## Evidence for a distribution of discrete energy gaps in polycrystalline high- $T_c$  superconductors

J. E. Núñez-Regueiro

Instituto Venezolano de Inuestigaciones Cientigcas, Apartado 21827, Caracas 1020-A, Venezuela and Departamento de Fr'sica, Facultad de Ciencias, Universidad del Zulia, Apartado 526, Maracaibo, Venezuela

## J. M. Aponte

## Instituto Venezolano de Investigaciones Científicas, Apartado 21827, Caracas 1020-A, Venezuela (Received 24 January 1992)

We present measurements of the differential conductance  $dI/dV$  versus V of normal-metal-high- $T_c$ ceramic point contacts. We have found that the measured curves can be interpreted by considering two effects: an asymmetric tunneling barrier and a set of distinct superconducting energy gaps. We show that the BCS density of states, modified by introducing a Gaussian discrete distribution of gap values, yields a very good fit of the experimental curves.

Tunneling measurements have been one of the most widely performed tests on high- $T_c$  superconducting ceramics. However, in these materials, the results have been more difficult to interpret than in conventional low- $T_c$  superconductors. In fact, a large number of effects have been introduced to explain the data, such as charging effects<sup> $1-3$ </sup> and modifications in the standard BCS density of states<sup>4,5</sup> in order to account for an equally large number of nonideal features observed in the  $dI/dV$ versus V curves of normal-metal-high- $T_c$  superconducting ceramics (X-HTSC) point contacts.

In this paper we present measurements of the differential conductance  $dI/dV$  of N-HTSC point contacts and we show that the curves can be fitted quite well by assuming an asymmetric tunneling barrier and a BCS density of states with a discrete distribution of values of superconducting energy gap. The measurements were performed on ceramic samples of  $YBa_2Cu_3O_7$  and we used either copper or tungsten as normal contacts.

The  $YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub>$  pellets were prepared by the wellknown mixing-grinding-pressing-annealing method. The Cu points were fabricated by sandpapering a copper wire. The W points were prepared by chemical etch of tungsten wires in a solution of NaOH. The contact was achieved by pressing, by means of a screw, the metal point on the pellet. In this manner, contacts with resistances at room temperature ranging from 10 to 1000  $\Omega$  were obtained.

Two wires, one for the current and one for the voltage, were attached to each of the metal point and the pellet. The I-V curves were taken at fixed temperatures above and below  $T_c$ , controlled by a temperature controller. The current, provided by a programmable current source, was swept at discrete steps (between 500 and 1000 steps) while the voltage values were measured 100 times and averaged for each current value. The differential conductance was then calculated numerically.

Figure 1 represents the differential conductance  $dI/dV$ for three of our contacts, called samples 1, 2, and 3, respectively. We can distinguish several common features present in all our contacts. First, we observe a background conductance, with the same shape at all temperatures. We have fit this conductance to a polynomial expression and we have found that the voltage dependence of  $dI/dV$  is very close to a parabola. This is true for curves taken both above and below  $T_c$  down to the lowest temperature. This can be seen in Fig. 2 where we plot  $dI/dV$  for a Cu-Y-Ba-Cu-O contact (sample 1) at  $T=110$  K. The parabolic shape of  $dI/dV$  versus V curves is one of the basic properties of good tunnel junctions<sup>6</sup> (the other being a negative  $dR/dT$  above  $T_c$  which was also observed in the samples reported in this paper).

Another feature present in all the curves is that the background conductance is not symmetric with respect to the voltage, in fact, the minimum of the parabola is always shifted towards the side where the pellet is positive with respect to the metal point. For the curves presented in this paper,  $dI/dV$  has its minimum for approximately  $V = -10$  mV for the copper points and for  $V = -1$  mV for the tungsten points. This asymmetry is independent of the temperature and therefore it is intrinsic to the barrier and not to the state either normal or superconducting of the electrodes. This asymmetry has been previously observed in  $N$ -HTSC point contacts<sup>1,2,7</sup> and it has been ascribed to rectifying effects<sup>1,2</sup> at the interface between the semiconducting phase and the superconducting phase of the Y-Ba-Cu-0 compound not only close to the normal point but throughout the sample.

