PHYSICAL REVIEW B VOLUME 36, NUMBER 1 1 JULY 1987

Tunneling measurements on the high-T_c superconductors $La_{1.85}Sr_{0.15}CuO_{4-\delta}$ and $YBa_2Cu_3O_{7-\delta}$

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(Received 13 April 1987)

We have measured the superconducting properties of the new high-T_c superconductors $La_{1.85}Sr_{0.15}CuO_{4-\delta}$ and $YBa_2Cu_3O_{7-\delta}$ by electron tunneling. The measured *I-V* curves can be described neatly with simple tunneling theory, using the Bardeen-Cooper-Schrieffer density of states for the superconductor. For the La-Sr compound we find an energy gap ranging between 10 and 20 meV. By exerting a large pressure on the sample the gap value could be increased to 35 meV. From these measurements we extract $2\Delta/k_BT_c=3.5-4$. For the Y-Ba compound we find a pressure-independent gap $2\Delta=23\pm2$ meV or $2\Delta/k_BT_c=3.2\pm0.4$.

The new class of superconductors described for the first time by Müller and Bednorz[1] poses a new challenge to solid-state physics. The elucidation of the underlying mechanism for the extremely high critical temperatures is of the utmost importance for progress in this new field of research. A basic, and as yet unanswered, question is whether or not the BCS theory[2] is applicable to describe the fundamental properties of these superconductors. Recent far-infrared reflection measurements[3] of $La_{1.84}Sr_{0.16}CuO_4$ yielded a ratio E_g/k_BT_c of 1.8, substantially below the BCS prediction $2\Delta/k_BT_c=3.53$. In contrast, far-infrared transmission measurements[4] indicate a ratio $E_g/k_BT_c\approx5$. It must be noted, however, that both results allow for an appreciable uncertainty in E_g.

One of the most powerful tools to study the density of states in the superconducting state is electron tunneling. In addition, tunneling might provide detailed information on the electron-phonon coupling parameters. In this paper we report measurements of the energy gap, using a piezo-driven microprobe which is quite similar to a scanning-tunneling microscope.[5] Using a relocatable microprobe has the advantage that the properties of individual grains can be measured, which is especially useful in inhomogeneous superconductors with multiple phases, as is often the case in these sintered ceramic samples. In addition, the microprobe can be used to apply a large local pressure, so that pressure-induced changes of the gap can be studied.

We prepared the La-Sr compounds in a way similar to that described in Ref. 6. The critical temperature $T_c=33$ K was taken as the point where the resistance was 0.5 of the normal state resistance R_N. The tunneling measurements were done on samples with a narrow resistive transition ($0.2R_N$ to $0.8R_N$ less than 2 K). The onset of superconductivity occurs near 38 K. Measurements in high magnetic fields (up to 15 T) confirmed that the samples consisted predominantly of only one phase.[7] The Y-Ba compounds were made in different compositions. Samples with a nominal composition given by $Y_{1.2}Ba_{0.8}CuO_4$ as described in Ref. 8 yielded rather broad transitions ($\Delta T>10$ K) with the onset of superconductivity near 90 K. Measurements in high magnetic fields showed that these samples consisted of at least two phases with different critical temperatures.

Very sharp transitions were obtained with the nominal composition $YBa_2Cu_3O_{7-\delta}$. The oxygen content was determined by gravimetric measurements during the annealing process. The critical temperature ($0.5R_N$) varied from 84 to 91.2 K, depending on the annealing temperature and oxygen deficiency. The highest T_c was obtained for $\delta\approx0.7$. The 20%–80% transition of the best samples occurred within 1.4 K. High magnetic field measurements up to 25 T confirmed that at least 80% of these samples were of a single phase.[7]

We will first describe the experimental results on $La_{1.85}Sr_{0.15}CuO_{4-\delta}$. The tunnel junctions were formed by gently lowering either a superconducting Nb:Zr or a normal-tungsten whisker onto the oxidized surface of the sample. The coarse adjustment was made by deforming a steel base plate, using a differential screw mechanism. Fine adjustment in the *x*, *y*, and *z* direction was made possible with a set of piezoelectric crystals, allowing a scan region of $\pm1\ \mu m^3$. The entire setup was immersed in a pumped liquid-helium bath at 1.2 K. *I-V* characteristics were taken by sweeping the bias voltage. The use of a digital storage oscilloscope allowed us to scan the relevant voltage range in typically 0.5 s. No curve smoothing or digital averaging was used.

