Measurements of the absolute value of the penetration depth in high-T_c superconductors using a low-T_c superconductive coating

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A method is presented to measure the absolute value of the London penetration depth, $\lambda(T=0)$, from the frequency shift of a resonator. The technique involves coating a high-T_c superconductor with film of low-T_c material of known thickness and penetration depth. The method is applied to obtain $\lambda(YBa_2Cu_3O_{7-\delta}) \approx 1460 \pm 150$ Å, $\lambda(Bi_2Sr_2CaCu_2O_{8+\delta}) \approx 2690 \pm 150$ Å and $\lambda(Pr_{1.85}Ce_{0.15}CuO_{4-\delta}) \approx 2790 \pm 150$ Å. $\lambda(YBa_2Cu_3O_{7-\delta})$ from this method is very close to that obtained by several other techniques. For both $Bi_2Sr_2CaCu_2O_{8+\delta}$ and $Pr_{1.85}Ce_{0.15}CuO_{4-\delta}$ the values exceed those obtained by other methods. © *2000 American Institute of Physics*. [S0003-6951(00)00148-0]

The London penetration depth, $\lambda(T)$, is a basic parameter of the superconducting state. For the sub-mm sized crystals characteristic of high-T_c work, measurements of the resonant frequency shift of a microwave cavity[1,2] or tunnel diode oscillator[3–5] currently provide a resolution of order 0.2 Å for changes in the penetration depth, $\Delta\lambda \equiv \lambda(T) - \lambda(T_{\min})$. However, the usual resonator approach does not provide the absolute magnitude of λ. This shortcoming arises from various experimental uncertainties and is not an inherent limitation of the technique. As we show in this paper, by plating samples with a thin superconducting film, it is possible to use a resonator to obtain both $\Delta\lambda(T)$ and $\lambda(T_{\min})$ simultaneously. The zero-temperature penetration depth, $\lambda(0)$ can be obtained by extrapolation to $T=0$. Together, $\lambda(0)$ and $\Delta\lambda(T)$ determine the normalized superfluid density $\rho_s(T) = [1 + \Delta\lambda(T)/\lambda(0)]^{-2}$, the quantity directly related to the electromagnetic response of the superconductor. No new physical model is required to obtain $\lambda(T)$ from the data, unlike the case with techniques such as μSR or reversible magnetization. Our method has been tested on single crystals of $YBa_2Cu_3O_{7-\delta}$ (YBCO), $Bi_2Sr_2CaCu_2O_{8+\delta}$ (BSCCO), and $Pr_{1.85}Ce_{0.15}CuO_{4-\delta}$ (PCCO) and compared with values of $\lambda(T)$ obtained from other techniques.

It is worthwhile first discussing why resonator methods cannot normally determine the absolute penetration depth. We focus on a lumped LC resonator but the ideas also hold for a distributed device such as a microwave cavity. In the absence of a superconducting sample the empty resonant frequency is $2\pi f_0 = 1/\sqrt{LC}$. When a superconducting sample is inserted into the resonator, flux is expelled from the sample volume, thus decreasing the stored magnetic field energy $W_m = LI^2/2$ and, therefore, inductance L. For a platelet sample of thickness $2d$ in the z direction and mean planar dimensions $2w \times 2w$ in the $x-y$ plane, this leads to an increase of the frequency by an amount $\Delta f \equiv f(T) - f(0)$ given by[9]

$$\frac{\Delta f}{f_0} = \frac{V_s}{2V_0(1-N)}\left[1 - \frac{\lambda}{R}\tanh\frac{R}{\lambda}\right]. \tag{1}$$

Here V_s is the sample volume, V_0 is the effective volume of the resonator, N is the demagnetization factor, and the field is applied along the z direction. R is the effective sample dimension which depends upon field orientation relative to the sample and sample geometry.[4] For the standard ''Meissner'' configuration in which the field is applied parallel to the surface of an infinite slab, $N=0$ and $R=d$. For the geometry used here, in which the alternating current (ac) field is normal to the face of a platelet, $R \approx w/5$.[4]

