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Point-contact tunneling study of $HgBa_2CuO_{4+\delta}$: BCS-like gap structure

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We report on point-contact tunneling into polycrystalline $HgBa_2CuO_{4+\delta}$ superconductors with a T_c onset of 97 K. Tunneling data follow the general trend of the best curves seen in cuprate high-temperature superconductors, but these show significantly *lower* and *flatter* subgap conductance values than any other cuprate and they can be reproducibly obtained. By fitting the data to a smeared BCS density of states, the energy gap has been determined and the smearing parameter is so small that it is comparable to thermal smearing.

I. INTRODUCTION

Tunneling measurements have been an important probe of high-temperature superconductors (HTS), providing a direct measure of the superconducting energy-gap parameter Δ and giving insight into the quasiparticle density of states. In this paper, we report point-contact tunneling on the recently discovered $HgBa_2CuO_{4+\delta}$ (Hg 1:2:0:1) compound[1] using a Au tip onto two polycrystalline samples[2] which have T_c (onsets) of 97 and 95 K. In comparison with previous tunneling into HTS cuprates,[3] the conductances are unusually symmetrical, display well-defined peaks, and have *lower* and *flatter* subgap ($V < \Delta/e$) values. The zero-bias conductance is typically 6% of the normal state and can be reproducibly obtained.

The above T_c is unusually high for a compound with only one CuO_2 layer per unit cell. The simple crystal structure of $HgBa_2CuO_{4+\delta}$ consists of single CuO_2 layers separated by BaO-Hg-BaO insulating regions: the distance between CuO_2 layers[1,2] is only 9.5 Å. Such a small spacing between CuO_2 layers may lead to better intrinsic superconducting properties in a magnetic field.[4–6] This compound is the first member of the homologous series $HgBa_2Ca_{n-1}Cu_nO_{2(n+1)+\delta}$, similar to the single-thallium-layer series $TlBa_2Ca_{n-1}Cu_nO_{2n+3}$. The Hg 1:2:2:3 phase was first synthesized as a multiphase component[7,8] (T_c above 130 K) and subsequently as a single phase with a record high T_c of 135 K.[9] Further enhancements of T_c up to 150 K are reported[10] in an applied pressure of 150 kbar.

II. EXPERIMENT

Samples of Hg 1:2:0:1 were synthesized from stoichiometric mixtures of HgO and Ba_2CuO_3, as previously described,[2] and then annealed at 300 °C in oxygen. X-ray diffraction studies confirmed all samples to be single phase without any detectable impurities. ac susceptibility was measured in a 1 G, 100-Hz field using a Lake Shore Cryotronic susceptometer. Curve *A* in Fig. 1 shows the susceptibility of a bulk sample (tunneling sample *A*) which was powdered before susceptibility measurements. It exhibits a sharp transition with an onset T_c of 97 K and a full shielding fraction, consistent with the isolated grains of the polycrystalline material being uniform and fully superconducting. Susceptibility data (Fig. 1, curve *B*) on the bulk tunneling sample *B* (i.e., not ground to a powder before measuring), exhibits two transitions typical of that found in polycrystalline materials with weak coupling between grains.[11] It is suggested[11] that the first transition reflects the intragranular superconductivity of the decoupled grains in the polycrystalline sample, followed by a broader transition resulting from the intergranular coupling across the sample.

All low-temperature measurements were done with the apparatus cooled by exchange gas to liquid ^{4}He. After transferring liquid ^{4}He, we waited about 5 h for thermal stability so that stable junctions could be maintained. Raising the temperature above the cryogen boiling point with a heater was possible, but resulted in much poorer stability of the tunneling contact. As in previous cases,[12–16] an insulating surface prevented vacuum tunneling and the Au tip was used mechanically to cleave and/or scrape the surface before a measurable current was obtained. The resistance of resulting junctions could be varied by adjusting the force of the tip on the sample.

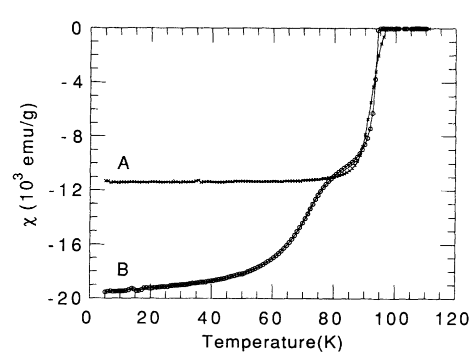

FIG. 1. ac susceptibilities for two samples of $HgBa_2CuO_{4+\delta}$ with T_c(onset)=97 and 95 K. Curve *A* (corresponds to tunneling sample *A*) is for a bulk sample, which was powdered before susceptibility measurements. Curve *B* corresponds to the bulk tunneling sample *B*.

