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Thermodynamic fluctuations in the superconductor $Y_1Ba_2Cu_3O_{9-y}$: Evidence for two-dimensional superconductivity

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The electrical resistance and the magnetoresistance of the ceramic superconductor $Y_1Ba_2Cu_3O_{9-y}$ have been precisely measured in order to extract the temperature derivative and to analyze the fluctuation regimes. The logarithmic dependence at high temperature is followed near the transition by a two-dimensional fluctuation regime in agreement with Aslamazov-Larkin theory (critical exponent $\lambda_{2D}=1$), and a complex behavior below T_c. Our findings disprove the observation of a three-dimensional fluctuation regime near and above T_c.

The study of dimensionality effects in granular superconductors has a long history.[1] They differ from those found in conventional type-II superconductors because of the geometrical connectivity of the superconducting regions. The "percolation transition" at which the resistivity falls to zero has been studied already, and we have found a quasi-one-dimensional regime above the temperature T_R (Ref. 2) when the superconducting coherence length becomes shorter than the connectivity length. Below the temperature T_M, i.e., near the so-called "midpoint temperature," Ginley *et al.*[3] have pointed out dimensionality (or size) effects in terms of the ratio between the penetration depth and the layer thickness of superconducting shells. Dimensionality plays a role there since the "shells" are anisotropic due to the inherent crystalline (orthorhombic) structure.[4,5] Single-crystal measurements would show evidence for "dimensionality" effects in various resistivity or conductivity regimes,[2] but measurements of critical exponents can also provide information on the nature of the conductivity state, in particular near phase transitions.

In this respect, a recent paper by Freitas, Tsuei, and Plaskett[6] discusses fluctuations in a granular superconductor oxide compound ($Y_1Ba_2Cu_3O_{9-x}$) and presents some evidence for three-dimensional (3D) (super-) conductivity fluctuations above T_M.[6] Our study shows strong evidence for two-dimensional (2D) superconductivity fluctuations near and below T_M. Our data approach T_c much more closely. We also examine the magnetic field influence. In all cases, the number of data points taken is large and allows us to study the temperature derivative itself on a log-log scale. The interplay between conducting and resistive regions is delicate and emphasized.

Due to thermal fluctuations, Cooper pairs have a finite existence probability above the so-called "critical temperature" for superconductivity. This leads to an "excess conductivity" $\Delta\sigma=\sigma-\sigma_0$, where $\sigma_0=1/\rho_0$ is the normal-state conductivity at some "high temperature." Aslamazov and Larkin (AL) (Ref. 7) have obtained theoretical expressions, which can be written

$$\Delta\sigma_3=(e^2/32\hbar)\xi_0\varepsilon^{-1/2} , \tag{1}$$

$$\Delta\sigma_2=(e^2/16\hbar)d\varepsilon^{-1} , \tag{2}$$

for the 3D and 2D excess conductivity, respectively. In Eqs. (1) and (2), ε is the deviation from the critical temperature in reduced units. ξ_0 is the zero temperature coherence length, while d is a characteristic length of the 2D system. Notice that the "critical temperature" is undefined up to here. It is convenient to take as a rough guess for temperature T_c that at $d^2R/dT^2=0$. It is usually observed that $T_c\simeq T_M$, but T_c would be better determined as a free parameter in data fitting as has been done at other second-order phase transitions.[8-10] We have also presented elsewhere a discussion on the relation between T_c and T_M as a function of the applied field.[2] In principle, independent measurements of the superconductivity coherence length or of the energy-gap temperature dependence are necessary to define T_c.

To keep the interpretation of results in terms of fluctuations, it is clear that we call the temperature T_c that at which the macroscopic coherence length diverges, i.e., where critical fluctuations dominate the scattering process.[11]

