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Anisotropy of the penetration depth in $YBa_2Cu_3O_{7-\delta}$: Josephson-tunneling studies

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The modulation of the Josephson critical current between two superconductors in a magnetic field provides a direct, low-field dc measurement of the penetration depth of the two superconductors. To measure the in-plane penetration depth anisotropy of $YBa_2Cu_3O_{7-\delta}$, we have fabricated low leakage Pb/Insulator/$YBa_2Cu_3O_{7-\delta}$ tunnel junctions on detwinned $YBa_2Cu_3O_{7-\delta}$ single crystals with tunneling along the c axis. By applying the magnetic field along the b and a crystalline axes of $YBa_2Cu_3O_{7-\delta}$, λ_a and λ_b are extracted from the respective field modulation patterns. We find a large anisotropy $[(\lambda_a/\lambda_b)^2 \geqslant 2]$ of the penetration depths along the a and b axes.

The vast majority of experiments aimed at studying the high-T_c cuprates, especially the symmetry of the order parameter, have been performed on the compound $YBa_2Cu_3O_{7-\delta}$. This is in part due to our ability to grow reproducible $YBa_2Cu_3O_{7-\delta}$ and fabricate structures. However, $YBa_2Cu_3O_{7-\delta}$ has a pronounced orthorhombic structure, containing Cu-O chains in addition to the CuO_2 planes that are present in all the other cuprates and are generally thought to be responsible for superconductivity. It is widely believed that the Cu-O chains serve as reservoirs that supply charge carriers for the CuO_2 planes. The cuprates are also often thought of as "approximately tetragonal" with the orthorhombicity regarded as minor. Recent far-infrared spectroscopy studies on untwinned $YBa_2Cu_3O_{7-\delta}$ crystals, however, showed an in-plane anisotropy of the penetration depth of about 2,[1] much larger than earlier experiments[2] had indicated, and sufficiently large that the orthorhombicity has to be seriously considered. The large in-plane anisotropy in the superconducting state implies that the Cu-O chains could play a more important role in $YBa_2Cu_3O_{7-\delta}$, and detailed studies might eventually resolve the seemingly contradictory results of experiments on $YBa_2Cu_3O_{7-\delta}$ (Refs. 3–7) designed to determine the symmetry of the order parameter of high-T_c cuprates. In this paper, we present our measurements of the in-plane penetration depth anisotropy in detwinned $YBa_2Cu_3O_{7-\delta}$ single crystals. We have previously shown[7] that Josephson coupling exists between Pb and $YBa_2Cu_3O_{7-\delta}$ along the c axis, and the magnetic-field modulation of the Josephson critical current provides a direct, low field dc measurement of the penetration depth λ_{ab} of $YBa_2Cu_3O_{7-\delta}$. By making c-axis Pb/Insulator/$YBa_2Cu_3O_{7-\delta}$ Josephson tunnel junctions on detwinned $YBa_2Cu_3O_{7-\delta}$ crystals, and applying the magnetic field along the b and a directions, we have been able to measure λ_a and λ_b, respectively, and our data show a large anisotropy ratio, $(\lambda_a/\lambda_b) \geqslant 2$. This result implies substantial electronic anisotropy in the a-b plane.

The $YBa_2Cu_3O_{7-\delta}$ crystals were grown using a "self-flux" technique. First, stoichiometric amounts of Y_2O_3, $BaCO_3$, and CuO were ground and sintered at 900 °C for 4 days with 3 intermediate grindings to form a polycrystalline precursor of $YBa_2Cu_3O_{7-\delta}$. Next, the precursor was mixed with extra $BaCO_3$ and CuO in a molar ratio of 1:7:21 to form a flux. Finally, the mixture was placed in yttria-stabilized zirconia trays, heated slowly to 980 °C, cooled from 980 °C to 880 °C at 6 °C per hour, and then cooled from 880 °C to room temperature at 100 °C per hour. This process yielded flat platelets of $YBa_2Cu_3O_{7-\delta}$ single crystals embedded in the flux, approximately 1 mm×1 mm×100 μm in size, which were removed mechanically from the crucible. The crystals were then annealed in gold crucibles in a tube furnace at 420 °C for 10 days, under flowing high-purity oxygen, in order to fully oxygenate the crystals ($\delta \approx 0.05$). The crystals were characterized by magnetic susceptibility, with typical transition temperature T_c and transition widths of approximately 90 and 2 K, respectively.