The measurement process is sketched in Fig. 1. The superconducting sample is inserted into the resonator, resulting in a change in frequency Δf_0. For typical high-T_c superconductor (HTSC) samples $\Delta f_0 \sim 10^4$ Hz. In principle, if R were known precisely then one could use Eq. (1) together with the measured Δf_0 to determine $\lambda(0)$. Unfortunately, there are several sources of error. First, the accuracy with which Δf_0 can be determined is limited by repeatability. Extracting and re-inserting the sample *in situ* typically leads to an error of $\delta f_0 \sim 10$ Hz out of a total $\Delta f \sim 10^4$. This ''static'' uncertainty is shown by the gray band in Fig. 1. According to Eq.

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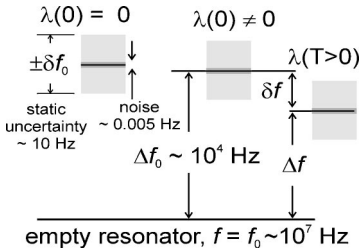

FIG. 1. Frequency shifts encountered in resonance measurements. The static uncertainty δf_0 is usually greater than the frequency shift due to finite $\lambda(0)$. Relative frequency shift δf is independent of δf_0 and permits accurate measurements of $\Delta\lambda$.

(1), the difference between a perfectly diamagnetic sample and one with finite λ is only $f_0(1-\lambda/R)\approx 30$ Hz for a typical YBCO crystal where $R\geqslant 50\,\mu$m and $\lambda(0)=0.15\,\mu$m which is comparable to the static uncertainty, $\delta f_0\approx 10$ Hz. Furthermore, inserting and extracting the sample gives the value of Δf_0 already reduced by finite $\lambda(0)$. Other methods for estimating Δf_0 using a ball[3] or a replica of the sample made from low-T_c material[2] lead to ''static'' uncertainty between different runs as well as errors related to differences between the real sample and the substitute. Realistic samples are irregular and have dimensions which are uncertain to much more than $\lambda(T)$. They may also have demagnetizing effects which are taken account of by Eq. (1) but which itself involves approximations for R. It is therefore not feasible to measure $\lambda(T)$ using resonator frequency shifts in the straightforward manner outlined. Despite this limitation on accuracy, the precision with which changes in λ can be measured is much higher. This is illustrated in Fig. 1 by the change δf upon warming the sample from low to intermediate temperature. In this case the sample stays fixed so the temperature-independent static uncertainty is irrelevant. Only the oscillator noise matters, which is typically 2000 times smaller than the static uncertainty. It is therefore imperative to adopt a technique that keeps the sample fixed.

Our method is illustrated in Fig. 2. The sample under study is plated with a conventional low T_c superconductor, in this case an Al film. The film thickness t should be larger than λ(Al) but much smaller than the normal state skin depth of Al ($\approx 3\,\mu$m at the operating frequency of 10 MHz).

Using $\lambda=H^{-1}\int_0^\infty B(x)dx$ we find that for $T<T_c$(Al) the magnetic field penetrates to $\lambda[T<T_c(\mathrm{Al})]=\lambda(\mathrm{Al})+\exp[-t/\lambda(\mathrm{Al})][\lambda(\mathrm{HTSC})-\lambda(\mathrm{Al})]$. Above T_c(Al) the penetration depth is $\lambda[T>T_c(\mathrm{Al})]=t+\lambda(\mathrm{HTSC})$. Converting the frequency change $\Delta f=f[T>T_c(\mathrm{Al})]-f[T<T_c(\mathrm{Al})]$ to a change in the effective penetration depth, $\Delta\lambda$, using Eq. (1), we obtain

$$\lambda(\mathrm{HTSC})=\lambda(\mathrm{Al})+\frac{\Delta\lambda-t}{1-\exp[-t/\lambda(\mathrm{Al})]}. \qquad (2)$$