The current-voltage characteristic $I(V)$ was monitored while the tip was maneuvered to obtain an acceptable junction. First derivative data dI/dV were obtained using a Kelvin bridge with the usual lock-in techniques.[13]

III. RESULTS AND DISCUSSION

Experimentally, no differences were observed for the two samples. Typical experimental conductances ($\sigma_s = dI/dV$) are shown in Figs. 2(a)–2(c). The high degree of symmetry is unusual for tunneling between asymmetrical electrodes of a normal metal and a cuprate HTS, although it is generally found for conventional superconductors. For all such stable superconductor-insulator-normal (SIN) junctions, the high-voltage (background) conductance usually increases with voltage [see Fig. 2(a)] as is commonly found in HTS, and can be attributed[15,16] to the ordinary parabolic tunneling conductance.[17] Several junctions showed a flat background conductance [see Fig. 2(b)] and one junction showed a decreasing background conductance [see Fig. 2(c)]. Such a decreasing background has also been observed previously in other cuprates.[18] Here in all plots, positive voltage means that the Au tip is at a positive voltage relative to the Hg 1:2:0:1 sample.

In order to proceed, the conductances need to be normalized (i.e., divided by the normal-state value σ_n). Direct measurements of σ_n can be made at $T > T_c$, but for HTS this is difficult due to the different thermal expansions in the tip assembly materials. However, σ_s is expected to approximate σ_n at high voltages, so we fit the high-voltage data to a fourth-order polynomial to obtain an estimate of σ_n [dashed lines in Figs. 2(a)–2(c)]. The tunneling data shown in Fig. 2(b) are interesting because of the very flat background and the data can be normalized by a constant (over the narrow voltage range), which ensures that no additional structure is introduced by this procedure.

When σ_s is normalized by σ_n, estimated in this way, we arrive at the normalized conductances [solid lines in Figs. 2(d)–2(f)], which can be compared to theory. Usually, HTS tunneling results are compared to the smeared BCS density of states, first proposed by Dynes, Narayanamurti, and Garno,[19] in which

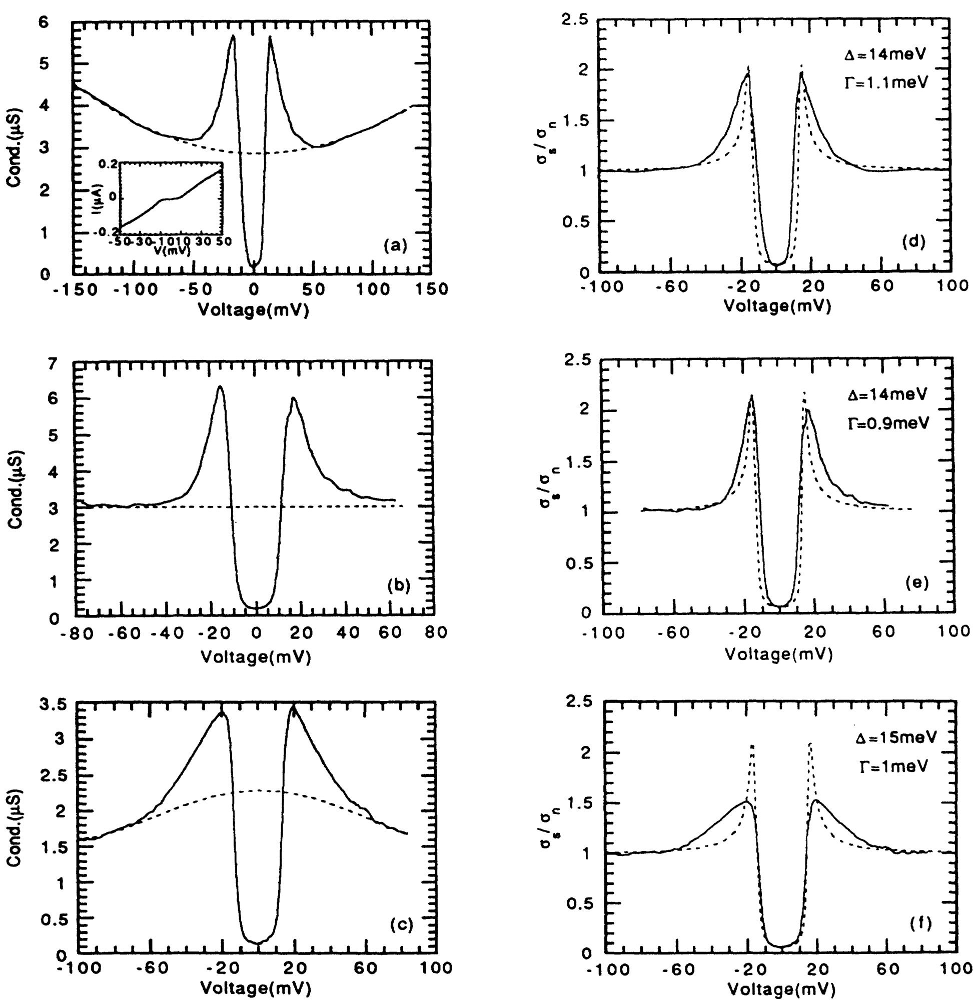

FIG. 2. (a)–(c): Experimental conductances (solid lines) with the fitted normal-state conductances (dashed lines) for junctions No. 1, No. 2 of sample *B* and junction No. 1 of sample *A*, respectively; (d)–(f): The solid lines are the corresponding normalized conductances, while the dashed lines are the smeared BCS fits including thermal smearing. Experimental $I(V)$ curve for junction No. 1 of sample *B* is shown in Fig. 2(a) as an inset. All tunneling curves here were measured at $T = 4.2$ K.

