Abstract

We have magnetically imaged interlayer Josephson vortices emerging from an ac face of single crystals of the single layer cuprate high-\(T_c\) superconductor \((\text{Hg,Cu})\text{Ba}_2\text{CuO}_{4+\delta}\). These images provide a direct measurement of the \(c\)-axis penetration depth, \(\lambda_c \sim 10 \, \mu\text{m}\). This length is a factor of 10 longer than predicted by the interlayer tunneling model for the mechanism of superconductivity in layered compounds, indicating that the condensation energy available through this mechanism is 100 times smaller than is required for superconductivity.
In the interlayer tunneling (ILT) model for superconductivity in layered superconductors such as the cuprates, transport of carriers between the planes is incoherent in the normal state, but coherent interlayer transport is allowed for Cooper pairs. The coherent pair tunneling lowers the c-axis kinetic energy, supplying the superconducting condensation energy, \( E_c \). Anderson, Leggett, and Chakravarty have each argued that the comparison of the experimentally measured c-axis penetration depth, \( \lambda_c \), and the value determined within the ILT model from the condensation energy, \( \lambda_{ILT} \), is an important test of the ILT mechanism. In all presently published versions of the theory, \( \lambda_c \approx \lambda_{ILT} \).

The calculation of \( \lambda_{ILT} \) and its comparison with experiment are most straightforward in materials with a single copper oxide layer per unit cell, such as La\(_{2-x}\)Sr\(_x\)CuO\(_{4+\delta}\) (La-214), HgBa\(_2\)CuO\(_{4+\delta}\) (Hg-1201), and Tl\(_2\)Ba\(_2\)CuO\(_{6+\delta}\) (Tl-2201). Different experiments on these materials have not provided a single answer to the question. In La\(_{2-x}\)Sr\(_x\)CuO\(_{4+\delta}\) (La-214), measurements of the Josephson plasma frequency \( \omega_p = c\lambda_c^{-1}\epsilon^{-1/2} \) (where \( c \) is the speed of light and \( \epsilon \) is the dielectric constant of the interlayer medium) are in good agreement with the predictions of the ILT model. In Tl\(_2\)Ba\(_2\)CuO\(_{6+\delta}\) (Tl-2201), Moler et al. observed interlayer Josephson vortices with \( \lambda_c = 17 - 21 \) microns. Measurements of the plasma resonance \( \omega_p \) and dielectric constant \( \epsilon \) by Tsvetkov et al. indicate \( \lambda_c = 17 \) microns. In HgBa\(_2\)CuO\(_{4+\delta}\) (Hg-1201), Panagopoulos et al. obtained a value of \( \lambda_c(T = 0) = 1.36 \pm 0.16 \) \( \mu m \) from magnetic susceptibility data on oriented powders. As both Anderson and Leggett have pointed out, a confirmation of the c-axis penetration depth in Hg-1201 seems essential to resolving this issue.

In this Letter we directly measure the c-axis penetration depth in Hg-1201 by magnetically imaging interlayer Josephson vortices emerging from the ac face of single crystals. These measurements give \( \lambda_c \approx 10 \mu m \), much longer than the value \( \lambda_c \approx 1 \mu m \) reported by Panagopoulos et al. Our value for \( \lambda_c \), when combined with previous results from Tl-2201, make it appear unlikely that presently published versions of the ILT model are the correct description for superconductivity in the cuprate high-\( T_c \) superconductors.

The Hg-1201 crystal growth has been described previously. Our method consists
in preparing a Ba/Cu/O precursor in flowing oxygen, and then mixing it with HgO to make a 0.8:2:1.2 ratio of Hg:Ba:Cu. An alumina crucible containing this powder is then sealed in a silica tube. After 48 hours of thermal treatment, black, platelet-like crystals are extracted. The largest crystals (typical dimensions ∼1×1×0.08mm) are selected for transport and SQUID imaging measurements. EDX and electron diffraction measurements lead to the formula Hg\textsubscript{0.8}Cu\textsubscript{0.2} for the mixed mercury layer. The copper position is displaced with respect to the Hg site in the mixed layer as shown from structural refinements. No intergrowth nor extended defects have been shown by high resolution microscopy \[15\]. The Hg-1201 single crystals were mounted in epoxy so that the ac face was aligned vertically, and then the sample and epoxy were polished to make a flat, smooth surface to scan.

