Fluctuation conductivity of Tl-Ba-Ca-Cu-O thin films

D. H. Kim and A. M. Goldman
School of Physics and Astronomy, University of Minnesota, Minneapolis, Minnesota 55455

J. H. Kang, K. E. Gray, and R. T. Kampwirth
Materials Science Division, Argonne National Laboratory, Argonne, Illinois 60439
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Measurements of the in-plane fluctuation-enhanced conductivity of c-axis oriented Tl-Ba-Ca-Cu-O thin films have been performed over the temperature range T_c to 240 K. The results were consistent with two-dimensional fluctuation theory and with a linear dependence of the normal resistivity on temperature down to $(T-T_c)/T_c \approx 0.03$. A crossover to three-dimensional fluctuations close to T_c was not found. The width of the superconducting transition appears to be a measure of a distance over which layers fluctuate in a correlated manner.

The planar structures and anisotropies of in-plane and out-of-plane components of resistivity and critical field H_{c2} of high-temperature superconductors[1,2] raise the issues of reduced dimensionality and anisotropy of the mechanism for superconductivity and of the order parameter. The study of the temperature dependence of the fluctuation-enhanced conductivity above T_c, or paraconductivity, is a way to probe the dimensionality of the order-parameter fluctuations. Most previous studies have concentrated on $YBa_2Cu_3O_{7-x}$ and the conclusions relating to the dimensionality include assertions that they are three dimensional (3D),[3,4] only two dimensional (2D),[5,6] 3D with a two-component order parameter,[7] or involve a crossover from 2D to 3D.[8] Measurements of resistivity and H_{c2} anisotropy for the Bi-Sr-Ca-Cu-O system[2] suggest 2D characteristics. In this article we report the results of a study of the paraconductivity of the Tl-Ba-Ca-Cu-O system which has even larger anisotropy in H_{c2} than the other materials.[9]

The main results of the Aslamazov-Larkin[10] theory for the paraconductivity σ' in different Euclidean dimensions D are

$$\sigma' = \begin{cases} \dfrac{e^2}{16\hbar d}\epsilon^{-1}, & D=2, \\ \dfrac{e^2}{32\hbar \xi(0)}\epsilon^{-1/2}, & D=3, \end{cases} \tag{1}$$

where $\xi(0)$ is the zero-temperature coherence length, d is the film thickness, and $\epsilon=(T-T_c)/T_c$ is the reduced temperature. For purposes of subsequent discussion, it is useful to define the parameters $C=e^2/16\hbar d$ and $C'=e^2/32\hbar\xi(0)$. The Maki-Thompson corrections,[11] which increase the apparent width of the transition and add parameters, have been ignored because they are probably absent in high-T_c superconductors as a consequence of the pair-breaking effect of strong inelastic electron scattering.[12]

Films of $TlBa_2Ca_2Cu_3O_9$ (1:2:2:3), $Tl_2Ba_2CaCu_2O_8$ (2:2:1:2), and $Tl_2Ba_2Ca_2Cu_3O_{10}$ (2:2:2:3) were deposited onto ZrO_2-9%Y_2O_3 substrates using a three-gun dc-sputtering system equipped with Tl, Cu, and Ba/Ca composite targets. Film thicknesses ranged from 0.3 to 0.7 μm. Post-deposition annealing treatments in flowing oxygen with the film and a sample of the Tl compound wrapped together in Au foil, were carried out at temperatures of 850–890 °C for about 5 to 30 min. Scanning electron micrographs of the films appear to indicate interconnected backbone structures. The chemical composition of films measured by energy dispersive x-ray analysis agreed well with structure determinations made using x-ray-diffraction analysis. The 1:2:2:3 and 2:2:1:2 films were single-phase, highly c-axis oriented structures, while for the case of the 2:2:2:3 film, although the majority phase was the 2:2:2:3 phase, there was a small 1:2:2:3 impurity phase.

Standard dc four-probe methods were used to determine $R(T)$ at measuring current densities of 1–3 A/cm^2. Silver print or silver-print coated evaporated gold electrodes were used to make contacts. The characteristics of the five samples are summarized in Table I, where T_{c0} is the temperature where resistivity vanishes and T_c^A is the temperature where $d^2\rho/dT^2=0$.

