6th International Conference on Materials and Mechanisms of Superconductivity and High-Temperature Superconductors
Feb. 20-25, 2000 - Houston, Texas
To be Published in Physica C

submitted to M2S
ANL/CHM/CP-101212

Images of Interlayer Josephson Vortices in Single-Layer Cuprates

K. A. Moler,[a] J. R. Kirtley,[b] Ruixing Liang,[c] D. A. Bonn,[c] W. N. Hardy,[c] J. M. Williams,[d] J. A. Schlueter,[d] D. Hinks,[d] G. Villard,[e] A. Maignan,[e] M. Nohara,[f] and H. Takagi[f]

[a]Department of Applied Physics, Stanford University, Stanford, CA 94305-4045

[b]IBM T.J. Watson Research Center, P.O. Box 218, Yorktown Heights, NY 10598

[c]Dept. of Physics and Astronomy, U. of British Columbia, Vancouver, BC, V6T1Z1, Canada

[d]Chemistry and Material Science Division, Argonne National Laboratory, Argonne, IL 60439

[e]CRISMAT-ISMRA, 6 Boulevard du Maréchal Juin, 14050 Caen Cedex, France

[f]Inst. Solid State Physics, University of Tokyo, Roppongi, Minato-ku, Tokyo 106, Japan

The interlayer penetration depth in layered superconductors may be determined from scanning Superconducting QUantum Interference Device (SQUID) microscope images of interlayer Josephson vortices. We compare our findings at 4 K for single crystals of the organic superconductor κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ and three near-optimally doped cuprate superconductors: $La_{2-x}Sr_xCuO_4$, (Hg,Cu)$Ba_2CuO_{4+\delta}$, and $Tl_2Ba_2CuO_{6+\delta}$.

1. INTRODUCTION

Each vortex in a type II superconductor carries a single quantum of magnetic flux, $\Phi_0 = hc/2e = 20.7$ Gauss-μm^2. The spatial extent of a vortex in the direction perpendicular to the vortex axis is determined by the magnetic penetration depth, λ. Because of the layered nature of cuprate superconductors, the penetration depth is very anisotropic. The in-plane penetration depths, λ_a and λ_b, are usually much less than a micron. The interlayer or c-axis penetration depth, λ_c, ranges from microns to hundreds of microns for different cuprates with different dopings. The c-axis penetration depth is the length scale on which a magnetic field parallel to the layers can change in the direction parallel to the layers (figure 1). A larger λ_c indicates a smaller out-of-plane super-current density.

Superconducting QUantum Interference Devices (SQUIDs) can have magnetic flux sensitivities in the $10^{-6}\Phi_0$ range [1,2]. In

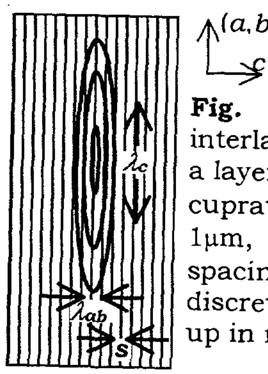

Fig. 1. Cross-section of an interlayer Josephson vortex in a layered superconductor. For cuprates, $\lambda_{ab} \approx 0.1$-$0.2\mu m$, $\lambda_c >$ 1μm, and the interlayer spacing s ≈ 10Å, so the discrete layers do not show up in magnetic images.

practical SQUID microscopes, the pickup loops have so far been microns or tens of microns in size [1,2]. By imaging interlayer Josephson vortices emerging parallel to the planes on a crystal face, it is possible to determine λ_c quantitatively. Results in two single-layer cuprate superconductors disagree with predictions [3,4] of the Inter-Layer Tunneling (ILT) model proposed by P.W. Anderson and co-workers as a mechanism for superconductivity [3].

DISCLAIMER

2. SQUID MICROSCOPY

The I-V characteristic of a SQUID is periodic in the total magnetic flux through the SQUID loop with periodicity Φ_0. SQUID controllers feed back current to a modulation or "mod" coil to keep the total flux through the SQUID pickup loop constant. The fields from the modulation coil perturb the sample. In order to study vortices, it is desirable to use SQUIDs with a pickup loop which is some distance from the modulation coil [2,5] (figure 2). The leads to the pickup loop should be well-shielded to prevent parasitic lead pickup. Fabrication considerations are the current limitation on the state-of-the-art pickup loop size [5].

