Magnetic ordering and magnetization in the high-temperature superconductor $ErBa_2Cu_3O_{7-x}$

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(Received 11 August 1987)

Low-temperature heat-capacity, electrical resistivity, and magnetization measurements are performed to investigate the superconducting and magnetic behavior of $ErBa_2Cu_3O_{7-x}$. This compound exhibits a superconducting transition at 84 K as determined from zero resistance and Meissner effect. A well-defined magnetic transition is seen at 0.87 K from heat-capacity measurements. We deduce the coexistence of superconductivity and antiferromagnetic order in this compound; however, magnetization data show that the paramagnetism from Er^{3+} and superconductivity are independent, leading to indirect evidence of extremely anisotropic superconductivity in this class of compounds.

I. INTRODUCTION

The compounds $RBa_2Cu_3O_{7-x}$ (R = Y, La–Lu, except Ce, Pr, and Tb) are all found[1–5] to undergo a superconducting transition at almost the same temperature of about 90 K. Due to the presence of rare-earth elements, these materials are good candidates for studying the interplay between superconductivity and magnetism. Among them, $GdBa_2Cu_3O_{7-x}$ is the only reported[4,8,9] compound to order antiferromagnetically above 1.6 K. In the rare-earth metal ternary compounds which exhibit superconductivity and magnetism, erbium is the rare-earth metal most often involved in the phenomena of coexistence or reentrant superconductivity;[6] therefore, $ErBa_2Cu_3O_{7-x}$ was chosen for further investigation. Low-temperature neutron-diffraction and small-angle scattering studies[7] indicate that the Er moments in $ErBa_2Cu_3O_{7-x}$ become ordered in a two-dimensional array, where chains of spins are aligned ferromagnetically, while adjacent chains are coupled antiferromagnetically. In this work, we provide heat-capacity data to show the bulk property of magnetic ordering at low temperatures in $ErBa_2Cu_3O_{7-x}$. Furthermore, the magnetization data between 5 and 300 K are also presented to study the magnetic properties in the normal and superconducting states.

II. EXPERIMENTAL METHODS

The sample preparation and heat treatment have been described elsewhere.[5,7] Magnetization data were taken in a commercial SQUID magnetometer equipped with a 5.5-T superconducting solenoid and automatic temperature control. The temperature dependence of the magnetic susceptibility was measured by first cooling the sample in zero field down to 5 K and then measuring its magnetic moment up to 350 K at a constant magnetic field of 2 T. Low-temperature heat-capacity measurements were performed using a semiadiabatic heat-pulse-type ^3He calorimeter. Electrical resistivity was measured using a four-probe method with silver paint as an electrical contact between the sample and the leads. Temperatures below 1.2 K were obtained in a commercial dilution refrigerator system where the resistivity and ac susceptibility were measured to about 60 mK.

III. RESULTS AND DISCUSSION

Heat capacity of $ErBa_2Cu_3O_{7-x}$ was measured between 0.8 and 24 K. A plot of C/T vs T^2 is shown in Fig. 1. The high-temperature part ($T > 11$ K) shows a linear behavior, which is characteristic of the lattice contribution to the heat capacity. This also confirms that the magnetic order seen in neutron scattering experiments[7] at 12 K is due to an impurity. The upturn in C/T at low temperatures is a magnetic contribution from the superconducting oxide. The magnetic ordering peak at 0.87 K is clearly shown in the inset of the C vs T plot in the Fig. 1. This is consistent with the temperature where the magnetic peak intensity starts to rise in neutron scattering experiments.[7] Comparing $T_N = 2.24$ K for $GdBa_2Cu_3O_{7-x}$,[4,8,9] and our result of $T_N = 0.87$ K for $ErBa_2Cu_3O_{7-x}$, we observe that these ordering temperatures do not follow the ratio of the de Gennes factors (15.75 for Gd and 2.55 for Er). This is consistent with the insensitivity of T_c to substitution of magnetic rare-earth ions for Y in this class of compounds. Thus, we conclude that RKKY interactions are not the predominant mechanism giving rise to magnetic order in this system. Another probable interaction in a system which orders at such low temperatures is the dipole-dipole interaction. Redi and Anderson[10] showed that antiferromagnetism can result from such an interaction at about 1 K. The fact that other compounds in this system (for example, R = Tb, Ho, Tm,...) are reported not to order in this temperature range ($T > 0.5$ K) may be due to oth-

