Measurement of the Intrinsic Josephson Coupling Energy between Cu-O Bilayers in a High-T_c Superconductor: Possibility of an Unusual Electron Transmission Rate

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The c-axis dissipation in a magnetic field for oxygen-deficient $YBa_2Cu_3O_{7-\delta}$ single crystals is shown to agree with that of a series stack of Josephson tunnel junctions. The intrinsic Josephson coupling energy, thus obtained, indicates the possibility of increased c-axis coherence below T_c.

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Recent experiments on $Bi_2Sr_2CaCu_2O_8$ [1,2] and $Tl_2Ba_2Ca_2Cu_3O_{10}$ [2] crystals related to the c-axis conduction in highly anisotropic, high-temperature superconductors (HTS) and a subsequent interpretation [3] of the field dependence found in Ref. [1] with the model of Ref. [4] have strongly suggested that these HTS materials behave as a stack of Josephson junctions between well-coupled, bilayers or trilayers of Cu-O. As such, a detailed study of the c-axis transport in the *superconducting* state (by Josephson coupling) will probe the c-axis transmission probability of pairs. It can address c-axis coherency issues in a complementary way to optical reflectivity in the superconducting state [5], in which a plasmon has been linked to more coherent c-axis transport. However, extrapolations to low frequencies determine the conductivity for the quasiparticles, not for the pairs.

Conventional Josephson coupling considers the same tunneling matrix element for both pairs and quasiparticles and is probed with the critical current, I_{cj}, or coupling energy, $E_j \equiv \hbar I_{cj}/e$, through the Anderson-Ambegaokar-Baratoff [6] model as

$$E_j(T) = \frac{\pi \hbar \Delta(T)}{2e^2 R_N} \tanh\left[\frac{\Delta(T)}{2k_B T}\right], \qquad (1)$$

where $\Delta(T)$ is the energy gap and R_N is the normal-state junction resistance, for $T > T_c$ or $B > B_{c2}$ (below T_c). Anderson [7] first suggested that superconducting *pair* transmission along the c-axis should be enhanced relative to the normal state in layered HTS cuprates. This was followed by other proposals of enhanced c-axis coherency [8]. If these ideas [7,8] are correct, then R_N (interpreted now as the normal-state resistance of *each* junction between Cu-O bilayers) in Eq. (1) should be replaced by an *effective resistance for pair transmission* which is smaller than the normal-state value for quasiparticles. We test this idea by measuring E_j from thermal activation rather than I_{cj}, which is too large in these materials to avoid heating effects. We find the c-axis transmission for pairs enhanced over that for quasiparticles.

The Josephson energy of Eq. (1) is extrinsic, depending on the junction resistance or area. This poses a difficulty if Cu-O bilayers are not coherent across the entire crystal. In addition, the thermally activated broadening of Josephson coupling will be very small in zero field and comparable to broadening by inhomogeneities (e.g., oxygen content). Fortunately both problems are overcome by applying a magnetic field parallel to the c-axis which introduces a new length scale, $\sqrt{\Phi_0/B}$, so that the *intrinsic* E_j, per unit area, can be obtained (as described below).

We used reasonably anisotropic [9], oxygen-deficient $YBa_2Cu_3O_{7-\delta}$ single crystals ($T_c = 32$–75 K), grown to a size ~ 1 mm $\times$ 1 mm $\times$ 50 μm using a self-flux method [10]. The oxygen content was reduced by annealing for 200 h at 500 °C in appropriate mixtures [11] of O_2 in N_2 such that 0.095%, 3.7%, and 8.3% O_2/N_2 yielded $T_c = 32$, 65, and 75 K, respectively. For such large resistivity anisotropies, currents redistribute rapidly in the a-b planes creating a uniform c-axis current. This uniformity and the absence of heating was confirmed by exchanging current and voltage leads on the bottom (only) of the crystal and getting identical results. For all data reported here, we used $J \sim 1$ A/cm^2 for which the current-voltage characteristics were linear.

