Comparison of the superconducting pairing between the oxide superconductors $Ba_{1-x}K_xBiO_3$ and $YBa_2Cu_3O_{6.6}$ using phase-sensitive Josephson tunneling

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We compare the Josephson tunneling of a suspected d-wave superconductor, $YBa_2Cu_3O_{6.6}$, with that of another oxide superconductor $Ba_{1-x}K_xBiO_3$. For comparison the critical current I_c vs applied field of a single-crystal Josephson junction injecting supercurrent in the [110] direction is measured for both superconductors. For the $YBa_2Cu_3O_{6.6}$ single crystal (T_c=60 K), the critical current is a minimum at zero applied field, while for $Ba_{1-x}K_xBiO_3$, the critical current reveals a maximum at zero field. The results are consistent with a d-wave order parameter for $YBa_2Cu_3O_{6.6}$, and conventional s-wave superconductivity for $Ba_{1-x}K_xBiO_3$. [S0163-1829(97)08205-2]

A great deal of attention has recently been focused on determining the symmetry of the superconducting order parameter of cuprate superconductors. This interest has stimulated some phase sensitive tunneling experiments and their results are compatible with an order parameter of d-symmetry for $YBa_2Cu_3O_{6.9}$.[1–7] A d-wave order parameter was originally suggested as a possible explanation for the complex superconducting phase diagrams of some heavy-electron superconductors.[8] Subsequent work dealing with t-J and Hubbard models, which are often considered appropriate for the high-T_c cuprates, generally favor a d-wave order parameter for their superconducting ground state.[9] Later theoretical work showed that many experimentally determined physical properties of cuprate superconductors are compatible with a d-wave order parameter.[10] At this time almost all the phase sensitive tunneling experiments have been performed on $YBa_2Cu_3O_{6.9}$ (Ref. 11) with the exception of $YBa_2Cu_3O_{6.6}$ (Ref. 12) and $Tl_2Ba_2CuO_{6+\delta}$.[13]

In this article we describe phase sensitive tunneling experiments on the suspected d-wave superconductor $YBa_2Cu_3O_{6.6}$ and for comparison, the same experiments using $Ba_{1-x}K_xBiO_3$. Such experiments with $Ba_{1-x}K_xBiO_3$ should serve as a crucial comparison, because it is again an oxide superconductor with, conventionally viewed, a rather high critical temperature and cubic crystal symmetry. Because of a sizeable isotope effect, the lack of any local magnetic moments, and the rather conventional features of its physical properties, (see, e.g., Ref. 14) most would agree that $Ba_{1-x}K_xBiO_3$ is an s-wave superconductor. In our measurement layout (Fig. 1) we used a small Nb block as a counter-electrode and the tunneling was performed into the [110] direction of both $YBa_2Cu_3O_{6.6}$, where a node is supposed to exist in the d-wave gap function and $Ba_{1-x}K_xBiO_3$, where a nonzero gap in this direction is anticipated. To a first approximation, one might expect that Josephson tunneling would not be allowed into the d-wave cuprate in this direction. Nevertheless, because of details of the geometry of the measurement described below and a corresponding decomposition of the current into the [100] and [010] directions, a nonzero supercurrent I_c was observed. The magnetic flux dependence of the critical current $I_c(\Phi)$ for this tunneling geometry was found to closely resemble the classical Fraunhofer pattern with a maximum I_c for $\Phi=0$ for $Ba_{1-x}K_xBiO_3$. In contrast, for $YBa_2Cu_3O_{6.6}$ the pattern is characteristic of d-wave superconductivity.

