Experimental Evidence for Intra- and Inter-Unit-Cell Josephson Junctions in a $YBa_2Cu_3O_{7-\delta}$ Single Crystal

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Modulation of the dynamic resistance versus the magnetic field of $YBa_2Cu_3O_{7-\delta}$ single crystal shows two groups of peak structures with nearly uniform magnetic field spacings. The spacings suggest two kinds of Josephson junctions: one formed by the CuO_2-Y-CuO_2 atomic planes within each unit cell and the other formed by the CuO_2 bilayers between different unit cells. The intra-unit-cell junctions appear first as the temperature is lowered from T_c. The existence of CuO_2-Y-CuO_2 Josephson junctions implies two-dimensional superconductivity in the CuO_2 atomic planes.

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High-T_c superconducting cuprates, due to the perovskite structures of these materials, have several physical properties, such as coherence length and resistivity, showing extremely large anisotropies between the *a-b* plane and *c*-axis directions [1]. It is generally believed that the high-T_c superconductivity is intrinsically two dimensional [1–4] in the CuO_2 bilayers or trilayers coupled together by the Josephson currents along the *c*-axis direction [4]. Theories and experiments concerning several phenomena and properties, such as angular dependence of the critical current [5], upper critical field [6], vortex motion and coupling [7], and far-infrared conductivity [8], all support this picture. Recently, Kleiner *et al.* [9,10] have shown that the current-voltage (*I-V*) characteristics of a single crystal of $Bi_2Sr_2CaCu_2O_{8+\delta}$ (BSCCO) as well as that of other high-T_c cuprates consisted of numerous branches indicative of a stack of Josephson tunnel junctions connected in series. According to the magnetic-field periods derived from the magnetic-field dependences of the critical current, the Josephson junctions are formed by the inter-unit-cell CuO_2 bilayers. To our knowledge, there is no direct experimental evidence showing that two CuO_2 atomic planes within a unit cell can also form a Josephson junction. Evidence for such intra-unit-cell Josephson junctions would have a strong implication for atomic-plane 2D superconductivity.

Furthermore, the previous attempts [9] to search for a Josephson-junction-like *I-V* characteristic in $YBa_2Cu_3O_{7-\delta}$ (YBCO) have not been fruitful, possibly due to the lack of an energy gap in the *c*-axis direction [11]. This raises the question of whether the superconductivity in YBCO is bulklike as opposed to a stack of Josephson junctions as in BSCCO. In this Letter, we shall describe experimental results which indicate not only the existence of inter-unit-cell Josephson junctions in YBCO but also intra-unit-cell Josephson junction formed by the CuO_2-Y-CuO_2 (CYC) atomic planes.

The samples are YBCO single crystals grown by a self-flux method. Nonstoichiometric starting powders of Y_2O_3, BaO_2, and CuO with a Y:Ba:Cu ratio of 1:18:45 were first thoroughly mixed, then heated in air in an Al_2O_3 crucible in several steps to 1010 °C. In the cooling process, the melt was cooled to 970 °C in 6 h and then to 890 °C in 99.5 h. Finally, it was cooled to 500 °C in 30 h followed by nature cooling to room temperature. The single crystals were postannealed at 500 °C in flowing oxygen at 55 cm^3/min for 4 weeks. Two Ag pads of thickness about 10^3 nm were first *e*-beam deposited on the top and the bottom surfaces of each single crystal platelet and subsequently annealed at 500 °C for 10 h. Electrical leads were then attached to these Ag pads by Ag epoxy. Normally, each contact resistance is less than 0.1 Ω. Typically, these annealed single crystals have the $YBa_2Cu_3O_{7-\delta}$ structure according to the x-ray diffraction patterns and a resistive onset temperature of 93 K in the *a-b* plane. In the *c*-axis direction, they have the commonly observed inverse temperature dependence with a resistive peak occurring in the range of 84 to 89 K.

