Correlation of electronic and magnetic properties of La_2CuO_4

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The orientational transition in La_2CuO_4 crystals in strong magnetic fields has been studied. The magnetization and magnetroresistance curves both exhibited hysteresis. The width of the hysteresis loops increased as a result of cooling, reaching fairly high values of $\Delta H = 15$ kOe at liquid-helium temperature. The experimental results are compared with the theoretical (H, T) phase diagram of La_2CuO_4. Microscopic reasons for the correlation between the magnetization and magnetoresistance curves are discussed.

1. INTRODUCTION

The possibility that a magnetic mechanism may account for the high-temperature superconductivity has recently been under active investigation along with other nonphonon mechanisms. Therefore, the relationship between the electronic and magnetic properties of copper oxides is important. One of the examples of this relationship is the orientational phase transition in a magnetic field in insulating La_2CuO_4; according to Refs. 1–3, in fields $H > H_c$, the antiferromagnetic (AFM) phase changes to weakly ferromagnetic (WF). The appearance of the ferromagnetic moment is accompanied by a reduction in the magnetoresistance $R(H)$ (Ref. 3).

Although the correlation between the magnetization curves $\sigma(H)$ and $R(H)$ is established in Ref. 3, it still leaves a number of unanswered questions. For example, the authors of Ref. 3 reported considerable hysteresis of the $R(H)$ curves, but not of $\sigma(H)$. A later study[4] revealed the hysteresis of $\sigma(H)$, but the magnetoresistance was not investigated on that occasion. Moreover, these studies[1–4] did not provide information on the low-temperature magnetization because of the limited range of magnetic fields used.

Here we report the results of an investigation of the orientational transition in La_2CuO_4 crystals in strong magnetic fields. We observed complete correlation between the $\sigma(H)$ and $R(H)$ curves: both exhibited hysteresis and the width of the hysteresis loops increased as a result of cooling, reaching fairly high values $\Delta H = 15$ kOe at liquid-helium temperature. The experimental results are compared below with the theoretical (H, T) phase diagram of La_2CuO_4 (Ref. 5). Microscopic explanations for the correlation between the magnetization and magnetoresistance curves are also discussed.

2. SAMPLES AND MEASUREMENT METHODS

Our experiments were carried out on La_2CuO_4 single crystals grown at the Institute of Crystallography of the USSR Academy of Sciences by crystallization from nonstoichiometric melts with an excess of CuO. The details of the procedure were given in Ref. 6. The temperature dependence of the electrical resistivity was determined by the usual four-probe method and it was found that $\rho(T)$ resembled the resistivity of a semiconductor in the temperature range 4.2 K $< T <$ 300 K.

Our investigation of the magnetic properties was made using an automated vibrating-sample magnetometer and a superconducting solenoid. We determined the dependence of the magnetization on an external field applied at a fixed temperature as well as the temperature dependence of the magnetization in fixed external fields. The temperature of a sample could be varied within the range 4.2–300 K and the magnetic field within 0–75 kOe. The sensitivity of the magnetometer in respect of the magnetic moment was 10^{-4} emu/g and temperatures were kept constant to within 0.1 K.

The electrical resistivity was determined by the four-probe method under constant-current conditions. The potential and current contacts were formed by a silver paste and were arranged so that the current flowed along the c crystal axis. The magnetic field of up to 100 kOe intensity generated in a Bitter solenoid coincided with the direction of the current. The sample together with the heater and a thermocouple were placed inside a helium cryostat and the voltages from the potential contacts as well as from a shunt across the solenoid were recorded using an X-Y potentiometer.

3. RESULTS OF THE MAGNETIZATION AND MAGNETORESISTANCE MEASUREMENTS

According to Ref. 3, the orientational transition is observed when an external magnetic field is oriented parallel to the tetragonal c axis (across the CuO_2 layers). The dependences $\sigma(H)$ were obtained at different temperatures (Fig. 1). As expected, cooling increased the critical field H_c and reduced the width of the superconducting transition. Similar changes in the magnetization curves were reported earlier for weak ferromagnets (see, for example, Ref. 7 for the data on $CoSO_4$). An increase in the width of the hysteresis loop ΔH as a result of cooling was less usual. This was reported also in Ref. 4 descending in temperature to $T > 45$ K, whereas in the present study it was observed to $T \geqslant 4.2$ K. The value $\Delta H(4.2\text{ K}) = 15$ kOe was obtained. It should be pointed out that the large width of the hysteresis loop at 4.2 K had been deduced earlier from hf measurements[8] and our value of ΔH agreed with that given in Ref. 8.

