Electron tunneling in the high-T_c superconductor Y-Ba-Cu-O

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Electron-tunneling measurements were performed in granular and homogeneous samples of Y-Ba-Cu-O, taking advantage of the presence of natural insulating barriers between grains. The results show supercurrents, the superconducting gap, and sharp peaks that can be related to a new mechanism for superconductivity. A phenomenological interpretation of these results is also outlined.

The recent discovery of high-T_c superconductors represents not only an unexpected technological revolution but has also attracted the interest of scientists from a wide range of fields of basic research. In spite of the enormous amount of data taken in the last few months, important questions remain unanswered. In particular it is not clear which are the basic mechanisms that produce superconductivity in these systems. Among the ceramics that have been found to be high-T_c superconductors is Y-Ba-Cu-O for which various experiments[1] and theoretical results[2] have been reported. It is well known that electron tunneling is a useful probe to investigate important characteristics of the superconducting state such as the superconducting gap, the electronic density of states, and, in some cases, the electron-phonon coupling, the electron-electron interaction, and the phonon structure through the McMillan procedure.[3]

There have been tunneling results in Y-Ba-Cu-O (Ref. 4) using point-contact junctions, resulting in very noisy spectra. In this paper we report tunneling measurements performed in granular samples, fabricated in our laboratory, taking advantage of the fact that the superconducting grains are imbedded in an insulating matrix and coupled through Josephson interactions in a natural way when the Josephson coupling energy exceeds the thermal energy (k_BT). Therefore, by changing the temperature one can couple the superconducting grains at will. The results reported here show very clear features that provide unquestionable quantitative characteristics of this superconducting state that contribute toward a better basic knowledge of the mechanisms responsible for high-T_c superconductors.

Samples were prepared by mixing high-purity powders of CuO, $BaCO_3$, and Y_2O_3 with a nominal composition given by the formula $(Y_{1-x}Ba_x)_2CuO_{4-\delta}$. The correct proportion of oxygen was obtained by the usual calcination and sintering processes.[1]

By varying x from 0.37 to 0.4 one finds that the R vs T characteristics of the samples change dramatically. In Fig. 1 two types of samples are shown. Curve a corresponding to $x=0.37$, shows a large difference between the onset and critical temperatures, which we shall call a granular sample, and curve b, corresponding to $x=0.4$, shows a more stepped transition, and we shall call it a homogeneous sample.

The reasons by which the samples are called granular and homogeneous, respectively, although strictly speaking, both are in a granular state, are that the R vs T curves show changes in the onset temperature for superconductivity and electrical resistance variations which resemble the description of a granular state in a percolation model.[5] In the optimized $Y_1Ba_2Cu_3O_{7-\delta}$ samples the superconducting phase overwhelmingly occupies most of the volume and, therefore, one surpasses the percolation limit, and the sample shows an abrupt fall of resistance at $\sim$90 K. In the sample with $x=0.4$, it should be relatively easy to form the superconducting phase, while in the samples with $x=0.37$ the nonsuperconducting phase should be more abundant.

The experimental setup differs from the conventional one in that there is not one artificially created barrier but a distribution of barriers between grains and, therefore, some anisotropy is expected, due to the privileged direction in the sintering process. The samples were sintered

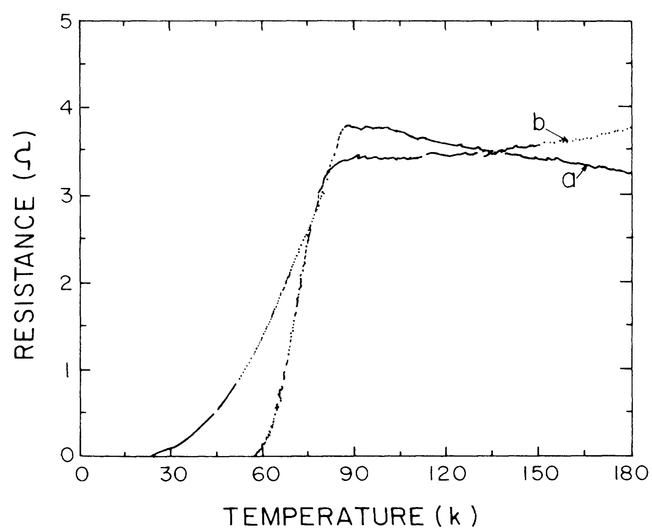

FIG. 1. Resistance vs temperature plots. Curve a is for a granular sample of Y-Ba-Cu-O, and curve b is for a homogeneous sample of Y-Ba-Cu-O.

disks with two flat faces. Therefore, the four contacts for the measurements can be made in the same flat face of the disk or in pairs in opposite faces of the disk. The differences detected in the two geometries will be shown below. The tunneling experiments were performed at $T=10.3$ K, using the conventional modulation technique,[6] that consists in injecting a modulated current (1000 Hz) and a polarization voltage from 0 to 800 mV and detecting the differential resistance (dV/dI) with a lock-in amplifier and a resistance bridge.

