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Magnetostriction of single-crystal La_2CuO_4

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Longitudinal magnetostriction ($\Delta l/l$) experiments have been used to probe the magnetic phase diagram of single-crystal La_2CuO_4 at 4.2, 77, 145, and 191 K. The geometry used is ($l \parallel \mathbf{H} \parallel \hat{\mathbf{b}}$), where $\hat{\mathbf{b}}$ is a unit vector along the orthorhombic b axis, perpendicular to the CuO_2 planes. The observed magnetic transition is associated with an induced alignment, along the field direction, of the small out-of-plane canting of the Cu^{2+} spins. At all temperatures, the transition is marked by a small increase in l, with $\Delta l/l$ about 2 to 3×10^{-8}. A large hysteresis was observed at 4.2 K, with the transition field, H_t, in the up trace being 60.1 kOe, and in the down trace 45.8 kOe. This hysteresis strongly decreases with increasing temperature. At 191 K the jump in l is still quite sharp indicating that the transition is still first order. Our values of H_t are in good agreement with those obtained on similar crystals using different techniques.

INTRODUCTION

The magnetic properties of La_2CuO_4 have been of interest recently due to the suggestion that magnetic interactions may provide a mechanism for high-temperature superconductivity in the doped compounds.[1–5] Neutron scattering experiments on powder samples[6] have shown that, below about 230 K, the Cu^{2+} spins order antiferromagnetically, with a magnetic structure similar to that of the well-known two-dimensional antiferromagnet K_2NiF_4. The spins lie in the CuO_2 planes and are pointed in the [001] or [00$\bar{1}$] directions.[6] Here, we are referring to orthorhombic axes, where the ac plane contains the CuO_2 sheets, and the b axis is the longest crystallographic direction. Peculiar to this compound is the unusually high antiferromagnetic coupling J between nearest-neighbor spins, of the order of 1300 K.[7] Furthermore, a small rotation of the CuO_6 octahedra in the orthorhombic phase, generates an antisymmetric exchange term J^{bc} (of about 7 K) that causes a small canting of the spins out of the CuO_2 planes.[8] A weak antiferromagnetic interaction between the CuO_2 planes[8] (of about 0.03 K) then determines that the direction of the canted moments alternate along the b direction.

The antiferromagnetic order of the spin canting can be modified by a magnetic field. In particular, a field applied along the b axis can align all the canted moments, changing the antiferromagnetic canting arrangement into a ferromagnetic one. This process has been investigated by magnetization, magnetoresistance,[8,9] and neutron scattering[10] experiments, and a detailed theory is presented in Ref. 8. The observed phase transition is similar to the one that occurs in metamagnets.[11] The associated phase boundary, $H_t(T)$, is expected to be of first order at low temperatures, eventually changing to second order at a tricritical point, as the temperature approaches the Néel temperature.[11] The magnetization jump observed in previous work[8] indicates a first-order transition at 150 K, but is less conclusive at higher temperatures. In this paper we report magnetostriction results associated with this transition. These results were obtained from measurements at a constant temperature of the fractional change in length l

along the b direction, caused by a varying applied magnetic field aligned along the same direction:

$$\Delta l/l = [l(H) - l(0)]/l(0) . \quad (1)$$

Magnetostriction measurements provide an important and complementary tool to determine phase boundaries and to characterize the order of the transition.[12]

EXPERIMENT

The investigated sample was a specially oriented piece cut from a larger single crystal grown at MIT by the top seeded solution method.[13] The magnetic behavior of this sample was checked prior to the magnetostriction experiment. The maximum of the dc susceptibility vs temperature curve, measured in an applied field of 5 kOe, gives a Néel temperature $T_N \sim 235$ K. Magnetization measurements with $\mathbf{H} \parallel \hat{\mathbf{b}}$ accurately reproduced previous published data[8] on similarly grown single crystals.