One of the hallmarks of a Josephson junction is its oscillatory magnetic-field-dependent critical current $I_c(H)$. For a junction with a uniform current density, i.e., in the short-junction regime, it has the usual Fraunhofer diffraction pattern given by $I_c(H) = I_0|\sin(\pi H/H_0)/\pi H/H_0|$, where I_0 is the Josephson current for $H = 0$ and H_0 is the magnetic field equivalent to one flux quantum Φ_0 enclosed by the junction. Thus $I_c(H)$ can be reduced to zero whenever an integer number of magnetic flux quantum enters the junction. If, however, the Josephson current density j_0 is too large so that the length of a junction L becomes greater than $2\pi\lambda_J$, where λ_J is the Josephson penetration depth ($\lambda_J \propto j_0^{-1/2}$), then it is in the long-junction regime. In this case, Owen and Scalapino have shown [12] that $I_c(H)$ typically has complex overlapping branches depending on the L/λ_J ratio, and the zeros of $I_c(H)$ are no longer simply related to $n\Phi_0$. Qualitatively speaking, the larger the L/λ_J ratio, the greater the threshold magnetic field needed for the entry of one flux quantum due to more effective screening of magnetic field. Thus, in order to observe intra-unit-cell CYC Josephson junctions in the

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short-junction regime and that are not coupled to one another, the experimental temperature should not be too far below T_c. In this temperature range the experimental measurements of $I_c(H)$ are extremely difficult and ambiguous, since the sample is high up in its resistive transition, typically near the midpoint of the transition. Thus an alternative method [13] is needed to observe the effect of oscillatory $I_c(H)$. We have used a small ac bias current, typically a few μA at 17 Hz, and a lock-in technique to monitor the small change in its dynamic resistance dv/di as a function of magnetic field. Whenever $I_c(H)$ of a junction or a group of identical junctions in a series reaches a local zero or minimum below the level of the bias current, a peak should develop in dv/di. This technique permits us to see a Josephson junction even if it is connected to a large resistance background. The noise level of our detecting system is 0.2 nV, which allows us to detect a resistance change of 0.5 mΩ on top of a 50 mΩ background with an ac current as small as 1 μA. Another important experimental consideration is the temperature stability. Since the experiment is done in the transition region, a small temperature fluctuation can lead to an erroneous signal. For our samples, this requires a temperature stability of $\Delta T < 5$ mK. To avoid liquid nitrogen bubbling and gas fluctuation which often can complicate the results, we enclosed the sample in a vacuum or in a stable gas environment surrounded by a can cooled by liquid nitrogen from the outside. For measurements at each temperature, thermal equilibrium was reached first to ensure temperature stability.

Four YBCO single crystals, each from a different batch, all have shown oscillatory magnetic field dependences indicative of Josephson junctions. In order to show a correlation between the inter- and intra-unit-cell Josephson junctions at various temperatures, we shall describe only one sample on which we have the most complete results. The dimensions of this single crystal are 0.65 mm × 0.85 mm in the *a-b* plane and about 50 μm thick in the *c*-axis direction. The relevant dimension, i.e., the length facing the external magnetic field, is 0.65 ± 0.10 mm. The resistance in the *c*-axis direction R_p is about 29 mΩ at 295 K and has roughly an inverse temperature dependence reaching a resistance peak R_p of 96 mΩ at 84 K. At this peak value, each unit-cell layer on average has a resistance of about 2 $\mu\Omega$ in the *c*-axis direction. For a CYC triple-plane junction, the share is much less, say, only 30% [see Fig. 1(a)]. Thus a change of 0.5 mΩ in the total resistance means a simultaneous involvement of hundreds of CYC junctions connected in series. This number, although large, only represents a small fraction of the total crystal.

Figures 1–3 are dv/di vs H plots for several different temperatures, all in the resistive transition region. Actually, a more important parameter may be the resistance in comparison with the peak value. Figure 1(b) was taken at 80.50 K, where R_c is $0.51R_p$, near the midpoint of the

