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# Tunneling measurements of the energy gap in the high-$T_c$ superconductor $Tl_2Ba_2Ca_2Cu_3O_{10+\delta}$

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The tunneling $I$-$V$ characteristic curves have been studied for a ceramic sample of $Tl_2Ba_2$-$Ca_2Cu_3O_{10+\delta}$ using a scanning tunneling microscope. We have found two kinds of curves which are interpreted due to the tunneling in superconductor-insulator-normal-metal and superconductor-insulator-superconductor junctions. Although the $T_c$ of the intervenient superconducting regions are unknown, values of $2\Delta/k_BT_c$ are around the BCS value 3.5 using the $T_c$ obtained by resistance measurements.

Following the discovery of high-critical-temperature ($T_c$) superconductivity in the Bi-Sr-Ca-Cu-O system,[1] Sheng and Hermann[2] replaced Bi by Tl, finding superconducting transitions with onset temperatures of up to 120 K in a Tl-Ca-Ba-Cu-O compound.

The Tl-based high-$T_c$ superconductor with three Cu-O planes per unit cell $Tl_2Ba_2Ca_2Cu_3O_{10+\delta}$ so far has the highest critical temperature among the well-characterized superconducting materials. Some reports claiming higher $T_c$ in other systems have been produced, but in the generality of cases the reported superconductivity shows peculiar features such as dependence of $T_c$ on thermal cycling, very small critical current suggesting filamentary superconductivity, small Meissner signal, etc.

Different superconducting phases have been isolated and identified[2,3] for the Tl-Ba-Ca-Cu-O systems, which can be expressed by the general formula $Tl_2Ba_2Ca_{n-1}$-$Cu_nO_y$ ($n=1,2,3$). Depending on preparation conditions, critical temperatures $T_c$ have been reported[3] to range from 95 to 108 K for the $n=2$ phase and from 118 to 125 K for the $n=3$ phase.

Studies and measurements of relevant physical properties for this new high-$T_c$ superconducting system appear to be necessary in order to better understand the underlying mechanisms of high-$T_c$ superconductivity.

One of the most outstanding properties of BCS superconductors is the energy gap $2\Delta$ which opens at the Fermi level, when they are cooled below their critical temperature $T_c$. As is well known, there exist several techniques which can provide information about the value of $\Delta$ and its dependence on other parameters such as temperature or applied magnetic field, tunneling spectroscopy being one of the most important of them. This kind of spectroscopy, based on the current-voltage ($I$-$V$) characteristics given by the tunneling of quasiparticles, is being widely used in order to measure the energy gap and the quasiparticle density of states for high-$T_c$ superconductors.

There is a mass of experimental results at present, with important problems of interpretation and serious disparities between different experiments. High-$T_c$ materials are to some extent "troublesome" because of difficulties in maintaining the bulk chemical composition on the surfaces or in the interfaces of tunnel junctions. These problems have already been pointed out by other groups working in this field.[4-7] The result is that there is a lack of agreement in the tunneling measurements.

Most tunneling data have been analyzed within the framework of the BCS theory of superconductivity, but there are many features in the experimental curves which are far from those found in the "low-$T_c$ superconductors." Anyhow, this kind of analysis allows us to obtain the important $2\Delta/k_BT_c$ ratio, which in most cases is higher than the predicted value of 3.5 for weak-coupling BCS superconductors. Ratios ranging from 4.5–6 are usually reported[6-9] from tunneling measurements, although some authors have reported values higher than 10.[4,5] On the other hand, ratios of 3.5 have also been obtained[10] for some high-$T_c$ superconductors, and values as low as 2 have been measured[6,11] in some infrared spectroscopy experiments.

If there is no agreement for the gap region, according to what has been said above, the situation is even more confusing at higher voltages. Asymmetric and increasing conductivity, and often structures more or less correlated with the value of $\Delta$, are usual features of the tunneling current-voltage characteristics.

In this work we report our results in a sample of $Tl_2Ba_2Ca_2Cu_3O_{10+\delta}$, using a low-temperature scanning tunneling microscope (STM). Four-probe dc resistivity measurements showed zero resistance at 105 K. The midpoint of the transition was at 110 K.

The experimental setup of our low-temperature STM has been described elsewhere.[12] However, significant changes have been introduced for the present experiment. Sweeping of the tip-sample bias voltage is made by a function generator HP 3314 A. The tunnel current signal is measured by a digital voltmeter Keithley 194 A which stores data and transfers them to an IBM PC computer. $I$-$V$ curves are also displayed in real time in a digital oscilloscope Philips PM 3350. We have usually taken $I$-$V$ curves of 600 points in time intervals shorter than 10 ms for the whole curve. Both W and Pt-Rh tips have been used. In most of our measurements the tip was touching the surface of the sample. That was necessary in order to obtain significant current at low voltage.

