Ferroelectric anomalies and superconductivity in metal–oxide compounds

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The ferroelectric anomalies in systems $YBa_2Cu_3O_{6+x}$ $(0 \leqslant x \leqslant 1)$ and $La_{2-x}Sr_xCuO_4$ $(0 \leqslant x \leqslant 0.4)$, which we have observed previously at frequencies of ~ 1 GHz, are studied. A correlation between the broadening of superconducting transitions and the microwave field resonance absorption frequencies observed by us is discussed.

In studying the rf properties of conducting metal–oxide compounds belonging to the class of high-T_c superconductors we have observed[1–3] an unusual phenomenon: the coexistence of ferroelectric anomalies at finite frequencies and a metallic conductivity. In this letter we report the results of an experimental study of the ferroelectric anomalies as they relate to the superconducing characteristics of the systems $YBa_2Cu_3O_{6+x}$ $(0 \leqslant x \leqslant 1)$, YBCO and $La_{2-x}Sr_xCuO_4$ $(0 \leqslant x \leqslant 0.4)$, LSCO.

The procedure for synthesizing the samples is described in Ref. 3, the oxygen content in YBCO was determined in Ref. 4, and the strontium content in LSCO was determined in Ref. 5. The dc conductivity $\sigma(0)$ was measured by a standard four-contact method. The effective conductivity $\sigma(\nu)$ was analyzed in the frequency range $10^8 \lesssim \nu \leqslant 10^{10}$ Hz by measuring the electromagnetic loss in the resonator.[1,2] As in Refs. 1–3, we measured $\sigma(\nu)$ at room temperature.

As was discussed in Refs. 1–3, the absorption of power due to the ferroelectric anomalies seems to be a resonance absorption at the frequencies ν_0 (since the excitation frequency of the resonator changes in a descrete manner, ν_0 is determined with an accuracy of about 20% at $\nu \sim 1$ GHz and with an accuracy of about 10% at $\nu \gtrsim 100$

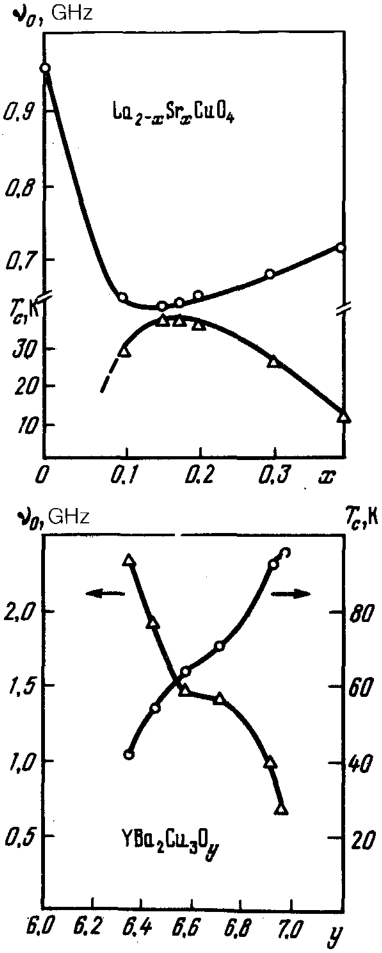

FIG. 1. Concentration dependences of the resonance frequencies $\nu_0(0)$ and T_c (Δ) in $La_{2-x}Sr_xCuO_4$ and $YBa_2Cu_3O_y$.

MHz). As can be seen in Fig. 1, the concentration dependence of ν_0^{-1} on the oxygen content in YBCO or strontium content in LSCO demonstrates an obvious correlation with the superconducting transition temperature T_c: The higher the ν_0 in the given system, the lower is the critical temperature T_c. This situation suggests that there may be a correlation between the ferroelectric anomalies and the large values of T_c.

The YBCO superconductors with $x \lesssim 0.9$ are characterized by highly diffuse transitions. The temperature at which the transition begins, T_c^+, and the temperature at which it ends, T_c^-, can be clearly identified. T_c^+, the temperature at which a superconducting pairing begins, can be defined experimentally as the temperature below which the resistivity in strong magnetic fields is appreciably higher than the resistivity at $H = 0$. T_c^- is the temperature at which the coherence in the Cooper pairs is established and hence the resistivity vanishes. Such experiments in 75-kOe magnetic fields were carried out for all samples of the YBCO series (a typical example of the tempera-

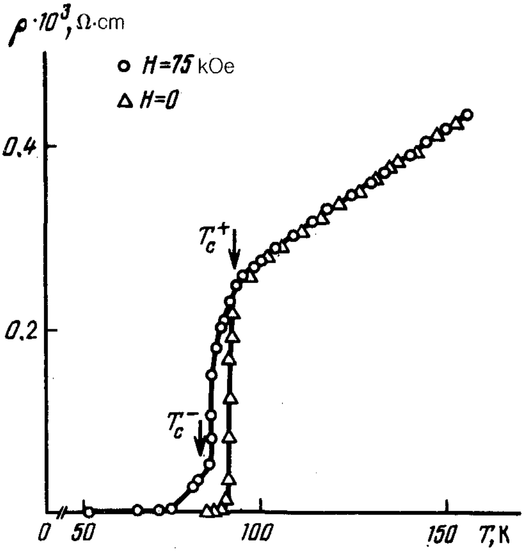

FIG. 2. Temperature dependence of the resistivity $\rho(T)$ in a magnetic field $H = 75$ kOe (O) and in the absence of a field (Δ).

ture dependence is shown in Fig. 2). The results for T_c^+, T_c^-, the transition width $\Delta T_c = T_c^+ - T_c^-$, and T_c, which was determined from the half-maximum of the transition, are shown in Fig. 3. A comparison of Fig. 3 and Fig. 1 shows that the concentration dependence of ν_0^{-1} correlates best with T_c^-. The link between ferroelectric anomalies and superconductivity, therefore, is probably determined by the effect of Cooper pairs on the establishment of coherence in the system, rather than by the effect of individual pairs on the energy characteristic.