Figure 1 represents a typical *I-V* characteristic of a W-$La_{1.85}Sr_{0.15}CuO_{4-\delta}$ junction. The superconductive gap structure is clearly visible. The solid dots are calculations of the basic tunneling integral, where we used the BCS density of states for the superconductor. The order parameter Δ was treated as a fit parameter. The calculations in Fig. 1 were done with $\Delta=7.6$ meV. The good general agreement indicates that this new class of high-T_c superconductors can be readily described by the elementary BCS equations for the density of states.

Small deviations from theory are visible at low voltages. This is probably due to electromagnetic disturbances, for which these very high impedance, low capacitance junctions are extremely sensitive. Also, the whisker acts as an antenna for very high frequencies, which leads to a rounding of the *I-V* curves.

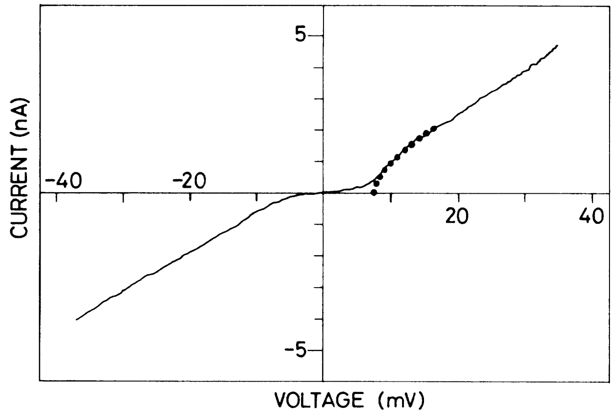

FIG. 1. *I-V* characteristic of a superconducting-normal tunnel junction between a tungsten microprobe and a $La_{1.85}Sr_{0.15}CuO_{4-\delta}$ sample. The solid dots are calculations using the BCS density of states and $\Delta = 7.6$ meV.

The slightly asymmetric shape of the curve is also noteworthy. This is probably due to the local density of states at the tip, as is commonly observed in scanning-tunneling microscopy. In addition to the expected superconducting-normal tunneling we frequently find evidence for superconducting-superconducting tunneling as well. A typical example is reproduced in Fig. 2. We propose that in this case one measures the tunneling between two grains, separated by an oxide layer. Again, the solid dots are calculated using the BCS density of states, but now for both sides of the junction. The order parameter for both grains was taken to be 8.7 meV.

The values for the order parameter, measured at different positions on the sample and for different samples of the same composition were found to vary from 5 to 10 meV. The resistance measurements, however, do not indicate an associated variation of T_c for these samples. We therefore propose that a large portion of the scattering is due to the local pressure exerted by the tip on the surface. Although the force on the surface is very small, the corresponding microscopic contact area does lead to a nonnegligible pressure. Postmortem examination of the tip indicates that the deformation pressure of tungsten whiskers, approximately 100 kbar,[9] can easily be reached.

In Fig. 3 we have displayed a set of *I-V* characteristics where we have deliberately increased the pressure P at the junction area. The whisker was formed in an S shape, with an experimentally determined spring constant of $8.6 \pm 0.3 \times 10^{-7}$ N/nm. The voltage over the piezo is directly proportional to the pressure at the contact area. Although the normal parallel tunneling prohibits a detailed comparison with theory, a clear gap structure is visible which shifts to higher voltages with increasing pressure. The inset shows the width of the gap as a function of the piezo displacement. At low pressures we find a linear increase of the gap. At very high pressure, a saturation takes place, which is probably due to the deformation of the tungsten probe. Assuming a deformation limit for the tungsten whiskers of $P_d = 100$ kbar, we obtain a contact area of 0.07 μm^2, which is reasonable for the probes that we used. In this way we can estimate the pressure dependence of T_c, yielding $dT_c/dP \approx 0.7$ K/kbar, in good agreement with the results of Chu *et al.*[10] It would be interesting to measure superconducting properties of these samples at high temperatures to check whether there is a related increase of the critical temperature. Our present setup is not well suited for this purpose due to thermal drifts.