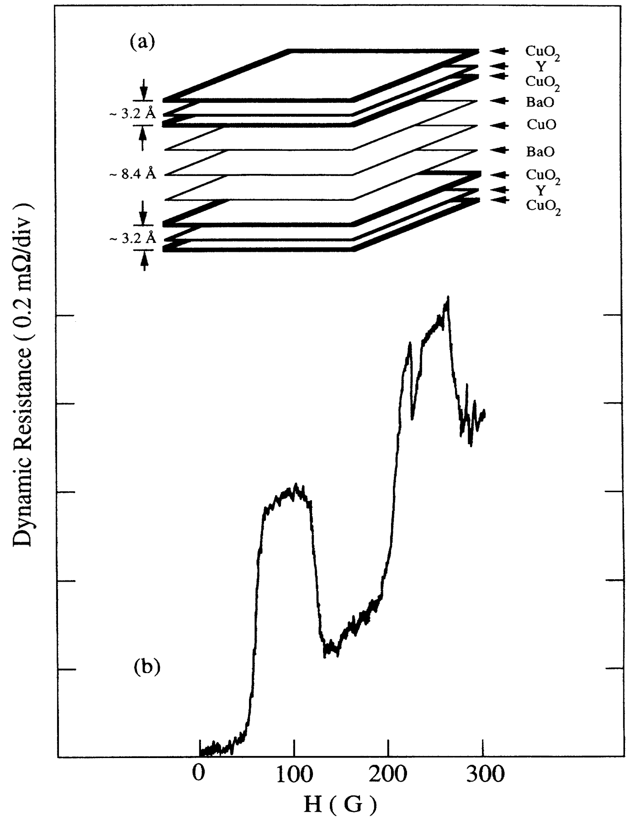

FIG. 1. (a) The relative positions of various atomic planes in $YBa_2Cu_3O_{7-\delta}$. (b) Modulation of the dynamic resistance vs magnetic field taken with a 17 Hz ac current of 10 μA at $T = 80.54$ K. The ac current is in the *c*-axis direction and the magnetic field is parallel to the *a-b* plane. The resistance value for $H = 0$ is 49 mΩ ($0.51R_p$).

transition. Two peaks can be seen: one centered at about 90 G and the other is close to 220 G. For larger currents, the first peak becomes broader and the second peak becomes a step, not returning to the lower level. These bias-current dependences can be understood by considering the relative magnitudes of the bias current and the oscillatory field dependence of I_c. For a larger bias current, the range of $I_c(H)$ to fall below the bias current becomes wider making the peaks broader. When the amplitude of the ac bias current exceeds the second maximum of $I_c(H)$, the junction cannot fall completely back to the zero-resistance state. This is why the second peak becomes a step. The peak positions allow us to estimate the dimensions of the junctions involved. For a Josephson junction with two thick electrodes, the first peak in the dv/di vs H corresponding the first zero in $I_c(H)$ should occur at a magnetic field H_1, which satisfies $(l + 2\lambda)LH_1 = \Phi_0$, where l is the tunneling barrier thickness and λ the magnetic penetration depth. On the other hand, for a junction with two thin electrodes, the condition is $dLH_1 = \Phi_0$, where d, replacing $l + 2\lambda$, is the distance between the cen-

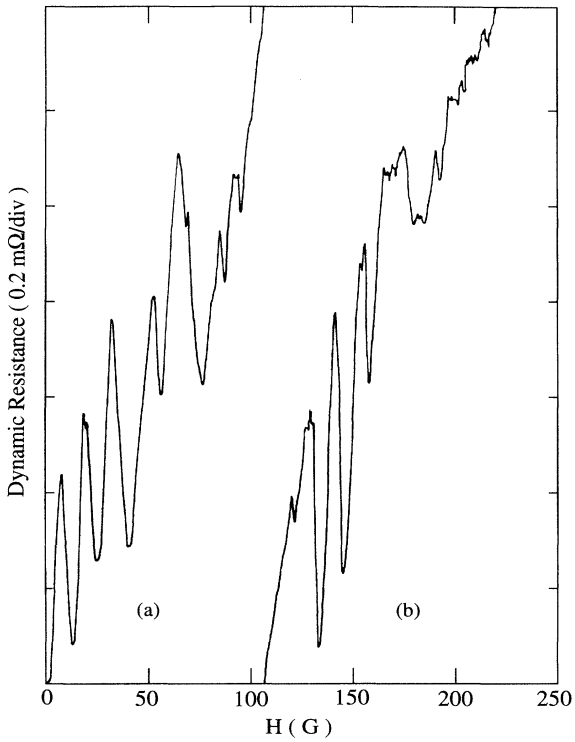

FIG. 2. Same as Fig. 1(b), except $T = 79.80$ K and $R_c = 39$ mΩ ($0.41 R_p$). Curve (b) is a continuation of curve (a) shifted down from the upper end of (a).