The tunneling experiments capable of measuring the phase of the Josephson supercurrents in single-junction or multiple-junction loops provide the most direct determination of the symmetry of the superconducting order parameter. The idea for such experiments, aimed at investigating heavy-electron superconductors, was first suggested by

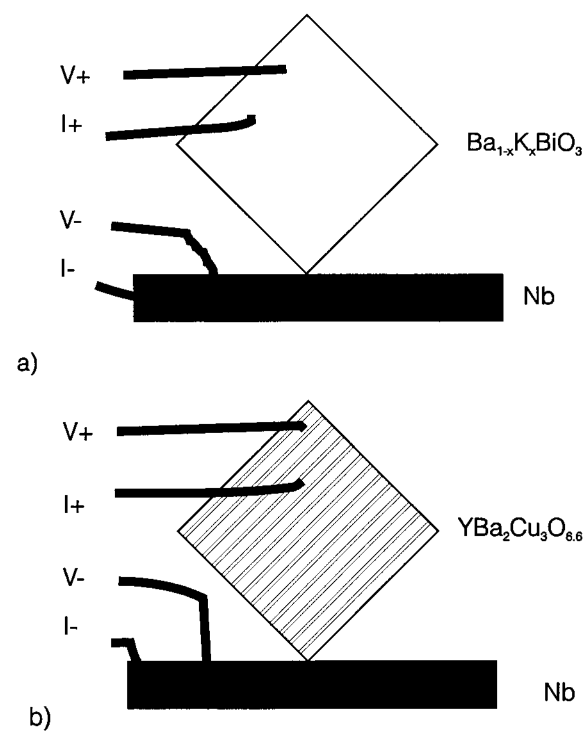

FIG. 1. Schematic diagram of the experimental arrangement. The superconducting single crystals of $Ba_{1-x}K_xBiO_3$ (a) and $YBa_2Cu_3O_{6.6}$ (b) were pressed against a Nb block to create the small corner junction. The critical current of the Josephson junction was measured as a function of the magnetic field applied along the direction into the page.

Geshkenbein and Larkin.[15] This suggestion was adapted to the typical features of cuprate superconductors with the proposal of Sigrist and Rice,[16] that Josephson supercurrents from an s-wave superconductor into a $d_{x^2-y^2}$-wave superconductor will have a current-phase relation given by

$$I=I_0(n_x^2-n_y^2)\sin\,\Delta\phi. \tag{1}$$

Here, n_x and n_y are the x and y components of the unit normal vector of the junction interface. The phase difference between the superconducting condensates on either side of the junction is expressed as $\Delta\phi$. For the case of the cuprate superconductors, this normal vector is chosen to lie in the CuO_2 ab plane with $x=a$ and $y=b$. If the superconducting order parameter was $d_{x^2-y^2}$-wave symmetry then from Eq. (1) it is clear that Josephson tunneling currents into the x direction will have an opposite sign compared with the tunneling into the y direction. The first tunneling results that were reported for $YBa_2Cu_3O_{6.6}$ (Ref. 1) were consistent with Eq. (1), implying that this material might be a d-wave superconductor. More complex order parameter symmetries such as $d+id$ or $s+id$ could not be ruled out, but the results of Ref. 1 were not consistent with a simple s-wave order parameter. This first indication got support from other work[2–4] as well as from the results of Refs. 5, 6, and 7. All of those results are in favor of a non-s-wave order parameter, of which pure d-wave symmetry is the simplest possibility. We also note additional work in Refs. 13 and 14, which indicated s-wave superconductivity in cuprate materials, however. A more complete description of the status of these types of experiments is given in the review article by van Harlingen.[11]

We begin by describing the experiment using the $T_c=23$ K superconductor $Ba_{1-x}K_xBiO_3$. These crystals were grown using an electrodeposition technique in a teflon crucible. Superconducting crystals of approximate size $400\times400\times400$ μm^3 were obtained with a transition temperature of 23 K. They possessed flat surfaces and sharp corners which reflect the cubic crystal symmetry. The current and voltage leads were attached directly to the $Ba_{1-x}K_xBiO_3$ crystals with silver epoxy and the crystal was embedded in stycast 1266 epoxy for mechanical support. The corner of the crystal was pressed against a block of polished Niobium at 4.2 K [Fig. 1(a)] and the pressure was adjusted until Josephson supercurrents were observed. Since the corners of the crystals are very sharp, i.e., with a radius of curvature on the order of microns, a very small Josephson junction could be made. After the experiment was performed, the crystals were examined and it was observed that the pressure required to obtain a junction eroded the corner enough to create a flat contact area on the order 25×25 μm^2. For a pure d-wave order parameter one would expect no tunneling permitted in this [110] direction if, as is most likely, the order parameter lobes extend along the [100] and the [010] axis, respectively, because the superconducting gap exhibits a node in this direction. For our particular tunneling geometry, one would expect that the tunneling along the [110] direction would only be possible if it is decomposed into components, not necessarily of the same magnitude, pointing along the [100] and the [010] directions respectively. In this way, a finite tunneling current may be achieved, in spite of the node in the order parameter. The phase difference between the two component directions would, due to Eq. (1), cause a destructive interference and a phase-shifted, Fraunhofer-like $I_c(\Phi)$ pattern. For a common s-wave superconductor, the tunneling direction should be irrelevant and no difference from the conventional tunneling behavior should be observed. Therefore, using this tunneling configuration, distinct differences between the tunneling into an s-wave superconductor, $Ba_{1-x}K_xBiO_3$, and an alleged d-wave superconductor, $YBa_2Cu_3O_{6.6}$, should be observed.