The measured *I-V* characteristics frequently showed additional structure at higher bias voltages. Reproducible

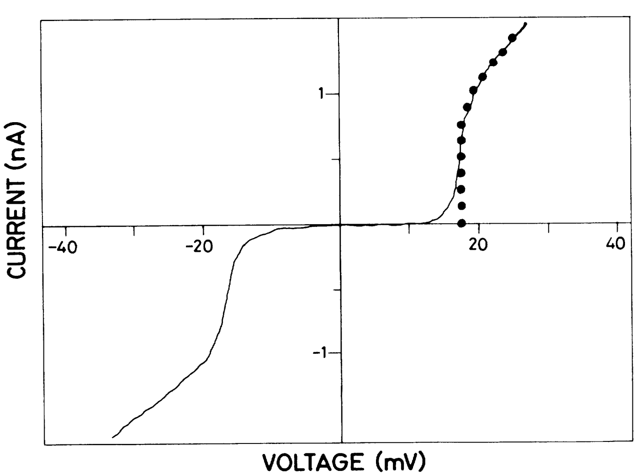

FIG. 2. *I-V* characteristic of a superconducting-superconducting tunnel junction between two grains embedded in a $La_{1.85}Sr_{0.15}CuO_{4-\delta}$ sample. The solid dots are calculations using the BCS density of states and $\Delta = 8.7$ meV.

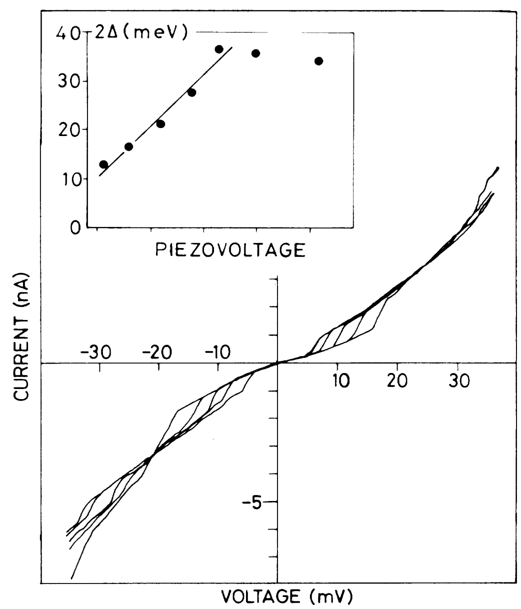

FIG. 3. *I-V* curves for a normal-superconducting junction for different values of the pressure, exerted by the microprobe. The inset shows the energy gap 2Δ as a function of the voltage over the piezo, which is proportional to the force exerted by the tip.

steps were observed at multiples of the gap energy. As yet, no reproducible structure due to the electron-phonon interaction could be determined.

As pointed out by Hor *et al.*,[11] the high-T_c compound Y-Ba-Cu-O has a much smaller pressure dependence of the critical temperature. This is confirmed by our tunneling measurements on a $YBa_2Cu_3O_{7-\delta}$ sample, which gives a rather coherent picture of the energy gap and density of states, independent of pressure. In Fig. 4 we have plotted three *I-V* curves, characteristic for these samples. Figure 4(a) represents a superconducting-normal tunneling junction between the tungsten whisker and the superconducting sample. Again, the solid dots are calculations of the elementary tunneling integral, using the BCS density of states, and $\Delta = 11.5$ meV. Figure 4(b) shows a superconducting-superconducting junction between grains embedded in the sample. The solid dots represent a fit to the data using the same value for $\Delta_1 = \Delta_2 = 11.5$ meV as in Fig. 4(a). Finally, in Fig. 4(c) we have plotted a typical *I-V* curve, where the dc-Josephson effect dominates the structure. This is a clear indication that we are dealing with a superconducting-superconducting junction. The values for the order parameter were found to vary between 10 and 13 meV. With $T_c = 84$ K $(0.5R_N)$, we get for the ratio $2\Delta/k_BT_c = 3.2 \pm 0.4$.

