

# Magnetoresistance and metamagnetic transition in $\text{La}_2\text{CuO}_{4+\delta}$ with a low Néel temperature

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(Submitted 5 June 1991)

*Pis'ma Zh. Eksp. Teor. Fiz.* **54**, No. 1, 32–35 (10 July 1991)

An anomalous increase in the hysteresis in the magnetoresistance and magnetization in the course of a metamagnetic transition has been observed in  $\text{La}_2\text{CuO}_{4+\delta}$  single crystals with  $T_N \simeq 165$  K as the temperature was lowered below 20 K. At the same time, the size of the jump in the magnetoresistance was observed to decrease. Both effects are interpreted as manifestations of the reentrant transition from an antiferromagnetic phase.

Theoretical<sup>1,2</sup> and experimental<sup>3,4</sup> research on the  $\text{La}_2\text{CuO}_{4+\delta}$  system shows that there is an interval of the concentration  $\delta$  (and thus of the Néel temperature  $T_N$ ) in which a reentrant transition from an antiferromagnetic phase to a paramagnetic phase, or to a phase of a so-called spin glass, can occur at low temperatures. It is possible that a transition of this sort was observed in Ref. 5 in samples with  $T_N < 170$  K. At low temperatures, there is a substantial increase in the hysteresis in the  $M(H)$  curves upon a phase transition to a state with a weak ferromagnetism. The curves of  $H_C(T)$  [where  $H_C$  is the critical field for the metamagnetic antiferromagnet–(weak ferromagnet) transition] for samples with  $T_N < 170$  K and  $T_N > 200$  K are very different. A Néel temperature in the interval  $170 < T_N^* < 200$  K appears to be critical, separating samples which exhibit different types of  $M(H)$  behavior in the course of the metamagnetic transition. In samples with  $T_N > T_N^*$ , in which a long-range magnetic order appears to exist for all  $T < T_N$ , the magnetoresistance and the magnetization undergo jumps.<sup>6</sup> These jumps were attributed in Ref. 7 to a change in the magnetic structure in the course of the metamagnetic transition. In the case  $T_N < T_N^*$ , on the other hand, the situation concerning the electron kinetics is not clear, particularly at low temperatures.

Our purpose in the present study was to learn about the magnetoresistance of  $\text{La}_2\text{CuO}_{4+\delta}$  single crystals with  $T_N < T_N^*$  in the geometry  $H \parallel c, j \parallel c$  (we are using the tetragonal designation system for the axes) for assistance in studying the metamagnetic transition from the antiferromagnetic state to the weak ferromagnetic state. The same samples were used on a magnetometer<sup>8</sup> for a study of the  $M(H)$  dependence over the temperature range  $T = 4.2\text{--}200$  K, in order to determine the relationship between magnetic and transport effects.<sup>1)</sup>

The samples for the resistance measurements were single-crystal parallelepipeds (with dimensions  $\sim 1 \times 0.5 \times 2.5$  mm; the  $c$  axis ran along the long side; the original crystals were pyramids). The samples for the magnetic measurements were truncated

pyramids (the larger base was  $2.5 \times 2$  mm, and the height was 3 mm). The original crystals had  $T_N \sim 260$  K; heat treatment in oxygen lowered  $T_N$  to 165 K (below  $T_N^*$ ). The height and width of the peak on the temperature dependence of the magnetic susceptibility  $\chi(T)$  did not change substantially at  $T = T_N$ . This result is evidence of a fairly uniform distribution of the excess oxygen. Contacts were applied to the samples through cathode sputtering of copper in certain areas. We found a resistivity  $\rho_{300K}^c = 18 \Omega \cdot \text{cm}$  with  $\rho_{4.2K}^c / \rho_{300K}^c = 4 \times 10^3$ .

