Homogeneous and fractal behavior of superconducting fluctuations in the electrical resistivity of granular ceramic superconductors

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Experimental data show that the electrical resistivity temperature derivative diverges in a different manner in the granular ceramic superconductors $YBa_2Cu_3O_{7-y}$ and $Bi_{1.75}Pb_{0.25}Ca_2Sr_2Cu_3O_{10-y'}$. Critical exponents are extracted and explained. The dimensionality of anomalous superconductivity fluctuations is deduced. A homogeneous case and a self-similar (fractal) behavior can occur and are found. They correspond to different values of the superconductivity ξ_s and some percolation ξ_p coherence-length ratio. The results indicate that superconductivity percolation is achieved through surface connection rather than through path connection.

Behavior *universality* should be searched among a variety of different well-controlled experimental properties and systems to ascertain *change* in features related to a given process. We have examined granular superconducting ceramic transport properties in order to observe whether universal behavior can be seen in superconducting fluctuations, and deduce their dimensionality and nature if possible.

In this Brief Report we report precise measurements of the electrical resistivity ρ as a function of temperature taken on samples from the same batch for two different types of granular ceramic oxides ($YBa_2Cu_3O_{7-y}$ and $Bi_{1.75}Pb_{0.25}Ca_2Sr_2Cu_3O_{10-y'}$). Experimental details on the measurements and synthesis have been presented elsewhere.[1-4] As explained in Refs. 1-4, data have been taken in quasiequilibrium conditions: sweeping rate less than a few K/h, almost no thermal gradient, and very small current intensity (10^{-5} A/cm^2). Data are reproducible on a couple of runs (heating and cooling); sensibility is some mK on the temperature and corresponds to some nV on the resistance determination. Data resolution is thus such that temperature derivatives can be numerically taken and analyzed. The electrical resistivity ρ data are similar to those reported in the literature by other authors[5] for the same kind of compounds. The temperature dependence of $d\rho/dT$ (inset of Figs. 1 and 2) over a temperature range encompassing the so-called "critical temperature" T_c presents a λ peak, i.e., a sharp maximum which usually identifies T_c (Ref. 6) (even though some discussion may arise[7] whether such a value is identical to that derived from static properties).

It should be emphasized that the displayed data do not rely on the previous determination of a particular value for T_c, but data have to be compared at the same ϵ, not at the same T, thus also removing any calibration error on the temperature scale. Comparison to specific-heat data, e.g.,[8-10] to observe whether a Fisher-Langer[6] prediction holds on such superconductors, is not possible at this time. Severe criticisms on any conclusion resulting from such a comparison would bear on the fact that the compared samples are of a different chemical origin and received varied treatments indeed.

Actually, even with the high-precision data we nevertheless doubt, in contrast to other reports,[7] that we reach the true scaling regime. Intrinsic chemical and physical inhomogeneities do not allow confidence for data statistically analyzed below $\epsilon = 0.0010$. On the other hand, fluctuation-fluctuation correlations are expected to be extremely important. It could be thought that a Maki-Thompson (MT) theory[11,12] rather than a Aslamazov-Larkin (AL) theory[13] would describe the region close to T_c, but the MT prediction only leads to a log divergence near T_c [in two dimensions (2D) and to a very small contribution in 3D]. However, such a MT term is in fact more important than a AL term at *high temperature*. Therefore, the dimensionality of fluctuations found through a critical exponent analysis will only be em-