The crystals were then detwinned using a design similar to that reported by Welp *et al.*[8] A uniaxial pressure of $\sim 10^8$ N/m^2 was carefully applied to the crystal with a spring. The displacement of the spring was measured with a calibrated micrometer head attached to the press. The crystal under pressure was annealed at temperatures between 450 and 490 °C for several hours. Preheated oxygen gas was supplied through a capillary quartz tube to maintain the oxygen content of the single crystal during the detwinning process. The uniaxial stress and oxygen supply were maintained as the crystal was cooled at 4 °C per minute to room temperature. The crystals were scanned for twin domains using a high-resolution metallographic microscope with polarized light. The oxygen concentration was checked indirectly by comparing the magnetization curves of the sample before and after detwinning. The Cu-O chains run perpendicular to the direction of applied uniaxial pressure, and the crystalline orientation is thereby determined.

The Josephson tunnel junctions were fabricated using a previously reported technique.[7] Each junction was rectangular in shape (we attempted to make them nearly square), with one dimension defined by epoxy and the other defined by the shadow mask through which the Ag diffusion barrier and the Pb stripe were evaporated, see Fig. 1. The sides of the rectangle were made so that they were parallel to either the a or the b direction. The junction was put into a vacuum can with leads attached for four-point measurements, and cooled down quickly to 77 K with the help of He exchange gas. This was done within half an hour after the Ag and Pb deposition,

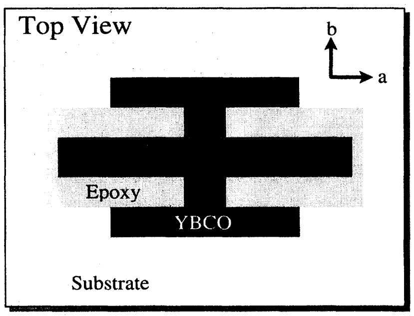

FIG. 1. Top view of the junction. The junction area is rectangular (almost square) in geometry, with sides parallel to either the a or b crystalline axis of $YBa_2Cu_3O_{7-\delta}$.

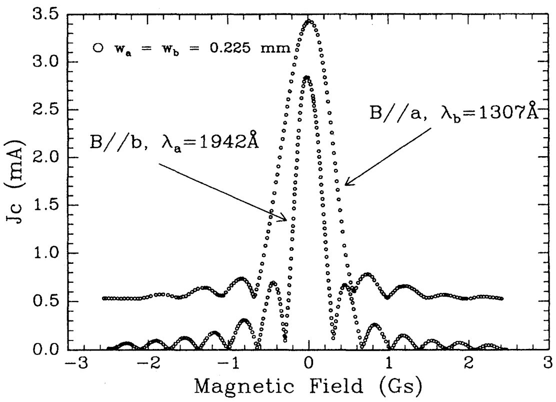

FIG. 2. $I_c(B)$ patterns of junction No. 6 with magnetic field along a (upper curve) and b (lower curve) directions. The upper curve is offset by 0.5 along the y axis.