The typical magnetic-field dependence of the normalized resistance $R(H)/R(0)$, where $R(H)$ is the resistance in a magnetic field and $R(0)$ is the corresponding value in the absence of the field, (plotted in Fig. 2), was determined for

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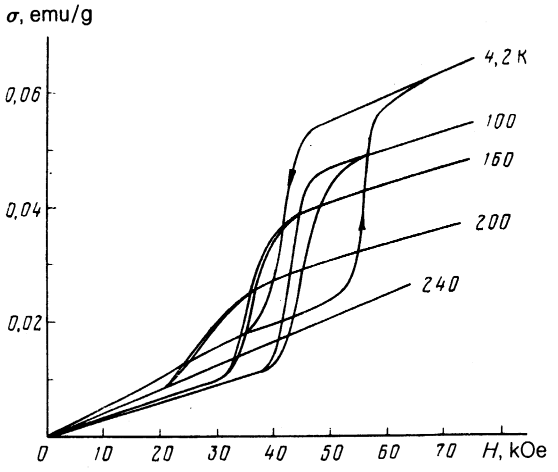

FIG. 1. Field dependence of the magnetization of La_2CuO_4, measured parallel to the c axis at various temperatures.

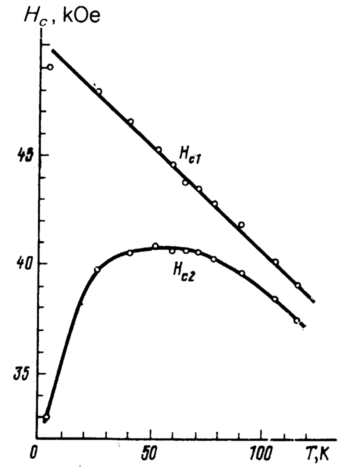

FIG. 3. Temperature dependence of the critical fields H_{c1} and H_{c2}.

two temperatures. Hysteresis was clearly observed and a partial hysteresis loop was also obtained (dashed line). The critical fields H_{c1} and H_{c2}, determined for half the change in the magnetoresistance as a function of temperature, are plotted in Fig. 3: it was found that H_{c2} varied nonmonotonically with low temperatures. The existence of partial loops indicated that the orientational transition was a first-order phase transition.

4. DISCUSSION OF RESULTS

The noncollinearity of the magnetic structure of the antiferromagnet La_2CuO_4 and the possibility of appearance of weak ferromagnetism were considered theoretically in Ref. 9 using symmetry analysis. Neutron-diffraction investigations[10] showed that below H_c a crystal of La_2CuO_4 was a noncollinear antiferromagnet with canted sublattices and above H_c a weak ferromagnetic phase appeared. The hysteresis of the magnetization curves and the field dependence of the magnetoresistance[3] can be interpreted as evidence of a first-order orientational transition.

An alternative explanation of the broad phase transition in La_2CuO_4 is suggested in Ref. 5, where it is shown that a transition can take place to canted phases in which the spins in the adjacent layers are in the basal plane and are rotated by an angle $\theta(H_z) < 180°$ relative to one another.

We shall analyze the phase diagram using the results of Ref. 5 and the fact that the transition is of the first order.

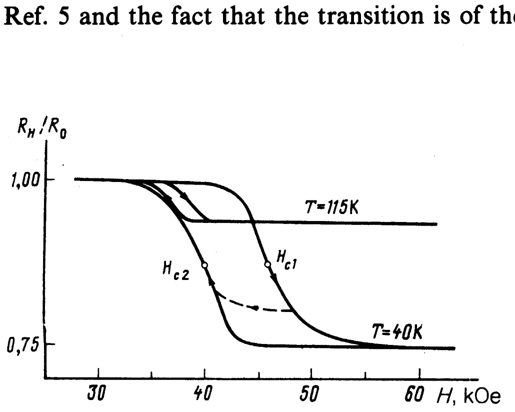

FIG. 2. Field dependence of the resistance of La_2CuO_4.

According to Ref. 5, magnetic phase diagrams change fundamentally depending on the ratio of the parameters H_{1e}, H_3; for $H_{1e} < H_3$ one first-order transition takes place from the AFM to the WF phase and in the range $H_{1e} > H_3$ we can expect canted phases (Fig. 4). Using the notation of Ref. 5, we now have

$$H_{1e}^2 = (2H_e/H_D)^2 (2\tilde{h}_e' - \tilde{H}_{Ay})^2,$$
$$H_3^2 = (2H_e/H_D)^2 (2\tilde{h}_e' + \tilde{H}_{Ay})(2\tilde{h}_A' + \tilde{H}_{Ay}), \qquad (1)$$

where H_e is the intralayer exchange field; H_e' and h_e' are the interlayer exchange fields; H_D is the Dzyaloshinskii field; H_{Ay} is the anisotropy field within a layer; h_A' is the field of the tetragonal interlayer anisotropy. The renormalized fields occurs as follows:

$$\tilde{h}_e' = h_e' + (H_D/2H_e)^2 H_e'/2,$$
$$\tilde{h}_A' = h_A' + (H_D/2H_e)^2 H_e'/2, \qquad (a)$$
$$\tilde{H}_{Ay} = H_{Ay} + H_D^2/2H_e.$$