The magnetic imaging measurements were made with a scanning SQUID microscope \[16\], in which a sample is scanned relative to a superconducting pickup loop oriented nearly parallel to the sample surface. The data are represented as the magnetic flux $\Phi_s$, the integral of the normal component of the magnetic field over the pickup loop, vs the position of the pickup loop in the $x, y$ plane. The pickup loop is fabricated with well-shielded leads to an integrated Nb-Al\textsubscript{2}O\textsubscript{3}-Nb SQUID. For these measurements, we used a SQUID with our smallest pickup loop, an octagon $L = 4\mu$m in diameter with 0.8$\mu$m linewidth. The silicon substrate carrying the SQUID was polished to a sharp tip a few microns from the edge of the pickup loop, the substrate was oriented an angle $\theta \sim 20^\circ$ from parallel to the sample, and scanned with the tip in direct contact with the sample. Fits to data on Abrikosov vortices indicate that the pickup loop is about $z_0=1\mu$m above the surface of the sample under the experimental conditions used here. Our images were made with both sample and SQUID immersed in liquid helium at 4.2K.

Figure 2 shows a SSM image of a 512$\mu$m×256$\mu$m area of the crystal and surrounding epoxy. The outer edges of the crystal face are indicated by dashed lines. The sample was cooled and imaged in a field of about 10mG. Visible in this image are about 50 interlayer Josephson vortices. These vortices are remarkably uniform in shape. Some are sufficiently isolated from their neighbors that overlapping fields do not interfere with detailed modelling.
Although one must consider that the vortices may be pinned in areas with unusually weakly coupled planes, the uniformity of the vortex shapes over a large area of the crystal face makes us believe that we are measuring an intrinsic penetration depth. This belief is supported by the good agreement between bulk plasma resonance measurements of $\omega_j$ and local interlayer Josephson vortex imaging measurements of $\lambda_c$ in Tl-2201 [12]. Three vortices chosen for further analysis are shown in figure 2. Cross-sections through the image data parallel to the layers are displayed in Figure 3.

The decay of the observed vortex magnetic flux perpendicular to the layers (along the c-axis) is determined by the size of the pickup loop. In contrast, the extent of the images of the vortices along the layers provides a direct measure of $\lambda_c$.

For quantitative modelling of the shape of the vortex we use the results of Clem and Coffey [17]. Neglecting the influence of the surface on the fields, the $z$-component of the magnetic field of an interlayer vortex is given by [17]:

$$B_z(x, y, z = 0) = \frac{\Phi_0}{2\pi \lambda_{ab} \lambda_c} K_0(\tilde{R}),$$

where $\lambda_{ab}$ is the in-plane penetration depth, $K_0$ is a modified Bessel function of the second kind of order 0, $\Phi_0 = hc/2e$ is the superconducting flux quantum, $h$ is Planck’s constant, $e$ is the charge on the electron, $\tilde{R} = ((s/2\lambda_{ab})^2 + (x/\lambda_{ab})^2 + (y/\lambda_c)^2)^{1/2}$, $s$ is the interplane spacing, $x$ is the distance perpendicular to the planes, and $y$ is the distance parallel to the planes. Since for our experiments $s \ll \lambda_{ab} \ll (L, z_0) \ll \lambda_c$, we neglect both $s$ and $\lambda_{ab}$.

