Competition between antiferromagnetism and superconductivity in $RBa_2Cu_3O_{6+x}$ (R = Lu, Tm)

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The antiferromagnet–superconductor transition in $LuBa_2Cu_3O_{6+x}$ single crystals, in which the concentration of current carriers increases continuously, has been studied experimentally. It was shown that the superconducting state competes with the three-dimensional magnetic ordering as the system undergoes a partial transition to an insulting state. © *1995 American Institute of Physics.*

A general property of all high-T_c cuprate superconductors is the presence in their structure of one or several CuO_2 planes, with which the superconducting state is ordinarily associated. According to the charge-transfer model,[1] the role of the other structural elements is to provide the required degree of electronic (hole) doping of the superconducting CuO_2 planes. The compounds $RBa_2Cu_3O_{6+x}$ are of special interest because of the unusual stepped dependence of the their physical properties on the oxygen content x. The function $T_c(x)$ exhibits two plateaus — at 60 K and 90 K — and sharp transition regions — from 0 to 60 K at $x \approx 0.4-0.5$ and from 60 K to 90 K at $x \approx 0.7-0.8$ (Refs. 1 and 2). These features are ordinarily explained[1] by the nonmonotonic dependence on the oxygen content of the carrier concentration in the CuO_2 planes.

The region of nucleation of superconductivity is of special significance for understanding the nature of high-T_c superconductors. Investigation of the evolution of the physical properties in the immediate vicinity of the threshold for the appearance of superconductivity can yield information about the mechanisms which determine the appearance of superconductivity and make it possible to check existing ideas about the correlation of T_c with the carrier concentration.

In the present study we performed accurate measurements of the anisotropic conductivity of $RBa_2Cu_3O_{6+x}$ (R = Lu, Tm) single crystals with oxygen content near the antiferromagnetic–superconductor (AFM–SC) boundary. The methods used to grow the crystals, to prepare the samples, and to measure the components of the resistivity are described in Ref. 3. The crystals were held at room temperature in an atmosphere of pure helium.

According to the ideas developed in Ref. 1, charge carriers do not enter the CuO_2 planes up to an oxygen content of $x \approx 0.4$. As x increases further, the carrier concentration n increases in a jump-like manner. It is thought that the appearance of superconductivity coincides with the appearance of carriers in the CuO_2 planes.[1] However, the presence of structure features on the $n(x)$ dependence must necessarily manifest itself in the kinetic characteristics. Specifically, one would expect a jump-like change in the conduc-

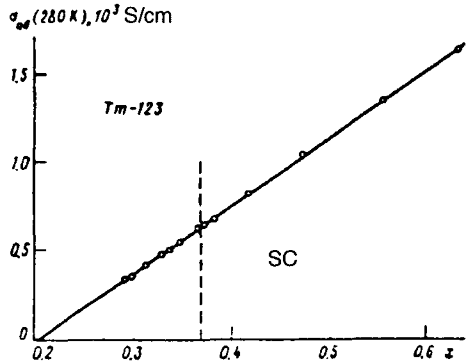

FIG. 1. σ_{ab}(280 K) of a $TmBa_2Cu_3O_{6+x}$ single crystal as a function of oxygen content x. The dashed line separates the regions of superconducting and nonsuperconducting compositions.

tivity σ and Hall coefficient R_H as x increases above the threshold concentration $\sim$0.4, which has not been observed experimentally.[3,4] Measurements performed on a $TmBa_2Cu_3O_{6+x}$ single crystal showed (Fig. 1) that for oxygen content in the range $0.29 \leqslant x \leqslant 0.60$ the conductivity σ_{ab} in the ab planes changes continuously and monotonically and does not exhibit significant features on going over to the superconducting compositions. We note that when the oxygen content x is increased from $\approx$0.37 to 0.5–0.6, which results in an increase of T_c from 0 to $\approx$55 K, the conductivity σ_{ab} (280 K) increases by only a factor of 2; see Fig. 1. The fact that superconductivity appears only when the carrier density n is sufficiently high indicates that at lower values of n (lower oxygen content) there exists a mechanism which suppresses superconductivity.

