Effects of quasiparticle bound states in the tunneling characteristics of $YBa_2Cu_3O_7$ readjustable junctions

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By carefully adjusting the distance between the two fractured electrodes, we have investigated the in-plane conductance characteristics of $YBa_2Cu_3O_7$ break junctions in different tunneling regimes. In the *q-p* regime we have found that the conductance spectra strongly depend on the misorientation angle between the tunneling current and the $YBa_2Cu_3O_7$ crystal axis. When the tunneling current along the *a-a* or *b-b* axis was favored, conductance maxima and shoulders around ±40 and ± 20 mV have been measured at low temperatures. For different misorientation angles, we have found the presence of a zero bias conductance peak (ZBCP) that develops at the junction T_c as well as gap related structures at about ± 20 mV. The amplitude of the ZBCP diverges as $1/T$ for temperatures below 40 K. All these features are consistent with a *d*-wave model for the high-T_c superconductors.

I. INTRODUCTION

Recently, many tunneling experiments have been devoted to the study of the symmetry of the pair potential in the high-T_c superconductors (HTS), since important information can be achieved to elucidate the mechanism responsible for superconductivity in these materials. Indeed, tunneling spectroscopy is sensitive to the material properties close to the surface and results in being very effective when studying the anisotropic structure of the superconducting order parameter. In early approaches, quantum phase interference devices were used to directly probe the unconventional symmetry of the superconducting state since Josephson coupling directly probes the phase difference across the boundaries.[1–4] The majority of these experiments, including SQUID interferometer, corner Josephson junction, and grain boundary flux quantization measurements, seem to indicate a predominant $d_{x^2-y^2}$ wave symmetry in these compounds.[5]

On the other hand, in the *d*-wave model for cuprate superconductors, surface bound states are predicted that can be observed as a zero bias conductance peak (ZBCP) in the HTS/*I*/normal-metal junction characteristics for *a-b* plane tunneling.[6–9] Quasiparticle bound states appear in the region where the bulk order parameter is suppressed or changes sign with momentum direction. The presence of these zero-bias anomalies (ZBA) has been reported in point contact[10] and scanning tunneling microscopy (STM) measurements[11] as well as in film-oriented $YBa_2Cu_3O_7$ junctions.[12–14] Splitting of the low-energy peak has been observed in magnetic fields of a few tesla or in zero field at low temperatures.[15] Recently, one of the authors of the present study (A.M.C.) has discussed the different behavior of the ZBCP due to unconventional symmetry of the superconducting order parameter or to Kondo-type scattering from isolated magnetic moments at the tunnel barrier.[16]

It has been reported that the sign change responsible for the ZBCP in oriented HTS/*I/N*junctions is also responsible for the π phase shift in the Josephson interference experiments.[5,17] Moreover, theoretical analyses have studied the relation between the Josephson critical current and the presence of the zero-energy peak in the *q-p* density of states (DOS) of anisotropic superconductors. Indeed, the midgap energy states lead to an anomalous behavior of the Josephson current at low temperatures. It has been predicted that the singularity at low energies dominates the dc Josephson effect with a rapid $1/T$ increase of the current amplitude for decreasing temperatures.[18,19] Recently, these studies have been extended and the dependence of the anomaly on the barrier roughness and transparency as well as on the misorientation angle between the tunneling current and the *a, b* crystal axes has been studied.[20,21]

In this paper, we show that the ZBCP expected for tunneling along the *a-b* planes is indeed observed in the conductance characteristics of readjustable break junctions realized with highly oriented $YBa_2Cu_3O_7$ films. We have found that the effect depends on the fracture direction with respect to the $YBa_2Cu_3O_7$ *a,b* axes and for temperatures less than 40 K we have measured a $1/T$ divergence of the ZBCP amplitude.

