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Comments

Comment on "Double resistive superconducting transition in $Sm_{2-x}Ce_xCuO_{4-y}$"

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Recently, E. A. Early *et al.* [Phys. Rev. B **47**, 433 (1993)] presented resistivity measurements on $Sm_{2-x}Ce_xCuO_{4-y}$ polycrystalline samples exhibiting double-step resistive superconducting transitions, and drew general conclusions about the behavior of granular electron-doped compounds. We point out that the behavior observed in this family of compounds is very sample dependent, and that many different and richer cases of this can be found, the one described by the authors being merely a particular example. We also discuss unusual signatures of the beginning of superconducting transitions in inhomogeneous samples.

In a recent article[1] Early *et al.* have presented an experimental study of the resistive superconducting transition of $Sm_{2-x}Ce_xCuO_{4-y}$ polycrystalline samples. The most salient feature of the $R(T)$ curves they obtained is a double-step transition consisting of, when the temperature is lowered, a first partial drop at T_{c1}, followed by a plateau and then by the complete disappearance of resistivity at a temperature T_{c2}, below which macroscopic diamagnetism is also observed. The authors attribute the first drop (T_{c1}) to the superconducting transition of the grains of the sample, and the disappearance of resistivity below T_{c2} to the establishment of Josephson coupling between the grains.

In this paper we would like to discuss some basic features of superconductivity in granular samples based on our observations, and comment on the authors' attempt to generalized their findings concerning the occurrence of single-particle tunneling and the onset of intragranular superconductivity.

We have extensively studied the resistive superconducting transition of polycrystalline samples of the same family of compounds as the authors: $L_{2-x}M_xCuO_{4-y}$ where L stands for Pr, Nd, Sm, Eu and M stands for Ce and Th.[2] A careful analysis of the $R(T)$ curves measured under different ranges of magnetic field, together with superconducting quantum interference device magnetization measurements, lead us to the following conclusions: the main ingredients governing the resistive transition of polycrystalline n-doped superconductors are the width of the grains T_c distribution and the mechanisms of charge transfer across grain-boundary junctions (Josephson tunneling, and Andreev reflection or thermally activated single-particle tunneling, depending on the junction).

The important point is that the interplay between these phenomena is very sample dependent and we have indeed observed a great variety of behaviors, including the ones described by the authors.

As an especially richer case, we observed nonmonoton-

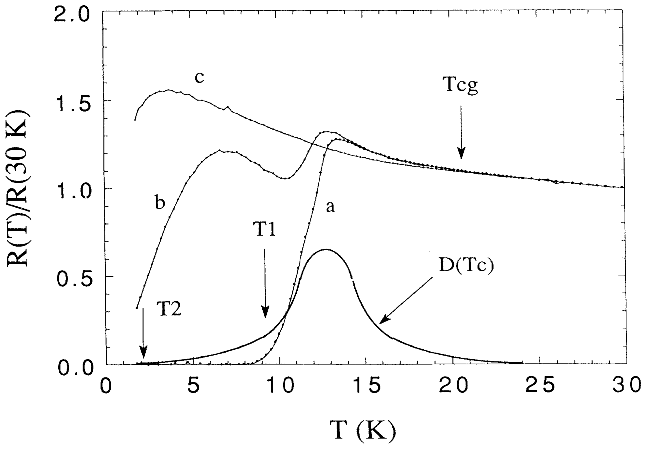

FIG. 1. Resistivity of a $Sm_{1.85}Ce_{0.15}CuO_{3.97}$ under (a) zero field, (b) 2kOe, and (c) 60 kOe. The sample is well coupled at zero field and presents a double-step transition under magnetic fields smaller than 5 kOe. The presumed grain's T_c distribution is schematically indicated.

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ic "double-peak" $R(T)$ sets of curves, in which an increase of resistance above the extrapolated normal-state value appears when T is lowered below T_{cq}, preceding the first drop of resistivity (at T_{c1} according to the authors' notation) and a marked peak instead of a plateau exists between T_{c1} and T_{c2} when a moderate magnetic field is applied (Fig. 1). Our qualitative interpretation, which has been comforted by effective medium calculations,[3] relies on the balance, when T is lowered, between the reduction of the *intragranular* resistance due to the superconducting transition of the grains, and the increase of the *intergranular* resistance because of the appearance of thermally activated single-particle tunneling between superconducting-normal and superconducting-superconducting pairs of grains. We have shown that this balance depends on the shape of the grains critical temperature distribution function, and that it can cause an *increase* of the global resistance in the upper temperature range of the distribution, and a reduction of the global resistance in its central part (Fig. 1). The resistance increase disappears if superconductivity in the grains is suppressed, giving rise to a negative magnetoresistance between T_{cg} and T_{c1} when fields comparable to the grains critical fields are applied.