$$N_s(E)=\mathrm{Re}\{(E-i\Gamma)/[(E-i\Gamma)^2-\Delta^2]^{1/2}\}\ , \qquad (1)$$

where Γ is a smearing parameter to account for lifetime effects, such as inelastic scattering. The normalized conductance is then given by [17]

$$\sigma_s/\sigma_n=\int_{-\infty}^{\infty} N_s(E)[-\partial f(E+eV)/\partial(eV)]dE\ , \qquad (2)$$

where the Fermi function f accounts for thermal smearing. The fits using Eq. (2), shown as dashed lines in Figs. 2(d)–2(f), provide values of Δ and Γ. All tunneling curves here were measured at $T\sim4.2$ K. Note that all subgap conductances [Figs. 2(d)–2(f)] are very low and flat, resulting in small values of Γ. The small Γ (~1 meV) means that thermal smearing ($k_BT=0.36$ meV) cannot be ignored in the above procedure. In our previous work[15,16] on other cuprate HTS as well as other tunneling studies,[3] the normalized conductance was fitted to Eq. (1) directly because Γ was much larger than k_BT.

Note that all the junctions of Fig. 2 are fit with $\Gamma/\Delta=6-8\,\%$, making this the *lowest reproducible* value of all cuprate HTS. The subgap conductance in Fig. 2(f) is quite flat and agrees extremely well with the BCS expression [Eq. (2)] with only small smearing. It is thus much closer to what would be expected for *s*-wave superconductivity than is typically found in other cuprates.[20] A flat subgap conductance was also reported by Hasegawa *et al.*[21] using a scanning tunnel microscope on a single junction of $YBa_2Cu_3O_{7-\delta}$, but the preponderance of data on $YBa_2Cu_3O_{7-\delta}$ suggests more paraboliclike subgap conductances.[22]

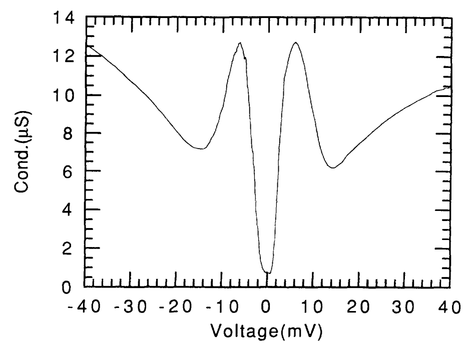

FIG. 3. Experimental conductances (measured at 4.2 K) of junction No. 3 of sample *B* showing a smaller gap value.

Figure 3 is an example of σ_s (measured at 4.2 K) showing well-defined conductance peaks with an energy gap, $\Delta\sim5-6$ meV. The tunneling characteristics of this junction showed changes when the temperature was raised from 4.2 to ~11 K using a heater. This indicates that the gaplike features are indeed due to the superconducting density of states and not a charging effect which would be insensitive to this temperature change.

The energy gaps Δ determined according to the above procedure for 24 junctions range from ~5 to 20 meV. The origin of the spread in Δ is unknown, but this could be an indication that there is a considerable variation in T_c, which possibly results from variations in oxygen content of $HgBa_2CuO_{4+\delta}$ near the sample surface. It is known[2] that T_c can be increased up to 97 K for $\delta\sim0.06$ on annealing in oxygen at 300 °C for 24 h. On annealing in argon at 500 °C for 24 h, the T_c drops to 59 K for $\delta\sim0.01$. Further reduction decreases the T_c to ~44 K, after which the compound decomposes.[2] Also the Hg 1:2:0:1 samples are found to be electromagnetically granular.[5] Thus it is quite possible that we are probing local properties of individual grains with different δ values and hence different T_c. We suspect, therefore, that the small gaps ($\sim5-6$ meV) observed in some cases are associated with this minimum value of T_c that can be obtained before decomposition. Although surface deterioration upon exposure to moisture causes no observed change in the bulk T_c (with only minor changes in x-ray pattern),[11] it may reduce the T_c of some grains on the surface which are probed by the tunneling tip.

Broadened conductance peaks [e.g., see Fig. 2(c)] suggest a small distribution of gap values, and our fitted value of Δ is within this distribution. It is difficult to obtain an accurate value of $2\Delta/k_BT_c$ due to the spread of the energy gaps and the unknown T_c associated with each junction. Using Δ for our best junctions (~15 meV) together with the onset T_c of 97 K, implies a lower limit of $2\Delta/k_BT_c\sim3.6$. This is significantly lower than the usual tunneling results for HTS cuprates.

In summary, the SIN tunneling data of Hg 1:2:0:1 are quite symmetrical and follow the general trend of the best curves seen in cuprate HTS, but with the *lowest* and *flattest* subgap conductances compared to other cuprates, and both features are obtained reproducibly. Energy gaps Δ are determined using a smeared BCS expression *including* thermal smearing.

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