II. SAMPLE PREPARATION AND CHARACTERIZATION

Due to the low controllability of the HTS surfaces and boundaries, it is difficult to realize high quality HTS-*I*-HTS junctions and in the literature, no numerous examples of *a,b* tunneling in artificial heterostructures can be found. To avoid interface problems and to favor the *ab*-plane contribution to the tunneling current, we have used a different approach. We report here the extension of the break junction technique, earlier applied to break polycrystals or single crystal samples, to epitaxial $YBa_2Cu_3O_7$ thin films. This technique pioneered by Moreland[22,23] is particularly appropriate when dealing with HTS. Indeed, junctions are realized at low temperatures with freshly fractured surfaces and an inert tunnel barrier is created by helium gas or liquid. In addition to this, different tunneling regimes can be investigated by mechani-

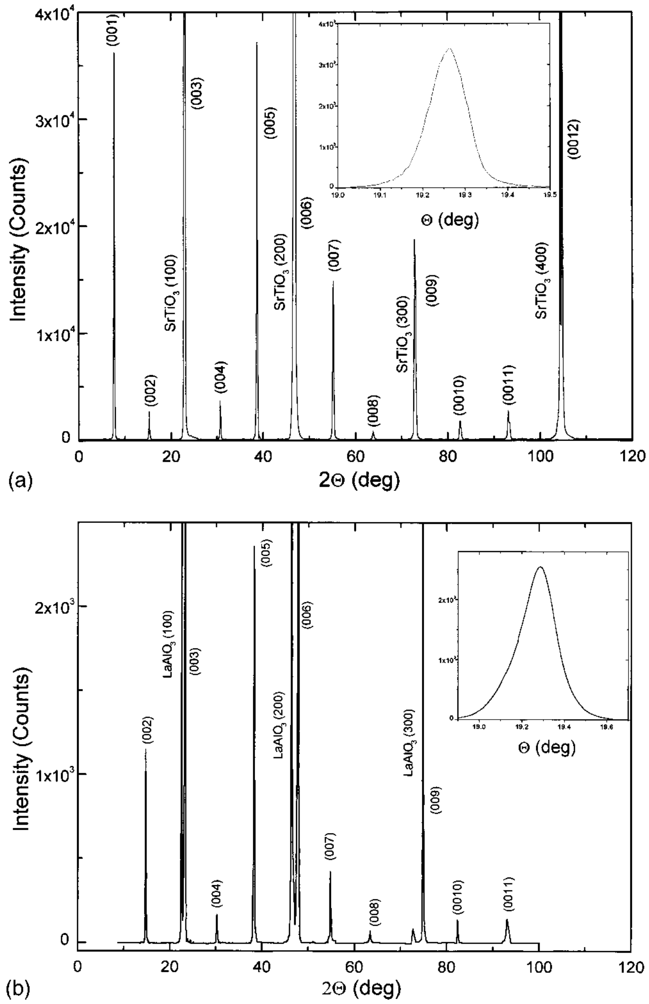

FIG. 1. X-ray, θ-2θ diffraction patterns and rocking curves of the (005) peaks for two $YBa_2Cu_3O_7$ films on a $SrTiO_3$ (a) and a $LaAlO_3$ (b) substrate.

cally adjusting the distance between the two electrodes.[24]

The c-axis-oriented $YBa_2Cu_3O_7$ films used for this study were deposited on (100)-oriented $SrTiO_3$ or $LaAlO_3$ substrates by a high oxygen pressure, dc sputtering technique. Details on the fabrication process and on the growth conditions have been reported elsewhere.[25] The structural and electrical characterization confirmed equivalent high-quality properties of the films on both substrates. As an example, in Figs. 1(a) and 1(b) we report the x-ray diffraction patterns of two $YBa_2Cu_3O_7$ films deposited on different substrates. Only (001) reflections are observed with the peaks of major intensity corresponding to the $SrTiO_3$ and $LaAlO_3$ substrate reflections, respectively. The rocking curves of the (005) peaks show a FWHM $\leqslant 2°$, indicating a strong c-axis orientation in both cases.

Only high-quality $YBa_2Cu_3O_7$ films with $T_c(\rho=0)=91\,\mathrm{K}$ and $\Delta T \leqslant 0.5\,\mathrm{K}$ were selected to realize the break junctions. The film thicknesses were about 1500 Å and photolithography was used to reduce to 100 μm the junction width across the fracture. Four in-line Ag contacts were thermally evaporated for current and voltage contacts and gold leads were attached by indium soldering. In our experiments, the resistive T_c was monitored during the sample cooldown and the substrate and film fractures were produced at temperatures below 20 K by applying an external force parallel to the c axis, by means of a differential screw.

