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Scattering of normal excitations by superconducting fluctuations in single-crystal and granular copper oxide superconductors

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By using an unified empirical procedure, the intrinsic paraconductivity in the *ab* plane, $\Delta\sigma^i$, is extracted from different available data for resistivity rounding in single-crystal and granular $YBa_2Cu_3O_{7-\delta}$ samples, and from data in $Bi_{1.5}Pb_{0.5}Sr_2Ca_2Cu_3O_y$ granular samples. These results strongly suggest that both high-temperature superconductor systems will have relatively strong pair-breaking effects, and that $\Delta\sigma^i$ may be explained by direct superconducting order-parameter-fluctuation effects alone.

Due mainly to the shortness of their superconducting correlation-length amplitudes of the order of the interatomic distances, the copper oxide superconductors will present strong thermodynamic fluctuations of the superconducting order parameter (SCOPF).[1] In particular, it is now widely accepted that these intrinsic SCOPF effects are the chief cause of the observed rounding of the critical behavior, near the superconducting transition, of observables such as the heat capacity,[2] the electrical resistivity,[3] or the magnetic susceptibility.[4] However, various central issues of SCOPF in these so-called high-temperature superconductors (HTSC's) are still open. One of them concerns the scattering of normal excitations by SCOPF above the mean-field-like superconducting transition temperature, T_{c0}, an intrinsic mechanism first proposed by Maki for isotropic BCS superconductors.[5,6] The intrinsic rounding of the resistivity above T_{c0}, characterized by the so-called intrinsic excess conductivity or paraconductivity,[3] $\Delta\sigma^i$, should, therefore, contain two different contributions: One is directly associated with SCOPF, $\Delta\sigma_F$, and the other, called "anomalous" and noted here as $\Delta\sigma_A$, is due to the scattering of normal excitations by SCOPF. By assuming, as usual,[6] that both contributions are linearly additive, we may write

$$\Delta\sigma^i = \Delta\sigma_F + \Delta\sigma_A \ . \qquad (1)$$

In contrast with $\Delta\sigma_F$, whose importance in HTSC is a direct consequence of the shortness of the order-parameter correlation length, $\Delta\sigma_A$ will crucially depend on the type of interactions giving rise to the superconductivity and on the pair-breaking effects of each material. Moreover, whereas for *layered superconductors* $\Delta\sigma_F$ was calculated twenty years ago by Lawrence and Doniach (LD),[7] $\Delta\sigma_A$ in *layered superconductors* has been calculated only recently, on the grounds of the BCS-like theory for *s*-wave pairing superconductors.[8,9] These mean-field results for $\Delta\sigma_A$ have been compared in detail with the magnetoresistance measurements in $YBa_2Cu_3O_{7-\delta}$ samples by Matsuda and co-workers[10,11] and by Hikita and Suzuki,[12,13] but various important ambiguities remain. For instance, in their first papers[10,12] these authors conclude that $\Delta\sigma_A$ contributes negligibly to the measured excess conductivity and that, therefore, the $YBa_2Cu_3O_{7-\delta}$ system has relatively strong pair-breaking effects. However, some time later both groups proposed just the opposite conclusion:[11,13] relatively weak pair-breaking effects and important $\Delta\sigma_A$ contributions. This leads to SCOPF being two dimensional (2D) in the mean-field region (MFR) in $YBa_2Cu_3O_{7-\delta}$ materials, which strongly disagrees with other paraconductivity analyses.[3,14] Further, their proposal for $\Delta\sigma_A$ disagrees with our preliminary analysis of this contribution in granular $YBa_2Cu_3O_{7-\delta}$ compounds.[15] (Other groups[16,17] have analyzed $\Delta\sigma_A$ in copper oxide superconductors by using the conventional 3D or 2D Maki-Thompson[5,6] approach for isotropic *s*-wave BCS superconductors, but such an approach is not well adapted to these layered materials.)

