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MAGNETIC FIELD DEPENDENCE OF THE 3D ORDERING IN $La_2CuO_{4-\delta}$

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The magnetic field dependence of the three dimensional magnetic ordering transition has been examined in single crystalline $La_2CuO_{4-\delta}$ by magnetization and magnetoresistance measurements. We establish a phase diagram in the magnetic field-temperature plane and point out similarities to metamagnetic transitions observed in other magnetic systems.

The unusual magnetic properties of $La_2CuO_{4-\delta}$ have been explored thoroughly by magnetic susceptibility[1], neutron diffraction[2] and nuclear resonance[3] studies but the detailed nature of the various correlations still is not understood completely. The static magnetic susceptibility χ measured at a relatively low magnetic field has a broad minimum around room temperature, followed by a sharp maximum at T_N with decreasing temperature[1]. The temperature dependence of χ above T_N can not be understood in terms of simple antiferromagnetic interactions and signals the presence of ferromagnetic interactions in the material. Neutron diffraction studies[2] above T_N clearly indicate the presence of strong two dimensional antiferromagnetic correlations within the copper-oxygen planes and at T_N three dimensional ordering is established with a significant ferromagnetic component. T_N also strongly depends on the oxygen content and it is believed that the largest value, $T_N \sim 290$ K, corresponds to $\delta = 0.03$, with T_N depressed as the oxygen deficiency decreases[4]. The onset of 3D ordering leads to a rather sharp decrease in the electrical resistivity which may be due to a loss of spin disorder scattering.

We report the observation of a magnetic field induced transition at temperatures $T < T_N$ from magnetic susceptibility and magnetoresistance(R(H)) studies and establish an H-T phase diagram. We also point out the similarities of our findings to those made on $MnAu_2$[5] or $FeCl_2$[6] and suggest that our observations are consistent with a field induced metamagnetic transition.

Cubicly shaped crystals of $La_2CuO_{4-\delta}$ were grown from a CuO-based flux. Electron microprobe analysis showed La : Cu = 1.85 : 1 and X-ray crystallography gave orthorhombic lattice constants a = 5.358 Å, b = 13.181 Å and c = 5.403 Å. The excess Cu may be associated in part with CuO flux at the sample surface.

The magnetic susceptibility was measured with a Quantum Design SQUID susceptometer using magnetic fields to 5 Tesla. Magnetization vs. temperature curves with various applied fields are displayed in Fig. 1 along with the magnetic susceptiblity plotted for two different magnetic fields in the inset of the figure. The low field magnetization and susceptibility are essentially identical to those measured by

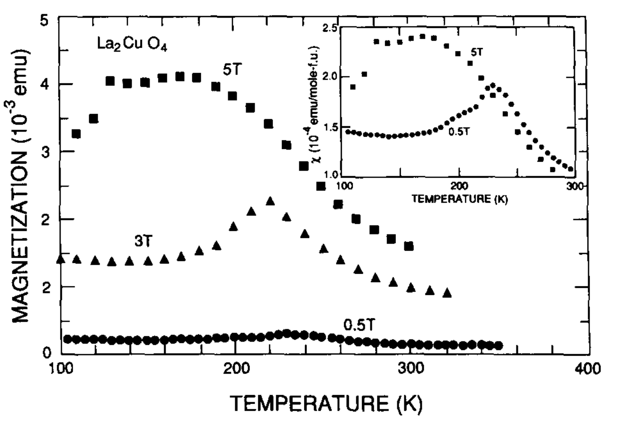

Fig. 1 Temperature dependence of the magnetization of $La_2CuO_{4-\delta}$ at various applied fields(sample mass = 139.3 mg). The inset shows the magnetic susceptibility for two different field values.

others earlier($T_N \sim 235$ K at low field) and we also observe a shift of the magnetization maximum to lower temperatures with increasing fields. The behavior shown in Fig. 1 suggests a suppression of the three dimensional antiferromagnetic ordering temperature with increasing field. However, the very broad maximum of magnetization in a 5 Tesla magnetic field in the temperature range between 130 K and 200 K is unusual and is not observed in the previous work[7] by others on sintered powders. This behavior is indicative of a magnetic field induced transition. We have, further, measured isothermal magnetization curves(M(H)) at three different temperatures and the results are displayed in Fig. 2. The transition, indicated by the deviation from linear M-H behavior, shifts progressively to lower magnetic field values with increasing temperature and the M(H) curve at 250 K, which is above T_N, is linear within experimental error. (The zero field intercept of this curve is essentially zero, which indicates there are negligible ferromagnetic impurities). We note that the magnetic field range($0T < H \leq 5T$) in our susceptometer prohibits further study of the magnetic field induced transition in the M(H) curves below 80 K.