III. DIFFERENT TUNNELING REGIMES

By fine adjustment of the mechanically controlled gap between the two pieces of substrate, we have obtained highly stable, readjustable junctions. In the majority of our samples the evolution from the weak-link to q-p tunneling regime and vice versa was clearly observed in a range of variation of the junction resistance of 10^4 order of magnitude. For higher resistance junctions, the normal-state resistance R_N changed about 15% in the temperature range between 4.2 and 100 K. For low resistance junctions, the temperature stability was more critical. In the majority of the junctions the fracture direction, inspected by scanning electron microscopy (SEM), was parallel to one of the substrate edges.

A. Weak-link regime

In the weak-link regime, normal junction resistance less than 1 Ω was obtained and point-contact-like tunneling characteristics were measured. In contrast to the case of a heliun barrier between the two electrodes, weak contacts between the two pieces of film were much easier to achieve; however, in these cases neither effective reproducibility with changes of the contact pressure nor a good thermal stability of the data was obtained.

In Fig. 2(a), we show the $G(V)$ vs V curve of a $YBa_2Cu_3O_7$ junction at $T=13\,\mathrm{K}$. No gap-related structures (GRS) are observed, while a highly asymmetric, increasing background for high biases and a huge ZBCP are measured. This last feature develops soon below T_c. The width of the low bias anomaly is less than 1 mV and the relative amplitude is more than 200% at the reported temperature.

In Fig. 2(b), the low bias I-V curves are reported for the same junction and a suppression of the critical current in a magnetic field $H=1$ G is observed. Such a low value of the modulation field seems to indicate that we are dealing with the Josephson effect; however, we do not have a definitive explanation for the finite slope of this feature. Indeed also in Ref. 24 a finite slope of the critical current was observed for Nb-based break junctions but the question remained about the mechanism responsible for this effect. Due to the thermal instability of the contact junctions and to the resistive contribution to the low bias tunneling current, a quantitative comparison between these experiments and the predicted Josephson temperature behavior[18,19] does not appear very meaningful.

B. Quasiparticle tunneling regime

For these junctions, with resistances ranging between a few Ω and $10^4\,\Omega$, we have found that the tunneling characteristics strongly depend on the misorientation angle between the tunneling direction and the $YBa_2Cu_3O_7$ crystal axis. Indeed in Refs. 9, 26, 27 a strong dependence of the a-b plane, quasiparticle conductance spectra on the misorientation angle α between the normal to the interface and the crystalline axes is predicted. For $\alpha=0$, no zero bias anomalies are formed since there are no zero energy bound states at the material surface. For increasing misorientation angles, more