Experimental studies of nonsuperconducting compositions showed that a marked anomaly is observed in the interplanar resistivity ρ_c at the AFM ordering temperature T_N.[3] Figure 2 shows a typical temperature dependence $\rho_c(T)$ obtained for a $LuBa_2Cu_3O_{6+x}$ single crystal at $x \approx 0.29$. We see that as the temperature decreases below 170–180 K, the resistivity ρ_c increases in a jump-like manner; this indicates that the system undergoes a partial transition to an insulating state. The anomaly in the derivative $d\rho_c/dT$ has a negative λ-peak characteristic of phase transitions of this kind; see the inset in Fig. 2. The conductivity mechanisms in strongly correlated systems, which the compounds $RBa_2Cu_3O_{6+x}$ are (especially for low degrees of doping), have not been adequately studied. This circumstance makes it impossible to perform a detailed analysis of the resistivity anomaly. Nonetheless, the general conclusion can be drawn that the establishment of long-range magnetic order results in substantial changes in the energy spectrum of the quasiparticles which determine charge transport.

It is well known that short-range AFM order is present in CuO_2 planes in superconducting compositions,[5] and that the interaction with AFM correlations is considered to be a possible mechanism of high-T_c superconductivity. It is possible, however, that the suppression of superconductivity for $x<0.4$ is due to the establishment of *long-range* AFM order in the CuO_2 planes, which causes the system to undergo a partial transition to an unsulating state. More definite information about the interaction of antiferromagnetism

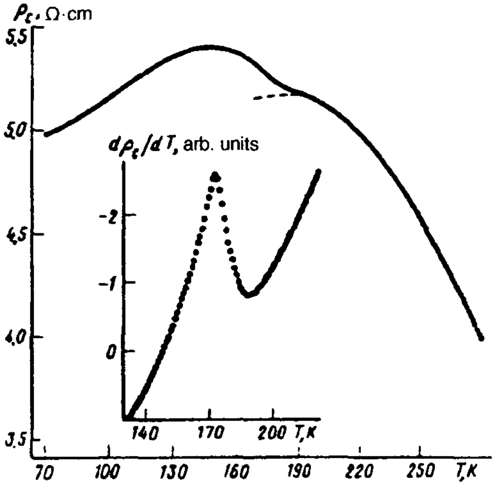

FIG. 2. Temperature dependence of the resistivity ρ_c of a $LuBa_2Cu_3O_{6+x}$ single crystal ($x \approx 0.29$). The dashed line represents the qualitative extrapolation of the high-temperature curve. Inset: Temperature dependence of the derivative $d\rho_c/dT$ in the region of the anomaly (for clarity, the coordinate axis is directed downward).

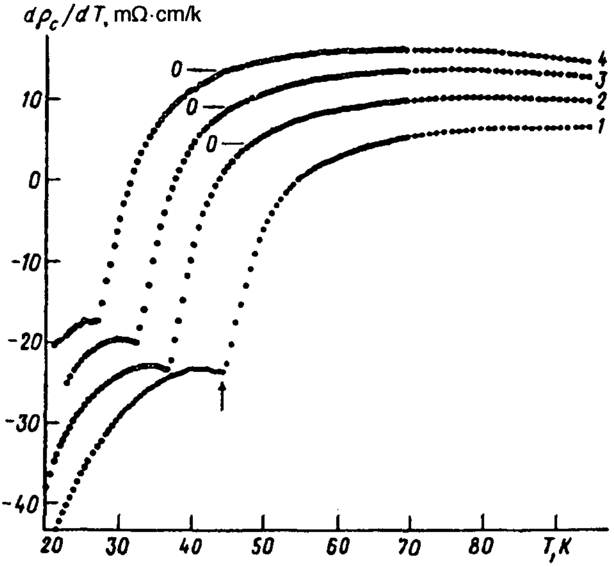

FIG. 3. Temperature dependences of the derivative $d\rho_c/dT$ for $LuBa_2Cu_3O_{6+x}$ ($x \approx 0.34$) single crystal. The series of measurements were performed immediately after quenching of the sample (●) and after different holding periods at room temperature (○) up to 5 days (curve *4*). The curves *2–4* are displaced for convenience (the zero point is indicated for each curve).