At 4.2 K the junctions for both superconductors had critical currents on the order of 1 μA, and thus a critical current density on the order of 1600 A/m^2. From these parameters the Josephson penetration depth $\lambda_J=\sqrt{\Phi_0/\mu_0 J_c\lambda_L}$ can be calculated to be on the order of 1000 μm. Since the size of our junctions is much less, this would imply that these junctions are not in the long junction limit. This is important because previous authors[5,17] have shown that for junction sizes greater than λ_J vortices may nucleate in the junction and make order parameter symmetry measurements impossible.

Any magnetic fields present in the junction area will affect the critical currents measured and therefore several precautions were taken to reduce the ambient fields. The experiments were performed in an rf shielded room and the leads were filtered for frequencies above 10 kHz. A three-axis Helmholtz coil was used to cancel the earth's field to within several mOe. A μ-metal cylinder and two concentric Pb superconducting cylinders with caps surrounded the apparatus to provide additional shielding. Subsequent examination of the sample space with a flux-gate magnetometer showed that the resulting background field was less than 1 mOe at room temperature. Care was also taken to keep magnetized objects away from the glass cryostat. The critical-current values were read directly from the IV curves of the junctions, as monitored with an oscilloscope. A small superconducting solenoid was placed next to the sample to provide the magnetic induction used in the experiment.

The Josephson critical current vs applied field pattern in Fig. 2 indicates that the period for a complete oscillation is of the order of 0.6 G. Due to the geometry of the samples they have a demagnetizing factor of approximately 3 which implies that the field at the junction is actually 1.8 G. From this field, the cross sectional area of the junction can be estimated to be approximately $\Phi_0 B\sim1.1\times10^{-7}$ cm^2. Since the width of the junction is approximately 25 μm this area indicates that the sum of the penetration depths of the superconductors plus the thickness of the tunneling barrier is 4400 Å which is the expected order of magnitude. As can be seen from Fig. 2, the critical current density is a maximum at zero applied field.

If one considers single junction Josephson tunneling between two s-wave superconductors, the equation for the relation between the critical current and the applied field is

$$I_c=I_m\left|\frac{\sin(\pi\Phi/\Phi_0)}{\pi\Phi/\Phi_0}\right| \tag{2}$$

and is independent of the tunneling direction. Here I_m is the maximum critical current, Φ is the flux through the junction, and Φ_0 is the flux quantum; a plot of I_c as in Eq. (2) is given