The magnetostriction measurements were carried out in a capacitive dilatometer, the details of which have been described previously.[14] To properly install the sample in the dilatometer, a pair of parallel ac faces were produced by lapping. The normal to these faces was within $\pm 0.5°$ from the b axis, as checked by x-ray diffraction measurements. The total length of the sample along the b axis was 1.9 mm, with the area of the ac faces being roughly 4 mm^2. The sample was held in place by a spring loaded brass plate, that produced a uniaxial pressure of about 70 bar. The capacitance, at the gap size of 0.08 mm, was $C = 13.5$ pF, and was measured by a bridge driven with an amplitude of 10 V and a frequency of 1000 Hz. The magnetic field was generated by a superconducting magnet for which the field varied by less than 0.01% across the sample and dilatometer. To check for the possibility that the results were influenced by magnetic torques, the dilatometer was deliberately tipped by about 1.5° relative to $\mathbf{H}$, in several directions. No significant differences were found.

To measure the small values of $\Delta l/l$, associated with the very small jumps of magnetization observed at the transition,[8] a high degree of stability in temperature has to be achieved. This is necessary to avoid the spurious signals caused by the thermal expansion. For this reason we have measured $\Delta l/l$ only at temperatures where the dilatometer could be immersed in a boiling liquid thermal bath, that is, 4.2 K (liquid helium), 77 K (liquid nitrogen), 145 K (Freon-14), and 191 K (Freon-13). The higher temperatures were checked using a platinum resistance thermometer.

RESULTS AND DISCUSSION

At all temperatures a clear step in $\Delta l/l$ was observed at the transition field H_t, with l always increasing with increasing H. Figures 1 and 2 show the data for $T = 4.2$ and $T = 191$ K. The upper parts of the figures show the raw traces, as they were recorded from the capacitance bridge. Figure 1 shows traces for the field going both up and down, while Fig. 2 shows just the trace for the field going down. The steps are quite evident. To determine the

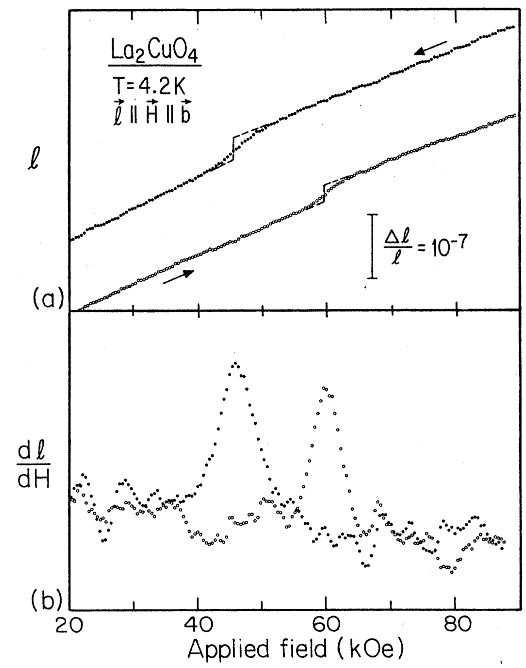

FIG. 1. (a) Raw longitudinal magnetorestriction data of length l vs applied field H for a single crystal of La_2CuO_4 at 4.2 K. The values of the transition fields of the up and down traces (see arrows) were determined from the derivatives shown in (b). The dashed lines illustrate the determination of the value of Δl at H_t.