To test the above ideas, we must establish that the c-axis dissipation ($J \parallel c$) is Josephson related, and not, e.g., due to flux flow. This we show the measured c-axis resistance, R_c, of the $YBa_2Cu_3O_{7-\delta}$ crystal with $T_c \sim 65$ K in Fig. 1 as a function of the angle, ϕ, between the field direction and the c axis for a temperature of 60 K and a field, B, of 1 T. The maximum R_c occurs for $B \parallel c$, which corresponds to zero macroscopic Lorentz force on the Abrikosov vortices, and it drops over 3 orders of magnitude into the noise for $B \parallel ab$ which is the *maximum* Lorentz force and *minimum* pinning direction for the applied flux. This rules out dissipation from external-field vortices [12,13] (e.g, a-b plane components of the c-axis field due to fluctuations), but it is consistent with interbilayer Josephson junctions, as will be shown below.

We also measured $R_c(T)$ at $\phi = 0$ for various B obtained the activation energy, U, from Arrhenius plots. These are shown in Fig. 2 for two $YBa_2Cu_3O_{7-\delta}$ crystals ($T_c = 65$ and 75 K), together with identical determinations for two discrete, thin-film Nb junctions [14]. The striking qualitative similarity leads us to identify U with

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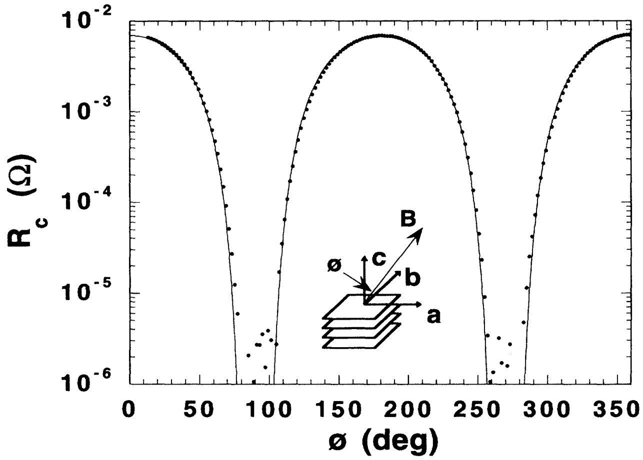

FIG. 1. The c-axis resistance of a $YBa_2Cu_3O_{7-\delta}$ crystal (T_c = 65 K) as a function of the angle between the field direction and the c axis taken at 60 K and 1 T. The solid line is a fit by Eq. (4) for purely Josephson dissipation, using $R_{Nc}^* = 0.1\ \Omega$ and e_j (60 K) $= 2.6 \times 10^{-6}\ \mathrm{J/m^2}$.

E_j for the $YBa_2Cu_3O_{7-\delta}$ crystals and implies consistency with Ref. [14] in which the effective Josephson area, A_{eff}, of the Nb junctions is experimentally shown to be Φ_0/B at high fields. At low fields, U may be limited by inhomogeneity broadening of T_c or A_{eff} may be limited by junction dimensions or defects; we empirically model these by

$$A_{\mathrm{eff}} = \Phi_0/(B + B_0). \tag{2}$$

Then the fits shown in Fig. 2 indicate $B_0 \sim 0.05$ T for $YBa_2Cu_3O_{7-\delta}$. Therefore, high-field data determine the *intrinsic* Josephson coupling energy, $e_j = E_j/A_{\mathrm{eff}}$, which does not depend on the extrinsic area nor resistance of the bilayer junctions.