The establishment of the dimensionality of superconducting fluctuations from conductance measurements involves two things: precise determination of the normal-state resistivity, since the excess conductivity is the difference between the actual conductivity and the normal-state one, and determination of the actual transition temperature which may not be that at which $d^2\rho/dT^2=0$, or some other arbitrary criterion is satisfied. The fact that resistivities of high-temperature superconductors exhibit linear-temperature dependences for $T>2T_c$ should in principle provide a means of determining $\rho_n(T)$ by simple extrapolation of the high-temperature behavior down into the transition region. However, there is a small curvature in $\rho(T)$ of 1:2:2:3 and 2:2:2:3 samples at the higher temperatures, possibly due to the effect of the fluctuations themselves, or the presence of small impurity phases or grain boundaries, which makes this approach ambiguous. Consequently, the

TABLE I. Properties of Tl-Ba-Ca-Cu-O films. T_{c0} and T_c^A are the temperatures at which $\rho=0$, $d^2\rho/dT^2=0$. T_c is the mean-field transition temperature from Eq. (1). The quantity $C=e^2/16\hbar s$ and $C_{\rm exp}=e^2/16\hbar s_{\rm eff}$.

Sample	T_{c0} (K)	T_c^A (K)	T_c (K)	$C_{\rm exp}$ $(\Omega\,{\rm cm})^{-1}$	C $(\Omega\,{\rm cm})^{-1}$	$s_{\rm eff}$ (Å)	ρ (270 K) (mΩ cm)	$d\rho_n/dT$ ($\mu\Omega$ cm/K)
2:2:1:2	98.0	101	100.8	119	104	12.8	0.50	1.7
2:2:1:2	99.5	102	101.1	69.9	104	21.8	1.22	4.7
2:2:1:2	100.3	103	102.4	16.5	104	92.2	2.13	7.4
1:2:2:3	111.5	115.5	115.3	12.6	95.7	120.8	1.23	4.1
2:2:2:3	113.5	117	116.0	32.7	84.1	46.5	1.10	4.8

method used by Dubson *et al.*[3] and Ong *et al.*,[8] in which conductivity data from about 2 K above T_c to 240 K $> 2T_c$ were fit by

$$\sigma(T)=1/\rho_n(T)+\sigma'(T)\,, \tag{2}$$

was used. Here, $\rho_n(T)=aT+b$ is the normal-state resistivity. It is necessary to include σ' in the fit self-consistently as the fluctuations can contribute to $\sigma(T)$ over the entire temperature range of the fit. Fitting with the $D=3$ form of the theory was unsuccessful.

The range of temperatures over which the fit was carried out was varied to make sure that the parameters determined for the normal-state resistivity were independent of the contribution to Eq. (2) of σ'. The results were found to be more sensitive to the choice of the lowest temperatures than the highest of the range, and were range independent from 2 K above T_c to $2T_c$. This procedure ensured that the parameters determined from the fit, which employs only the 2D form of the theory, were not affected by either a crossover to the 3D form or critical behavior near T_c.

The parameters of five samples are summarized in Table I. The transition temperature determined from the fit T_c is close to T_c^A estimated from the condition $d^2\rho/dT^2=0$. Using the parameters describing the normal-state resistivity, the excess conductivity σ' is then determined from 240 K down to T_c. The 2D character of σ' for a representative sample (2:2:1:2*A*) can be clearly seen on the log-log plot of σ' vs ϵ in Fig. 1. The slopes of the excess conductivities of the other four Tl-Ba-Ca-Cu-O thin films are -1 over most of the region where $\epsilon > 0.03$ and increase towards 0 as T approaches T_c. This might be interpreted as a crossover to 3D near T_c since the slope would be -0.5 for 3D fluctuations. However, the shape of the plot near T_c is sensitive to the choice of T_c. Thus, an erroneous choice of T_c could result in incorrect conclusions regarding the dimensionality.

To avoid the above problem a procedure for determining the dimensionality of σ' in which T_c is not an adjustable parameter[13] was adapted for use in the case of temperature dependent $\rho_n(T)$. Equation (1), for the excess conductivity, is rewritten as

$$(\rho_n-\rho)/\rho=A\rho_n/\epsilon^\alpha\,, \tag{3}$$

where A is an appropriate constant, and α is either 1 or 0.5, depending on whether $D=2$ or 3. Then taking derivatives with respect to temperature on both sides of Eq. (3) and rearranging

$$\frac{1}{\rho^2}\frac{d\rho}{dT}-\frac{1}{\rho_n^2}\frac{d\rho_n}{dT}=\frac{\alpha}{T_c}(A)^{-1/\alpha}\left[\frac{\rho_n-\rho}{\rho\rho_n}\right]^{1+1/\alpha}. \tag{4}$$