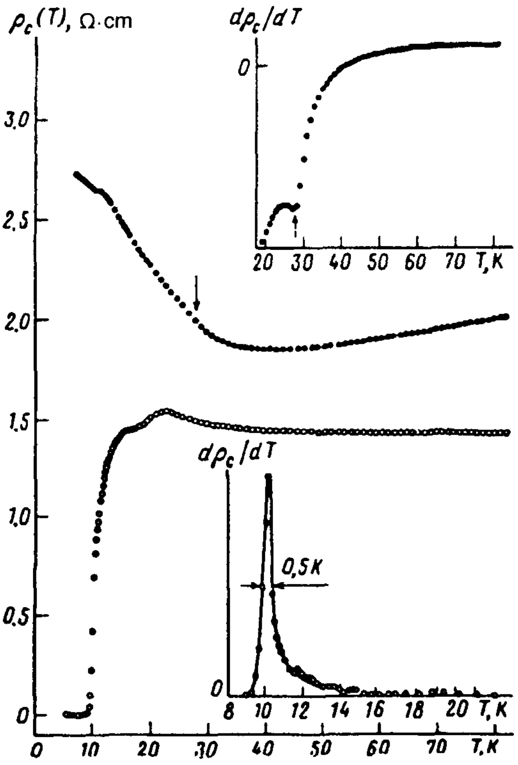

FIG. 4. Temperature dependences of the resistivity ρ_c of a $LuBa_2Cu_3O_{6+x}$ ($x \approx 0.36$) single crystal. The data were obtained immediately after quenching (●) and after a 5-day holding period at room temperature (○). Insets: Temperature dependences of the derivatives.

and superconductivity in $RBa_2Cu_3O_{6+x}$ can be obtained by investigating the particular features of the AFM–SC transition and the relative arrangement of the AFM and SC regions in the x–T phase diagram. The published data on the arrangement of the boundaries of the AFM and SC phases are contradictory; see, for example, Refs. 2, 6, and 7. The lack of detailed information stems from the fact that the standard methods of preparation and treatment of high-T_c samples, including changing the cationic and anionic stoichiometry, make it possible to perform only discrete measurements of their properties with a relatively large step. In this situation the low-temperature ordering of oxygen, which has been studied extensively, holds great promise.[8–10] The local ordering of oxygen, which occurs as a result of holding quenched samples at temperatures near room temperature, leads to a monotonic increase in the hole concentration in the CuO_2 planes.[8–10] This phenomenon provides a unique possibility of continuous variation of the properties of the sample.

Figure 3 shows the evolution of the properties of an $LuBa_2Cu_3O_{6+x}$ single crystal ($x \approx 0.34$) as it is held at room temperature. The increase in the carrier concentration in CuO_2 planes as a result of oxygen ordering results in the suppression of AFM ordering.

This is manifested as a monotonic shift of the negative λ-peak on the $d\rho_c/dT$ curve in the direction of low temperatures. Such a decrease of the Néel temperature with the samples held at low temperature was observed for all AFM compositions investigated. The shape of the anomaly does not change (see Fig. 3), indicating that the sample remains uniform.

The oxygen-ordering effects enabled us to observe how the phase transition from a three-dimensional antiferromagnetic ordering into the superconducting state occurs in $RBa_2Cu_3O_{6+x}$ single crystals with a continuous increase in the carrier concentration. For the $LuBa_2Cu_3O_{6+x}$ crystal an oxygen content near the boundary of the region of AFM compositions was chosen: $x \approx 0.36$. For this oxygen content the Néel temperature of the crystal in the quenched state was $T_N = 27-28$ K, as determined from the position of the peak on the $d\rho_c/dT$ curve (Fig. 4, filled dots). Subsequent holding of the crystal at room temperature gave rise to an AFM–SC transition, and after 5 days T_c reached 10 K (Fig. 4, open dots). We underscore the fact that all qualitative changes — decrease in T_N; change in the type of ground state, and increase of T_c up to 10 K — occur with a very small change in the transport characteristics. The total change in the resistivity ρ_c (50 K) is only 20–25% [the change in σ_{ab}(280 K) is even smaller — 12–15%]. Therefore, the suppression of long-range AFM order immediately leads to the appearance of superconductivity. This indicates that the competition between superconductivity and AFM order determines the threshold for the appearance of superconductivity.

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