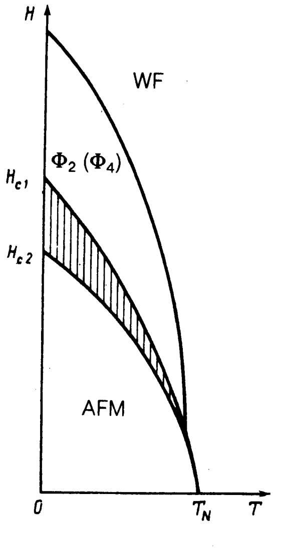

FIG. 4. Magnetic phase diagram of La_2CuO_4 in the range $H_{1e} > H_3$, for $h_e' > h_A'$ (taken from Ref. 5). Measurements were made parallel to the z axis.

It is demonstrated in Fig. 4 that different canted phases Φ_2 (for $h'_e > h'_A$) or Φ_4 (for $h'_e < h'_A$) are possible, and in the shaded region we have coexisting AFM phases and Φ_2 (Φ_4) in the range of fields $\tilde{H}_{Ay} > 0$ and a first-order transition or two second-order phase transitions in the range $\tilde{H}_{Ay} < 0$.

The existence of a first-order transition demonstrates that either $H_{1e} < H_3$ or $H_{1e} > H_3$ and $\tilde{H}_{Ay} > 0$. If we assume that all the interactions are of the same order and postulate $\tilde{h}'_e = \tilde{h}'_A = \tilde{H}_{Ay}$, then the condition $H_{1e} < H_3$ is satisfied and there are no intermediate phases.

We can easily show that the condition $H_{1e} < H_3$ is equivalent to

$$\tilde{H}_{Ay} > 2\tilde{h}_e'(\tilde{h}_e' - \tilde{h}_A')/(3\tilde{h}_e' + \tilde{h}_A'). \tag{2}$$

We shall consider four possible cases:

1) $\tilde{H}_{Ay} > 0$, $\tilde{h}'_e > \tilde{h}'_A$, when both variants of phase diagrams with canted media and without them are possible;

2) $\tilde{H}_{Ay} > 0$, $\tilde{h}'_e < \tilde{h}'_A$, when canted phases cannot appear if there is only a first-order AFM—WF transition;

3) $\tilde{H}_{Ay} < 0$, $\tilde{h}'_e > \tilde{h}'_A$, when there are two second-order transitions (this is in conflict with the experimental results);

4) $\tilde{H}_{Ay} < 0$, $\tilde{h}'_e < \tilde{h}'_A$, as in the first case, both variants of the phase diagrams are possible.

Therefore, the occurrence of the first-order phase transition imposes some limitations on the possible range of the parameters.

As for the relationship between the jumps in the wings of $R(H)$ and $\sigma(H)$, a similar correlation is known to occur in narrow-gap magnetic semiconductors.[11] Transition from the homogeneous ferromagnetic phase to the AFM state is accompanied by spin–polaron narrowing of the band and by a reduction in the carrier mobility, since in an inhomogeneous magnetic structure a hop of a carrier without spin flip is difficult. In this case an increase in the magnetic field results in the converse process: a transition to a more homogeneous phase increases the carrier mobility and reduces the resistivity.

This spin mechanism of the negative magnetoresistance can be expected in the case of band carriers within narrow bands or in the case of hopping in semiconductors containing deep small-radius impurity states. In La_2CuO_4 we can expect Mott variable-length hopping. A theory of the spin magnetoresistance of La_2CuO_4 given in Ref. 12 postulates this mechanism for the dependence of the probability of a jump of an electron on the symmetry of the magnetic state.

5. CONCLUSIONS

Magnetic and electrical measurements in strong magnetic fields and at low temperatures established that the changes in the magnetic structure and the electrical conductivity are fully correlated. The appearance of a homogeneous component of the magnetization reduced the resistivity and this behavior was typical of antiferromagnetic semiconductors with narrow bands. The low-temperature hysteresis loops of the magnetization curves and of the field dependences of the resistance were quite wide. The observed magnetic phase transition was of first order. Using this conclusion we established some limitations on the permissible parameters of the anisotropy and exchange fields used in the phenomenological theory.

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