A parabolic conductance can be derived from a WKB approximation. In fact, for tunneling between normal metals, this approximation yields  $I-V$  curves that are linear with small cubic corrections at low voltages, becoming exponential at high voltages.<sup>8</sup> The offset to the parabolic dependence of  $dI/dV$  is the result of an asymmetric barrier and can be obtained by numerical calculations. $9$  The origin of this asymmetric parabolic background conductance is, therefore, independent of the superconducting state and it contributes to the total conductance in the same manner above and below  $T_c$ . Consequently, this background conductance should be subtracted from the measured curves in order to enhance the features that are intrinsic to the superconducting properties of the ceramic. The remaining curves (clean curves), which are now symmetric, preserve all the features inherent to the superconducting properties of the samples.

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FIG. 1. Differential conductance of three contacts: (a) Sample 1, Cu point, at  $T=6$  K; (b) sample 2, W point, at  $T=4$  K; (c) sample 3, Cu point, at  $T=6$  K. The solid lines represent the experimental data, the dashed lines are the corresponding polynomial fits. Note that the units in the vertical axis of (a) and (c) are  $10<sup>3</sup>$  times larger than the units of (b).



FIG. 2. Parabolic conductance measured in sample <sup>1</sup> above  $T_c$ ,  $T=110$  K.

(b) These curves exhibit a large number of peaks in contrast to the smooth conductance curves of N-HTSC point contacts found in the literature, where the superconductor is either a single crystal<sup>10,11</sup> or a thin film.  $12,13$ 

> We have fitted our curves to a BCS density of states, modified by introducing a discrete Gaussian distribution of energy gaps, namely,

$$
\rho(E) \propto \sum_{\Delta} \frac{E}{(E^2 - \Delta^2)^{1/2}} e^{-(\Delta - \Delta_0)^2/2\sigma^2}, \qquad (1)
$$

where E is measured relative to the Fermi level, and  $\Delta_0$  is chosen between  $3.5k_BT_c$  and  $5k_BT_c$ , which is the most widely reported range for the energy gap of this compound. In order to write (1) we are assuming that the electrons are tunneling into many regions with different gap values, due to either local variations of the temperature or to the fact that the metal point is touching many grains, each one formed by many monocrystals with various orientations relative to the metal point. This effect was suggested by Pan et  $al.$ <sup>4</sup> By taking three different gap values they obtained only a fair fit to their experimental curves.

Both the modified experimental curves [curves shown in Figs. 1(a) and 1(b) after the parabolic background has been subtracted] and the curves calculated using (1) are shown in Fig. 3. The insets represent the corresponding discrete distribution of energy-gap values used in each one of the fits. The number of terms in (1) that yielded the best fits was around 200. The values of  $\Delta_0$  are 28 meV for sample <sup>1</sup> and 32 meV for sample 2 and the values of  $\sigma$  are 10.2 meV for sample 1 and 11.3 meV for sample 2.

A Gaussian distribution of energy gaps has been previously used by Kirtley et  $al$ .<sup>5</sup> By using a continuous distribution they obtained smooth curves, in contrast to our results in which even the peaks observed in the experiments are reproduced theoretically.

The main difference between the measured curves and the fit is the peak at zero bias observed in the low resis-





FIG. 3. Comparison of the experimental curves (continuous lines) and the curves calculated by using expression (1) (dashed lines). The experimental curves (a) and (b) correspond to the curves shown in Figs. 1(a) and 1(b), respectively, after the parabolic conductances have been subtracted. The insets represent the corresponding Gaussian distributions used for each fit.

tance contacts such as samples 1 and 3 [note that the units in the vertical axis of Figs. 1(a) and 1(b) are  $10<sup>3</sup>$ times larger than the units of Fig. 1(b)]. This peak has been associated with Andreev reflection processes at the interface in contacts with metallic character.