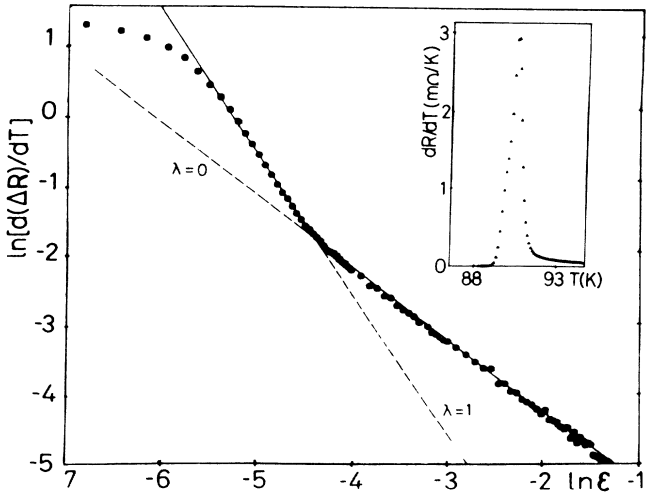

FIG. 1. Analysis of singular behavior of the excess resistivity (ΔR) above T_c for Y-Ba-Cu-O. Critical exponents are indicated. Inset: data of temperature derivative as taken.

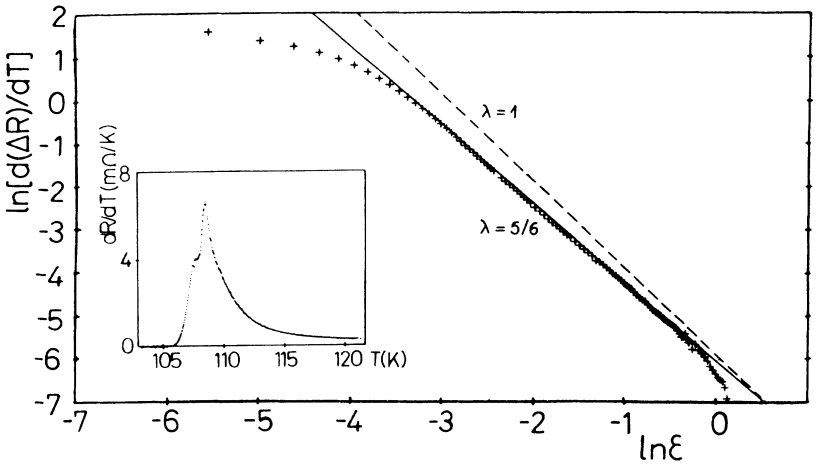

FIG. 2. Same as Fig. 1, but for $Bi_{1.75}Pb_{0.25}Ca_2Sr_2Cu_3O_{10-y'}$. A line of slope -2 (corresponding to a critical exponent $\lambda=1$) has been shown to contrast the results with those of Fig. 1. Inset: data of temperature derivative as taken.

phasized in the mean-field regime, but close to the Ginzburg temperature T_G. A crossover temperature may be expected to arise from several causes, e.g., it might be the Ginzburg temperature itself separating the mean-field and the scaling regimes, or the temperature at which different superconductivity mechanisms supersede each other in the mean-field regime, or even intrinsically result from data acquisition or analysis.

After a log-log analysis of

$$\frac{d}{dT}(\Delta R)\simeq A\epsilon^{-(\lambda+1)}, \tag{1}$$

where ΔR is an "excess resistance" (see below), values of λ are obtained. Values of critical exponents for ΔR are exactly those for the excess conductivity $\Delta\sigma$,[14] in the examined temperature range.

The excess resistance has been obtained from the data as follows. (The length and cross section of the samples are supposed to remain temperature independent, and the electrical current to uniformly flow through the bar shape sample). The slope near 250 K is estimated in both cases from a linear fit. To obtain an as-smooth-as-possible (but realistic) data fit, the slope is multiplied by a factor of C. This is equivalent to choosing σ_0. The statistical data analysis then leads to the smallest rms deviation within the largest possible temperature interval (i.e., as a function of the number of data points in the interval). We have found $C=0.9$ and 1.0 to be good values in these (Y-Ba-Cu-O)- and Bi(Pb)-based samples, respectively. The "background" is then subtracted from dR/dT. We consider that excellent straight-line fits (as obtained) are *a posteriori* positive indications on the fit quality and on the reliability of the critical exponent.