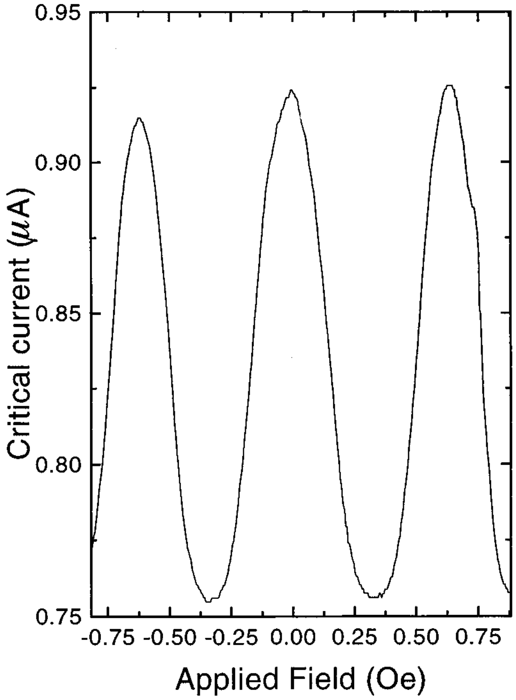

FIG. 2. Critical current vs applied field for a Josephson junction between $Ba_{1-x}K_xBiO_3$ and Nb as depicted in Fig. 1(a). The periodic nature of the critical current can be seen with a maximum at zero applied field. This picture is consistent with conventional (s-wave) pairing for the $Ba_{1-x}K_xBiO_3$.

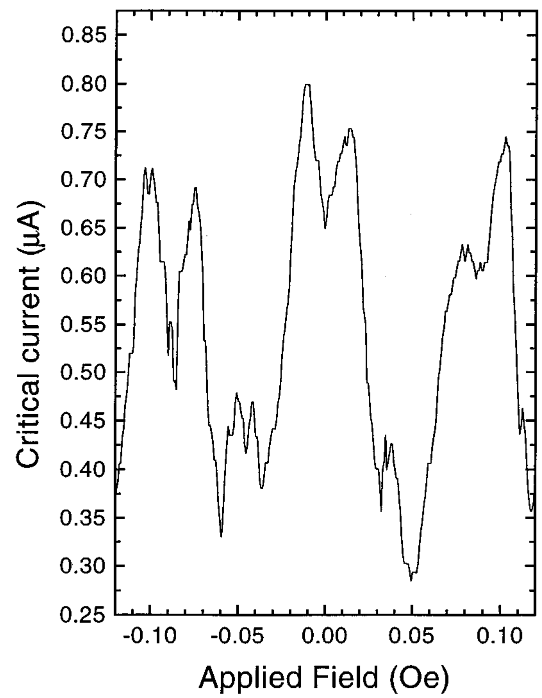

FIG. 3. Critical current vs applied field for a Josephson junction between $YBa_2Cu_3O_{6.6}$ and Nb as depicted in Fig. 1(b). The critical current has a local minimum at zero applied field which is evidence that $YBa_2Cu_3O_{6.6}$ is a d-wave superconductor. This data is consistent with the notion that the tunneling is divided into the a,b directions producing a destructive interference at zero applied field.

in Ref. 11. Within errors, the amplitude of the oscillations does not noticeably decrease with increasing field in our case. This implies that the tunneling current is not uniform across the junction area. The possibility exists that the tunneling current is primarily concentrated on two small spots on opposite sides of the junction, and in this case the junction would resemble a dc/superconducting quantum interference device. Most importantly, the critical current is a maximum for zero applied field indicating that there are no extra phases entering the tunneling behavior. Therefore, this behavior is most consistent with an s-wave order parameter for $Ba_{1-x}K_xBiO_3$.

We now consider the case of a tunneling junction between $YBa_2Cu_3O_{6.6}$ and Nb as illustrated in Fig. 1(b). The single crystals of $YBa_2Cu_3O_{6.6}$ used in this study were grown by the slow-cooling method.[18] The microscopic directionality of the tunneling is important to control if evidence for a d-wave order parameter is to be gained. As previously described, tunneling should not be permitted in the [110] direction due to the presence of a node in the order parameter. Instead the tunneling, recalling our geometry, will necessarily be decomposed into components in the a and b directions. Therefore, according to Eq. (1), there will be interference effects in the case of a d-wave order parameter. This tunneling geometry makes the experiment comparable to those previously done with single ''corner'' junctions.[6,12] These corner junctions consisted of a thin film of a low-T_c superconductor covering both faces of the corner of the high-temperature superconductor. The critical current of this junction as a function of field for a d-wave order parameter may be calculated and according to Ref. 6 one obtains

$$I_c=I_0A\left|\frac{\sin(\pi\Phi/2\Phi_0)}{(\pi\Phi/2\phi_0)}\right|. \tag{3}$$