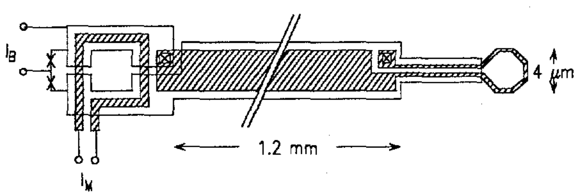

Fig. 2. Sketch of a SQUID designed for scanning SQUID microscopy. The 4 μm octagonal pickup loop is separated from the junctions and mod coil (I_M).

The fact that the output signal of a SQUID controller is simply proportional to the local magnetic field averaged over the area of the SQUID loop aids in the quantitative interpretation of images. The shielded leads can complicate the interpretation, since some flux is pushed into the pickup loop by the shield.

3. IMAGES OF VORTICES

We imaged two single crystals of κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ [6], one single crystal of $La_{1.83}Sr_{0.17}CuO_4$ [7], two single crystals of (Hg,Cu)$Ba_2CuO_{4+\delta}$ [8], and three single crystals of $Tl_2Ba_2CuO_{6+\delta}$ [9,10,11]. The cuprates were all near optimal doping. Measurements in κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ were made with 8 micron square pickup loops. Measurements in (Hg,Cu)$Ba_2CuO_{4+\delta}$ and $La_{2-x}Sr_xCuO_4$ were made with 4 micron octagonal pickup loops. Measurements in $Tl_2Ba_2CuO_{6+\delta}$ were made with both types of pickup loops. Typical results are shown in figure 3, along with ab-plane vortices in $YBa_2Cu_3O_{7-\delta}$ imaged with similar pickup loops. Since the ab-plane vortices are much smaller than the pickup loop, their images serve to illustrate the resolution of the SQUID microscopy.

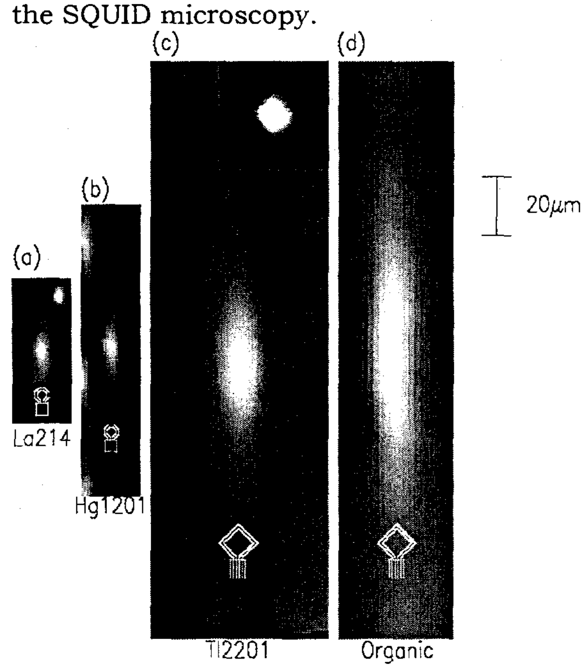

Fig. 3. Images of interlayer Josephson vortices in (a) $La_{1.83}Sr_{0.17}CuO_4$, (b) (Hg,Cu)$Ba_2CuO_{4+\delta}$, (c) $Tl_2Ba_2CuO_{6+\delta}$, and (d) κ-(BEDT-TTF)$_2$Cu(NCS)$_2$. In each case a sketch of the pickup loop used to make the measurement is shown. The insets in (a) and (c) show ab-plane vortices in $YBa_2Cu_3O_{7-\delta}$ imaged with a four micron octagonal pickup loop and an eight micron square pickup loop respectively.