In AL theory,[13] the critical exponent λ of the (excess) resistivity is related to the superconductivity fluctuation dimensionality D by

$$\lambda=2-D/2. \tag{2}$$

Therefore, from values of slopes on Figs. 1 and 2, it is found that $D=4$ for this $YBa_2Cu_3O_{7-y}$ sample above T_c (between $\epsilon=2$ and 0.07) and $D=2$ for $YBa_2Cu_3O_{7-y}$ between $\epsilon=0.01$ and 0.003. On the other hand, $D=\frac{7}{3}$ for those $Bi_{1.75}Pb_{0.25}Cu_2Sr_2Cu_3O_{10-y'}$ samples between $\epsilon=1$ and 0.002.

A fluctuation dimension $D=2$ is easily understood from the *anisotropic* crystallographic structure of the Y-Ba-Cu-O compound. A $D=4$ value seems awkward, but from Eq. (2) it corresponds to $\lambda=0$. It is known from the singular function analysis that such a value indicates a "logarithmic singularity," e.g. Ref. 6. In fact, Lawrence and Doniach[15] have predicted such a logarithmic term for $\Delta\sigma$ (thus for $d\Delta\sigma/dT$ and $d\Delta R/dT$) as arising from pair-breaking mechanisms, e.g., twins, grain boundaries, and other intrinsic or extrinsic defects can thus account for such a behavior. It extends up to 180 K, as seen in Fig. 1.

The most anomalous value ($D=\frac{7}{3}$) in a Bi(Pb)-based compound can be understood when generalizing Eq. (2) to noninteger D as well, i.e., when allowing for a fractal description of a nonhomogeneous (granular) material. It has often been pointed out that the superconducting system is indeed a percolation network, i.e., superconductivity is restricted to regions where microscopic mechanisms are active in inducing the phenomenon.[16]

According to well-known results in a fractal description of percolation networks,[17,18] a structurally quasi-3D network may behave as a lower-dimensionality system from the *dynamical* point of view, e.g., in transport properties. The dynamical behavior should scale as the so-called fraction or spectral dimensionality which governs energy diffusion. The (length, and consequently, temperature) validity range of this description is usually unknown. However, the lower cutoff is connected to a length of the order of the mean interatomic spacing, but the maximum is as some correlation length ξ_p of the percolation network. This effect can evidently only apply if the superconductivity correlation length ξ_s is of the order of *or less* than ξ_p: for larger distance scales, the system has to appear homogeneous and fractal effects disappear.

It is interesting to point out that the simple condition $\xi_s<\xi_p$ for the observation of fracton contribution explains the difference between Y-Ba-Cu-O and $Bi_{1.75}(Pb)_{0.25}Ca_2Sr_2Cu_3O_{10-y'}$ compounds. On one hand, ξ_s

has to be temperature dependent, while in granular materials ξ_p should depend on the grain and pore size distribution. The length scale can be set also by the dimensions of oxygen-defective regions, i.e., by superconducting layers. Thus samples can have distinct behavior. The granularity is indeed quite different in both cases,[19,20] and the superconductivity coherence lengths as well.[21–23] It remains for other investigations to discuss how such results depend on sample preparations or whether they are intrinsic properties.

The most striking results of the above analysis are thus (i) the validity of AL law but with noninteger dimensionality as well (where the fractal dimension D_F replaces D), and (ii) the dimension of the fractal network. Notice that the value $D=\frac{7}{3}$ found above is "nothing more" than $1+\frac{4}{3}$, where $\frac{4}{3}$ is known as the fractal dimension for percolating clusters.[17,18,24] Therefore, we emphasize that the relevant phase-space dimensionality of the fluctuation spectrum of such superconductors is markedly higher by a single unit than in usual cases. Such a result indicates that the percolation backbone is *surfacelike* rather than *pathlike* (Fig. 3). This confirms the recent report[10] based on specific-heat data.