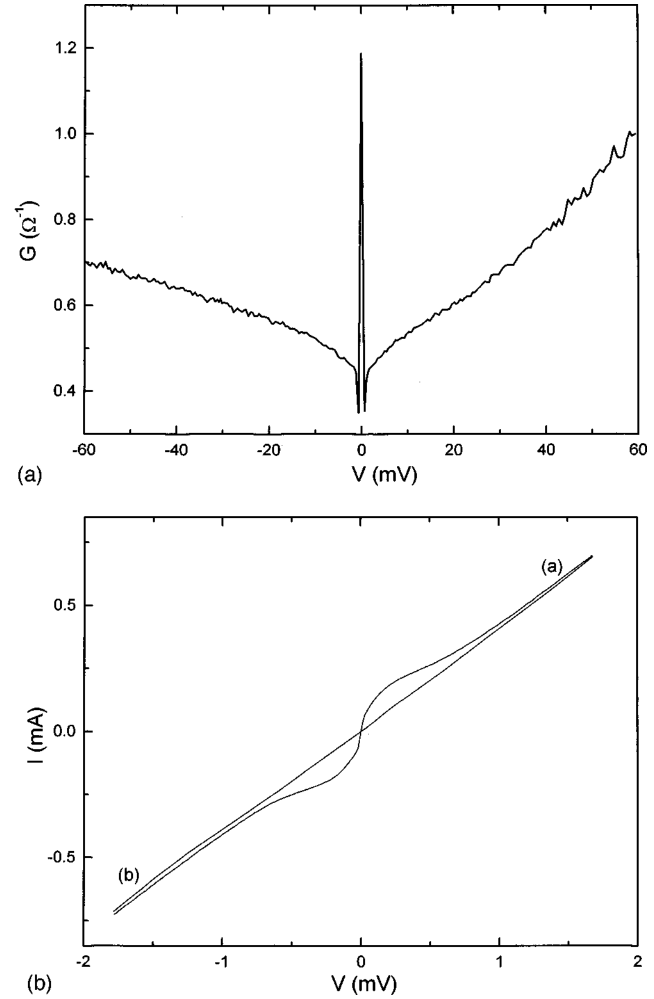

FIG. 2. (a) The $G(V)$ vs voltage characteristic at 13 K for a $YBa_2Cu_3O_7$ break junction in the weak-link regime. (b) The I-V characteristics for the same junction in magnetic fields (a): $H=0$ G and (b): $H=1$ G.

and more bound states are created and the ZBCP amplitude shows a maximum for $\alpha=\pi/4$.

In order to determine the lateral lattice alignment of our films, the x-ray pole figure analysis was used.[28] By this technique, the relative azimuthal angular relation between the film and the substrate can be obtained to quantify the in-plane misorientation of the samples. In Figs. 3(a) and 3(b) as an example, the (101) pole figure of a $SrTiO_3$ substrate and the (103) pole figure of a $YBa_2Cu_3O_7$ film are reported. These data confirm the biepitaxiality of our films and the alignment between the crystal axis and the substrate edges.

Unfortunately, after breaking, from the above information and from the knowledge of the fracture direction, we can only estimate an ''average misorientation angle'' since, due to the microscopic faceting of the fracture, different orientations are present at the junction interface. Indeed, the microscopic random distribution of the grain orientation at the interface has been reported also for grain boundary junctions.[29] This fact, together with the intrinsic irreproducibility of the tunnel barrier for different break junctions, prevented us from carrying out a quantitative analysis of the dependence of the ZBCP amplitude on the misorientation angle. Nevertheless, important information about the symmetry of the superconducting order parameter can be achieved from the study of the differences and similarities of the conductance characteristics in the two limiting conditions, that is, for tunneling close to the *a-a* (*b-b*) axis, $\alpha\simeq 0$, and for tunneling close to the node directions, $\alpha\simeq\pi/4$.

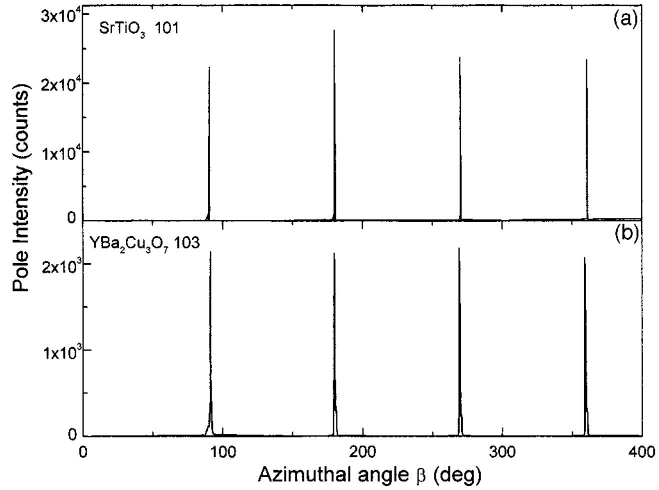

FIG. 3. X-ray line profiles of (101) poles for a $SrTiO_3$ substrate (a) and of the (103) poles for a $YBa_2Cu_3O_7$ film (b).

1. Tunneling along the a-a (b-b) directions

In our previous experience of tunneling in HTS, we have studied both HTS/*I*/N and HTS/*I*/HTS junctions. The $YBa_2Cu_3O_7/I/Pb$ planar junctions with natural barriers[30,31] and the $YBa_2Cu_3O_7/PrBa_2Cu_3O_7/YBa_2Cu_3O_7$ heterostructures[32,33] showed consistent conductance maxima around $\pm$ 20 and $\pm$ 40 mV, respectively. Indeed, in the present study we have found conductance curves that show similar structures for misorientation angles close to $\alpha=0$.

