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Thermodynamic fluctuations in the superconductor $Y_1Ba_2Cu_3O_{9-\delta}$: Evidence for three-dimensional superconductivity

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The rounding of resistivity near and above the superconductivity transition temperature T_c, observed in the $Y_1Ba_2Cu_3O_{9-\delta}$ superconductor, is found to be a manifestation of thermodynamic fluctuations. A quantitative agreement between the experiment and the three-dimensional (3D) Aslamazov-Larkin theory was achieved near T_c. Low-field measurements of the excess diamagnetism are also in agreement with the 3D theory of Prange. These results strongly suggest that superconductivity in high-T_c copper oxides ($Y_1Ba_2Cu_3O_{9-\delta}$) is basically 3D in nature.

The recent discovery of superconductivity in copper oxides has generated a great deal of experimental and theoretical interest.[1-5] The $Y_1Ba_2Cu_3O_{9-\delta}$ compounds are of particular importance from the point of view of a very high T_c, and from the fact that a well-characterized single-phase specimen can be made. This allows a systematic and fundamental probing of the nature of superconductivity in this new class of materials. In terms of the crystal structure and models of oxygen deficiency,[6-9] high-temperature superconductivity has been associated with a planar or linear-chain atomic arrangement of the structure, suggesting superconductivity of low-dimensional nature,[9-13] although high-T_c superconductivity is also expected in three-dimensional (3D) systems.[11]

Experimentally, the observed rounding of the resistivity above T_c has been interpreted frequently as a possible signature of the onset of a higher-temperature superconducting phase. In this Rapid Communication we point out that this effect can be described as a manifestation of thermodynamic superconducting fluctuations above T_c. Furthermore, we show that superconductivity in these materials is 3D-like rather than 2D or 1D, suggesting significant electronic coupling between the low-dimensional structure entities such as layers and chains containing oxygen atoms.

As a result of thermal fluctuations, there is a finite probability of Cooper-pair formation above T_c. This gives rise to an excess conductivity $\Delta\sigma$ for $T > T_c$. This effect has been studied in detail, in particular by Aslamazov and Larkin (AL).[14] They obtained the following:

$$\Delta\sigma_{3D} = \frac{e^2}{32\hbar}\frac{1}{\xi(0)}\epsilon^{-1/2} , \tag{1}$$

$$\Delta\sigma_{2D} = \frac{e^2}{16\hbar}\frac{1}{d}\epsilon^{-1} , \tag{2}$$

where ϵ and $\xi(0)$ are, respectively, the reduced temperature $(T - T_c)/T_c$, and the zero-temperature coherence length, and d is a characteristic length of the two-dimensional system. Quantitative agreement with this theory is well documented in the literature.[15] The quantity $\Delta\sigma/\sigma_0$ (where σ_0 is defined as the normal state conductivity at 294 K) is an experimental measure of the magnitude of the fluctuation effect on conductivity. In a conventional 3D superconductor this effect is extremely difficult to observe because $\Delta\sigma/\sigma_0$ is of the order of 10^{-6}–10^{-8}. High-T_c oxides are generally characterized by a normal-state resistivity ranging from 0.5 to 3 mΩ cm at room temperature, and a short zero-temperature coherence length of the order of 20 Å. Therefore, in the Cu oxide system the expected values of $\Delta\sigma/\sigma_0$ are roughly 10^4 times larger than that of a conventional superconductor and should be readily observed. In the case of a lower-dimensional system, the effect should be even larger. Such thermodynamic fluctuations will also produce excess diamagnetism as a function of temperature and field, above T_c, and can serve as a complementary check of the conductivity results.[15]

The samples used in this investigation were prepared by mixing high-purity powders of Y_2O_3, BaO or $BaCO_3$, and CuO, followed by sintering around 950 °C and oxygen annealing at 650 °C. The samples were then slow cooled to room temperature. According to x-ray diffraction analysis, all samples are single phase (within 5%) with an orthorhombic unit cell. The oxygen deficiency is undetermined, with $\delta \simeq 2$. The samples were cut into spheres and bars for magnetization and resistivity measurements. The resistivity measurements were done using a standard dc four-probe method, capable of detecting changes of resistivity of 1 part in 10^4. We used current densities from 0.1 to 1 A/cm^2. The sample temperature is varied quasistatically, at a rate no greater than 0.1 K/min, near T_c, and measured with 10 mK resolution. Electrical contacts were were made by soldering fine copper wires to the sample with pure indium. The magnetization measurements were made in a SHE superconducting quantum-interference device magnetometer, with a resolution of $(2-3)\times10^{-7}$ emu, in magnetic fields ranging from 1 to 1000 Oe. Below T_c our samples show near complete diamagnetic shielding (85–100%) and a Meissner effect of 30–37% in an applied field of 1 Oe. If the porosity of the samples is taken into account, the Meissner effect can be as large as 60–80%. The dc susceptibility of three of the samples studied shows a Pauli-like behavior with

$$\chi(T > 2T_c) \simeq (0.5-1)\times10^{-6}.$$

However, sample B shows a temperature-dependent paramagnetic susceptibility.