In conclusion, we find that the new class of high-T_c superconductors have an energy gap and a density of states that is, within our experimental accuracy, consistent with the BCS theory.

The tunneling results on $La_{1.85}Sr_{0.15}CuO_{4-\delta}$ indicate an energy gap ranging from 10 to 20 meV. The measured gap can be enhanced by increasing the local pressure at the contact area between tip and sample, with an upper limit of approximately $2\Delta_{max} = 35$ meV. For the ratio $2\Delta/k_BT_c$, we find values ranging between 3.5 and 7. When the pressure is increased intentionally, the apparent $2\Delta/k_BT_c$ ratio can be as high as 12. With our technique, a moderate amount of pressure on the surface cannot be avoided. Therefore we believe that the lower bound is the more reliable value, yielding $2\Delta/k_BT_c = 3.5$ to 4.

For the $YBa_2CuO_{7-\delta}$ compound we find an energy gap $2\Delta = 23 \pm 2$ mV, which is independent of pressure, and a ratio $2\Delta/k_BT_c = 3.2 \pm 0.4$. These gap-$T_c$ ratios do not indicate an exceptionally strong electron-phonon coupling for the new class of high-T_c superconductors.

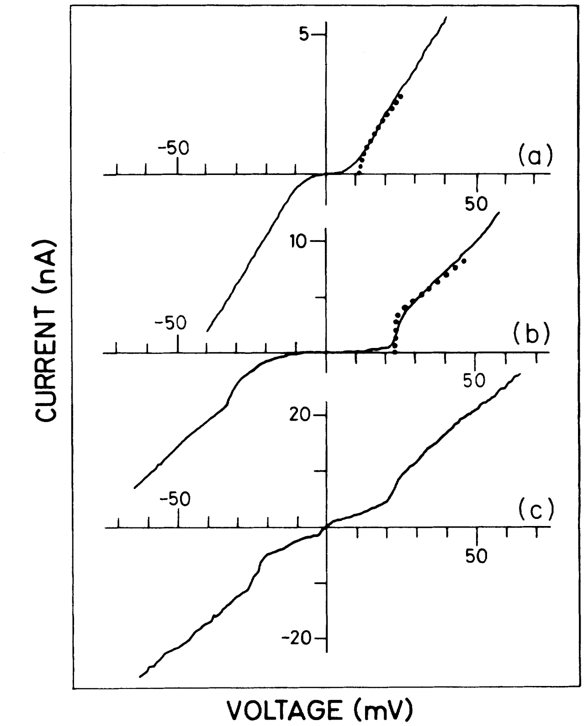

FIG. 4. Tunneling characteristics of a $YBa_2Cu_3O_{7-\delta}$ compound with $T_c = 84$ K. (a) Superconducting-normal junction between the tungsten microprobe and the Y-Ba-Cu-O sample. The solid dots are calculations from the BCS theory with $\Delta = 11.5$ meV. (b) Superconducting-superconducting junction between two grains embedded in the sample. The solid dots are calculations from the BCS theory with $\Delta = 11.5$ meV. (c) Superconducting-superconducting junction showing the dc-Josephson effect.

Useful discussions with R. A. de Groot and J. C. Fuggle are gratefully acknowledged. We also acknowledge the technical support of J. G. H. Hermsen and J. W. Gerritsen. This work was part of the research program of the Stichting voor Fundamenteel Onderzoek der Materie (FOM) and was made possible by financial support from the Nederlandse Organisatie voor Zuiver Wetenschappelijk Onderzoek (ZWO) and from Nederlandse Philips Bedrijven B.V.

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