Let us consider the relationship between the diffuse superconducting transitions

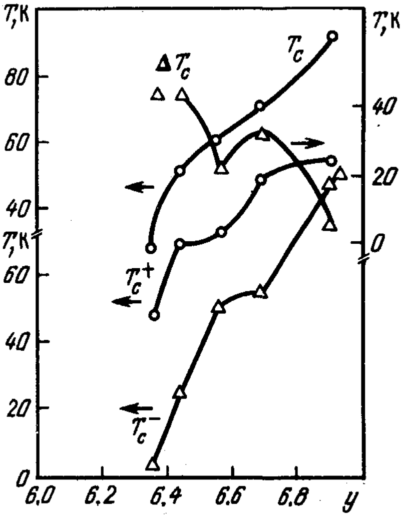

FIG. 3. Concentration dependences of T_c^+, T_c^-, ΔT_c, and T_c in $YBa_2Cu_3O_y$.

and the existence of a low-frequency dipole-active dynamics. As was discussed in Refs. 1–3 and 6, this phenomenon is attributable, in particular, to the presence of sufficiently large regions of radius $R \gg l$, where l is the mean-free path of electrons, with a roughly uniform dipole polarization. In view of this circumstance, these regions have an electrostatic potential $V(\mathbf{r},t)$ which fluctuates in space (with a scale dimension on the order of R) and in time (with a time scale of $\tau_c \gtrsim \nu_0^{-1}$). Let us assume, for simplicity, that $R \gtrsim \xi$, where ξ is the correlation length. The spatial fluctuations of the superconducting order parameter ψ in the polarized region can then be ignored and the temporal fluctuations can be described by a time-dependent linearized Ginzburg-Landau equation[7]

$$\partial \psi/\partial t + (2ieV/\hbar)\psi + [8(T-T_c)/\pi\hbar]\psi = 0 \tag{1}$$

($|T-T_c| \ll T_c$). To be specific, we assume that V is a Gaussian stray field with a correlation function $\langle V(t)V(0)\rangle = V_c^2 K(t/\tau_c) K(0) = 1$, where V_c is the fluctuation amplitude. For the order-parameter correlation function $G(t) = \langle\psi(t)\psi(0)\rangle$ we will then find from (1)

$$G(t) = G(0)\exp[-\frac{8(T-T_c)}{\pi\hbar}t - \frac{4e^2V_c^2}{\hbar^2}\int_0^t d\tau (t-\tau)K(\tau/\tau_c). \tag{2}$$

For $t \gg \tau_c$ we have

$$G(t) = G(0)\exp[-\frac{8(T-T_c^*)}{\pi\hbar}t], \tag{3}$$

where $T_c^* = T_c - (e^2V_c^2/2\hbar^2)\int_0^\infty dx K(x)$. For $t \ll \tau_c$ we have

$$G(t) = G(0)\exp[-\frac{8(T-T_c)}{\pi\hbar}t - \frac{2e^2V_c^2}{\hbar^2}t^2]. \tag{4}$$

Let us assume

$$|e|V_c\tau_c/\hbar \ll 1 \tag{5}$$

(weak fluctuations).

Asymptotic relation (3) will then be valid for all real times and by virtue of (3) the effect produced by fluctuations reduces to a shift of T_c by the amount

$$T_c - T_c^* \sim e^2V_c^2\tau_c/\hbar \ll \hbar/\tau_c \lesssim 2\pi\hbar\nu_0 \ll T_c, \tag{6}$$

consistent with the arguments based on the Anderson theorem[8]: T_c changes only slightly in the parameter $\hbar\nu_0/T_c$. This situation does not occur, in our view, in high-T_c superconductors, because ferroelectric anomalies have a strong effect on T_c, as we have indicated above.

In the case of strong (or slow) fluctuations

$$eV_c\tau_c/\hbar \gg 1 \tag{7}$$

we must use expression (4), which can be written in the form

$$G(t) = \int_{-\infty}^{\infty} d\Delta T_c \exp\left[- \frac{(\Delta T_c)^2}{2\sigma^2} - \frac{8(T - T_c - \Delta T_c)}{\pi \hbar} t \right] / (2\pi\sigma^2)^{1/2}, \qquad (8)$$

if we set $\sigma = \pi|e|V_c/2$, consistent with the broadening of the superconducting transition by an amount $\sim|e|V_c$. Disorder in this case is in fact static. Its effect on T_c (which manifests itself, however, in the transition broadening, rather than in the shift of T_c) may nonetheless be appreciable. There is no inconsistency here with the Anderson theorem,[8] since that theorem deals with a random potential which is not correlated on the order-parameter correlation length scale ($R_c \ll \xi$), and we are analyzing the case in which $R_c \gtrsim \xi$.

In connection iwth high-T_c superconductors, trivial reasons such as sample inhomogeneity are customarily used to explain broad transitions. According to our analysis, such transitions may be an intrinsic property of these systems. In this connection, we wish to recall an old problem involving broad superconducting transitions in alloys of titanium-based transition metals and in nitrides of the transition metals,[9] where the ferroelectric anomalies have also been detected.[6] In the case of these systems it was shown[9,10] that the large values of $\Delta T_c/T_c$ [$T_c^+ \sim 20$–35 K (Ref. 9)] are intrinsically characteristic of these materials and that the existence of ferroelectric anomalies identifies them with the high-T_c superconductors. In view of this circumstance, a report concerning the observation of superconductivity with $T_c \approx 40$ K in a rapidly cooled alloy merits attention.[11]

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