As mentioned above, the dc resistivity shows a rather sharp decrease at T_N due probably to reduced spin scattering. The derivative of resistivity with respect to temperature normalized by resistivity, which is, in some circumstances, identical[8] to the specific heat anomaly, is shown in the inset of Fig. 3 and exhibits a feature indicative of a broad antiferromagnetic transition around 235 K. The magnetoresistance, measured as a function of magnetic fields up to 9 Tesla, decreases sharply at the field value where the M(H) curve is also anomalous and three representative curves are displayed in Fig. 3. The critical fields(H_C) at various temperatures defined by the sharp decrease of magnetoresistance (i.e., the maximum of $|dR(H)/dH|$) are displayed as squares in Fig. 4. For comparison, in Fig. 4 the solid circle shows the ordering temperature determined by resistivity in zero field or magnetic susceptibility at low field, the triangle is determined by the sharp decrease of magnetic susceptibility in a 5 Tesla field(see Fig. 1), and the sharp increase of isothermal magnetization at 170 K(see Fig. 2) defines the diamond. Thus all results in our measurements are suggestive of the phase diagram shown in Fig. 4.

Similar behavior was found earlier[9] in specimens grown from PbO-flux that exhibited a weak magnetic susceptibility anomaly around 130 K. In that study, the anomalies in the M(H) and R(H) curves were less pronounced than those reported here and the magnetic fields at which the transition appeared were found to be small, with a fractional reduction from values given here roughly comparable with the fractional differences in T_N.

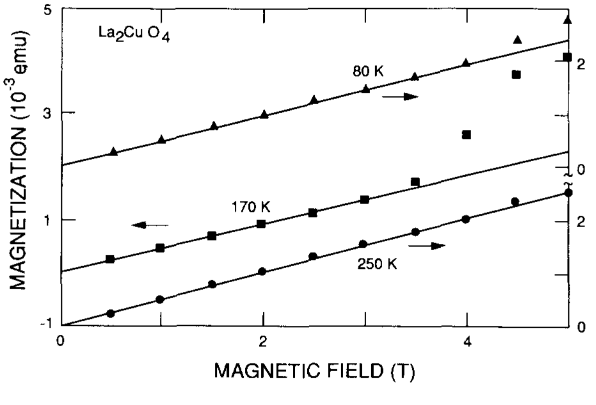

Fig. 2 Isothermal magnetization vs. field curves at various fixed temperatures. The solid lines are drawn through the low field data to demonstrate the nonlinear behavior observed.

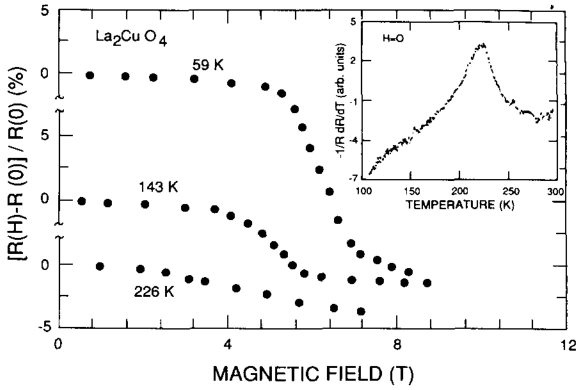

Fig. 3 Fractional change of the resistivity vs. magnetic field at three different temperatures. -(dR(T)/dT)/R vs. temperature is displayed in the inset.