The effective shape of the pickup loop is anisotropic due to the shielded leads, as seen in the figure 3a inset. However, for all four of the interlayer Josephson vortices shown in figure 3, the in-plane penetration depth is much less than the instrumental resolution, and the interlayer penetration depth is larger than the instrumental resolution. The interlayer penetration depth may be roughly estimated simply by measuring the long axis of each vortex.

4. ANALYSIS

In order to obtain more quantitative results, the data were analyzed using an exact expression for the magnetic fields of a vortex at a superconductor-vacuum interface in an anisotropic London model, as described in reference 10. This analysis also accounted for the finite size of the pickup loops. The results are shown in table 1.

Table 1
Interlayer penetration depths determined experimentally by SQUID microscopy

Material	λ_{exp}
κ-(BEDT-TTF)$_2$Cu(NCS)$_2$	λ_a=63±15μm
$La_{1.83}Sr_{0.17}CuO_4$	λ_c=5±1μm
(Hg,Cu)$Ba_2CuO_{4+\delta}$	λ_c=8±1μm
$Tl_2Ba_2CuO_{6+\delta}$	λ_c=18±3μm

The measurements of (Hg,Cu)$Ba_2CuO_{4+\delta}$ and $Tl_2Ba_2CuO_{6+\delta}$ were motivated by the predictions of the ILT model [3,4]. The measurements of $La_{1.83}Sr_{0.17}CuO_4$ were made to check this technique for determining λ_c, which is known to be 4½ μm in $La_{1.83}Sr_{0.17}CuO_4$ [12].

The size of the vortices in $La_{1.83}Sr_{0.17}CuO_4$ is comparable to the size of the 4 micron octagonal pickup loop. We estimated that the systematic errors induced by the shape of the pickup loop were comparable to or smaller than our statistical errors, and we verified this estimate by measuring $La_{1.83}Sr_{0.17}CuO_4$ in two different orientations (figure 4).

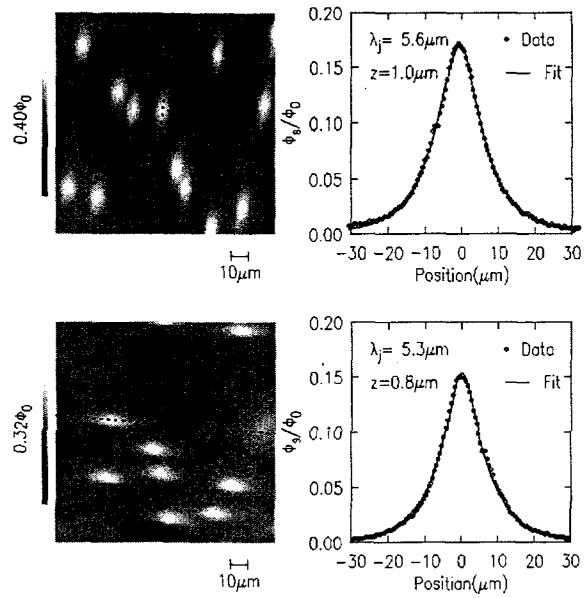

Fig. 4. Images of interlayer Josephson vortices in $La_{1.83}Sr_{0.17}CuO_4$ taken with two different relative orientations of the sample and pickup loop, to check for systematic errors induced by the pickup loop shape, and fits to one vortex from each image.

5. COMPARISON WITH OTHER EXPERIMENTS

Our value for $La_{1.83}Sr_{0.17}CuO_4$ agrees with the previously accepted value within statistical error. Panagopoulos *et al.* have conducted magnetic susceptibility studies of oriented powders of $HgBa_2CuO_{4+\delta}$, finding λ_c = 1.36±0.16μm in a slightly overdoped sample [13]. We are at a loss to explain this discrepancy. There have been two measurements of λ_c by other techniques in $Tl_2Ba_2CuO_{6+\delta}$. Tsvetkov *et al.* found λ_c=17μm [11], and Basov *et al.* found λ_c=12μm [14]. The discrepancy between our results and Basov's are within the range that could be explained by doping dependence.

