading to a change of their magnetic moments. A similar memory effect is observed also at $T < T_c$ (Fig. 4, the lower inset). In this case the thermal run shown in sequence (8) was used:

$$\begin{aligned}
&300-6 \text{ K in } 0 \text{ G(ZFC)}, \quad 6-300 \text{ K in } 100 \text{ G(FW)}, \\
&300-6 \text{ K in } 100 \text{ G(FC)}, \quad 6-20 \text{ K in } 0 \text{ G(ZFW)}, \\
&20-6 \text{ K in } 0 \text{ G(ZFC)}, \quad 6-300 \text{ K in } 0 \text{ G(ZFW)}, \\
&300-6 \text{ K in } 0 \text{ G(ZFC)}.
\end{aligned} \tag{8}$$

These results suggest that the magnetic structures above and below T_c do not compete, and the currents responsible for the measured moment above T_c (Fig. 4, main panel, red solid diamonds, crosses, and crossed squares and grey open squares and solid triangles) are those responsible for the moment below T_c (Fig. 1, inset, and Fig. 4, lower inset).

IV. SUMMARY

We report the magnetization of a $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ single crystal under various history protocols. Using a special combination of in-field and in-zero-magnetic-field cooling and warming experiments we showed that the observed hysteresis effects cannot be caused by spurious magnetic impurities. We have demonstrated that the same thermal hysteresis

effects in the normal state are obtained not only when the applied magnetic field is increased or decreased at $T < T_c$, but also when it is increased or decreased at $T > T_c$ —for as long as the temperature is below T_s . We showed that for $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ with $H \parallel ab$, $T_s \approx 250$ K represents a transition from a fluctuating to a pinned ordered state ($T \leq T_s$), in which long-lived weak persistent currents may exist. When the applied magnetic field is zero, crossing T_c —both by warming and cooling—results in a smooth reversible transition indicating that the paramagnetic moments at $T > T_c$ interact with the supercurrent at $T < T_c$. The unprecedented similarity found in the thermal and magnetic history behavior of the measured magnetization below and above T_c [Fig. 1 (inset) and Figs. 2 and 4 (upper inset), respectively] adds credence to our earlier suggestion² for a common cause of the hysteretic behavior in the two temperature regions.

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⁶X-ray diffraction, micro-Raman, scanning probe microscopy, and electron probe microanalysis found no traces of impurities in our samples—at least to the level of 90 ppm. Our earlier studies of different single crystals and polycrystals, which included both same and different Sr concentrations (prepared in three different laboratories and with two different preparation methods), showed similar hysteresis effects (Ref. 2). As we discussed in Ref. 2, for any given sample we confirm our results on smaller pieces cut from the original measured piece. Some of the smaller pieces are also polished and remeasured to confirm the absence of possible surface effects. Separate experiments like those discussed in Fig. 1 were performed on pure Fe and Fe-contaminated samples. The results showed that the data obtained by cooling the sample in zero magnetic field do not follow the

straight “zero line” but the magnetic moment increases with decreasing temperature. Moreover, the ratio of the width of the hysteresis at 100 K to the magnetic moment at 300 K is at least one order of magnitude lower for the iron and iron-contaminated samples than that obtained from Fig. 1. Furthermore, in our samples (both here and in Ref. 2) the onset of the thermal hysteresis has strong doping dependence and crystallographic anisotropy, and the values of T_s are incomparable to those expected for any composition of iron, iron oxide, or other ferromagnetic impurities in general. These experiments exclude the possibility that the hysteresis effects reported here and earlier (Ref. 2) are due to possible undetected magnetic impurities. Of course there is always the possibility that a tiny amount of impurity phase might still be present and be beyond detection by any of the aforementioned chemical, spectroscopic, and thermodynamic tests we performed. If that is the case, then that impurity will have to follow the stringent systematic trends discussed in Ref. 2 and here. To the best of our knowledge there is no such magnetic phase.

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