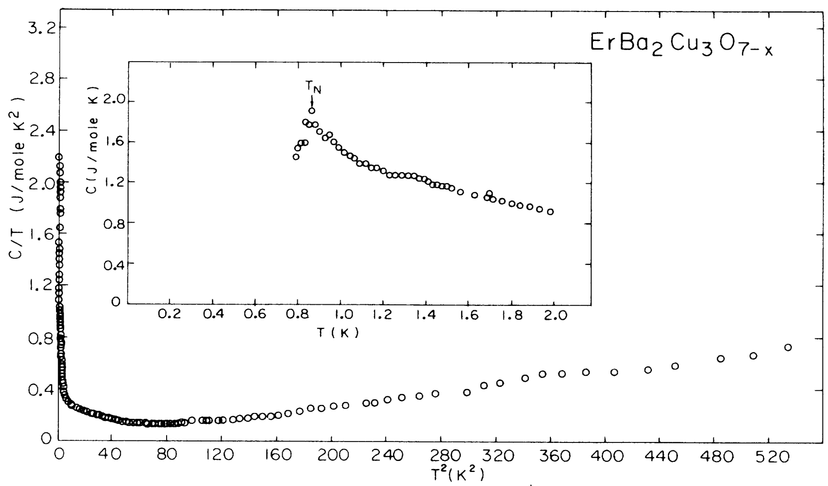

FIG. 1. A plot of C/T vs T^2 for $ErBa_2Cu_3O_{7-x}$. The inset shows the ordering feature plotted as C vs T for temperature between 0.8 and 2 K.

er origins, such as crystal-field effects. To further examine the ground state and associated entropy for magnetic order in Er, lower temperature and magnetic-field-dependent heat-capacity measurements are necessary. Moreover, similar to $GdBa_2Cu_3O_{7-x}$,[8] ac susceptibility and electrical resistivity were measured down to 60 mK showing no paramagnetic signals and reentrant behavior, respectively. Therefore, we conclude that the order is of an antiferromagnetic type which coexists with superconductivity below $T_N = 0.87$ K.

The superconducting transition temperature $T_c = 86$ K (midpoint) of $ErBa_2Cu_3O_{7-x}$, measured resistively, is shown in Fig. 2. This is consistent with a previous report[7] on Meissner effects in the same sample. We note that the onset and midpoint of the transition temperature are affected very little by an applied magnetic field at 2 T. On the other hand, the zero resistance temperature shifts from 84 to 60 K. At this point, the determination of the upper critical field will depend sensitively on the choice of the transition temperatures using the criteria of 90%, 50%, or 10% of a full resistive transition. The inverse molar magnetic susceptibility between 5 and 350 K at a constant field of 2 T is presented in the inset of Fig. 2. At $T > T_c$, we fit the data to the Curie-Weiss law

$$\chi_m = \chi_0 + C/(T+\Theta) \ .$$

The corresponding effective magnetic moment $\mu_{\text{eff}} = 9.67\mu_B$ is in good agreement with the Hund's rule ground state for the free ions Er^{3+}. The small and negative value of $\Theta = -10.8$ K then coincides with the low

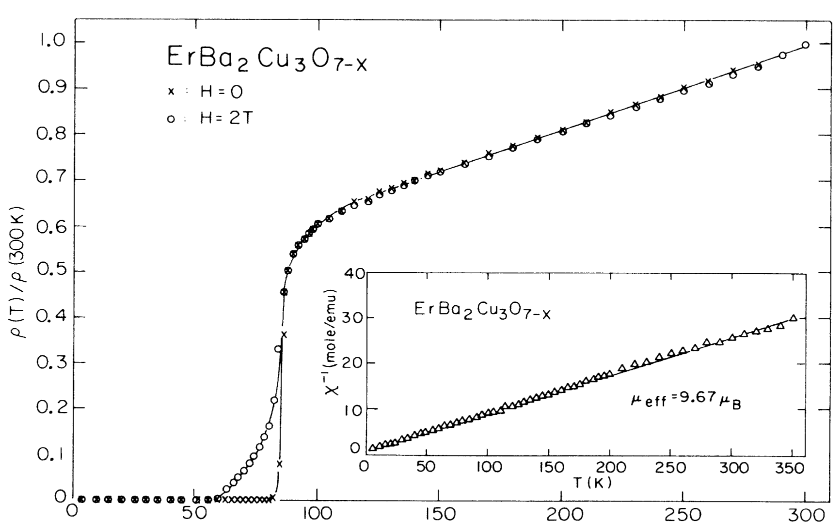

FIG. 2. Normalized resistivity vs temperature between 5 and 300 K at 0 and 2 T external magnetic field. The inset shows the inverse molar magnetic susceptibility vs temperature between 5 and 350 K.