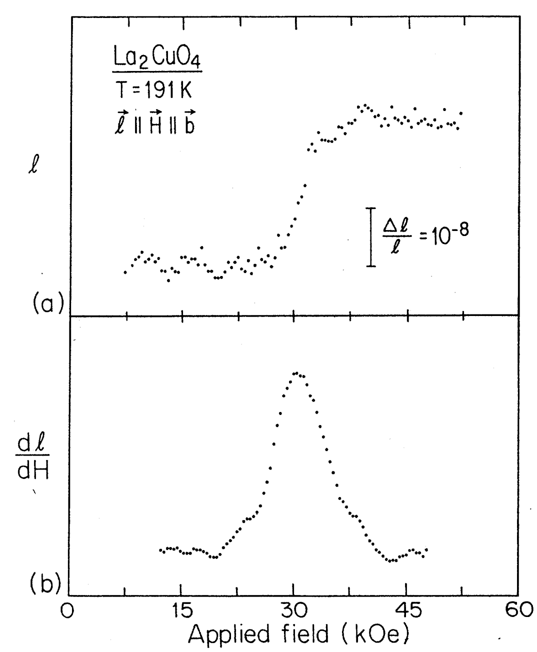

FIG. 2. (a) An example of raw data of length l vs a decreasing applied field H for a single crystal of La_2CuO_4 at 191 K. The value of the transition field was determined from the derivative shown in (b). Note that there is a clear first-order-like step at the transition field $H_t = 30.9$ kOe.

value of H_t the traces were differentiated using a standard computer program. The lower part of the figures show the corresponding derivatives. The transition field H_t was taken as the point of maximum derivative in Figs. 1(b) and 2(b), and these values of H_t were used in the interpretation of Figs. 1(a) and 2(a). The size of the jump in $\Delta l/l$ was obtained by using linear fits to the data above and below the transition, and by taking the mismatch of the two fits at H_t. The dashed lines in Fig. 1(a) illustrate the procedure.

Several traces were taken at each temperature, and Table I summarizes the averages obtained for each parameter and the corresponding uncertainties. Since the traces were obtained by sweeping the field, it was necessary to check for a possible dependence on sweep rate. Traces obtained at 120 and 240 Oe/sec, for the same temperature, showed no noticeable difference in the measured parameters for all temperatures that were investigated. Furthermore, no time-dependent effects were observed when the field sweep was stopped at a given field value and then resumed. Above 4.2 K, the values of H_t listed in Table I and in Fig. 3 are in good agreement with the phase boundary in similarly grown samples, as determined by magnetization and magnetoresistance,[8] and by neutron scattering[10] measurements. Our dc susceptibility result for $H_t=0$ (the square point) is plotted in Fig. 3 together with data from neutron scattering.[10] The transition fields $H_t(T)$ in Ref. 9 are about 20% higher than ours, which may be due to differences in samples.

Unique to the present data is the large hysteresis, of about 14 kOe, observed at $T=4.2$ K. References 8–10 do not quote a value for the hysteresis at 4.2 K. However, the value $H_t(0)=53\pm3$ kOe obtained in Ref. 8 by extrapolation of magnetization data, is in very good agreement with the average of our up and down values of $H_t=52.95$ kOe at $T=4.2$ K. At $T=77$ K, the hysteresis in our data is already much smaller, $\Delta H_t=2.8$ kOe, being comparable to what was observed at $T=80$ K in the neutron scattering experiment, $\Delta H_t \sim 2.3$ kOe.[10] It is noteworthy that, in our experiments, the maximum field of the superconducting magnet was always high enough to far exceed the upper values of H_t observed. This was not the case in the magnetization and neutron scattering measurements of Refs. 8 and 10 and this may have been the reason why the large hysteresis was not observed at $T=4.2$ K in these cases.

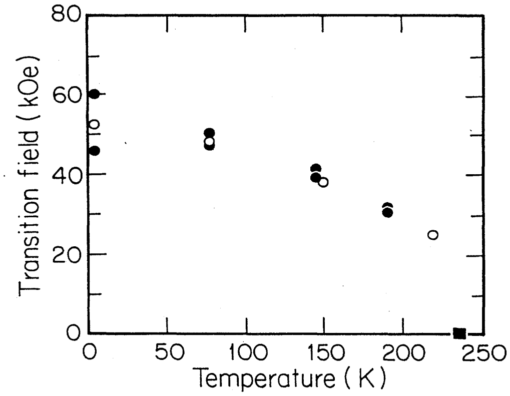

FIG. 3. The phase boundary $H_t(T)$, in the H-T plane, as determined by magnetostriction measurements (solid circles). The comparison points (open circles) are from the neutron scattering data of Kastner *et al.* (Ref. 10). The point (square) at 235 K was determined by dc susceptibility measurements on this sample.