The exponent α can be determined from the slope of a log-log plot of the left-hand side of Eq. (4) vs $\sigma'=(\rho_n-\rho)/\rho\rho_n$ and the transition temperature is not needed to obtain α in this scheme. A plot of this sort is shown in Fig. 2 for sample (2:2:1:2*A*). The slope should be 2 for 2D fluctuations ($\alpha=1$) and 3 for 3D fluctuations ($\alpha=0.5$). As can be clearly seen the slope is 2 over most of the region as expected from Fig. 1, and decreases to smaller values in the region $\epsilon<0.03$. This implies that the value of the exponent α increases from 1 only when T is very close to T_c, which is opposite to the behavior shown in Fig. 1 where T_c is an adjustable parameter. The inference of the existence of a dimensional crossover from an analysis similar to that in Fig. 1 may be a spurious consequence of having an additional adjustable parameter. From Fig. 2 there would appear to be no discernible dimensional crossover to 3D. However, the excess conductivity of Tl-Ba-Ca-Cu-O thin films does not exhibit a power-law dependence on ϵ in the temperature region $\epsilon<0.03$. Similar analyses, in which T_c is not a fitting pa-

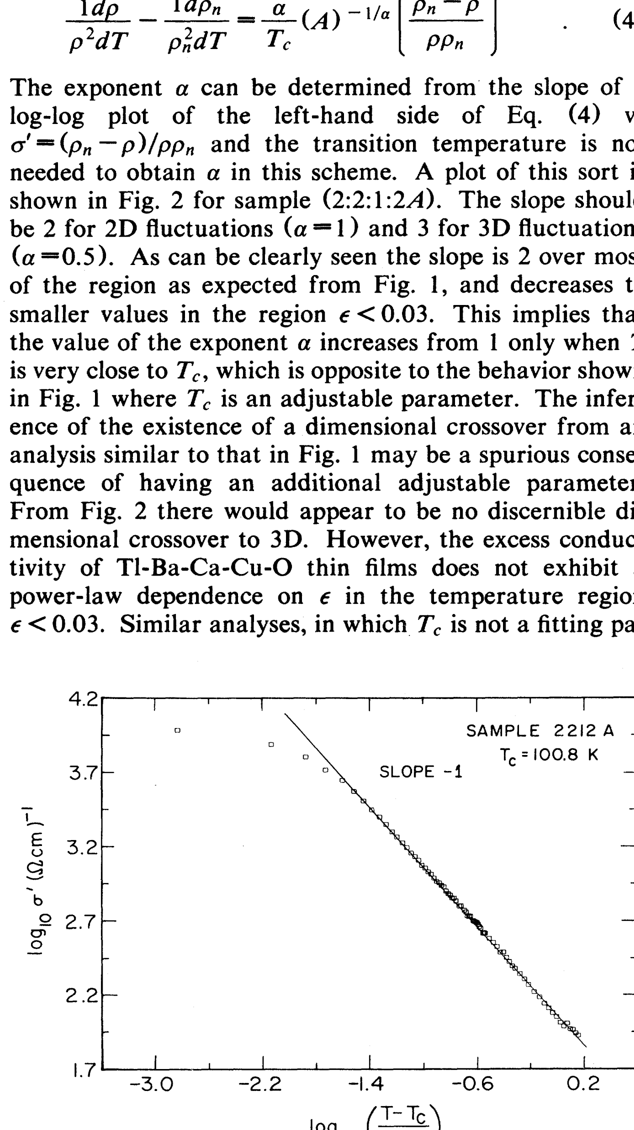

FIG. 1. Plot of excess conductivity σ' vs $\epsilon=(T-T_c)/T_c$ on a log-log scale for sample 2:2:1:2, where $T_c=100.8$ K. A slope of -1 corresponding to the 2D theory is observed over the regime $\epsilon>0.03$. The slope increases towards zero as T approaches T_c.

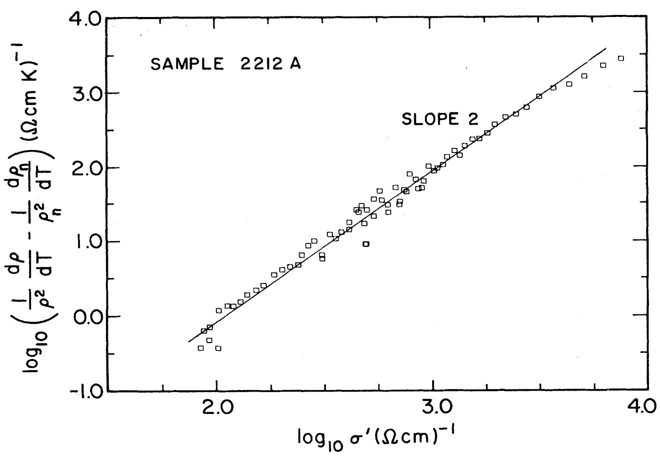

FIG. 2. Plot of the left-hand size of Eq. (4) as a function of excess conductivity σ' for sample 2:2:1:2A. A slope of two is observed over most of the region except for a very narrow region close to T_c, where the slope decreases [i.e., the exponent in Eq. (3) increases]. This result, which is independent of the fitting parameter T_c, is the opposite of the behavior shown in Fig. 1