Such fluctuations are known to exist below as well as above T_c just like at magnetic transitions. In a mean-field-like approximation, the appearance of a gap in the density of states implies different critical exponents below and above T_c.[12] However, correlated fluctuations (between spins at magnetic phase transitions, e.g., or of the wave functions considered to be the order parameter here) can be taken into account to calculate the contribution from the energy gap fluctuations, and its congruent effect on the number of conduction electrons temperature dependence near T_c.[13] In the case of magnetic transitions (and the argument is expected to apply here as well), it is observed that the same singularity occurs in the contribution from the number of conduction electrons and from the scattering mechanism by the relevant fluctuations below the critical temperature, but also above it, into the electrical resistivity temperature derivative. This does not imply that the same value for the critical exponent is found above and below T_c.[9] But it is then expected that a single critical exponent suffices to describe the anomaly in the excess electrical resistivity temperature derivative on both sides of T_c. Therefore, starting from the above formulae (1)-(2), we will investigate the behavior of the excess resistivity through a single power-law dependence:

$$\Delta\rho = B\varepsilon^{-\lambda} + C(T) \ , \tag{3}$$

where B is a (negative) critical amplitude, $C(T)$ a smooth temperature function, and $\varepsilon^{-\lambda}$ will determine whether the fluctuations are 1D, 2D, or 3D.

The samples have been obtained from the same batch as that used in previous investigations.[2] They are quasisingle phase and very porous. The oxygen deficiency is undetermined. The resistivity measurements were done using a standard dc four-probe method, capable of detecting changes in the electrical resistivity of 1 part in 5×10^{-7} Ω, with great care on eliminating spurious effects. Current density was 0.3 A/cm^2 (but was varied in order to observe any influence[14]). Quasistatic conditions were carefully obtained; the heating and cooling rates near T_c were, at most, of the order of 2 K/h. Temperature measurements are accurate with a few mK resolution over the whole range.

For the magnetoresistance measurements, the magnetic field (perpendicular to current direction) was applied below room temperature before cooling. The warming run was stopped at a moderate temperature above the superconducting onset temperature (and below a temperature of the order of 240 K, above which some aging effect was seen on other samples even in the absence of magnetic field). Then the field or the current was changed and another run started. Susceptibility measurements were made but are not reported here. They were not precise enough to bring any special argument for the following analysis on the nature of fluctuations near T_c.

All samples show similar behavior starting from "high temperature." A quasilinear law is followed by a sigmoid curve extending down to a "break" temperature T_S which is field dependent. We show our extensive data for such a "critical region" on Fig. 1, in the case of zero magnetic field and for 4150 G.

The "excess electrical resistivity" temperature derivative was obtained after subtracting a small "background" of the order of the high-temperature slope (10^{-5} Ω/K).

We have observed the behavior of R as a function of ε^{-1} and $\varepsilon^{-1/2}$ and noticed a change in the precision of the data fitting at $\varepsilon \simeq 10^{-2}$, just up to where AL theory has usually been verified,[6,7] but neither law gives a convincing, good fitting. Thus, in order to compare the experimental behavior to theoretical expressions (1) and (2), we plot the results on a log-log graph (Fig. 2). Similar analysis was made by Freitas *et al.*[6] We use their scale and data points for comparison. Our data run through theirs with a similar departure from linearity at high temperature, but without any substantial difference with respect to the amplitude (Fig. 2 inset). Equations (1) and (2) do not allow for free fitting parameters except T_c, which Freitas *et al.*[6] have chosen equal to T_M. This choice does not play an important role away from the critical region. However, the apparent disagreement between the amplitude of the "2D theory" and the data might be accidental. Such an amplitude depends on the normal-state conductivity, which is very anisotropic. The ratio between $\sigma_\parallel$ and $\sigma_\perp$ (i.e., within and perpendicular to the layers) is expected to be large (of the order of 100) indeed.[15,16] Freitas *et al.*[6] use the 294-K value (i.e., 2×10^{-3} Ω cm). To compare to 2D theoretical expressions, the value $\sigma_\parallel$, rather than σ_0, must be used. This leads to a scale shift of the order of $\ln(100/3) = 3.5$. Hence, a fitting of the data with the 3D line (slope $-\frac{1}{2}$) is as good (or as bad) as with the true 2D (AL) expression (slope -1). See Fig. 2.

If less emphasis is put on the amplitude than on the slope itself, then, taking into account data closer to the critical temperature, such a plot hardly shows whether fluctuations are 3D or 2D indeed (Fig. 2).