In this paper, the intrinsic $\Delta\sigma^i$ is first extracted from different available resistivity measurements in both single-crystal and granular $YBa_2Cu_3O_{7-\delta}$ samples, and from experimental results in $Bi_{1.5}Pb_{0.5}Sr_2Ca_2Cu_3O_y$ granular samples (with $T_{c0} \simeq 110$ K). The $\Delta\sigma^i$ extraction is a central issue of this paper. In fact, we will show that some of the ambiguities and contradictory results on $\Delta\sigma^i$ published so far are associated to a large extent with an inadequate $\Delta\sigma^i$ extraction. Then, both the amplitude and the reduced-temperature behavior of $\Delta\sigma^i$ are compared with the existing theoretical approaches for $\Delta\sigma_F$ and $\Delta\sigma_A$ in layered *s*-wave superconductors. Note here that our choice of the $Bi_{1.5}Pb_{0.5}Sr_2Ca_2Cu_3O_y$ system as a complement for the very good available resistivity results in $YBa_2Cu_3O_{7-\delta}$ samples was mainly motivated by the different temperature behavior of $\Delta\sigma$ in both systems, as first observed by Vidal *et al.*,[18] and later confirmed by several groups.[19] Our analysis strongly suggests that there exists a simple and unified scenario where $\Delta\sigma^i$ in copper oxide superconductors may be fairly well explained by direct ($\Delta\sigma_F$) SCOPF effects alone and that these materials will have relatively strong pair-breaking effects (i.e., a negligible $\Delta\sigma_A$ contribution). These are central issues of the HTSC physics. Finally, for completeness we will use our $\Delta\sigma^i$ experimental results to briefly comment on some other recent theoretical mean-field results on $\Delta\sigma^i$ which neglect these $\Delta\sigma_A$ contributions.[20,21]

A meaningful comparison between the theory and the experiments is possible only if, contrary to the process used, for instance, in Refs. 11, 13, 16, 17, or 22, the experimental $\Delta\sigma^i$ is extracted from the measured resistivity,

ρ^M, *independently* of any particular theoretical expression for $\Delta\sigma_F$ or $\Delta\sigma_A$ in Eq. (1). Our treatment may be summarized through the $\Delta\sigma^i$ definition itself,

$$\Delta\sigma^i(\epsilon)\equiv\frac{1}{\rho^i(\epsilon)}-\frac{1}{\rho_B^i(\epsilon)}\,, \tag{2}$$

where ρ^i and ρ_B^i are, respectively, the intrinsic total and the intrinsic background resistivities in the *ab* plane, and $\epsilon\equiv(T-T_{c0})/T_{c0}$ is the mean-field reduced temperature, as follows:

Intrinsic and measured resistivities. All the samples analyzed here are single phase within 4%. In addition, we have recently shown that the observed resistivity, and in particular its rounding above T_{c0}, is little affected by possible small critical-temperature inhomogeneities, associated for instance with local sample strain or oxygen-content variations.[23] We will thus suppose that the differences between intrinsic and measured resistivities above T_{c0} are mainly due to structural inhomogeneities at scales larger than the correlation-length amplitude (here with its geometrical meaning). In order to take into account these inhomogeneities, which may be associated with orientational mismatch between sample domains (for instance, grains, untwinned regions), or with the presence of nonsuperconducting domains (for instance, due to compositional inhomogeneities or sample porosity) we will use the empirical picture proposed in Ref. 3. In the absence of an applied magnetic field, and above T_{c0}, the basic relations are[3]

$$\rho^M(T)=\frac{1}{p}\rho^i(T)+\rho_c\,, \tag{3}$$

and

$$\rho_B^i=\frac{C_1^i}{T}+C_2^iT\,. \tag{4}$$

The notation is the same as in Ref. 3. In particular, the i (instead of g in Ref. 3), M, and B stand, respectively, for intrinsic (corresponding to an ideal single crystal in the *ab* plane), measured, and background, and C_1^i and C_2^i are the Anderson-Zou (AZ) background coefficients. C_1^i and C_2^i are obtained by fitting Eq. (4) to $\rho^M(T)$ in the *ab* plane of single crystals ($\rho_c\approx 0$, $p\approx 1$) in the background region (see below). Then, p (the effective cross section of the sample) and ρ_c (the overall intergrane resistance) are extracted by using again that fitting procedure, but with the values of C_1^i and C_2^i obtained before.