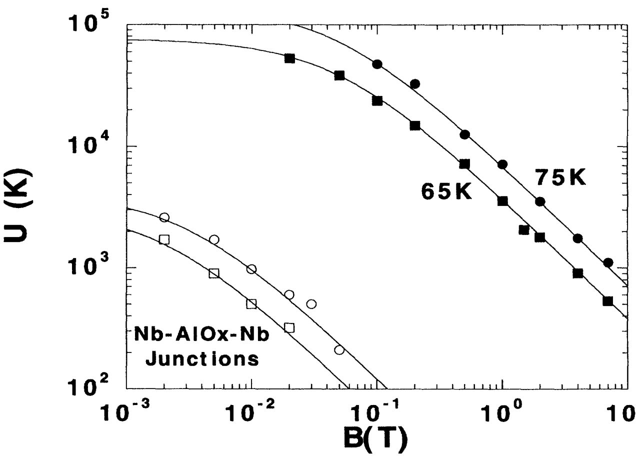

FIG. 2. The activation energy determined from Arrhenius plots of $R_c(T)$ as a function of B for $\phi = 0$. Two $YBa_2Cu_3O_{7-\delta}$ crystals (T_c = 65 and 75 K) and two discrete, thin-film Nb junctions [14] (open symbols) are shown to be strikingly similar with a $1/B$ dependence at high fields. Solid lines are fits by Eq. (2).

In Fig. 3, we show *experimental* evidence of the extent to which it is the c-axis component of B, i.e., $B\cos\phi$, which primarily determines R_c (as first described in Ref. [15] *for a-b plane transport*). The solid lines represent data for field sweeps with $B \parallel c$ from three $YBa_2Cu_3O_{7-\delta}$ crystals (T_c = 32, 65, and 75 K), while the symbols represent rotations at fixed field (of 4, 2, and 1.6 T) for the same crystals. This new result is consistent with Josephson dissipation of Refs. [3], [4], and [14], but not flux motion [16].

Encouraged by this excellent qualitative agreement with Josephson dissipation, we seek a more quantitative fit of the data of Fig. 1 by extending the Ambegaokar-Halperin model [17] for Josephson dissipation to the c axis of layered cuprates [18]

$$R_c(T) = R_{Nc}(T)\{I_0 E_j/2k_BT)\}^{-2}, \tag{3}$$

where I_0 is the modified Bessel function and, without considering possible differences between pair and quasiparticle transmission, R_{Nc} was shown [17] to be the same as the R_N of Eq. (1) for overdamped junctions. For $E_j \gg k_BT$, Eq. (3) reduces to

$$R_c(\phi) = R_{Nc}^* \exp\left(-\frac{e_j(T)\Phi_0}{(|B\cos\phi| + B_0)k_BT}\right), \tag{4}$$

which is consistent with the Arrhenius activation leading to Fig. 2, since $e_j \sim 1 - T/T_c$, and incorporates the results of Figs. 2 and 3. The fit parameters are $e_j \equiv E_j/A_{\mathrm{eff}}$ and R_{Nc}^*, since $B_0 = 0.05$ T (see above). For *conventional* overdamped junctions, R_{Nc}^* would be the normal-state c-axis resistance, R_{Nc}, extrapolated from above T_c, but not for extremely underdamped junctions [19]. Using Eq. (4), we find excellent agreement with the $R_c(\phi)$ data, as shown in Fig. 1: it strongly supports the

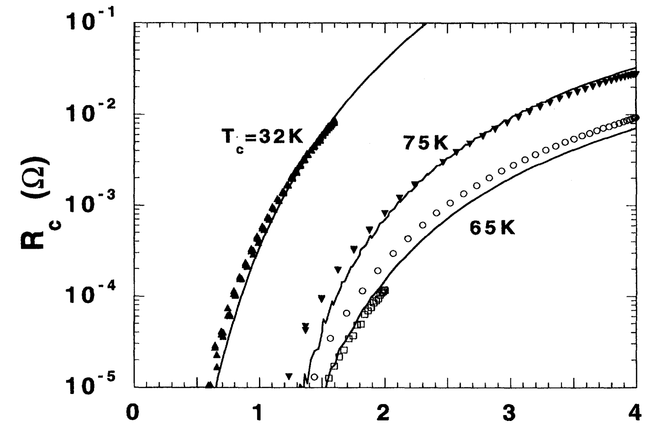

FIG. 3. The c-axis resistance as a function of the *c-axis component* of B. The solid lines are data for field sweeps (with $\phi = 0$), while the symbols represent rotations at fixed fields (of 4, 2, and 1.6 T) for the same crystals. Note that for the 65 K crystal, the rotation data for 4 and 2 T were taken at 51 and 49.5 K, respectively, while the field sweep was done at 50 K, leading to the slight systematic offsets shown.