6. COMPARISON WITH ILT

The Inter-Layer Tunneling (ILT) model proposed by Anderson and coworkers as a mechanism of superconductivity rests on the assumption that the interlayer coupling is much weaker in the normal state than it is in the superconducting state. Therefore, a layered cuprate will save c-axis kinetic energy in the superconducting state. The predictions shown in Table 2 result from the assumption that all of the condensation energy comes from this saved c-axis kinetic energy, and use experimental values for the measured condensation energy. It is best to test these predictions in a material with only one copper oxide plane per unit cell. It is also best to use a material with a high T_c. The two highest-Tc single-layer materials are $(Hg,Cu)Ba_2CuO_{4+\delta}$ and $Tl_2Ba_2CuO_{6+\delta}$.

Table 2
Interlayer penetration depths predicted by the ILT model [3,4]

Material	λ_{ILT}
κ-$(BEDT\text{-}TTF)_2Cu(NCS)_2$	$\lambda_{ILT}=11\mu m$
$La_{1.83}Sr_{0.17}CuO_4$	$\lambda_{ILT}=3\mu m$
$(Hg,Cu)Ba_2CuO_{4+\delta}$	$\lambda_{ILT}=1\mu m$
$Tl_2Ba_2CuO_{6+\delta}$	$\lambda_{ILT}=1\mu m$

The experimental results are not in good agreement with the ILT predictions. Ideally, the measurements of λ_c and the condensation energy should be made on the same sample, and Chakravarty *et al.* [15] has pointed out some of the subtleties of determining the condensation energy from the measured specific heat. However, the fraction of the condensation energy that could be supplied by the ILT mechanism is roughly $(\lambda_{ILT}/\lambda_c)^2$ [4], suggesting that only about 1% of the condensation energy of $(Hg,Cu)Ba_2CuO_{4+\delta}$ and $Tl_2Ba_2CuO_{6+\delta}$ could be supplied by the ILT mechanism.

This work was supported in part by IBM research and by the NSF through a seed grant from the Princeton MRSEC. Work at Argonne National Laboratory was supported by the US Department of Energy, Office of Basic Energy Sciences, Division of Materials Sciences, under Contract W-31-109-ENG-38.

REFERENCES

1. L.N. Vu, M.S. Wistrom, D.J. Van Harlingen, *Appl. Phys. Lett.* **63**, 1693 (1993).
2. J.R. Kirtley *et al.*, *Appl. Phys. Lett.* **66**, 1138 (1995); J.R. Kirtley, *et al.*, *IBM J. Res. Dev.* **39**, 655 (1995); J.R. Kirtley, *IEEE Spectrum* **33**, 40 (1996); J.R. Kirtley *et al.*, *Appl. Phys. Lett.* **74**, 4011 (1999).
3. P.W. Anderson, *Science* **268**, 1154 (1995), and references therein.
4. A.J. Leggett, *Science* **274**, 587 (1996); P.W. Anderson, *Science* **279**, 1196 (1998).
5. M.B. Ketchen and J.R. Kirtley, *IEEE Trans. Appl. Supercon.* **5**, 2133 (1995).
6. J. R. Kirtley *et al.*, *J. Phys. B.* **11**, 2007 (1999).
7. J. R. Kirtley *et al.*, *J. Phys. Chem. Sol.* (in press).
8. J. R. Kirtley *et al.*, *Phys. Rev. Lett.* **81**, 2140 (1998).
9. K. A. Moler *et al.*, *Science* **279**, 1193 (1998).
10. J. R. Kirtley *et al.*, *Phys. Rev. B* **59**, 4343 (1999).
11. A. A. Tsvetkov *et al.*, *Nature* **395**, 362 1998.
12. S. Uchida, K. Tamasaku, S. Tajima *Phys. Rev. B* **53**, 14588 (1996); T. Shibauchi *et al.*, *Phys. Rev. Lett.* **72**, 2263 (1994).
13. C. Panagopoulos *et al.*, *Phys. Rev. Lett.* **79**, 2320 (1997).
14. D.N. Basov, *et al.*, *Science* **283**, 49 (1999).
15. S. Chakravarty, Hae-Young Kee, E. Abrahams, *Phys. Rev. Lett.* **82**, 2366 (1999).