A much better examination of the critical fluctuations is to take the temperature derivative of (3) and to calculate λ from a log-log plot. This reduces the uncertainty in the "constants" and subtracted (or not) background. Such a plot is shown on Fig. 3 for the field-free case. One finds $\lambda = 1$ (slope -2) below $\varepsilon = 10^{-2}$, and $\lambda = 0$ (slope -1) above $\varepsilon = 10^{-2}$, since

$$\ln\left|\frac{d\Delta\rho}{dT}\right| \simeq -(\lambda+1)\ln\varepsilon \ , \tag{4}$$

assuming $\rho = a + bT + \Delta\rho$ and $\Delta\rho = R_0(1-(\varepsilon_0/\varepsilon)^\lambda)$. This indicates two-dimensional fluctuations closer to $T_M \simeq T_c = 90.51$ K and anisotropiclike behavior further away. Isotropy would give a slope of $\frac{3}{2}$.[17]

A 1D or 2D behavior would be expected throughout the

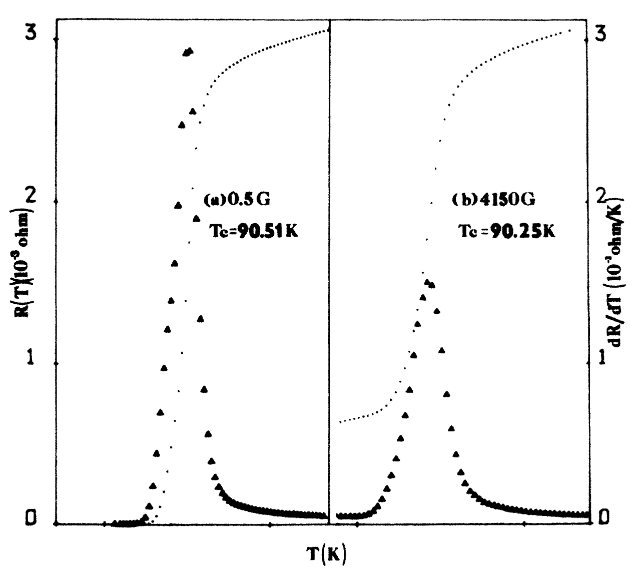

FIG. 1. Electrical resistivity (●) of granular ceramic oxide $Y_1Ba_2Cu_3O_{9-y}$ in the "critical region," and the temperature derivative (▲) of the electrical resistivity (a) without a magnetic field (earth's field = 0.5 G), (b) with 4150 G.

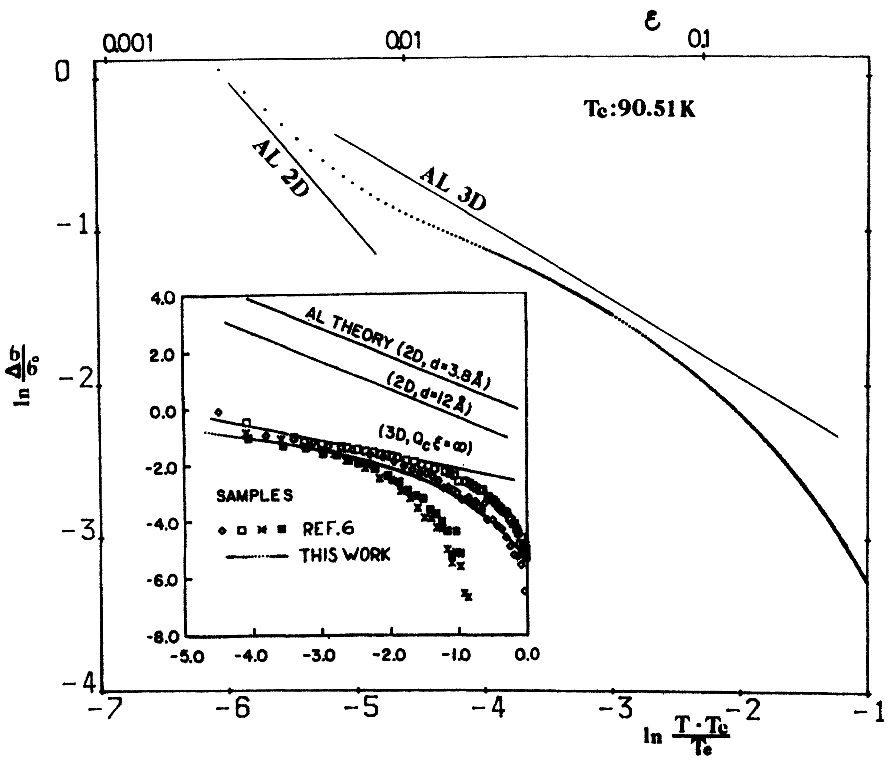

FIG. 2. Temperature dependence of the excess conductivity vs reduced temperature deviation ε with log-log scales. 2D and 3D AL-theory slope predictions are indicated. Inset: data and range from Ref. 6 (Freitas *et al.*, Ref. 6), and our data points.

whole temperature range if ξ was anisotropic, larger than a characteristic grain (or shell) linear dimension. The 3D-like behavior away from T_c in Ref. 6 could be due to polycrystallinity. Notice that the logarithmic behavior of ρ extends well above 180 K, but it is yet unexplained.

