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Weak coupling and anisotropy in the magnetic penetration depth of the high-temperature superconductor $Tl_2Ca_2Ba_2Cu_3O_{10+\delta}$

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Magnetic penetration depths $\lambda(T)$ have been determined as a function of temperature T for the high-temperature superconductor Tl₂Ca₂Ba₂Cu₃O_{10+ δ}. Magnetization measurements were performed on an aligned, composite powder sample ($T_c = 123$ K) with the magnetizing field parallel or perpendicular to the common c axes of the dispersed crystallites. The magnetization in the vortex state was analyzed using the theory of Kogan *et al.* [Phys. Rev. B 38, 11958 (1988)] to obtain values for λ . The results were best described by BCS weak-coupling theory in the clean limit, with deduced values $\lambda(0)$ of 173 and 480 nm for the components of the penetration-depth tensor.

INTRODUCTION AND EXPERIMENTAL ASPECTS

Despite intensive investigation during the past three years, an understanding of many aspects of high-temperature superconductors (HTSC) remains elusive, including the mechanism whereby Cooper pairs are formed. One clue can be provided by experiments that show whether the coupling is weak or strong, and a property that is sensitive to the coupling strength is the temperature dependence of the magnetic penetration depth λ . This quantity has fundamental importance, too, and should reflect the anisotropy of the layered HTSC's.

We have investigated the penetration depth $\lambda(T)$ in the Tl-based superconductor $Tl_2Ca_2Ba_2Cu_3O_{10+\delta}$ that remains the highest- T_c material known to date. Using a grain-aligned composite sample, we have performed experiments over a substantial range of reduced temperature to obtain λ for directions both perpendicular and parallel to the long c axis of the tetragonal unit cell. To our knowledge, this is the first such study of the anisotropic penetration depth in the Tl-based HTSC system. Earlier measurements of changes in $\lambda_{a-b}(T)$, made on single crystal YBa₂Cu₃O_z in the Meissner state^{1(a)} with the field parallel to the *a-b* plane, were interpreted in terms of BCS weak-coupling theory. Very recently, a study^{1(b)} employing methods similar to those used here reported values for the average penetration depth and components of the penetration depth tensor in Y-Ba-Cu-O and Bi-based HTSC's. In the present work, both components $\lambda_a(T)$

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and $\lambda_c(T)$ were determined and compared with theoretical expressions based on weak and strong coupling in the clean and dirty limits. It was found that the temperature dependence is best described by weak-coupling theory in the clean limit and that the two principal values of the penetration-depth tensor differ by a factor of 2.8 or more.

Bulk polycrystalline Tl₂Ca₂Ba₂Cu₃O_{10+ δ} material with relatively high-phase purity² was formed by repeated grinding and sintering of a stoichiometric mixture of constituents. An aligned composite sample was produced³ from fine monocrystalline powder of ~ 1 - μ m size, dispersed in "45 min" liquid epoxy, and aligned in a 5-T field. The volume fraction of superconductor (8.66 mm³) was 6.5%. X-ray diffraction with the scattering vector parallel to the alignment field direction revealed very strong (001) reflections, with a rocking curve width of $\sim 1^{\circ}$ for the (0022) line, indicating very good alignment of the *c* axes. The high transition temperature (123 K) indicates little intergrowth of one and two Cu-O layers in the Tl 2:2:2:3 compound.⁴

The magnetic investigations were carried out in a Quantum Design superconducting quantum interference device (SQUID) magnetometer, equipped with magnet current circuitry to ensure monotonic field changes, and in a vibrating sample magnetometer. The field was applied either parallel or perpendicular to the common c axes of the sample and the magnetization M(H,T) was measured at fixed temperatures T as a function of field H, with frequent checks to ensure that M was reversible. The data

were corrected for the background moment due to epoxy, core diamagnetism, etc. The main measurements were made in the superconducting mixed state with fields of 8-50 kG, i.e., with $H \gg H_{c1}$, the lower critical field. In this range, the mean spacing between fluxoids was much smaller than the particle size, so surface effects are unimportant; also, with $B \cong H$, the results are independent of variables like the grain size and shape and the demagnetizing factor that are difficult to control.

RESULTS AND DISCUSSION

In the vortex state of a uniaxial type-II superconductor, Kogan, Fang, and Mitra⁵ have developed a theory in the London limit that relates the magnetization M to applied field H as

$$M(H,T) = (\phi_0/32\pi^2 \lambda_{\rm eff}^2) \ln(\beta H_{c2}/H), \qquad (1)$$

where ϕ_0 is the flux quantum, β is a constant of order unity, and H_{c2} is the angularly dependent upper critical field. This relation is valid for $H_{c1} \ll H \ll H_{c2}$ when the magnetization is reversible, and applies to uniaxial materials such as tetragonal, layered Tl 2:2:2:3. There are two independent eigenvalues λ_a and λ_c in the penetration-depth tensor, where λ_i is explicitly defined as the depth of field penetration screened by supercurrent flow in the *i*th direction. With $H \parallel c$ axis, one has in Eq. (1) that $\lambda_{\text{eff}}^2 = \lambda_a^2$ so that M depends only on λ_a ; with H in the basal *a-b* plane, one has $\lambda_{\text{eff}}^2 = \lambda_a \lambda_c$.