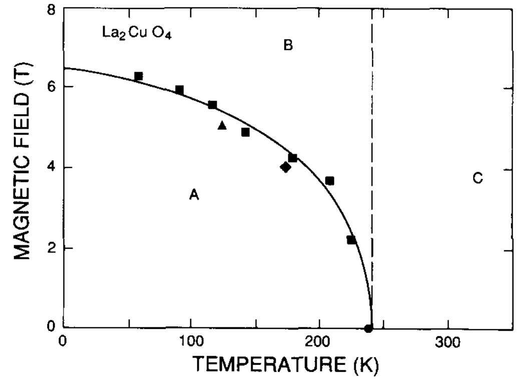

Fig. 4 H-T phase diagram suggested by our results. The dot, squares, triangle and diamond are from T_N at low field, R(H) vs. H, M(T) vs. T at 5 Tesla and M(H) vs. H at 170 K, respectively. (See text for details.) A-phase and C-phase are antiferromagnetic and paramagnetic states, respectively: B-phase could be a complex ferromagnetic-like state(see text for details). The vertical and dashed line dividing phases B and C could be more complicated than indicated.

We discuss the characteristics of these experimental findings. First, although the overall temperature dependence is complicated, just above the ordering transition(245 K < T < 320 K) the magnetic susceptibility measured in a 0.5 T field can be fit to $\chi_{mol} = \chi_o + C/(T-\theta)$, where $\chi_o = 4 \times 10^{-5}$ emu/mole-f.u.,$C = 7.9 \times 10^{-3}$ emu-K/mole-f.u. and $\theta = 46.4$ K, implying an effective magnetic moment per Cu ion of 0.25

μ_B if the Curie term is due solely to Cu ions. We note that even though the magnetic ordering is antiferromagnetic, Θ is positive, indicative of the presence of ferromagnetic correlations. Neutron diffraction experiments[10] suggest ordering due to antiferromagnetic coupling of ferromagnetically correlated layers. Second, the critical field(H_c) decreases with increasing temperature and disappears at T_N; furthermore, as shown in Fig. 2, the magnetization is linear with field below H_c: this behavior is rather different from a typical spin-flop transition such as observed in $(Cr_2O_3)_{1-x}(Al_2O_3)_x$[11]. We note, however, that the above properties of the field induced transition in La_2CuO_4 are general features[12] of the metamagnetic transition observed in systems such as $MnAu_2$ or $FeCl_2$, e.g., positive Θ(the presence of ferromagnetic sheets), a critical field that increases with decreasing temperature and a "kink" in M(H).[13] Because the metamagnetic transition is the magnetic field induced transition from an antiferromagnetically ordered state to a ferromagnetically ordered state, it leads to a large change in the magnetization at the transition. Even though the characteristics of the transition in La_2CuO_4 are similar to those of the metamagnetic transition, the magnetization jump shown in the 170 K-M(H) curve of Fig. 2 is small($\sim 1.2 \times 10^{-3}$ μ_B/Cu), i.e., almost two orders of magnitude smaller than that expected in the transition to a simple ferromagnetic state. This may indicate that, if the transition is metamagnetic, the high field state is a complex ferromagnetic-like state. Alternatively, sample inhomogeneity may be important in that it could lead to only parts of the specimen undergoing the transition[14].

In conclusion, we observe a strong field dependence of the 3D ordering in $La_2CuO_{4-\delta}$ with $T_N \sim 235$ K at small fields. The overall behavior is suggestive of a metamagnetic transition induced by the magnetic field. Neutron diffraction experiments in the presence of an applied field could help to understand the H-T phase diagram we describe. Finally, we point out that like the IBM group[7] we have observed in sintered $La_2CuO_{4-\delta}$ powders a field dependence of the temperature dependent susceptibility that is not as distinctive as that shown in Fig. 1. We attribute this difference to a distribution of sample compositions throughout the sintered compact.

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13. The zero field intercept of magnetization extrapolated from the magnetization curve at high fields is not zero and a small hysteresis in the magnetization curve below T_N was found in $La_2CuO_{4-\delta}$, which additionally suggest the transition to be metamagnetic rather than a spin-flop transition.
14. To check for the possibility of a metamagnetic transition in CuO, which exhibits an antiferromagnetic ordering transition at 240 K and could be a part of $La_2CuO_{4-\delta}$ crystals as impurities, specially as an incluson, we measured magnetization curves of CuO crystals at various temperatures but did not find any field induced transition in CuO similar to that reported here.