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In this paper we would like to discuss the most relevant features of our results. With this aim, we show in Figs. 1–3 three different $I$-$V$ characteristics representative of more than three hundred curves taken on different places of the sample and with different tips. We also show the conductance versus voltage curves obtained by numerical derivation of the experimental results. The curves of Figs. 1 and 2 were obtained with Pt-Rh and W tips, respectively. In both cases we obtain, within our resolution limits, zero current at low bias voltage. However, the curves are smeared, suggesting, from our point of view, a behavior similar to that reported by Fein *et al.* for $La_{2-x}Sr_xCuO_{4-y}$.[7] This smearing likely arises because $\Delta$ is not constant throughout the sample area probed by the tip. We have then fitted these curves using a Gaussian distribution of BCS gaps as described in Ref. 7

$$I(V)=C\int_{-\infty}^{\infty}d\Delta\int_{-\infty}^{\infty}dE[f(E)-f(E+eV)]N(E,\Delta)e^{-(\Delta-\Delta_0)^2/\delta\Delta^2}, \tag{1}$$

where $C$ is a constant, $f$ the Fermi-Dirac population factor, and $N(E,\Delta)=|E|/(E^2-\Delta^2)^{1/2}$ is the BCS density of states.

A combination of gap anisotropy, different local stoichiometry, and local defects (every one of them possibly present in these oxide ceramics) can produce a gap value distribution, as needed to fit our results. The fits are shown as curves in Figs. 1(b) and 2(b). From this analysis we obtain values of $\Delta=13$ meV, $\delta\Delta=6$ meV and $\Delta=15$ meV, $\delta\Delta=5$ meV, respectively.

We think that this model accounts well for our results, but we cannot exclude other possible effects to explain the smearing of the characteristic curves in other materials. For example, lifetime effects have been invoked by several authors[9,13,14] in the case of $I$-$V$ curves with nonzero subgap conductivity, but for our measurements and within our experimental resolution, that is not the case.

In Fig. 3 we show another kind of curve frequently obtained mainly with Pt-Rh tips. It is noteworthy that the conductance-voltage curve remarkably resembles those of classical superconductor-insulator-superconductor (SIS) junctions. Moreover, the separation between the conductance peaks for this type of curve is approximately twice that found in curves more likely representing normal-metal-insulator-superconductor (NIS) tunneling, as expected if this approach is correct. In fact, evidence of SIS type tunneling measurements have also been reported in other high-$T_c$ superconductors.[10] The occurrence of SIS tunneling may happen when the tip is touching one clean superconducting grain, separated from the rest of the sample by an insulating region. Hence the tunneling would occur between two superconducting areas instead

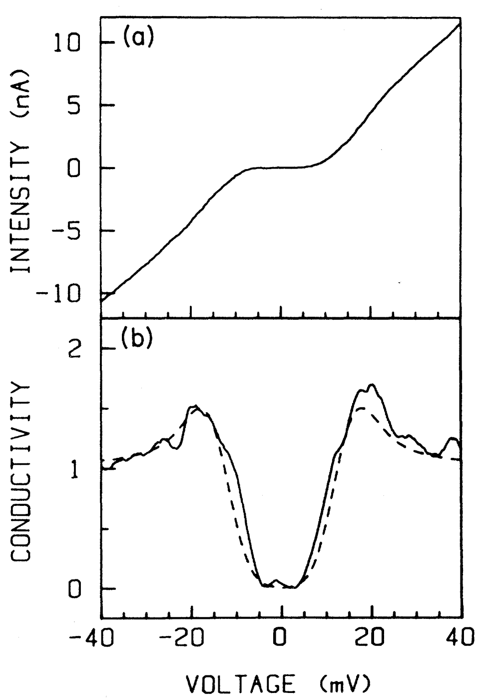


FIG. 1. (a) $I$-$V$ characteristic of a superconductor–normal-metal tunnel junction obtained with a Pt-Rh tip. (b) The solid line is the numerically calculated conductance from (a) normalized to the expected constant metallic conductance, guessed from the curve at $V>\Delta$. The dashed curve is a fit to the data as described in the text.