All samples show a sharp resistivity transition around 91 K, with a 10%-to-90% transition width around 2 K. The room-temperature resistivities of the samples range from 1 to 2.7 mΩ cm. The error in the determination of the absolute resistivity value is $\pm 10\%$ mainly due to uncertainty in the correction terms related to sample geometry.

In Fig. 1 we show the resistivity as a function of temperature for three of the samples studied, normalized to their room-temperature resistivity value. All the samples are characterized by a linear temperature dependence of resistivity from at least $2T_c$ to $3.5T_c$, and by a smooth rounding off above T_c, characteristic of superconducting thermodynamic fluctuations. The excess conductivity $\Delta\sigma$ above T_c can be unambiguously determined by measuring the deviation from linearity, as extrapolated from the resistivity data above $2T_c$. In Fig. 2, we show the temperature dependence of the excess conductivity normalized to the room-temperature conductivity. In order to compare the experimental data with the theoretical expressions for 3D and 2D fluctuations, we plot $\ln(\Delta\sigma/\sigma_0)$ vs $\ln\epsilon$. Near T_c, it is clear that the experimental results, for all the samples studied agree with the AL 3D theory [Eq. (1)], both in magnitude and temperature dependence. We note that Eq. (1) has no arbitrary fitting parameters. We use $\xi(0)=22$ Å as obtained by Cava *et al.*,[4] and from our own dH_{c2}/dT data near the critical temperature. T_c was chosen at the midpoint of the transition. As suggested by the theory, our results near T_c are essentially sample independent. We used the average value of the sample resistivities at room temperature, $\rho(294)=2.04$ mΩ cm to calculate $\Delta\sigma/\sigma_0$ from Eqs. (1) and (2). For higher temperatures, the experimental data fall below the Al-3D prediction. The deviations from the theory are larger for samples showing a longer mean free path (lower resistivity). This phenomenon has been observed in 3D amorphous superconductors[16,17] and was interpreted as a short-wavelength cutoff in the fluctuation spectrum. The need for a high-q cutoff can be understood from the following expression for $\Delta\sigma$:

$$\Delta\sigma=\frac{e^2\xi(0)^4}{6\pi\hbar\epsilon^3}\int_0^\infty\frac{q^4dq}{[1+\xi(0)^2q^2/\epsilon]^3} . \tag{3}$$

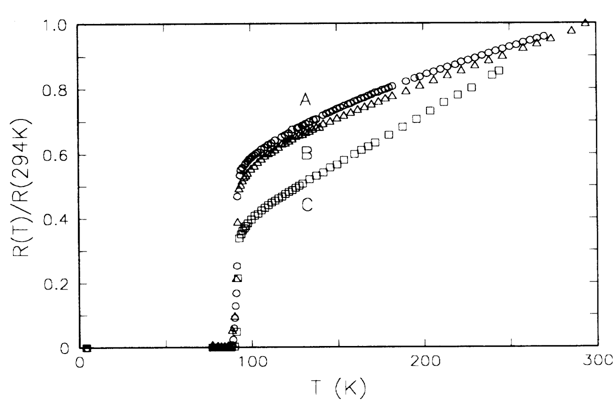

FIG. 1. Normalized resistivity vs temperature for three of our samples with different resistivities: $\rho_A=2.7$ mΩ cm, $\rho_B=2.4$ mΩ cm, and $\rho_C=2.1$ mΩ cm at 294 K.

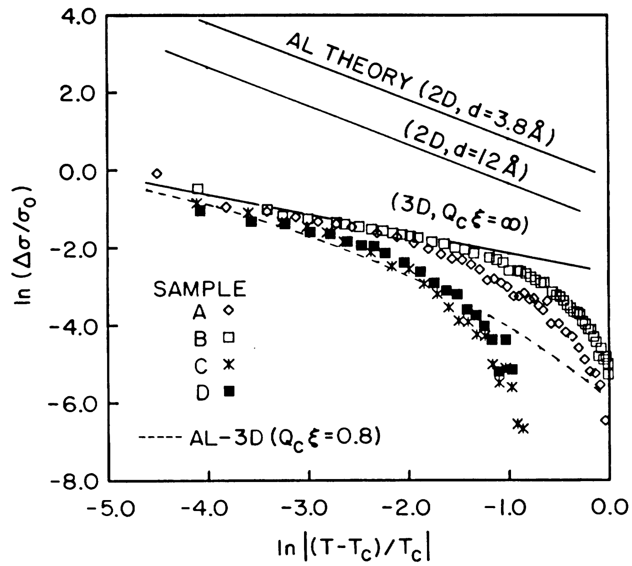

FIG. 2. Temperature dependence of the excess conductivity [$\ln(\Delta\sigma/\sigma_0)$ vs $\ln\epsilon$]. The solid lines are the predictions of the AL theory for 3D and 2D cases. The dashed line refers to the modified theory with short-wavelength cutoff.