The background region is empirically defined as the temperature region above T_{c0} where the critical effects become negligible, i.e., where the AZ functional form fits to better than 0.5% the experimental resistivity. For all the different samples studied here, we found this to be the case for T above $T_{c0}+60$ K and at least up to $T_{c0}+160$ K. In this way, $\rho_B(T)$ is, therefore, estimated in that temperature region *independently* of any theoretical expression for $\Delta\sigma^i$. This is not the case in other paraconductivity analyses, as, for instance, those presented in Refs. 11, 12, 16, and 22, where both ρ_B and the theoretical $\Delta\sigma^i$ are *simultaneously* fitted to ρ^M. In addition, in some of these works (see, e.g., Refs. 16 and 22) T_{c0} is also treated as a free parameter (see below). It is obvious that with such a procedure almost any theoretical $\Delta\sigma^i$ may fit reasonably well the experimental data in any ϵ region, the price of it being not only a considerable ambiguity in all the cases, but also the possible determination of unrealistic T_{c0} and ρ_B. Various examples of such a situation are detailed in Ref. 16. Another version of this type of procedure is used in Ref. 13: The values of the main parameters arising in the theoretical $\Delta\sigma^i$ are first obtained from magnetoconductivity data, by imposing the validity of the corresponding theoretical expressions. Then, $\Delta\sigma^i$ in zero applied magnetic field is fitted, simultaneously with a *free* $\rho_B(T)$, to the experimental data. The agreement is excellent, but the corresponding *ad hoc* background is appreciably different from the measured resistivity even well above $2T_c$, a temperature region where a substantial paraconductivity influence is hard to be justified. Moreover, as noted by the authors themselves, $\rho_B(T)$ will undergo an unreasonable drop near T_{c0}.

Mean-field temperature (T_{c0}) and region. As discussed in Ref. 3, one may approximate T_{c0} by T_{cI}, the temperature where $\rho^M(T)$ around the transition has its inflexion point. Our recent simultaneous measurements of $\rho^M(T)$ and the magnetic susceptibility confirm the adequacy of this choice.[24] Further, the T location of the MFR may be bounded by the empirical but general condition

$$0.1\lesssim[\rho_B^i(T)-\rho^i(T)]/\rho_B^i(T_{cI})\lesssim 0.4\,.$$

A more detailed analysis of these two features has been[3] or will be presented elsewhere.[24] However, let us just note here that (i) the lower-temperature limit agrees qualitatively with that obtained using the Ginzburg criterion.[1] (ii) Above the high-temperature limit not only the slow-variation condition of the TDGL-like theories will probably fail, but also any calculation of $\Delta\sigma$ cannot be trusted unless it takes full account of the dynamic local effects.[20] This makes the analyses in which the MFR is extended up to $2T_c$ very suspicious (see, e.g., Refs. 13 or 16).

$\Delta\sigma^i$ for $YBa_2Cu_3O_{7-\delta}$ superconductors is presented in Fig. 1. The initial data, from which circles and squares have been obtained, correspond to the resistivity in the *ab* plane of two single crystals measured by, respectively, Hikita and Suzuki[13] and Friedmann and co-workers.[16] The triangles have been obtained from the $\rho(T)$ data of sample A of Ref. 3, a granular sample. The $\Delta\sigma^i(\epsilon)$ data extracted from the other granular $YBa_2Cu_3O_{7-\delta}$ samples studied in Ref. 3, which have very different long-scale structural inhomogeneities, are well within the data dispersion of Fig. 1. The common *intrinsic* AZ background coefficients were $C_1^i=1$ mΩ cm K and $C_2^i=0.6$ $\mu\Omega$ cm K^{-1}. This good agreement among all these $\Delta\sigma^i$ results for both the amplitude and the reduced-temperature behavior is, indeed, an important test of the consistency of our extracting procedure. The present results for $\Delta\sigma^i$ strongly differ in both amplitude and reduced-temperature behavior from those proposed by other groups (see, e.g., Refs. 13 and 16), due to the very different extraction procedure used, as noted above. The consequences of these last differences on the theoretical interpretations of $\Delta\sigma^i$ in $YBa_2Cu_3O_{7-\delta}$ samples are dramatic, as we see below.

The solid line in Fig. 1 corresponds to the best fit of Eq.