and the increase of junction resistance at room temperature was thus minimized. Before the measurements, the exchange gas was pumped out of the vacuum can and the sample temperature was raised to about 120 K (well above the T_c of $YBa_2Cu_3O_{7-\delta}$) with a heater on the sample stage. The vacuum can housing the sample was then immersed in liquid He in a cryogenic Dewar shielded with μ-metal, and the sample was cooled down slowly (at a rate of 1 K/min) through the T_c of $YBa_2Cu_3O_{7-\delta}$ via a weak thermal link. If this procedure was carefully followed, it minimized trapped flux in the $YBa_2Cu_3O_{7-\delta}$ specimen. Trapped flux destroys the regular field modulation pattern of the Josephson critical current I_c and is easily detected because the $I_c(B)$ patterns are not symmetric in the four quadrants (symmetric in I and B). After 2 h when the sample had reached about 30 K (well below the T_c of $YBa_2Cu_3O_{7-\delta}$), exchange gas was transferred into the vacuum can to reduce the temperature to 4.2 K. The sample was then cycled a few times above the T_c of Pb to expel any trapped flux in the Pb. After this, the sample was ready for measurements. The junction was studied at different temperatures, the low temperature I_c was measured at 1.3 K and the field modulation patterns were taken at 4.2 K. The magnetic field was applied with a Helmhotz coil inside the μ-metal shield, with the field lines parallel to the CuO_2 planes. By rotating the sample around the c axis, we were able to align the field along different crystalline directions within the CuO_2 plane. Because the penetration depths in different directions are defined according to the directions of the screening current in the superconductor, λ_a was extracted from the $I_c(B)$ pattern when the magnetic field was applied along the b direction and vice versa.

In Fig. 2, we show a typical set of $I_c(B)$ patterns with field applied along a and b crystalline directions. The field dependence of the critical current I_c is given by

$$I_c = I_0 \left| \frac{\sin\left(\frac{\pi\Phi}{\Phi_0}\right)}{\frac{\pi\Phi}{\Phi_0}} \right|$$

for a rectangular uniform junction, where Φ is the flux through the junction and $\Phi_0 = hc/2e$ is the flux quantum. In the presence of some degree of spatial nonuniformity of the critical current in the junction, the minima of the Fraunhofer-like pattern may not reach zero. Nonetheless, the minima are almost equally spaced and each of them corresponds to an integral number of flux quanta enclosed in the geometry defined by the junction width w_a (or w_b depending on the field direction) in one direction and the sum of the penetration depths in Pb and $YBa_2Cu_3O_{7-\delta}$ as well as the barrier thickness in the other direction. In other words, the area $\Phi_0/\Delta B$ (ΔB being the spacing between adjacent minima) extracted from the I_c-B pattern equals $[w_a(\lambda_{Pb}+\lambda_a+t)]$ (or $[w_b(\lambda_{Pb}+\lambda_b+t)]$). The junction width w_a (or w_b) is defined by the geometric size of the junction, the barrier thickness t is 20 Å, and λ_{Pb} has been measured separately using the same technique on a Pb/PbO_2/Pb junction.[7] The value of λ_a (or λ_b) of $YBa_2Cu_3O_{7-\delta}$ can thus be determined with the above information.

Inspection of Fig. 2 suggests that the $I(B)$ patterns (especially for $B//a$) are not exactly of the form $|\sin\phi/\phi|$ which indicates a somewhat nonuniform current density. We emphasize that this minimally effects our estimates of λ_a and λ_b as we determine the quantities from the spacing of the maxima or minima. We do rely on the uniform spacing of these oscillations and that is verified. Nonuniform spacing or lack of symmetry in B or I suggest more serious problems and we reject those data.

To date we made measurements on eight junctions, each made on different $YBa_2Cu_3O_{7-\delta}$ crystals. Measurements on each junction were repeated several times, each after thermal cycling to above 120 K and a slow cooling, to establish reproducibility and eliminate flux trapping effects.

In Table I, we present the measured values of the penetration depths and the anisotropy ratio, along with other characteristics of the tunnel junctions. The values of the I_cR_n product in these junctions are on average a factor of 2–3 higher than those of the junctions made on heavily twinned $YBa_2Cu_3O_{7-\delta}$ crystals and will be discussed in a separate paper.[9] The last column shows the anisotropy ratio $(\lambda_a/\lambda_b)^2$ as the electronic properties are dependent on this quantity, i.e., $\lambda^{-2} \propto (n_s/m^*)$, where n_s is the superfluid density and m^* is the effective mass of an electron pair. There is significant variation in our measured values of λ_a and λ_b as well as $(\lambda_a/\lambda_b)^2$ from sample to sample, although $(\lambda_a/\lambda_b)^2$ is mostly around 2–3. The variation in λ is not

TABLE I. Measured values of penetration depth anisotropy and other characteristics of eight different junctions studied.