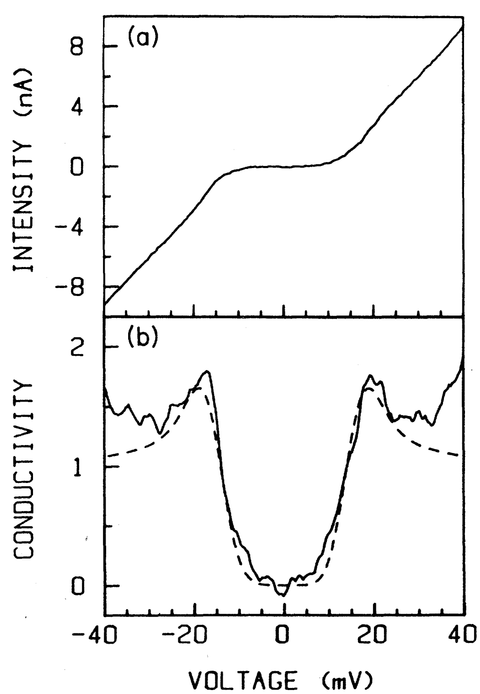


FIG. 2. (a) $I$-$V$ characteristic of a superconductor–normal-metal tunnel junction obtained with a $W$ tip. (b) The solid line is the numerically calculated normalized conductance from (a). The dashed curve is a fit to the data as described in the text.

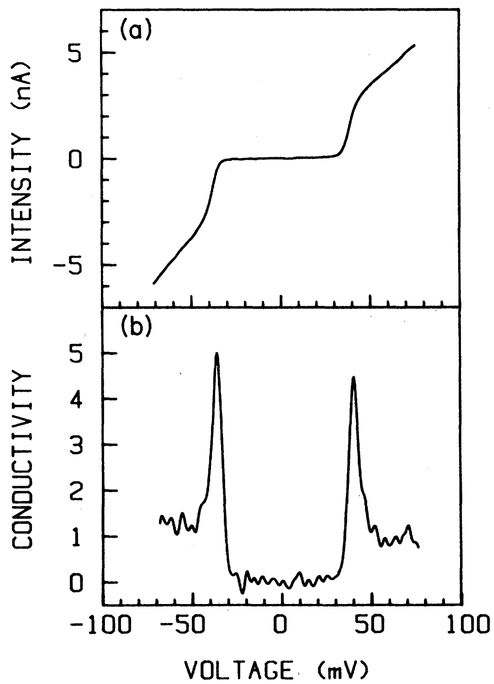


FIG. 3. (a) $I$-$V$ characteristic of a superconductor-superconductor tunnel junction, obtained with a Pt-Rh tip. (b) Numerically calculated normalized conductance from (a).

of between the normal-metal tip and a superconducting region. It is worthwhile to point out that the taking of SIS curves for NIS ones can partly explain the large spread of reported gap values for high-$T_c$ superconductors.

Therefore, if the tunneling results shown in Fig. 3 are interpreted as SIS tunneling, a value of $\Delta = 19$ meV is obtained, and $2\Delta/k_B T_c = 4$ close to the BCS ratio and within the range of the gap distributions in Figs. 1 and 2. It is interesting to note that if the superconducting material intervening in these SIS junctions were of the 2:2:3:3 phase, which has $T_c = 125$ K, the quoted ratio would be 3.5.

We would like to emphasize that it is difficult to obtain good tunneling characteristics, what is probably correlated to the short coherence length found in these high-$T_c$ oxides. This fact can produce a large dispersion of the superconducting properties, even inside the microscopic region probed by the tip. In addition, the uncontrolled nature and thickness of the insulating barriers can give a number of different kinds of $I$-$V$ curves. For example, if we take a look at the curve of Fig. 3(a), we can observe a slight asymmetry in the $I$-$V$ characteristics, the slope in the negative side being somewhat larger than that of the positive side. Such asymmetric features are not uncommon in tunneling characteristics.

In our opinion, they are not necessarily related to high-$T_c$ superconductivity or unusual density-of-states shapes. The "Ohmic" behavior (linear $I$-$V$ characteristics, in the normal-metallic state) is obtained only when the tunnel barrier height $\phi$ is much larger than the applied bias voltage ($\phi \gg eV$), and is well known in the STM research field, that it is normal to find $\phi$ values smaller than 1 eV in experiments on metallic samples.

In summary, we have obtained tunneling characteristics of $Tl_2Ba_2Ca_2Cu_3O_{10+\delta}$, showing clearly the existence of an energy gap at the Fermi level. We have found two distinct types of curves, associated in our opinion to NIS and SIS tunneling.

Although it is not possible to go further in our conclusions, we would like to stress that curves such as those in Fig. 3 remarkably resemble the SIS characteristics found in common BCS superconductors.

We have not found extremely high $2\Delta/k_B T_c$ values, and from our measurements, values around 3.5 are obtained for this high-$T_c$ superconductor.

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