In the regime of small $\xi(0)$ or large ϵ the integrand in Eq. 3 introduces significant high-q contributions, leading to a breakdown of the slow-variation approximation of the Ginzburg-Landau (GL) theory. We phenomenologically introduce a cutoff Q_c to remove the short-wavelength fluctuations, for which the GL theory does not apply. In Fig. 2 we show the modified AL 3D theory taking into account this effect. The observed deviations from the AL 3D theory ($Q_c=\infty$) are qualitatively explained by this model. In order to compare our results with AL 2D fluctuation theory, we also plot Eq. (2) for $d=12$ and 3.8 Å, corresponding to the lattice constants of the orthorhombic unit cell in these materials. Notice that this d value is always smaller than the experimentally observed ξ. This means that the 2D behavior should be found throughout the temperature range investigated. It is clear from Fig. 2 that this is not the case. The same conclusions apply to the 1D case. In the case of single-crystal layered superconductors, it is expected to find 3D behavior near T_c, and a 3D-to-2D crossover at higher temperatures.[18] In terms of this model, our results near T_c agree with the theory only in the isotropic limit, as a result of polycrystallinity. Further work is needed in single-crystal samples, where anisotropy is known to exist.[19]

In Fig. 3 we show low-field measurements of the excess diamagnetism as a function of temperature, for an applied field of 300 and 1000 Oe. This measurement involves a greater error than the excess conductivity one, due to difficulty in accurately subtracting the background. The experimental data are compared with Prange's theory,[20] the equivalent of the Al 3D theory without cutoff. Notice that at higher temperatures, the data fall below the theoretical prediction, as was also found in the conductivity data. Again, the temperature dependence and the magnitude of the effect agree with the 3D model. In the 2D

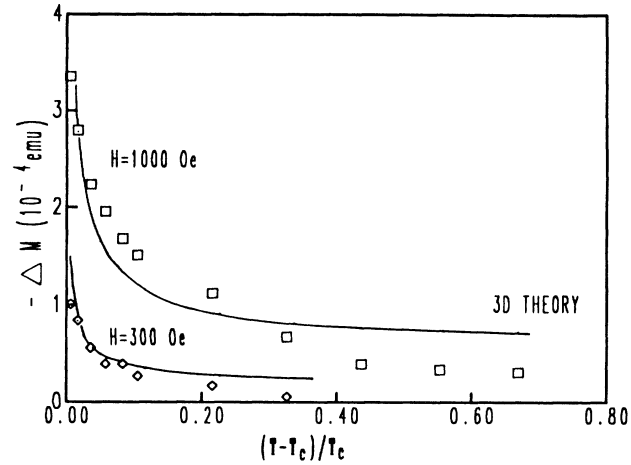

FIG. 3. Temperature dependence of the excess diamagnetism of sample A, in fields of 300 and 1000 Oe. The solid lines are the predictions of Prange's theory for the 3D case and both fields.

case, the excess diamagnetism is approximately $\xi(T)/d$ times larger than the 3D results.[21] As the coherence length diverges at T_c, the 2D contribution near T_c should be much larger than the 3D. We find no evidence for 2D or 1D behavior in our results.

We also performed preliminary studies of the influence of a magnetic field on the excess conductivity. The sample was oriented such that the magnetic field was perpendicular to the current direction. Our initial results show that in fields up to 4 T, the magnetoresistance is smaller than 0.1–0.2%, at a reduced temperature $\epsilon = 0.2$. This result is the same order of magnitude of that expected from fluctuation theory,[16] and rules out the possible existence of a higher-T_c phase. In the latter case, a large positive magnetoresistance would be expected. Further experiments with improved temperature stability and higher fields are planned.

In spite of the quantitative agreement between experiment and the AL 3D theory near T_c, we are not in a position to completely rule out the effect of inhomogeneities above T_c. This effect should give an additional contribution to $\Delta\sigma$, apart from that coming from thermodynamic fluctuations. We believe that most of the excess conductivity observed in our samples can be adequately accounted for by the fluctuation theory.

The small deviations from the AL 3D theory observed in some of the samples very close to T_c ($\epsilon < 5 \times 10^{-3}$) could be an indication of slightly higher-T_c regions, with T_c of the order of 0.5 K above the bulk value. Our experimental data do not indicate any contribution to $\Delta\sigma$ from the Maki-Thompson term.[13] A similar situation was observed in 3D disordered superconductors. Although this work was mainly concerned with single-phase Ya-Ba-Cu oxide, we expect such fluctuation phenomena to occur in other systems; we have observed similar resistivity behavior in Gd-Ba-Cu oxide and Sm-Ba-Cu oxide.

In conclusion, the deviation from linearity observed in the resistivity curve above T_c, can be explained in terms of 3D thermodynamic fluctuations with a short-wavelength cutoff in the fluctuation spectrum. A quantitative agreement between the experiment and the AL 3D theory near T_c was achieved without any fitting parameters. Low-field measurements of the excess diamagnetism are also in agreement with the 3D theory, giving additional support to our conclusions.

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