Figure 1, a and b, shows the reduced resistance  $R(H)/R(0)$  and the moment  $M(H)$  at  $T = 4.2$  K in the region of the metamagnetic transition. It is easy to see the obvious hysteresis in the antiferromagnet-(weak ferromagnet) transition. This transition is accompanied by sharp changes in both  $R(H)$  and  $M(H)$ , at critical fields  $H_{c1}$  (as the field is raised) and  $H_{c2}$  (as the field is lowered). These critical fields are conveniently taken as the fields corresponding to the maxima of  $|dR/dH|$  and  $|dM/dH|$ . Corresponding curves were recorded for temperatures 4.2–100 K. The temperature dependence of the jump in the magnetoresistance,  $r = [R(H = 8T) - R(0)]/R(0)$ , and that in the moment,  $\Delta M$  (Fig. 2a), were then plotted. The temperature dependence of the fields  $H_{c1}$  and  $H_{c2}$  was also plotted (Fig. 2b). It can be

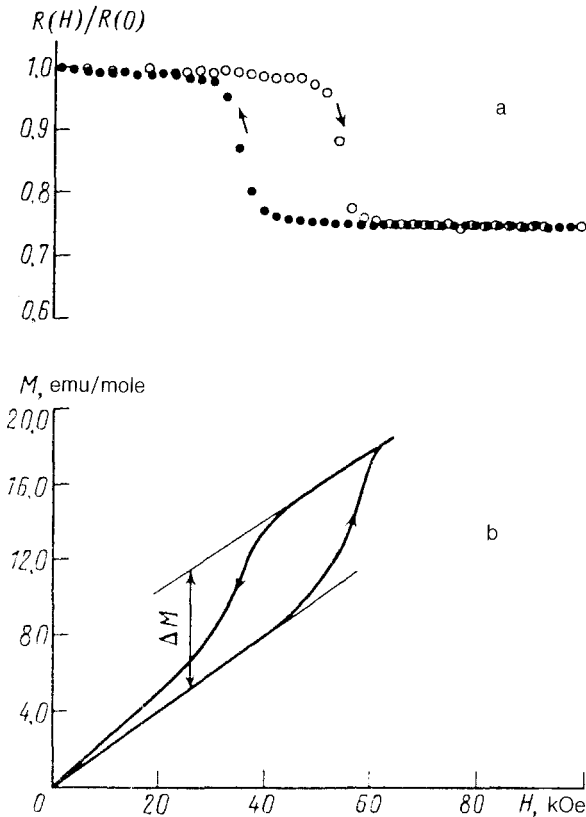


FIG. 1. The reduced resistance  $R(H)/R(0)$  (a) and the magnetic moment  $M(H)$  (b) versus the magnetic field.

seen from Fig. 2b that the curves of  $H_{c1}(T)$  and  $H_{c2}(T)$  found from  $M(H)$  and  $R(H)$  coincide completely. A noteworthy feature of these curves is that the anomalous increase in the hysteresis width ( $\Delta H = H_{c1} - H_{c2}$ ) and the decrease in the jump in the resistance,  $r$ , occur in the same temperature region. This correlation is illustrated in the inset in Fig. 2, which shows the relative changes in  $1/r$  and  $\Delta H$  in normalized form (the values are normalized to  $T = 30$  K) for the temperature interval 4.2–30 K. We observe a similar correlation at  $T_N > T_N^*$ ; whether this is a universal correlation (for  $T_N > T_N^*$ ) is an open question at this point.

This correlation suggests that the increase in the hysteresis and the decrease in the