The magnitude of these effects is of course smaller if a significant fraction of the junctions behave like normal metal weak links rather than pure tunnel junctions, which enables one to estimate the effective percentage of each type from the curves obtained.[3] Besides, T_{cg} and T_{c1} can coincide in the case of a very sharp distribution of T_c. But when these conditions are not gathered, the superconducting transition in a sample does not start at T_{c1}, as is commonly assumed, but at the temperature of appearance of the negative magnetoresistance. We found that this interesting phenomenon, although not general, was quite often present in the samples we studied. In all the cases we observed, the occurrence of the negative magnetoresistance at temperatures higher than T_{c1} *correlates with the appearance of diamagnetism.* The latter is significant, and, just above T_{c1}, it generally amounts up to 10% of the low-temperature (2 K) value [see Fig. 1-b of Ref. [4] or Fig. 9 of Ref. 2(b)]. This effect has also been observed in copper-free oxide superconductors.[5]

The two-step transition presented by the authors looks simpler: the intragranular and intergranular transitions are well separated, no effect of the grains critical temperature distribution is apparent, and the flat plateau between T_{c1} and T_{c2} suggests that quasiparticle tunneling is absent, which would indicate "weak-link-type" intergranular junctions as discussed above. But the absence of direct observation of tunneling by the authors *cannot rule out a priori its existence in other samples* of this family of compounds for the following main reason: The behavior observed is very sample dependent and sensitive to measurement conditions. We observed drastic changes upon aging and subsequent low-temperature (400 °C) reannealing of the samples, illustrating the sensitivity of the intergranular charge transfer mechanism to the precise characteristics of the junctions involved.

Moreover the authors seem to demonstrate the occurrence of single-particle tunneling in their samples with the use of high excitation currents (Ref. 1, Fig. 4). The steep increase of resistance observed at low temperature, well above the extrapolated normal state resistance (compare to the plateaus in Fig. 1), can only be obtained by this mechanism.

We also remark that in a sample containing only "tunnel-type" junctions, single-particle tunneling effects can cause the increase of resistance between T_{cg} and T_{c1} without necessarily giving a sharp peak between T_{c1} and T_{c2}. Indeed, when only a minority of the grains is superconducting (as between T_{cg} and T_{c1}), their first neighbors are normal grains and the charge transfer can only occur by thermally activated single-particle tunneling. But when most of the grains are superconducting, the charge transfer which occurs between superconducting grains through resistive junctions is much less temperature sensitive since excess supercurrents can add to the thermally activated one. This effect again depends on the precise characteristics of the junction.[6]

The interpretation of the negative magnetoresistance we observed[2] in terms of weak localization only, as proposed by the authors, is very questionable. It is not our goal to discuss here the experimental observations and claims reported in Ref. 7. We wish to emphasize that two-dimensional weak localization within intracrystalline a-b planes cannot be responsible for the overall behavior of randomly oriented granular systems in which the normal state resistance is mainly governed by intergranular boundaries, as claimed by the authors. In our samples, the variation of resistance under a 5-T magnetic field corresponds up to 30% of the *intragranular* resistance just above T_{c1} (~ 10 K), and to more than 100% at the lowest temperatures in samples whose resistivity does not vanish [see Figs. 4 and 8-a of Ref. [2(b)], respectively].

To conclude, we insist on the following points:

Since diamagnetism below T_{cg} is not always large enough to be observed (as the authors emphasized) the study of the superconducting transition in inhomogeneous granular samples should always involve careful comparisons of $R(T)$ curves measured under different magnetic fields.[4]

The behavior depicted by the authors is a particular case which enters the general framework of our qualitative interpretation, and which can easily be reproduced by effective medium calculations.

We are grateful to the authors of Ref. 1 for providing us with the series of samples.

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