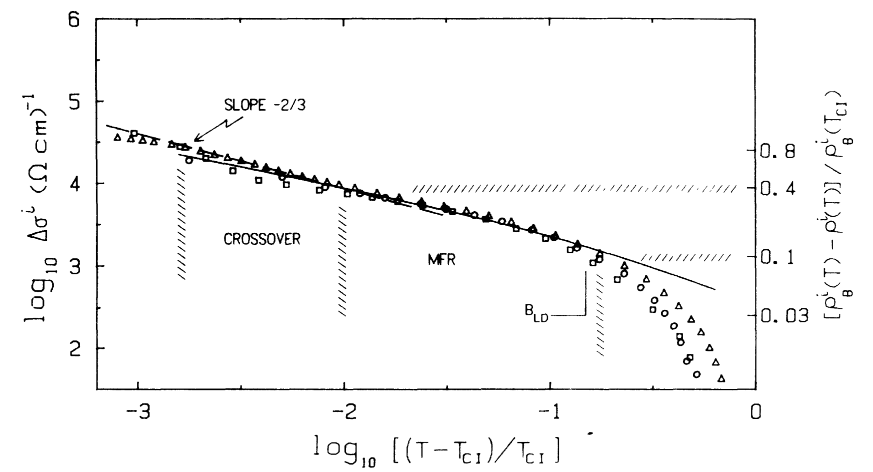

FIG. 1. Intrinsic paraconductivity in zero applied magnetic field of $YBa_2Cu_3O_{7-\delta}$ superconductors in the *ab* plane. For details see the main text.

(1) to the experimental data in the MFR, with $\Delta\sigma_F$ and $\Delta\sigma_A$ given by, respectively, the LD (Ref. 7) and the Larkin and co-workers[8] expressions. The fitting parameters are the LD amplitude $A_{LD} \equiv e^2/16\hbar d_e$, and 2D-3D LD crossover coefficient $B_{LD} \equiv [2\xi_\perp(0)/d_e]^2$, and the pair-breaking time, τ_ϕ. The resulting best-fits values (with a rms deviation of 0.8%) are $A_{LD} = (310 \pm 50)\ \Omega^{-1}\text{cm}^{-1}$, $B_{LD} = 0.16 \pm 0.05$, and $\tau_\phi \lesssim 5 \times 10^{-15}$ s. The main physical implications of these *intrinsic* values are (i) the smallness of τ_ϕ indicates relatively strong pair-breaking effects, in agreement with, for instance, normal reflectivity experiments.[25] With such a τ_ϕ, $\Delta\sigma_A$ will contribute less than 15% to the measured $\Delta\sigma^i$ over all the MFR. Recent theoretical works[20,21] on $\Delta\sigma^i$ seem to justify this result. (ii) As indicated in Fig. 1, the value of B_{LD} leads to 3D-SCOPF in most of the MFR. (iii) Beyond the MFR, closer to T_c, $\Delta\sigma^i$ in Fig. 1 confirms the existence of a crossover region with a critical exponent close to $-\frac{2}{3}$ (dashed line),[26] a result consistent with the 3D behavior found in the MFR.[27]

Using the original LD expressions for A_{LD} and B_{LD},[7] one obtains $\xi_\perp(0) = (1 \pm 0.3)$ Å, for the transverse correlation length amplitude, and $d_e = (5 \pm 1)$ Å, for the effective interlayer spacing (introduced here to take into account the double interlayer periodicity of these compounds). The value of $\xi_\perp(0)$ is somewhat smaller than that deduced from measurements of other properties.[28] In fact, the A_{LD} expression proposed in Ref. 21 ($A_{LD} \equiv rne^2/16\hbar d_e$) for *s*-wave pairing ($r = 1$) and a two-component superconducting order parameter ($n = 2$) leads to $\xi_\perp(0) = (2 \pm 0.6)$ Å and $d_e = (10 \pm 2)$ Å. These plausible different values will be commented on elsewhere. However, the important point here is that in all the cases the conclusions indicated in the previous paragraph remain unchanged.