With this assumption, the fields from Eq. 1 are propagated to a height $z = z_0$ [18] and then summed over the geometry of the pickup loop. This model has two free parameters: $\lambda_c$, which determines the length of the vortex, and $z_0$, which determines the magnetic amplitude of the vortex image. Fits to the three cross-sections (Figure 3) yield consistent values for the interlayer penetration depth $\lambda_c = 10 \pm 1 \mu m$.

This measured value is about ten times longer than the theoretical value, $\lambda_{ILT} = 1 \pm 0.5 \mu m$ [5]. Since the ILT condensation energy is proportional to $1/\lambda_c^2$, the ILT supplied condensation energy is about 100 times smaller than the estimated actual energy in Hg-
A more conventional estimate of $\lambda_c$ comes from the Lawrence-Doniach model [19]. For diffusive pair transfer (parallel momentum not conserved) between superconducting layers [19–21], the Josephson current between two identical superconducting sheets at $T=0$ is given by [21]

$$J_0 = \frac{\pi \Delta(0)}{2eR_{c,n}}$$

(2)

where $\Delta(0)$ is the zero temperature energy gap and $R_{c,n}$ is the normal state $c$-axis interplane resistance. The interlayer penetration depth is given by $\lambda_\perp = (e\Phi_0/8\pi^2 sJ_0)^{1/2}$ [17]. The application of this model requires at least two assumptions: that $R_n$ is temperature independent below $T_c$, or at least that the temperature dependence can be understood well enough for extrapolation [8], and that the gap is $s$-wave [22–24]. $d$-wave superconductivity would tend to increase $\lambda_c$ from this value: in a purely tetragonal $d$-wave superconductor, with purely diffusive pair transfer, the coupling would be reduced to zero [22–24]. With these considerations in mind, one would not expect the Ambegaoker-Baratoff model to apply quantitatively, but it is nevertheless natural to look for a correlation between $\lambda_c$ and $R_{c,n}$.

Figure 4a shows measurements of the $c$-axis resistivity of two of our Hg-1201 crystals as a function of temperature. These measurements were made by evaporating four Ag stripes, two on each $ab$ face of the crystal, as diagrammed in the inset of Fig 4a [25]. The contacts as prepared have high resistance, but after an annealing step at 400°C for 10 minutes the contact resistances drop to 1 to 2 Ohms. We have carefully checked that such annealing does not alter the superconducting transition by measuring the susceptibility of control crystals before and after such an annealing step. Because of the relatively high anisotropy of Hg-1201 and large thickness of our crystals ($\sim 80\mu m$), these crystals are electrically thick [26]. Therefore difficulties in separating the in- and out-of-plane conductivities such as reported by Hussey et al. [27] for YBa$_2$Cu$_4$O$_8$ should not occur. The midpoint $T_c=94K$ of the $\rho_c(T)$ curves is consistent with the onset $T_c=95-97K$ observed magnetically on such
crystals and confirms that the doping is close to optimal. Taking $\rho_{c,n} = \sigma_{c,n}^{-1} = 0.5$ Ohm-cm, and a BCS gap value $2\Delta = 3.54k_B T_c = 28$ meV, leads to a predicted penetration depth $\lambda_c = (\hbar c^2 / 4\pi^2 \Delta \sigma_{c,n})^{1/2} = 8 \mu m$.

Basov et al. [28] have previously noted a correlation between the values of the penetration depth $\lambda_c$ and the far-infrared $c$-axis conductivity, $\sigma_{c,FIR}$. In Figure 4b add our results for Hg-1201 and Tl-2201 [10], replacing $\sigma_{c,FIR}$ with $\sigma_{c,n}$ just above $T_c$. The dashed line is Eq. (2), assuming $2\Delta = 28$meV. This simple model, although it assumes the same $\Delta$ for all cuprates, and has the shortcomings outlined above, agrees with all of the measurements to within a factor of three.

It is not clear why our results for $\lambda_c$ are nearly a factor of 7 larger than those of Panagopoulos et al. There are possible systematic errors in both measurements. In the analysis of the imaging experiments, the spreading of the vortex near the surface is neglected. This neglect is justified within our experimental resolution on the basis of a consideration of the free energy associated with vortex spreading in highly anisotropic superconductors [29], as well as by the quantitative experimental agreement between $\lambda_c$ and $\omega_p$ in Tl-2201 [12].