In Fig. 4, an example of this kind of $G(V)$ vs V characteristics is reported that shows conductance maxima at $T=5$ K and shoulders around $\pm$ 40 and $\pm$ 20 mV. An increasing background for high biases and a small ZBCP at low energies is also observed. The amplitude of the anomaly is

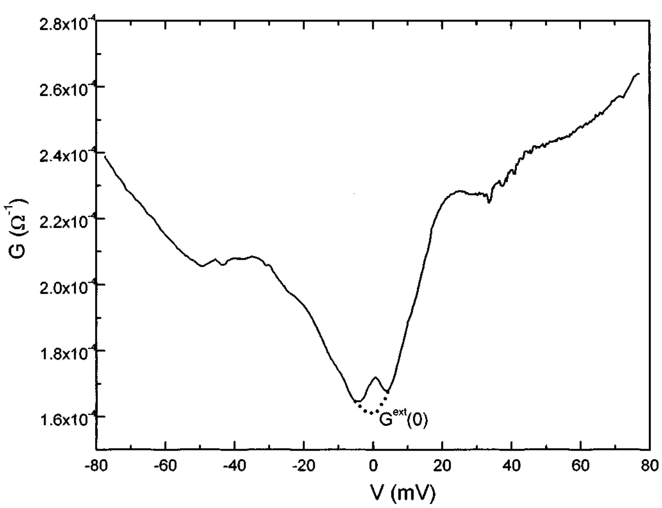

FIG. 4. The $G(V)$ vs voltage characteristic at $T=5$ K for a $YBa_2Cu_3O_7$ break junction in the q-p tunneling regime; probability of a-a(b-b) tunneling was maximized in this junction.

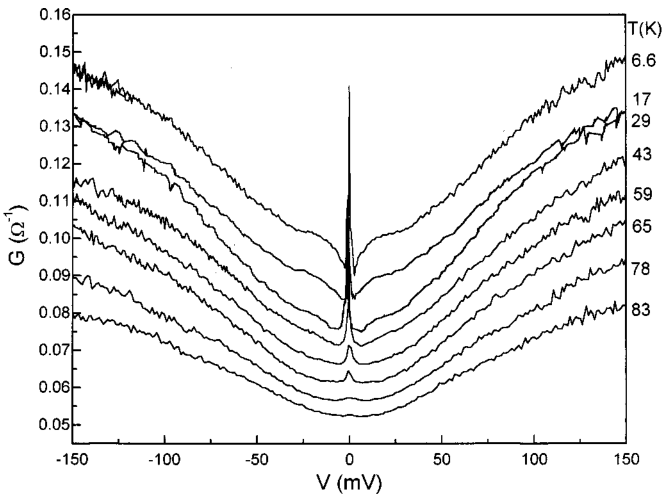

FIG. 5. The $G(V)$ vs voltage characteristics at different temperatures for a $YBa_2Cu_3O_7$ break junction in the *q-p* tunneling regime; probability of tunneling close to the node directions was maximized in this junction. The conductance scale refers to the lowest temperature data and the other curves have been shifted for clarity.

about 6% when compared with the extrapolated conductance value at zero bias, $G^{ext}(0)$, defined in the figure. The normal junction resistance, $R_N(100\,\mathrm{mV})=3700\,\Omega$, indicates that we were operating in the *q-p* tunneling regime in which there is physical separation of the two electrodes and the Josephson coupling is suppressed.[24] The data of Fig. 4 are consistent with the *d*-wave model for HTS, if small misorientation angles around $\alpha=0$ are supposed on a microscopic scale. A few junctions, with similar misorientation, showed featureless conductance characteristics for which *a-b* tunneling can be supposed.