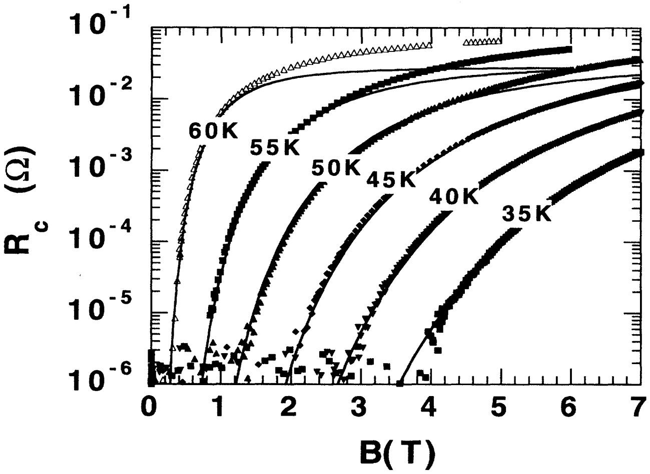

FIG. 4. The c-axis resistance of a $YBa_2Cu_3O_{7-\delta}$ crystal (T_c = 65 K) at various temperatures as a function of B, for $\phi = 0$. Solid lines are fits by Eq. (3).

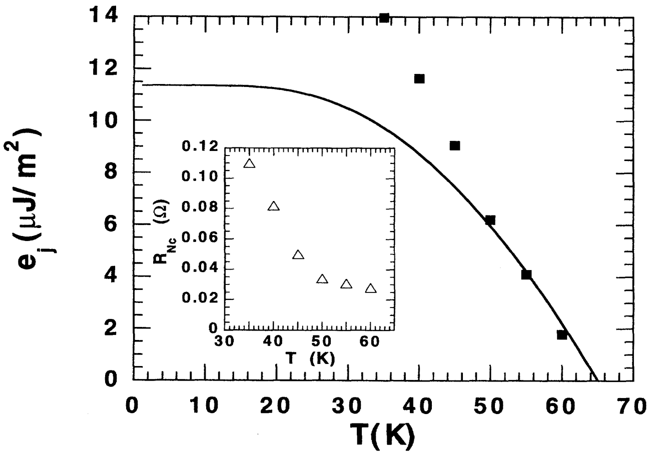

FIG. 5. The intrinsic Josephson coupling energy, e_j (squares), and prefactor resistance, R^*_{Nc} (inset), determined from the fits of Fig. 4. The solid line is e_j determined from Eq. (1) with the BCS temperature dependence for $\Delta(T)$ and a magnitude to match the data.

Josephson model of Refs. [3], [4], and [14] and provides further confirmations of the $1/B$ dependence of E_j and the significance of $B\cos\phi$. In Eq. (1), however, using the fit e_j with any reasonable extrapolation of R_{Nc} from above T_c for R_N, the low-temperature rotational data dictate a much larger $\Delta(T)$ than found by tunneling [20].

In order to systematically address this discrepancy, R_c was measured over a wide range of B and T for the crystal with $T_c = 65$ K. These are plotted in Fig. 4. A vast majority of the data (at lower R_c) fit nicely by Eqs. (3) and (4) (solid lines). Including the quasiparticle conductance, as in Ref. [3], made no noticeable difference in the fits or in the discrepancies at the highest R_c. The fit values of e_j and R^*_{Nc} are shown in Fig. 5. The temperature dependence of R^*_{Nc} is similar to data on extremely underdamped, discrete Nb-Pb junctions [19]. There, the prefactor resistance was related to *dissipation in the phase-slip process:* it was inversely proportional to the quasiparticle density, thus increasing dramatically as temperature decreased, and equaled the junction R_N well below T_c. Thus R^*_{Nc} of Fig. 5 (inset) is likely unrelated to e_j.