For the powder magnetization results, the analysis depends on a very accurate characterization of the distribution of particle sizes; it is possible that this analysis could be skewed by the presence of a slightly higher number of large particles than used in the modelling [30]. A more likely explanation is that the different samples do in fact have different $\lambda_c$ values. First, the doping in the two samples is not identical. It has been shown that the transport anisotropy of several materials can depend strongly on doping [31], but it has also been shown that the anisotropy in Hg-1201 is nearly constant near optimal doping [32]. Another possibility is the influence of the excess copper on the mercury layer. Neutron studies on ceramics [33,34] conclude they also have mixed Hg/Cu layers. It is not clear whether the ceramics of Panagopoulos et al. have similar substitutions on the mercury layer.

If the Hg-1201 crystals used in this study and the Tl-2201 crystals used in the previous study are true single-layer materials, then the long $c$-axis penetration depths $\lambda_c \approx 10$ microns for Hg-1201 and $\lambda_c \approx 20$ microns for Tl-2201 pose a serious challenge to the ILT model.
Although one can hypothesize structural explanations, perhaps based on the excess copper, to make these results consistent with the ILT model, such explanations must be found for both \((\text{Hg,Cu})\text{Ba}_2\text{CuO}_{4+\delta}\) and \(\text{Tl}_2\text{Ba}_2\text{CuO}_{6+\delta}\).

These two results fall into a previously unfilled range of penetration depths in the Basov correlation. This correlation, \(\lambda_c \approx (\hbar c^2/4\pi^2\Delta\sigma_{c,n})^{1/2}\), works surprisingly well given the simplicity of the theoretical model and considering that details of the gap anisotropy, the band structure, the scattering, and the temperature dependence of the normal-state \(c\)-axis resistivity are all unknown in these particular materials.

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FIGURES

FIG. 1. Scanning SQUID microscope image of a 256×512 micron area of the edge of a single crystal of Hg-1201, cooled in a field of about 10mG, and imaged at 4.2K. The false color lookup table corresponds to a total variation in flux through the SQUID pickup loop of 0.55Φ₀. The dashed lines indicate the top and bottom ab faces of the crystal. The box indicates the area that is expanded in Figure 2.

FIG. 2. Expanded view of a 54×100 micron area of the image of Figure 1. The dashed lines indicate the paths of cross-sections through the data parallel to the planes displayed in Figure 3.

FIG. 3. The symbols are cross-sectional data through the 3 representative vortices indicated in Figure 2, offset vertically for clarity. The solid lines are fits to the data, as described in the text. The fit penetration depths λ_c are as labelled in the figure, and the effective heights are z₀ = 0.6±0.1μ m(A), 0.8±0.1μ m(B), and 0.8±0.2μm(C). The error bars are assigned using a doubling of the best-fit χ² as a criterion.

FIG. 4. (a) c-axis resistivity vs temperature for two of our crystals. The contact geometry used is diagrammed in an insert. (b) Correlation plot between c-axis conductivity and c-axis penetration depth, following Basov et al. (Ref. 29). We have included our present data for Hg-1201 and previous data on Tl-1201. The dashed line is the prediction for diffuse tunneling between superconducting layers, assuming a gap value 2Δ=28meV. In this figure 124=YBa₂Cu₄O₈, 2212=Bi₂Sr₂CaCu₂O₈, ps₂=ps₃=Pb₂Sr₂RCu₃O₈, 214=La₁.₈₄Sr₀.₁₆CuO₄, 123=YBa₂Cu₃O₇.
A. $\lambda_c = 10 \mp 1 \mu m$

B. $\lambda_c = 10 \mp 1 \mu m$

C. $\lambda_c = 9 \mp 1 \mu m$