2. Tunneling close to the node directions

We have obtained a different kind of $G(V)$ vs V characteristic on samples that from the ϕ-scan analysis showed intermediate misorientation angles (close to $\pi/4$). In Fig. 5, we report the tunneling spectra of this kind of film for different temperatures. For this very stable junction, at low temperatures, shoulders around $\pm$ 20 mV are observed together with a well-developed ZBCP and an increasing background for high biases. The relative amplitude of the anomaly, that again develops just below T_c, is 55%. The normal junction resistance was $R_N(100\,\mathrm{mV})=7.5\,\Omega$. In Refs. 8, 9, 26, 27, coexistence of gaplike features and the ZBCP is reported for intermediate misorientation angles. In these cases, the conductance maxima at $V\neq 0$ are still related to the maximum value of the superconducting order parameter at the Fermi surface; however, the maxima shift towards lower biases. The data of Fig. 5 are consistent with this hypothesis.

In Fig. 6, the temperature behavior of the relative amplitude $[G^{max}(0)-G^{ext}(0)]/G^{ext}(0)$ of the ZBCP is reported as a function of $1/T$ for the same junction. The solid line in the figure is the least square best fitting that shows a correlation coefficient $r=0.99$ with the experimental data below 40 K. In other junctions with similar misorientation angles, the $1/T$ dependence extended over a wider temperature range.

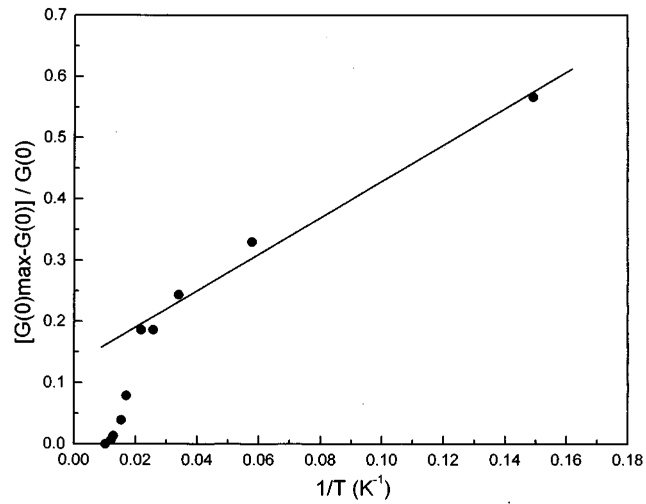

FIG. 6. The relative amplitude of the ZBCP vs $1/T$ for the same junction of Fig. 5. The full line shows a correlation coefficient $r=0.99$ with the data below 40 K.

IV. CONCLUSIONS

In summary, we have discussed a class of anomalies found in the conductance characteristics of $YBa_2Cu_3O_7$ readjustable junctions that reflect the sign changes of the superconducting order parameter at the Fermi surface. Highly anisotropic superconductors turn out to be very sensitive to inhomogeneities and interfaces and the break junction technique allows us to investigate the behavior of freshly created surfaces for different misorientation angles. The *d*-wave model for HTS predicts the presence of *q-p* bound states below the maximum energy gap that results in a different characteristic behavior of the $G(V)$ vs V curves. One of the most striking features expected for a,b tunneling is the presence of a large ZBCP in HTS-*I*-N junctions. This fact has important consequences on the tunneling characteristics of HTS-*I*-HTS junctions.

In agreement with recent models, we have found that for tunneling directions close to the *a-a* (*b-b*) axes, conductance maxima appear at $\pm 2\Delta_0$ and the ZBCP is not present or is highly suppressed. For different tunneling directions, the amplitude of the ZBCP increases while the conductance maxima at $V\neq 0$ move from $\pm 2\Delta_0$ towards lower energies. These GRS appear below the maximum energy gap or cannot be observed at all, as in the case of *a-b* tunneling. In fact, one also expects that gap anisotropy washes out the conductance maxima that would occur for isotropic *s*-wave superconductors.[34,35] Moreover, for very stable junctions, we have found a $1/T$ behavior of the ZBCP amplitude at low temperatures. By taking into account the further fact that, in the case of *c*-axis tunneling, in our as well as in other group experiments, the ZBCP is rarely observed, one can conclude that an impressive number of tunneling experiments are indicating a predominant *d*-wave component of the superconducting order parameter in the $YBa_2Cu_3O_7$ system.

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