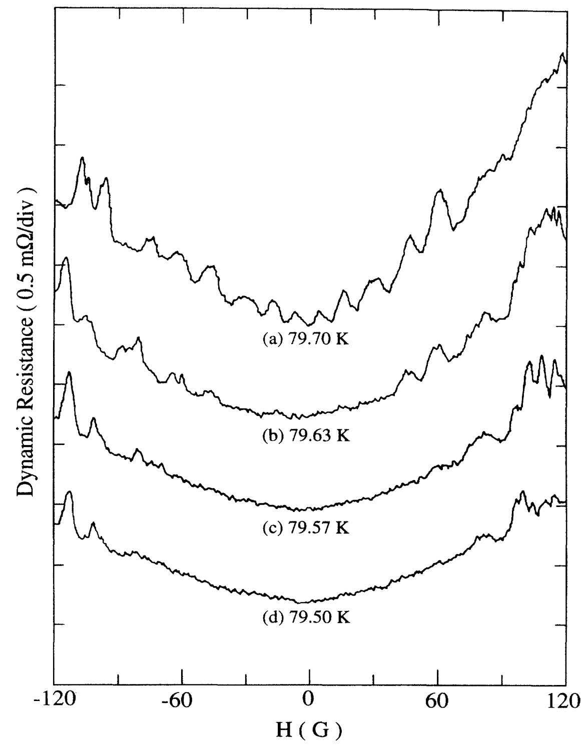

FIG. 3. (a)–(d) Modulation of dynamic resistance vs magnetic field showing an evolution from the short-junction to the long-junction regime as explained in the text. All curves were taken with $i = 10$ μA at the temperatures indicated, where $R_c \leq 0.4 R_p$. The curves are displaced from each other for clarity.

ters of the two electrodes [9]. In a CYC unit, d would be the separation between the two CuO_2 planes. Using $L = 0.65 \pm 0.10$ mm, $H_1 = 90 \pm 10$ G, and $\Phi_0 = 2.07 \times 10^{-7}$ G cm^2, we obtain $d = 3.4 \pm 0.9$ Å. Similar results have been found for all four single crystals studied. The H_1 vs $1/L$ data from these four samples are shown in Fig. 4, including two data points (squares) for two different dimensions of one rectangular sample. The slope yields $d = 3.5 \pm 1.0$ Å, which is in good agreement with the expected value of 3.2 Å for CYC. The position of the second peak is larger than the expected value of 180 G. However, this degree of deviation is commonly observed even in a conventional Josephson junction. A slight imperfection in the junction or the self-field effect due to the bias current can cause a spatial variation of the phase to deviate from linearity [14]. Another possibility is that the superconducting electrodes, being only atomic-layer thick, can be weakened somewhat after the entry of the first flux quantum. In any case, the significance of these results is that the second peak is observed, which indicates the oscillatory nature of the $I_c(H)$. The first peak position H_1 does not show an appreciable temperature dependence, although its magnitude is sensitive to the temperature. These peaks are observable in a narrow temperature range, equivalent to $0.45 R_p < R_c < 0.55 R_p$. Above this temperature, this phenomenon is not observable, understandably, because of the further weakening of the superconductivity near T_c. As the temperature is slightly reduced, the landscape of dv/di vs H (see Fig. 2) changes dramatically, signifying the occurrence of inter-unit-cell Josephson coupling.