In some of our samples and at certain temperatures, we observed periodic structures and we found that they can be reproduced (after the subtraction of the parabolic background) by using expression (1) with a small number of energy-gap values. Figure 4 shows one experimental curve and the plot of expression (1) in which only six terms were taken.

When we do the subtraction of the parabolic background from the total conductance, we are assuming that the total current can be written as  $I(V) = N_1(V) + S_1(V)$ , where  $S_1(V)$  contains all the dependence of the tunneling



FIG. 4. (a) Conductance  $dI/dV$  for sample 1, measured at 35 K, exhibiting periodic structures and (b) curve calculated by taking only six terms in expression (1).

current on the superconducting properties of the electrodes.

Among other possible alternatives to extract the information on the superconducting properties of the electrodes, we could choose either that the current is the product of two functions  $I(V) = N<sub>2</sub>(V)S<sub>2</sub>(V)$ , or that the conductance  $dI/dV$  can be written as the product  $(dN_3/dV)(dS_3/dV)$  where  $S_2(V)$  and  $S_3(V)$  have in each case all the information concerning the superconducting properties of the electrodes. We have done both calculations and we have found that the shapes of the curves  $dS_1/dV$ ,  $dS_2/dV$ , and  $dS_3/dV$  versus V are practically identical. Further measurements and theoretical calculations of the voltage dependence of the tunneling current are needed in order to determine which of the alternatives is the closest to reality.

In summary, we have performed measurements of the differential conductance  $dI/dV$  versus V of normalmetal-YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7</sub> point contacts and we have found that after subtracting the asymmetric parabolic conductance, the remaining curve can be fitted remarkably well by using a BCS density of states modified by a Gaussian discrete distribution of gap values.

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<sup>1</sup>R. Medina, J. Aponte, and M. Octavio, Physica B 165-166, 1595 (1990).

- <sup>2</sup>M. Octavio, R. Medina, and J. Aponte, in Progress in High Temperature Superconductivity. Transport Properties of Su perconductors, edited by Roberto Nicolsky (World Scientific, Singapore, 1990), p. 357.
- <sup>3</sup>P. J. van Bentum, H. van Kempen, L. E. C. van de Leemput, and P. A. A. Tunissen, Phys. Rev. Lett. 60, 369 (1988).
- 4S. Pan, K. W. Ng, A. L. de Lozanne, J. M. Tarascon, and L. H. Greene, Phys. Rev. B35, 7220 (1987).
- <sup>5</sup>J. R. Kirtley, R. M. Feenstra, A. P. Fein, S. I. Raider, W. J. Gallagher, R. Sandstrom, T. Dinger, M. W. Shafer, R. Koch, R. Laibowitz, and B. Bumble, J. Vac. Sci. Technol. A 6, 259 (1988).
- <sup>6</sup>John M. Rowell, Supercurrents 11, 1 (1991).
- 7M. E. Hawley, K. E. Gray, D. W. Capone, and D. G. Hinks,

Phys. Rev. B35, 7224 (1987).

- 8E. L. Wolf, Principles of Electron Tunneling Spectroscopy (Oxford University Press, London, 1985).
- <sup>9</sup>W. F. Brinkman, R. C. Dynes, and J. M. Rowell, J. Appl. Phys. 41, 1915 (1970).
- M. Gurvitch, J. M. Valles, Jr., A. M. Cucolo, R. C. Dynes, J. P. Garno, L. F. Schneemeyer, and J. V. Waszczak, Phys. Rev. Lett. 63, 1008 (1989).
- <sup>11</sup>J. R. Kirtley, R. T. Collins, Z. Schlesinger, W. J. Gallagher, R. L. Sandstrom, T. R. Dinger, and D. A. Chance, Phys. Rev. B35, 8846 (1987).
- <sup>12</sup>J. Takada, T. Terashima, Y. Bando, H. Mazaki, K. Iijima, K. Yamamoto, and K. Hirata, Phys. Rev. B 40, 4478 (1989).
- <sup>13</sup>I. Iguchi and Z. Wen, IEEE Trans. Mag. MAG-27, 3102 (1991).