We found that Eq. (1) described the experimental data quite well. The logarithmic slopes $dM/d[\ln(H)]$ obtained from plots of M vs $\ln(H)$ yielded values for $\lambda_a^2(T)$ (where $H||_c$ and for $\lambda_{\text{eff}}^2(T) = \lambda_a(T)\lambda_c(T)$ (where H||a-b). From Ginzburg-Landau theory, one has rather generally that λ^{-2} is proportional to $(T_c - T)$ near T_c , regardless of the form of pair coupling. The experimental results followed this linear dependence for both field orientations, and the T_c was defined as the temperature at which λ_{eff}^{-2} extrapolated to zero. The value of T_c chosen has a substantial impact on an analysis of the temperature dependence of λ ; an important feature of this study is the ability to determine T_c self-consistently from the penetration-depth measurements themselves, thereby helping to eliminate a major uncertainty in the analysis. The resulting values of $(122.8 \pm 0.3 \text{ K})$ were in good agreement with the 10% onset T_c obtained by dc measurement of M(T) in 10 G after cooling in zero field. The midpoints of the magnetic transition were 121.2 K for $H \parallel c$ and 116.5 K for $H \parallel a - b$. These lower values and apparent transition width arise mostly because $\lambda(T)$ is comparable to the size of the small superconducting particles.

From the Ginzburg-Landau relation $\lambda^{-2} \propto (T_c - T)$, similar values for T_c are obtained for both orientations of field, despite a difference in upper critical field slopes dH_{c2}/dT that is expected to be substantial. We attribute the similarity of observed T_c 's to the fact that the experiment determines values for the essentially *low field* quantity $\lambda(T)$ that diverges at the (low field) T_c . Very near T_c , application of large fields clearly violates an assumption of the theory that $H \ll H_{c2}$. Experimental results for the penetration depth with $H \parallel c$ are shown as symbols in Fig. 1(a), where the dimensionless quantity $[\lambda_a(0)/\lambda_a(t)]^2$ is plotted versus reduced temperature $t = T/T_c$. The data extend from t = 1, near which the linear dependence on T is evident, down to $t \sim 0.7$, below which temperature the magnetization became increasingly hysteretic and minority superconducting phases created additional contributions to M. The value at t = 0, $\lambda_a(0)$, was treated as a fitting parameter when comparing the data with various functional forms for $[\lambda(0)/\lambda(t)]^2$. These include the results from weak coupling, BCS theory in the clean and dirty limits, ⁶ strong-coupling calculations in the clean and dirty limits by Rammer, ⁷ and the empirical, two-fluid model with a $(1-t^4)$ dependence.

For the case with $H \parallel c$, the weak-coupling, clean-limit theory provided an excellent description of the experimental results. The quality of fit was clearly superior to that obtained for the other theoretical curves, as seen quantitatively in Table I, which lists values for $\lambda(0)$ and the goodness of fit R^2 obtained from linear regressions to five different temperature dependencies. The relative quality of the various descriptions is shown graphically in Fig. 2, a plot of the deviations $\delta([\lambda(0)/\lambda(t)]^2) = (ex$ perimental - theoretical values), as a function of t. Again



FIG. 1. The normalized penetration depth vs reduced temperature $t = (T/T_c)$. Full curves are theoretical and model relations as shown in the legend; symbols are experimental points normalized by $\lambda(0)$ values for best fits to the optimum temperature dependence given by BCS clean limit theory; see text and Table I. (a) For penetration depth λ_{a} , with $H \parallel c$. (b) For effective penetration depth $\lambda_{eff} = (\lambda_a \lambda_c)^{1/2}$, with $H \parallel a - b$. Statistical uncertainties: The symbol sizes correspond to three standard deviations vertically and one horizontally, respectively.





FIG. 2. Deviations in the quantity $[\lambda(0)/\lambda(t)]^2$ between experimental values and best fits to various theoretical functions, vs reduced temperature t, for Tl₂Ca₂Ba₂Cu₃O_{10+s} with H||c. Typical error bars show uncertainties of one standard deviation (STD) in t and three STD's in $\delta[\lambda(0)/\lambda(t)]^2$.

it is quite clear that the BCS clean-limit expression gives the best description of the data. For this temperature dependence, we have $\lambda_a(0) = 173$ nm.