Figure 2(a) shows the dV/dI characteristic from the granular sample in which the current and voltage were measured along one flat face of the disk. One notices the absence of noise that allows a clear identification of several features. (1) There is the appearance of a small superconducting current at zero voltage. (2) The asymmetric behavior with respect to the bias in agreement with previous results.[4] (3) The size of the energy gap (2Δ), which is difficult to determine, but roughly lies between 60 and 100 meV. (4) There are sharp peaks, also asymmetric, that, at positive bias, are found at 520, 360, and less clearly at 210 meV. Apparently similar features, although less well defined, attributed to phonons, have been recently found by Tanner.[7]

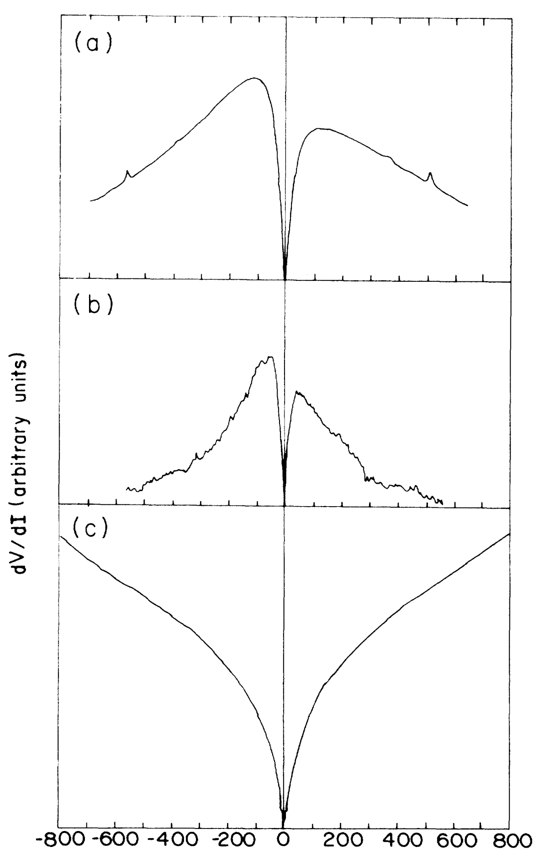

FIG. 2. Differential resistance (dV/dI) measured at 10.3 K, (a) from a granular sample of Y-Ba-Cu-O in the direction parallel to the disk faces, (b) from the same granular sample as (a) but in the direction perpendicular to the disk faces, and (c) from a homogeneous sample of Y-Ba-Cu-O in the same conditions as (a).

Figure 2(b) was taken from the same granular sample in identical conditions, except that the current and voltage were measured in the perpendicular direction of the disk faces. Again, a supercurrent at zero voltage and the asymmetric behavior are apparent, but the curve is much noisier than the previous one, although there is a hint of the same sharp peaks. The appearance of noise is attributed to the anisotropy in the distribution of grains.

For the sake of comparison, a similar measurement was performed in the homogeneous sample and the curve is shown in Fig. 2(c), in which it is clearly seen that tunneling is suppressed, since the grains are supposed to be touching. In this case the differential resistance was < 1 Ω, much smaller than in the tunneling junction (~ 270 Ω) in the gap region. There is an anomaly around zero voltage due to a small supercurrent, driven out of zero voltage by thermal fluctuations. This result reinforces the initial assumption of granular structure in the former sample. This behavior is the usual one in superconducting microbridges. There is a hint of decoration in this curve that is possibly related to subharmonic gap structure and nonequilibrium superconductivity processes,[8] but these are out of the scope of this communication.

A first attempt to interpret these experimental results could be made within the BCS framework. One immediately encounters difficulties. First, the ratio $2\Delta/k_B T_c$ calculated from the onset critical temperature is between 7 and 13, far too large for the Eliashberg equations to hold.[9] Second, although it is hard to envisage the presence of high-energy optical phonons, one could overlook this conceptual difficulty based on the existence of high-energy phonons in other oxides,[10] and estimate the critical temperature from the equations, appropriate for systems with Einstein peaks,[11] assuming that the sharp features in Fig. 2(a) derive from high-energy phonons responsible for superconductivity. A reasonable value of the electron-phonon coupling $\lambda=1.7$ was estimated from the energy bands,[2] the Coulomb interaction U^* was maximized (0.2), $\alpha(\omega)$ was assumed to be 1, and the phonon distribution $F(\omega)$ taken as a delta function at the experimental energy. With these assumptions, the critical temperature obtained was 34 K, exceedingly low with respect to the measured one (see Fig. 1).

It is worthwhile pointing out that the role of dimensionality is not included in the BCS theory and, as it is generally agreed that the superconducting state occurs in two dimensions in these high-T_c materials, the arguments above should not be taken as a proof of the failure of the BCS theory.

However, there is an alternative explanation of the origin of the high-energy peaks that no doubt are related to the appearance of superconductivity. A basic ingredient for the occurrence of superconductivity is the electron pairing from the attraction through the lattice. Phonons are not the only excitations that could produce this phenomenon; it has been suggested in a recent paper[12] that a possible source of negative U systems is a polarization caused in the lattice by charged localized impurities.

These polarization effects could account for the dramatic decrease of the effective U (U^*) in semiconductors such as dangling bonds in silicon. The presence of flat bands due to the oxygen lone pairs, perpendicular to the Cu-O planes[2] in these materials could play the role of charged states that polarize the lattice. A simple calculation using a two-band impurity model shows that $U^* = U - 2K$ where K is the polarization energy induced in the medium by a point charge and could be estimated.[13] Using reasonable values of the dielectric and geometrical parameters in these materials, it was found that the excitations due to this polarization mechanism are of the order of 0.6 eV, in agreement with the experimental features in Fig. 2(a). Polarization is also important in the case of transition-metal impurities in MgO. This picture is not alien to the old idea of bipolaronic superconductivity[14] and shares with other theories[15] the point of view that low dimensionality can lead to peculiar interactions between electrons and that phonons could not be of primary importance in this sort of superconductivity.

The important results in this paper are the following. (1) A superconducting current was clearly observed for the first time, (2) very sharply defined structures that yield information of the excitations coupled to the electrons are reported. (3) Clear evidence that the mechanism for superconductivity in these materials differs considerably from the conventional BCS theory is given.

This paper poses important questions to be tested by other experiments that could confirm our hypothesis, these being currently in progress.

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