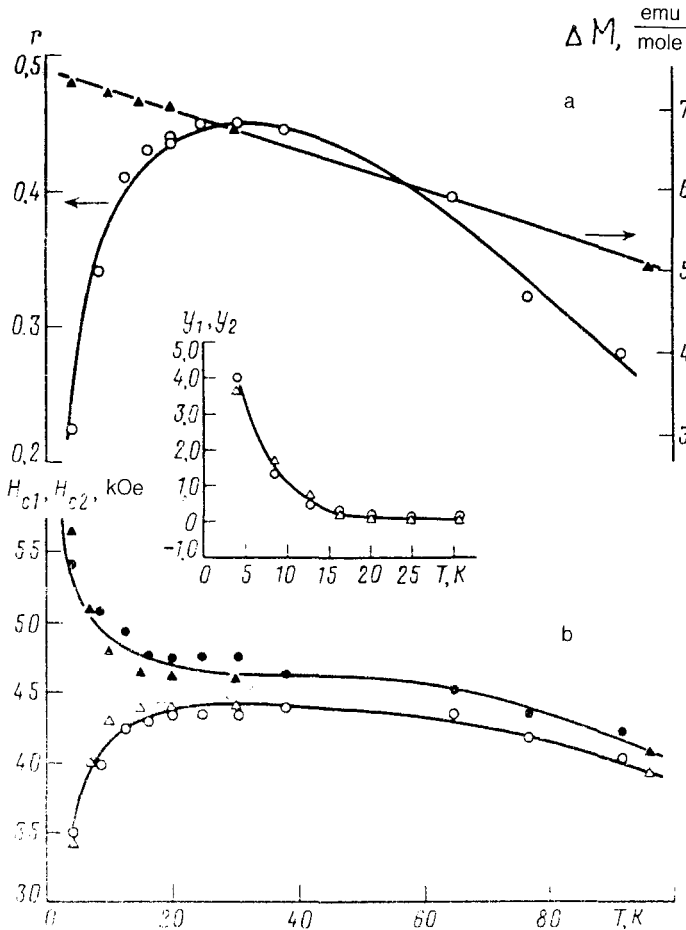


FIG. 2. a: Temperature dependence of the jump in the magnetoresistance,  $r = [R(H = 8T) - R(0)]/R(0)$ , and that in the magnetic moment,  $\Delta M$ , and of the magnetic fields. b:  $\bullet$ ,  $\blacktriangle$ — $H_{c1}$ ;  $\circ$ ,  $\triangle$ — $H_{c2}$ . The circles correspond to data on  $R(H)$ , and the triangles to data on  $M(H)$ . The inset shows the relative changes in  $1/r$  and the hysteresis  $\Delta H$  in normalized form.  $\circ$ — $y_1 = r(30 \text{ K})/r(T) - 1$ ;  $\triangle$ — $y_2 = \Delta H(T)/\Delta H(30 \text{ K}) - 1$  (the scale for  $y_1$  is larger by a factor of 4).

magnetoresistance jump are of the same nature. It is possible that a disruption of the antiferromagnetic order, which occurs with increasing oxygen content, is having an effect here.<sup>9</sup> Holes which localize between two copper atoms in Cu-O<sub>2</sub> planes (during the incorporation of superstoichiometric oxygen or during the replacement of lanthanum by strontium) cause a ferromagnetic interaction between them and form regions with a defective magnetic order in their vicinity.<sup>9</sup> These regions have a finite spin and are coupled with each other by a dipole-dipole interaction. At sufficiently low temperatures, this interaction can lead to a transition of the system of defects into a spin-glass state.<sup>1</sup> The defective regions with a frozen magnetic structure (with a size estimated<sup>1</sup> to be one or two cells) can act as pinning centers, retarding the propagation of the new phase during the first-order transition.<sup>10</sup> This effect would explain the increase in the hysteresis. At the same time, the interaction of defective regions with the antiferromagnetic matrix would amplify the fluctuations in the antiferromagnetism vector at low temperatures, thereby causing a reentrant transition from an antiferromagnetic state.<sup>1</sup> On the other hand, it would cause a decrease in  $r$  with decreasing temperature according to Ref. 7.

Going back to Fig. 2a, we see a surprising fact: The decrease in the jump in  $r$  is not accompanied by a decrease in the jump in the magnetic moment. In other words, the ferromagnetic moment of the planes does not decrease. It is possible that this fact means that the disorder of the magnetic structure after the reentrant antiferromagnetic transition involves only those components of the copper spins which are longitudinal with respect to the planes.

In summary, it can be concluded that an anomalous increase in the hysteresis in  $R(H)$  and  $M(H)$  is observed in the course of a metamagnetic transition, with a simultaneous decrease in the jump in the magnetoresistance, possibly because of a reentrant transition from the antiferromagnetic phase, as the temperature of samples with low Néel temperatures ( $T_N < T_N^*$ ) is lowered below 20 K.

We wish to thank A. N. Bazhan, A. S. Ioselevich, L. B. Dubovskii, and S. N. Burmistrov for discussions.

This study was carried out within the framework of Project 90347 of the State Program on High-Temperature Superconductivity.

<sup>1)</sup> We wish to thank A. N. Bazhan, of the Institute of Physical Problems, Academy of Sciences of the USSR, for studying the magnetic properties of La<sub>2</sub>CuO<sub>4+δ</sub>.

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Translated by D. Parsons