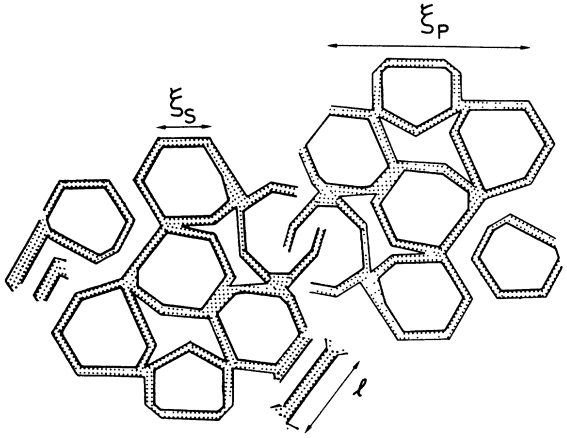

FIG. 3. Sketch of the superconductivity mechanisms in granular high critical temperature superconducting ceramics showing that for layerlike superconductivity the percolation ξ_p and superconductivity ξ_s coherence lengths can be similar (and of the order of a mean free path l). No scale is shown to emphasize the fractal nature of the granular system.

It is of interest to observe that ξ_p is apparently *not* of a macroscopic scale (e.g., the grain size): $\xi_s(0)\simeq 1.0$ nm, and we must have $\xi_s(T_c)\simeq\xi_p$. Thus ξ_p seems smaller than the "coherence-length backbone" of porous materials containing fluids or of polymer systems.[24] Thus we conjecture that ξ_p is more like the physical (rather than truly geometrical) measure of the backbone, hence it seems to be the carrier mean free path here. It is indeed easy to convince oneself that the latter enters through an exponential factor at the same level as the usual ξ_p in the electrical-resistivity expression. The smallest of either the mean free path l or the (usual geometrical) coherence length give the largest possible scale for the percolation network, and thus controls the type of singular behavior by monitoring the correlation function contribution to the energy integral defining $\rho(T)$.

Furthermore, it has been conjectured by others[25,26] that, due to chemical inhomogeneities and intrinsic anisotropic microstructures, the conductivity occurs mainly along shell-like structures (Fig. 3)—not necessarily only at grain interfaces. On the other hand, due to the greater anisotropy of the Bi(Pb) based ceramics, grain-conducting plane matching is harder. Therefore, we can understand that l rather than ξ_p is the quantity to which ξ_s has to be compared and both (l and ξ_s) are of the same order of magnitude, while their ratio controls this "extrinsic fractality" beyond the naturally occurring aggregated structure.

These arguments together with the randomness in the grain shape and/or size distribution can also finally serve to argue that the scaling regime is hardly obtained in these ceramics.

In conclusion, we have confirmed the occurrence of the strong enhancement of the conductivity fluctuations in *different* granular ceramic oxide superconductors. A similar (or "universal") behavior exists between superconductivity fluctuation contribution in different ceramic systems. A pair-breaking 3D regime followed by a 2D fluctuation regime is confirmed in YBaCuO. On the other hand, 2D fractal or homogeneous regimes can be found (even far away from the critical temperature). It appears that the fluctuation dimensionality and nature are controlled by microscopic sources over a very large temperature range.

Note added. A recent paper by Char and Kapitulnik[27] studying the fluctuation conductivity of inhomogeneous (*classical*) superconductors shows that the relevant dimensionality for the fluctuations spectrum can be the spectral dimensionality of the self-similar regime. The superposition of homogeneous and self-similar regimes in that case leads to good agreement with measurements on AlGe thin films that exhibit percolation structure. The present work has thus shown that the Char and Kapitulnik argument applies to *high-temperature superconductors* as well. Notice that Char and Kapitulnik also emphasize the competition between AL and MT terms and arrive at the same conclusion as ours (pair breaking) on the logarithmic behavior.

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