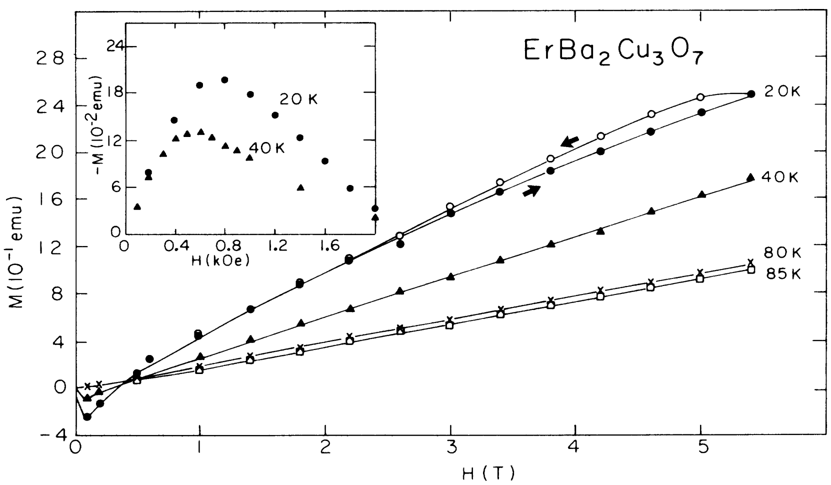

FIG. 3. The magnetization M vs applied field H between 0 and 5.4 T at 20, 40, 80, and 85 K. The inset shows the low-field magnetization at 20 and 40 K. The sample mass with irregular shape is 101.10 mg.

temperature antiferromagnetic transition. In general, when the applied field (2 T) is much smaller than the upper critical field ($\sim$100 T) a diamagnetic susceptibility is expected at temperature lower than T_c. An important feature here is that the magnetic susceptibility data can be fit to the Curie-Weiss law through the temperature range well below T_c, regardless of whether the sample is in the superconducting or normal state. Therefore, when using the magnetization versus field curves to determine the critical field, one should be very careful. This strongly indicates that the magnetic field induced paramagnetism in the Er subsystem is quite independent of superconductivity in this compound. This result may also imply that the superconducting electron pairs are localized in some directions (for example, in the Cu-O layers) which do not contain the rare-earth ions. We note that Tarascon *et al.*[4] destroyed superconductivity by annealing the sample under vacuum and reported that the magnetic susceptibility of several members of this series obeys the Curie-Weiss law from 10 to 300 K.

In Fig. 3 we show the magnetic field dependence of the magnetization up to 5.4 T at 20, 40, 80, and 85 K. At 80 and 85 K, the magnetization is proportional to the applied magnetic field. In contrast, at 20 and 40 K, the curves bend in low fields. These magnetization data illustrate a superposition of superconductive diamagnetism and high-field paramagnetism behavior. The low-field magnetization at 20 and 40 K is presented in the inset in Fig. 3. This indicates that $ErBa_2Cu_3O_{7-x}$ is a type-II superconductor.

In conclusion, we report the coexistence of superconductivity and antiferromagnetic order below 0.87 K in the high-T_c superconductor $ErBa_2Cu_3O_{7-x}$. Magnetic susceptibility shows a Curie-Weiss behavior through the temperature range from the normal state to the superconducting state. This implies that the high-field induced paramagnetism from the magnetic rare-earth ions and superconductivity are highly decoupled from one another.

ACKNOWLEDGMENTS

Ames Laboratory is operated for the U.S. Department of Energy by Iowa State University under Contract No. W-7405-ENG-82. Work at Ames was supported by The Office of Basic Energy Sciences, Grant No. WPAS-KC-02-02-02.

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