A source of ambiguity in this type of fitting analysis is the fact that by worsening somewhat the fitting quality it is possible when analyzing one HTSC system alone to impose rather different values of the fitting parameters. For instance, if in the original LD coefficients we impose (as in

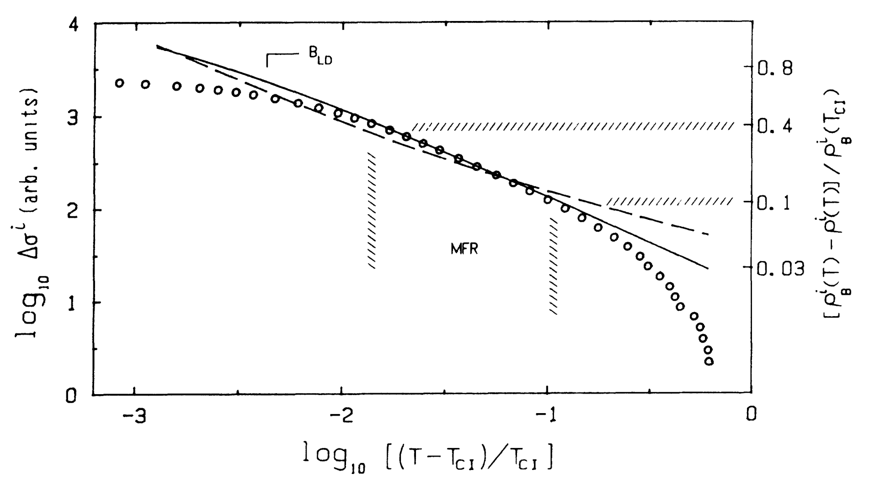

FIG. 2. Intrinsic paraconductivity in zero applied magnetic field, in arbitrary units, of a $Bi_{1.5}Pb_{0.5}Sr_2Ca_2Cu_3O_y$ superconductor in the *ab* plane. For details see the main text.

Refs. 11 and 13) $d_e = 11.7$ Å, the $YBa_2Cu_3O_{7-\delta}$ unit-cell size, we find $\tau_\phi \simeq (8 \pm 3) \times 10^{-14}$ s and $\xi_\perp(0) = (1 \pm 0.3)$ Å, the fit rms deviation in the MFR being 1%. This value of τ_ϕ, very close to those proposed in Refs. 11 and 13, leads to a completely different $\Delta\sigma^i$ behavior: $\Delta\sigma_A$ becomes the main contribution to the measured $\Delta\sigma^i$, whereas SCOPF will have a pronounced 2D behavior over all the MFR. This dramatic change in the SCOPF dimensionality suggests a basic question: Will the $Bi_{1.5}Pb_{0.5}Sr_2Ca_2Cu_3O_y$ materials, which have a more pronounced 2D behavior than the $YBa_2Cu_3O_{7-\delta}$ materials,[18,19] also support a weak pair breaking? The answer is presented in Fig. 2. The data points correspond to $\Delta\sigma^i$ (in arbitrary units) of a high-quality granular $Bi_{1.73}Pb_{0.50}Sr_{1.70}Ca_{1.80}Cu_3O_y$ sample,[29] with $T_{cI} = 108.3$ K. (The experimental details will be presented elsewhere.) The best fit (solid line) corresponds to $B_{LD} = 3.5 \times 10^{-3}$ (i.e., 2D-SCOPF in the MFR) and $\tau_\phi < 10^{-16}$ s (i.e., strong pair breaking). If $\tau_\phi = 10^{-13}$ s is imposed, one obtains the dashed line. These results rule out any important $\Delta\sigma_A$ contribution to $\Delta\sigma^i$ in $Bi_{1.5}Pb_{0.5}Sr_2Ca_2Cu_3O_y$ samples as well as (when combined with the results summarized above) in $YBa_2Cu_3O_{7-\delta}$ materials. Note, finally, that in $Bi_{1.5}Pb_{0.5}Sr_2Ca_2Cu_3O_y$ materials the B_{LD} location seems to suggest that the $\Delta\sigma(\epsilon)$ behavior for $\epsilon < 10^{-2}$ may be associated with the simultaneous presence of both a 2D-3D LD crossover and a mean-field full-critical regime crossover, as first indicated in Ref. 18.

In conclusion, the experimental results and the empirical analysis presented here strongly suggest that in absence of an applied magnetic field the copper oxide superconductors will have relatively strong pair-breaking effects (if the theoretical approach[8] for $\Delta\sigma_A$ is correct) and that their paraconductivity in the mean-field region may be explained *quantitatively* in terms of direct superconducting order-parameter fluctuations *alone* on the grounds of the LD theory. SCOPF in the MFR will be 3D and 2D in, respectively, $YBa_2Cu_3O_{7-\delta}$ and $Bi_{1.5}Pb_{0.5}Sr_2Ca_2Cu_3O_y$ compounds.

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