For H||a-b, the results were very similar, except that the slopes $dM/d[\ln(H)]$ were smaller by a factor of ~2.8, indicating that λ_c exceeds λ_a by this factor. Figure 1(b) shows λ_{eff} vs t for this case. The two weak-coupling theoretical functions gave essentially equal values for R^2 , as seen in Table I. (With H||a-b, the smaller signals produce somewhat more experimental scatter.) However, weak-coupling theory still provides a decidedly superior description. Similar conclusions were reached in the recent Ames study^{1(b)} on Y-Ba-Cu-O and Bi₂Sr₂CaCu₂O₈. For t=0, we obtain in the weak-coupling clean limit the value $\lambda_{eff} = (\lambda_a \lambda_c)^{1/2} = 292$ nm, so that $\lambda_c = 480$ nm. Thus, a major result of this work is that the Tl-based compound follows weak-coupling theory, and this supports the earlier argument of Little⁸ that the coupling is weak in the HTSC's.

In a Ginzburg-Landau treatment incorporating anisotropy of the effective mass,⁵ the components of the normalized mass tensor m_i are related by $\gamma = (m_c/m_a)^{1/2}$ $= (\lambda_c/\lambda_a)$. In this work, we obtain a value of 2.8 for γ . This is somewhat lower but comparable to that ($\gamma = 5.1$), obtained by Farrell *et al.*⁹ in torque magnetometry studies on aligned Tl 2:2:2:3. It is considerably smaller than the value of 70 deduced from resistive measurements¹⁰ of H_{c2} for Tl 2:1:2:2 near T_c . Similar anisotropy ($\gamma = 55$) was reported recently from torque studies^{11(a)} on single-crystal Bi₂Sr₂CaCu₂O₈, while H_{c1} data^{11(b)} give $\gamma \approx 3$ for this compound. The origin of these differences is not known.¹² We can rule out errors due to any angular misorientation $(<2-3^{\circ})$ of the sample, as their effect is much too small; e.g., with a hypothetical value of 64 for γ , then a misorientation of 2° would lead to an observed value $\gamma_{obs} = 26$. Angular averaging from the $\pm 1/2^{\circ}$ angular spread of crystallites would reduce γ from 64 to 56, assuming a Gaussian distribution. To reduce γ to 2.8 would require an angular spread of $\pm 25^\circ$, which is 50 times greater than the rocking curve width. Experimentally, the presence of unaligned Tl 2:2:2:3 particles would lead to a lower observed anisotropy in our measurements which sense the volume averaged bulk magnetization. While xray diffraction scans of θ -2 θ gave no clear evidence for significant amounts of unoriented superconductor, this possibility leads us to regard the value $\gamma = 2.8$ as a lower bound. We note, however, to reduce a hypothetical γ from 64 to 2.8 would require that fully one-half of the sample volume be unaligned. Another influence on the various experiments may be flux lattice melting.¹³ Here this phenomenon facilitated the study by widening the region of magnetic reversibility down to $t \approx 0.7$. In sum, we cannot account for the range of γ values from various investigations and believe that further experimental and theoretical progress will be required to resolve this issue.

There are relatively few other reports for the penetration depth in Tl-based superconductors. Lichti et al.¹⁴ have made muon spin relaxation measurements on polycrystalline Tl 2:2:2:3 ($T_c = 105$ K) and deduce an average λ_{avg} of 185 nm, comparable with our result for $\lambda_a(0)$ and about 25% smaller than the average $(\lambda_a \lambda_a \lambda_c)^{1/3} = 240$ nm. However, Lichti et al. found their results for Tl- and Bi-based materials to be best described by strong-coupling theory in the clean limit; for $R_1Ba_2Cu_3O_7$ materials (where R denotes rare earth), their results were described by strong-coupling theory in the dirty limit, implying an extremely short electronic mean free path. In NMR investigations on Tl 2:2:2:3, Lee *et al.*¹⁵ found $\lambda_{avg} = 176$ nm, but noted interpretive difficulties associated with anisotropies in random powders for both NMR and μ sr experiments, and this may affect adversely the functional form of $\lambda(t)$ deduced for the polycrystalline samples.

In summary, the current measurements of penetration depth in $Tl_2Ca_2Ba_2Cu_3O_{10+\delta}$ are best described by weak-coupling theory in the clean limit.

TABLE I. Goodness of fit to theoretical functions for $[\lambda(0)/\lambda(t)]^2$ and optimum $\lambda(0)$ for each case.

Theory or model	Coefficient of determination R^2		Optimum $\lambda_{eff}(0)^a$	
	Expt.: $H \parallel c$	Expt.: $H \parallel a - b$	H∥c	H a-b
Strong coupling, dirty limit ^b	0.9566	0.9755	220	384
Two-fluid (empirical) model	0.9741	0.9867	208	362
Strong coupling, clean limit ^b	0.9855	0.9920	198	340
BCS weak coupling, dirty limit ^c	0.9970	0.9972	189	322
BCS weak coupling, clean limit ^c	0.9994	0.9966	173	292

^cReference 6.

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