The errors in this method arise from uncertainties in the film thickness t, the resonator calibration constant and λ(Al). Literature values for the effective penetration depth of aluminum films, $\lambda(\mathrm{Al})\approx\lambda_L(\mathrm{Al})\sqrt{\xi(0)/\ell}$ range from 400 to 600 Å.[6] Here, the BCS coherence length $\xi(0)\approx 16\,000$ Å, the mean free path, $\ell\approx 1000$ Å, and the London penetration depth $\lambda_L(\mathrm{Al})\approx 160$ Å. We choose the commonly accepted value, $\lambda(\mathrm{Al})\approx 500\pm 100$ Å. The Al film was 800 ± 50 Å thick. Uncertainty in the calibration constant gives an additional error of about 10 Å giving a total error of approximately ± 150 Å. It can be further reduced by choosing different coating materials, which will give additional independent reference points and by varying the thickness of the coating layer. A related method, employing a SQUID magnetometer and a thin Cd layer, was used to determine λ in heavy fermion compounds.[7] However, the sensitivity was not sufficient for high-T_c samples.

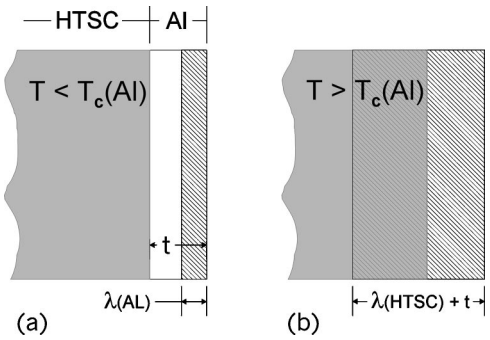

FIG. 2. Field penetration above and below transition temperature of Al film. (a) $T<T_c$(Al): magnetic field penetrates only λ(Al). (b) $T>T_c$(Al): magnetic field penetrates the Al layer and HTSC superconductor λ(HTSC).

λ was measured in three different high temperature superconductors, YBCO, BSCCO, and PCCO. YBCO crystals were grown in yttria stabilized zirconia crucibles as described[8] and annealed to achieve maximal $T_c\approx 93$ K.[9] BSCCO samples where grown using a floating zone process and had $T_c\approx 89.5$ K.[10] Single crystals of PCCO were grown using directional solidification technique and annealed in argon to achieve $T_c\approx 22.5$ K.[1] The aluminum coating was applied with a magnetron sputtering system with 5 cm rotated Al target (99.999% purity). Sputtering was conducted in an argon atmosphere and was homogeneous over 20 cm^2. The Al layer thickness, t, was calibrated using a Inficon XTC 2 with 6 MHz gold quartz crystal and later directly measured using scanning electron microscopy edge imaging of a broken sample.

The measurement technique utilized a 10 MHz tunnel diode oscillator whose specifications have been reported previously.[4] Samples were mounted on a movable sapphire stage whose temperature could be varied from 0.35 to 100 K. The low base temperature was crucial in order to obtain the full frequency shift due to the diamagnetism of the Al film.

We first present experiments in YBCO single crystals. Previous work has shown that $\lambda(0)$ is anisotropic with $\lambda_a(0)=1600$ Å and $\lambda_b(0)=800$ Å.[2] Since supercurrents for the $H\|c$ orientation flow along both a and b axes, we obtain an average of λ_a and λ_b. Two crystals, plated in separate evaporation runs, were measured. The first is shown in Fig. 3. Note that the contribution due to the Al film is subtracted using Eq. (2) and therefore the $\Delta\lambda(T)$ curve begins at negative values. Thus, at $T=T_c$(Al), $\lambda(\mathrm{HTSC})[T_c(\mathrm{Al})]$ is obtained. Linear extrapolation to $T=0$ yields λ(YBCO) ≈ 1390 Å. A second YBCO sample, shown in Fig. 4, gave $\lambda(0)\approx 1460$ Å. These values should be compared to those obtained from μSR, 1405 ± 92 Å,[11] 1550 Å,[12] 1586–1699 Å;[13] polarized neutron reflectometry, 1400 ± 100 Å;[14] magnetic susceptibility of grain-aligned powder, 1400 Å[15] and infrared spectroscopy, 1440 Å.[16] Since the Al plating in its normal state is transparent to the 10 MHz field, it is possible