An essential feature of this equation is that the critical current is *always* a minimum at zero applied flux Φ. A plot showing the general shape of $I_c(\Phi)$ according to Eq. (3) is given in Refs. 6, 11, and 12 and so needs not be reproduced here. We show $I_c(\Phi)$ measured with the configuration of Fig. 1(b) in Fig. 3. It may be seen that the central maximum of the critical current is split, leaving a local minimum at zero applied field. Qualitatively this agrees with $I_c(\Phi)$ given by Eq. (3). As mentioned before, the nonuniform current density across the junction causes a deviation from ideal behavior, nevertheless the most significant feature of Eq. (3) is reproduced, namely, a minimum of the critical current at zero applied field. A quantitative description that takes into account the inhomogeneity of the junction is given in Ref. 6. The authors consider the case when the tunneling is primarily along the a or b axis with a contribution of the other. This causes a reduction of the interference and thus produces a pattern similar to what is observed here. Similar calculations may be done as before, taking into account that the period of oscillations is 0.1 Oe. Using a demagnetization factor of 10 for this crystal ($500\times600\times40\ \mu m^3$) leads to a junction cross-section of $2\times10^{-7}\ cm^2$. Since the junction was observed to be approximately 30 μm in size, the penetration depths and the tunneling barrier must add up to a reasonable 6600 Å.

The effect of trapped flux on the situation can be modeled by adding an extra constant flux as described in Ref. 6. This results in an asymmetrical and aperiodic critical current with field and thus is readily recognizable by experiment. The effects of trapped flux found experimentally are illustrated, e.g., in Fig. 3 of Ref. 12. Because the presence of trapped flux produces this unmistakable signature and all the patterns presented here are seen to be symmetrical within error about zero applied field it may be concluded that any effects of trapped flux are not significant here.

A determination of a value for the superconducting gap is often done using the Ambegaokar-Baratoff (AB) formula, which at low temperatures simplifies to $I_c R = \pi\Delta/2$. Given that the critical currents are approximately 1 μA and that the resistances of all the junctions are on the order of a few Ohms, this would imply a gap of a few μeV. This is several orders of magnitude smaller than one would expect for both the materials $Ba_{1-x}K_xBiO_3$ and $YBa_2Cu_3O_{6.6}$. This inconsistency has been recognized before[19] and is most likely due to the difficulty of achieving highly resistive tunneling barriers.

The only reasonable conclusion for the observed minimum of the critical current at zero applied field is that there must be a phase difference close to π that occurs when tunneling into the corner of the $YBa_2Cu_3O_{6.6}$ crystal is attempted. A d-wave-type superconducting order parameter for $YBa_2Cu_3O_{6.6}$ is consistent with this observation according to Eq. (1). Other more exotic possibilities, such as mixtures of s- and d-type symmetries of the superconducting order parameter are also consistent with the data, provided that the d component dominates. An s-wave order parameter in the cuprate cannot provide the necessary phase shift, unless some other explanations can be found to explain the π phase difference between tunneling in the a and b directions.

Several authors have reported anomalies in tunneling characteristics of cuprate superconductors which are often ascribed to pair weaking by localized states in the junction[20] or pair breaking by spin flip scattering.[21] Spin flip and localized-state scattering may be capable of producing significant phase shifts at the junction interfaces. These effects may be material induced in $YBa_2Cu_3O_{6.6}$ but should not be observed for the nonmagnetic $Ba_{1-x}K_xBiO_3$, however, this provides additional evidence that spurious external flux trapped in crystal corners is quite unlikely to produce the persistent I_c minimia at zero flux for cuprate superconductors. Defect induced spin-flip tunneling, also, cannot explain the different results found on corner and edge junctions in previous experiments (see, e.g., Refs. 1, 6, and 12), because spin-flip scattering would occur in any direction. For this reason, we believe that spin-flip processes in the tunneling barriers are not responsible for the observed π phase shifts. To conclude, we have presented further evidence in support of a d-wave order parameter for $YBa_2Cu_3O_{6.6}$ and with a reasonable control experiment we show that $Ba_{1-x}K_xBiO_3$ may be classified as an s-wave superconductor.

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