	R_n (Ω)	I_c (mA)	I_cR_n (mV)	λ_a (Å)	λ_b (Å)	λ_a/λ_b	$(\lambda_a/\lambda_b)^2$
1	4.26	0.37	1.58	2360	1398	1.69	2.85
2	4.00	0.052	0.21	4171	1424	2.93	8.58
3	2.79	0.187	0.52	2668	1772	1.51	2.27
4	28.1	0.046	1.3	1609	904	1.78	3.17
5	0.72	1.32	0.95	1609	1139	1.41	2.00
6	0.44	3.2	1.4	1942	1307	1.49	2.21
7	4.88	0.202	0.99	2092	1749	1.20	1.43
8	0.60	1.5	0.9	2271	1368	1.66	2.76

directly related to the variation in the I_cR_n product, because unlike the I_cR_n product, which depends on pair coupling over a surface layer of thickness of the order of the coherence length at the junction interface, the penetration depth is more of a bulk property of the superconductor. We do not yet understand why we observe such a variation from sample to sample, but we suspect the variation in the measured values of the penetration depths comes from the difference in oxygen doping and disorder in the $YBa_2Cu_3O_{7-\delta}$ crystals, and the resulting chain integrity and planar carrier concentration. We do not have quantitative data confirming this suspicion, but there is circumstantial evidence; crystals No. 5 and No. 6 had the sharpest transitions, with onset at 92 K and a width of about 0.5 K, while crystal No. 2 had a broad transition, with a transition width of almost 15 K (onset still at 92 K). The rest of the crystals have transition widths around 2 K.

Questions remain as to the large range of variation in our measured penetration depth, especially in the values of λ_a. The penetration depth λ_a is usually attributed solely to the CuO_2 planes and thus thought to be less sensitive to small variations of oxygen concentration and disorder in the chains. Nonetheless, our data do show a large anisotropy between the *a* and *b* direction in the superconducting state of $YBa_2Cu_3O_{7-\delta}$, which indicates a large anisotropy in condensation density and the possibility of pair condensation along the chains. This is in agreement with recent infrared[1] and muon spin relaxation[10] results, and lends support to the picture that the normal-state transport anisotropy[11] can be accounted for by the anisotropy of the plasma frequency while the scattering time is more isotropic within the plane.

In summary, we have measured the penetration depths λ_a and λ_b by studying Josephson tunnel junctions made on untwinned $YBa_2Cu_3O_{7-\delta}$ crystals. Our results show a large anisotropy ratio, $(\lambda_a/\lambda_b)^2 \geq 2$, which suggests significant pair condensation on the Cu-O chains in $YBa_2Cu_3O_{7-\delta}$. This suggests that any theories of superconductivity in $YBa_2Cu_3O_{7-\delta}$ must take this orthorhombicity into account as it is a dominant consideration, and offers alternative explanations to the results of several experiments[3–5] which provided evidence for possible *d* symmetry of the order parameter in high-T_c cuprates. Similar experiments should be performed on tetragonal systems such as $Nd_{2-x}Ce_xCuO_{4-y}$ and $HgBa_2Ca_2Cu_3O_{8+\delta}$.

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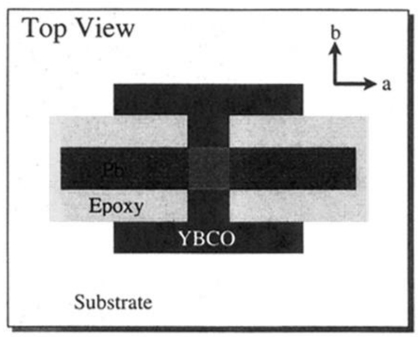

FIG. 1. Top view of the junction. The junction area is rectangular (almost square) in geometry, with sides parallel to either the *a* or *b* crystalline axis of $YBa_2Cu_3O_{7-\delta}$.