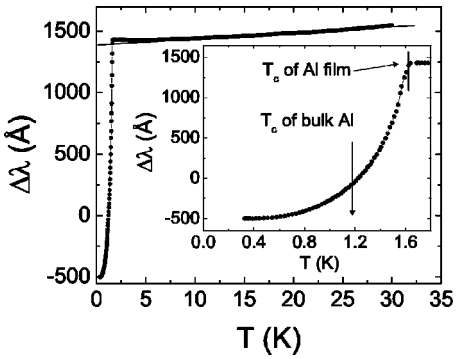

FIG. 3. Penetration depth in single crystal YBCO calculated from Eq. (2). Inset: Temperature range in which Al becomes normal.

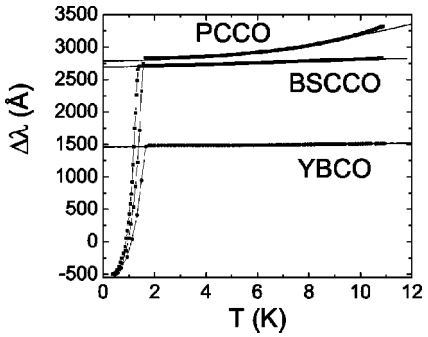

FIG. 4. Penetration depth in (from bottom to top) YBCO, BSCCO, and PCCO single crystals. For YBCO and BSCCO data were extrapolated using $\lambda(T) \propto T$, whereas for PCCO $\lambda(T) \propto T^2$ dependence was used.

to monitor $d\lambda(T)/dT$ of YBCO for $T > T_c(\mathrm{Al})$. This is an important check since the Al coating might change the surface properties of the cuprate enough to alter its penetration depth. For the samples in Figs. 3 and 4, $d\lambda/dT \approx 4.95$ Å/K, and 5.10 Å/K, respectively, which are somewhat larger than the reported value of 4.3 Å/K (Refs. 2 and 3) obtained in the $H \| ab$ configuration, but within the error of our measurements on these samples in the $H \| ab$ configuration without plating.

The inset to Fig. 3 shows the penetration depth variation while warming the sample above $T_c(\mathrm{Al})$. The measured $T_c(\mathrm{Al}) \approx 1.69$ K is significantly larger than the bulk value $T_c(\mathrm{Al}) \approx 1.18$ K. This increase could be due to a proximity effect,[17] but could also be caused by disorder and altered chemical composition of the aluminum film.

Figure 4 summarizes the measurements for all three cuprates. For BSCCO-2212 crystal we obtained $\lambda(\mathrm{BSCCO}) \approx 2690$ Å, which can be compared to data from: reversible magnetization, $\lambda \approx 2100$ Å;[18] μSR, $\lambda \approx 1800$ Å;[19] and lower critical field measurements, $\lambda \approx 2700$ Å.[20] We obtained a linear variation of $\lambda(T)$ with a $d\lambda/dT \approx 11.7$ Å/K, compared to $d\lambda/dT \approx 10.5$ Å/K in previous microwave and μSR.[21,22] To within our precision, it appears that the Al plating has no effect on the underlying cuprate superconductor.

The uppermost curve in Fig. 4 shows the results for the electron-doped cuprate superconductor, PCCO. This material has been cited as an example of a cuprate *s*-wave superconductor. Recent measurements with higher resolution and lower temperatures have shown that $\lambda(T) \propto T^2$, indicative of a nodal order parameter in the presence of impurity scattering.[5] This is shown in the figure with $d\lambda/dT \approx 4.38$ Å/K^2. We find $\lambda(0) \approx 2790$ Å. Measurements of $H_{c1} = \Phi_0/[4\pi\lambda(0)^2]\ln\kappa$ have reported $\lambda(0) \approx 1000$ Å.[23] Our approach is arguably more reliable since no vortices are involved, thus eliminating possible surface barrier effects.

In conclusion, we have developed a new technique to measure $\lambda(T)$ in high-T_c superconductors. The estimated accuracy of ± 150 Å is limited by uncertainties in the thickness and penetration depth of the Al coating.

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