Figure 5 also shows the main result of this paper, the fit values of e_j determined by Eqs. (3) and (4). Note that this fitted valued of e_j is strictly independent of Eq. (1), which can be rewritten for the present case of a Cu-O bilayer repeat distance of s, as

$$e_j(T) = \frac{\pi\hbar\Delta(T)}{2e^2\rho_{Nc}s}\tanh\left[\frac{\Delta(T)}{2k_BT}\right], \tag{5}$$

where ρ_{Nc} is the normal-state c-axis resistivity. However, using $\Delta(T) = \Delta_{\rm BCS}(T)$, these e_j do not agree with the temperature dependence [21] of Eq. (1) (solid line in Fig. 5), nor with its magnitude when the extrapolated normal-state resistivity is used for ρ_{Nc}. Agreement with Eq. (1) would require either a larger Δ than $\Delta_{\rm BCS}$ or a significantly smaller *effective resistivity for pair transmission* in Eq. (5) than the normal-state value for quasiparticles. The first case leads to unphysically large Δ (~100 meV for $T \ll T_c$), so we conclude that the *effective resistivity for pair transmission* must be significantly smaller than ρ_{Nc}. This difference is demonstrated graphically in Fig. 6 in which (1) quasiparticle transmission is represented by the incoherent, thermally activated normal-state resistivity ρ_{Nc} above T_c, and (2) pair transmission is represented below T_c by the coherent, metallic effective resistivity determined by Eq. (5) for the fit values of e_j, and plotted for both [20] $\Delta(T) = \Delta_{\rm BCS}(T)$ and $1.4\Delta_{\rm BCS}(T)$. Similar behavior was seen in fits [3] to the c-axis resistivity data [1] of $Bi_2Sr_2CaCu_2O_8$; using a constant ρ_{Nc}, the high-temperature fit by the Josephson model was consistent

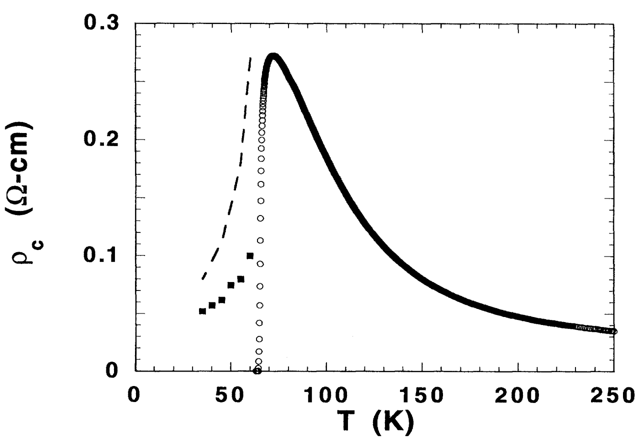

FIG. 6. The c-axis resistivity of a $YBa_2Cu_3O_{7-\delta}$ crystal (T_c = 65 K) in zero field as a function of T (open circles), together with the effective resistivity for pair transmission as determined for each e_j in Fig. 5 using Eq. (5). The latter are shown for $\Delta(T)$ equal to $\Delta_{\rm BCS}(T)$ (solid squares) and $1.4\Delta_{\rm BCS}(T)$ (dashed line).

with ρ_{Nc}, measured well above T_c, but to fit $\rho_c(T)$ at low temperatures needed a larger e_j.

The discrepancies in Fig. 4 at the highest resistances may be explained by the enhanced c-axis coherency. These deviations occur only as B becomes a sizable fraction of $B_{c2}(T)$ [using $B_{c2}(0) = 56$ T from Ref. [10]], for which the measured R_c is expected to approach an extrapolation of the larger, less-coherent $R_{Nc}(T \gtrsim T_c)$, and such behavior is consistent with Fig. 4.

In conclusion, our data undeniably show that Josephson dissipation dominates the c-axis transport, and the details suggest (1) the possibility of enhanced pair transmission, relative to normal-state quasiparticles, and (2) that these Josephson junctions are underdamped. The first point is consistent with recent theoretical models [7,8]. We also show that the c-axis dissipation is only dependent upon the magnetic field component directed along the c-axis [13].

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