Although Figs. 1(b) and 2 were taken with the same ac current at two slightly different temperatures about 0.7 K apart, they show very different structures. Figure 2 shows many more resistive peaks with smaller magnetic field spacings. Except for the first peak (to be discussed

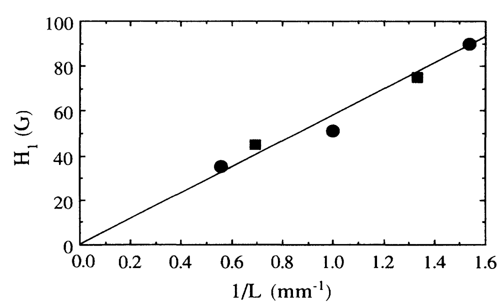

FIG. 4. Magnetic field for the first peak (as shown in Fig. 1) versus the inverse of sample width. See text for further details.

later), ΔH is about 15 $\pm$ 1 G on average. Using the same analysis as before, we get $d = 21 \pm 4$ Å. This value is roughly twice the c-axis lattice constant for YBCO. Closer inspection of many curves similar to those in Figs. 2 and 3(a) taken at different temperatures and with different bias currents have revealed that the peaks near 30 and 60 G are sometimes larger than those at 15 and 45 G. Also as can be seen in Figs. 2(a) and 3(a), they are broader or even split (not shown here). These results suggest that there is another group of peaks with a $\Delta H \approx$ 30 G overlapping with the group with a $\Delta H = 15$ G. The thickness of the junctions corresponding to this less distinguishable group would be about 11 Å.

Why do we get these peaks equivalent to a junction thickness of $d = 21$ Å? We offer several speculations. First, x-ray diffraction mainly probes the material near the surface and may not reveal other phases present in the interior of our single crystals. Other structures, such as $YBa_2Cu_4O_8$ (124) and $Y_2Ba_4Cu_7O_{15-\delta}$ (247), are known to exist in a 123 single crystal. However, the distance between the nearest inter-unit-cell CuO_2 bilayers for 124 is 13.6 Å, still shorter than the observed 21 $\pm$ 4 Å. Since 247 is a combination of the 123 and 124 phases, the distances between the two CuO_2 bilayers are either 11.7 or 13.6 Å. The second possibility is that there are oxygen deficient regions in the sample, causing some CuO_2 planes to be weaker superconductors or nonsuperconducting. In this case, the CuO_2 bilayers of the next unit cell are needed to form Josephson junctions, doubling the thickness of the junctions. The third possibility is that this is an inherent property of a stack of Josephson junctions coupled to one another. As two overlapping junctions are coupled together, they become essentially one junction with twice the original thickness. The first peak may be related to a case where even more junctions are coupled. Figure 2(a) shows this peak to be near 7 G. However, at slightly different temperatures, its field position would change or the peak would totally disappear even though the other peaks still remained. Thus we conclude that the first peak is not part of the group showing a ΔH of 15 G. Further investigation is still required to more fully understand the nature of the first peak.

Based on the condition $dLH_1 = \Phi_0$, one might argue that the peaks observed in Fig. 1(b) are due to some parasitic Josephson junctions with shorter lengths and thicker electrodes. If this were the case, H_1 would be very sensitive to temperature due to the temperature dependence of the penetration depth. However, this has not been observed. The temperature correlation between the two kinds of peak structures just described gives a natural explanation in terms of small thickness instead of short length. Another connection between the intra- and inter-unit-cell junctions is an anticorrelation between the two groups in their magnetic field positions. Figure 2 shows that the $\Delta H = 15$ G peaks occur in two magnetic field ranges: 0 to 75 G and 125 to 175 G. The field ranges where these 15 G peaks disappear or are drastically affected almost coincide with the field regions where the two peaks associated with the CYC junctions occur, as shown in Fig. 1(b). Small discrepancies are possibly due to the differences in temperature. This anticorrelation indicates that two groups of Josephson junctions share the same CuO_2 planes. It seems that when one complete flux quantum enters each CYC junction, it weakens the electrodes of the inter-unit-cell Josephson junctions.

We have also observed evidence that the intrinsic Josephson junctions in a YBCO single crystal evolve naturally from the short-junction into the long-junction regime as temperature is reduced. Figure 3 shows that the threshold field required for the entry of one flux quantum becomes larger at lower temperatures as discussed earlier.

In summary, using temperature as a varying parameter and dv/di vs H as a tool, we have shown how different Josephson junctions evolve in a YBCO single crystal. Furthermore, the evidence for uncoupled CuO_2-Y-CuO_2 Josephson junctions in these crystals suggests the existence of 2D superconductivity in the CuO_2 atomic planes.

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