The shape of the $\Delta l/l$ magnetostriction jump, as well as the hysteresis observed, are typical of a first-order transition. Although the hysteresis diminishes drastically with increasing temperature, the width of the transition remains approximately constant over the entire temperature range. A comparison between Figs. 2 and 1 shows that the transition at $T=191$ K is as sharp as at $T=4.2$ K. (Note that the field scale is not the same in the two figures.) The absence of any broadening at $T=191$ K is an indication that the transition remains first order up to that temperature.

Within the accuracy of our data, the size of the magnetostriction jump is approximately constant, $\Delta l/l \simeq 3\times10^{-8}$, from $T=4.2$ K up to $T=145$ K, with a small decrease being detected only at $T=191$ K. The fact that the sample expands along the b axis, as the sample goes through H_t with increasing H, is consistent with a decrease in the antiferromagnetic coupling between the CuO_2 planes, $J_\perp$, with increasing interplanar distance b.

We can relate the size of the magnetostriction jump to the change in the magnetization ΔM using a modified

TABLE I. Results obtained from longitudinal magnetostriction measurements performed on single-crystal La_2CuO_4 at different temperatures.

Temperature (K)	H_t^{up} (kOe)	H_t^{down} (kOe)	Other results[a] (kOe)	Size of jump $10^8\Delta l/l$
4.2	60.1 ± 0.3[b]	45.8 ± 0.4	52	3
77	50.1 ± 1.2	47.3 ± 0.7	48	3
145	41.2 ± 0.5	39.2 ± 0.9	38[d]	3
191	31.9[c]	30.9	···	2.5

[a]Transition fields from Ref. 10.
[b]Error corresponds to two standard deviations.
[c]Not enough traces for evaluating the statistical uncertainty.
[d]Measurement at 150 K.

Clausius-Clapeyron equation[15]

$$\left(\frac{\partial H_t}{\partial p_b}\right)_T = \frac{\Delta l}{l}\frac{1}{\Delta M}, \tag{2}$$

where p_b is a uniaxial pressure in the b direction. Reference 8 reports the value $\Delta\mu_F(0) = 2.1\times10^{-3}\mu_B/Cu^{2+}$, for the induced ferromagnetic moment at $T=0$ K. Taking the density as 7.1 g/cm^3 we obtained $(\partial H_t/\partial p_b)_{T=0} \simeq 0.15$ Oe/bar. Using the relation

$$H_t(0)\Delta\mu_F(0) \approx J_\perp s^2, \tag{3}$$

where $s=\frac{1}{2}$ and assuming that $\Delta\mu_F(0)$, and hence J^{bc}/J, are relatively insensitive to p_b, we can estimate $(\partial J_\perp/\partial p_b)_{T=0} \approx 7\times10^{-9}$ meV/bar. Assuming that the pressure coefficient of the lattice constant b does not vary much with temperature or doping, we can use the value of $\Delta p_b/(\Delta b/b) = 5.9\times10^6$ bar (Ref. 16) to obtain $(\partial J_\perp/\partial b)_{T=0} \approx 3\times10^{-3}$ meV/Å.

In conclusion, we have measured the magnetostriction that is associated with the field-induced alignment of the canted moments along the b axis of La_2CuO_4. A large hysteresis was observed, for the first time, in the magnetic transition at $T=4.2$ K and this hysteresis was followed up to $T=191$ K. The shape of the l vs H traces clearly indicates a first order transition up to the highest temperature investigated, $T=191$ K.

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