In Fig. 3, we also show the log-log plot of low-field (4150 G) measurements of the "excess resistivity" temperature derivative above (and below) T_c in order to observe the coupling between the magnetic field and the submicroscopic anisotropy. Indeed, the external magnetic field has a different and drastic influence on the carriers (Cooper pair) in the Cu-O layers or on the current parallel to the c axis. The background is also easily subtracted through the same (as for zero field) linear law. The magnetic field influence is neatly seen. A shift in T_M (or T_c) is of the order of 10^{-4} K/G, similar to that found at high "critical" field values. For this graph, we have $T_c = 90.25$ K, where $d^2R/dT^2 = 0$.

This low (4150 G) magnetic field reinforces the inherent 2D submicroscopic structure. If any exist, 3D fluctuations have to be reduced. It is seen that the 2D (AL) slope is also approached here as in the absence of field. At higher departure from T_c polycrystallinity dominates ($\lambda = 0$), and the slope $= -1$. The logarithmic behavior of ρ also extends as far as in the zero-field case (of course).

It is interesting to observe the change in regimes in the vicinity of T_c and at low temperature. The latter case has been studied in Ref. 2. Notice the very large slope ($\lambda > -7$) in the zero-field case when approaching the percolation transition. This was interpreted as an exponential law in Ref. 2, and has lead to 1D conductance channel interpretation.

In the presence of magnetic field, the transition is broadened due to the combined influence of the shift in T_c, lattice deformation, and the random distribution of anisotropic grains. This is marked by a normal flattening of the slope of $R(T)$ at T_c itself.

In conclusion, in opposition to the results of Freitas *et al.*,[6] we have proved by precise measurements near T_c,

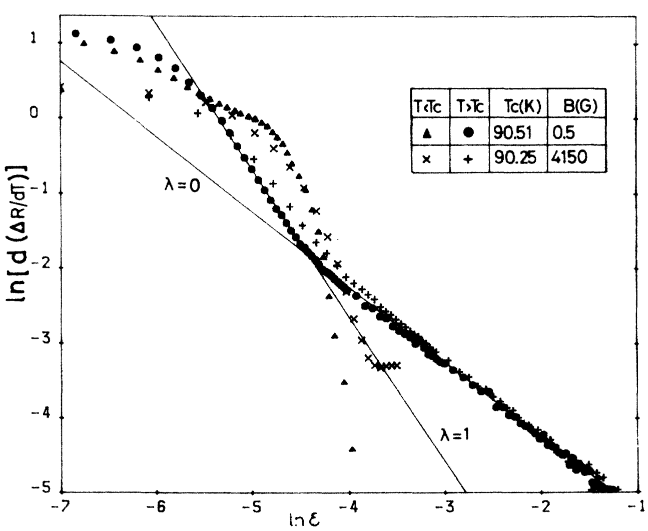

FIG. 3. Temperature derivative of the excess electrical resistivity in the presence of 0.5 and 4150 G magnetic field in the "critical region," on a log-log plot. The solid lines give the critical exponents $\lambda = 0$ and $\lambda = 1$. ΔR is expressed in mΩ and R(294 K) $= 5.72$ mΩ.

allowing for fine data analysis, that the deviation from linearity and the sigmoid behavior is due to 2D fluctuations. A quantitative comparison with 2D AL theory has been achieved near T_c. We have also observed a remarkable log-T dependence of $R(T)$ above the critical region over two orders of magnitude of ε.

The lack of matching between grains with *anisotropic* substructure is essential to understand the behavior of $R(T)$ in the "critical region," and to explain the magnetic field influence. Measurements on single crystals will better illuminate this conclusion.

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[17]It is well known that data fit around T_c are not infinitely precise when a univariate analysis is made because of the numerical derivative [cf. G. Sobotta, J. Phys. C **18**, 2065 (1985)]. However, this only occurs in Fig. 3 below $\varepsilon = 10^{-3}$, and we think that the uncertainty in our measurements is such that the slope changes in this figure are of physical origin.