rameter, have been carried out on data from other 2:2:1:2, 2:2:2:3, and 1:2:2:3 samples. The results are also consistent with 2D behavior down to $\epsilon \approx 0.03$ at which point the power-law dependence on ϵ breaks down. There is no way to determine whether the latter is an intrinsic result or a consequence of sample inhomogeneity.

It is useful to attempt a more quantitative comparison of experiment and theory, going beyond the identification of the dimensionality of the fluctuations. This can be done using the model of Lawrence and Doniach,[14] in which the paraconductivity parallel to the layers is given by

$$\sigma' = \frac{e^2}{16\hbar \epsilon s}\left[1+\left(\frac{2\xi_z}{s}\right)^2\right]^{-1/2}, \tag{5}$$

where s is the interlayer spacing and ξ_z is the coherence length perpendicular to the layers. For $\xi_z \ll s$, there is no observable 2D to 3D crossover. The fitting parameter C is then $e^2/16\hbar s$. Examination of Table I indicates that the values from the fit are smaller than estimates in which s is taken to be the distance between the centers of two adjacent Cu-O planes which are separated by a Tl-O plane. The CuO planes are presumably the sites of the superconducting electrons. The values of s used here were obtained from the structural studies and are 14.7, 18.1, and 15.9 Å for the 2:2:1:2, 2:2:2:3, and 1:2:2:3 compounds, respectively.[15]

The parameter C is actually a measure of the width of the resistive transition attributable to fluctuations. It is remarkable that it is smaller than the theoretical prediction. One possible cause of this may be the microscopic morphology of the films. When the conductivities are calculated from measured resistances the actual macroscopic dimensions of the films are used. On the other hand, scanning electron microscope photographs show an interconnected backbone structure. It is conceivable that the ratio of the effective cross section to effective length is smaller than the value obtained from the macroscopic dimensions of the film in such a manner as to result in an overestimate of the resistivity and an effective narrowing of the transition. This approach would imply an effective length of the conducting path of up to ten times the actual length, which is highly improbable. Furthermore, it should be noted that morphological inhomogeneities usually act to broaden transitions.

Another possibility is that the apparent narrowing of the transition is an additional intrinsic effect not contained in Ref. 14 in which it is assumed that the layers are coupled by the Josephson effect both above and below T_c. This explains the anisotropic critical fields below T_c. Above T_c, where the equilibrium order parameter in the layers vanishes, the theory may be invalid as the Josephson coupling constant is not really a fixed parameter above T_c, but must be treated self-consistently, as it is a function of the order parameter.

The smallness of the coherence length ξ_z (Ref. 9) within Ginzburg-Landau theory implies that the correlation function of the order parameter decays rapidly in space. It does not necessarily suggest that the interlayer coupling is weak, as is generally assumed, but that the system can tolerate rapid spatial variations of the order parameter without resulting in the superconducting state becoming unstable. This follows from the fact that the gradient term in the free energy is of the form $\xi^2|\nabla\phi|^2$. The ability to withstand rapid spatial variations of the order parameter may be a manifestation of a stronger rather than a weaker correlation between layers. Under such a circumstance one might interpret the reduced width parameter C_{exp} as resulting from an effective thickness s_{eff} greater than a single-layer spacing resulting from the correlated fluctuations of several electronic layers. This correlation might be enhanced by out-of-plane scattering produced by disorder which might explain the rough correlation of s_{eff} with ρ, as shown in Table I. These conclusions are the opposite of the expectations of a recent microscopic generalization of Ref. 14.[16] They should be considered speculative as any macroscopic theory must be applied cautiously since ξ_z is estimated to be the order of atomic dimensions.[9]

In summary, the temperature dependence of the excess conductivities of highly c-axis oriented Tl-Ba-Ca-Cu-O thin films are well represented by 2D fluctuation theory over the temperature region from $T_c + 2$ K to $2T_c$. In contrast with results on layered structures,[17] a dimensional crossover from 2D to 3D very near T_c is not observed. The apparent narrowing of the transition relative to the predictions of the Lawrence-Doniach theory of layered superconductors, if not due to morphology or the lack of self-consistency in the theory, may be a consequence of the correlation of superconducting order over distances of several electronic layers.

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