Measurements of the absolute value of the penetration depth in high-$T_c$ superconductors using a tunnel diode resonator

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A method is presented to measure the absolute value of the London penetration depth, $\lambda$, from the frequency shift of a resonator. The technique involves coating a high-$T_c$ superconductor (HTSC) with film of low-$T_c$ material of known thickness and penetration depth. The method is applied to measure London penetration depth in YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO) Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (BSCCO) and Pr$_{1.85}$Ce$_{0.15}$Cu$_4$O$_{7-\delta}$ (PCCO). For YBCO and BSCCO, the values of $\lambda(0)$ are in agreement with the literature values. For PCCO $\lambda \approx 2790$ Å, reported for the first time.

The London penetration depth, $\lambda(T)$, is a quantity of fundamental importance. Its temperature, field, and doping dependencies are directly related to the density of quasiparticle energy states, knowledge of which is crucial for testing models of pairing symmetry and mechanisms of superconductivity. $\lambda(T)$ is also a key parameter in determining the response and collective properties of the Abrikosov vortex lattice. For single crystals, measurements of the resonant frequency shift of a microwave cavity or tunnel diode oscillator can provide the highest resolution for changes of the penetration depth, $\Delta \lambda \equiv \lambda(T) - \lambda(T_{min})$, with respect to temperature. For the sub-mm sized crystals typically studied in high-$T_c$ work, resolution of better than 0.2 Å can be achieved. However, the usual resonator approach has the disadvantage that it does not provide the absolute magnitude of $\lambda$. This shortcoming arises from various experimental uncertainties and is not an inherent limitation of the resonator technique. As we show in this paper, by suitably plating superconducting crystals it is possible to exploit the extremely high sensitivity of the resonator to changes in frequency and thus obtain an absolute measurement of $\lambda(T)$.

The method described here permits a simultaneous measurement of $\lambda(T_{min})$ and $\Delta \lambda(T)$ on the same sample. 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. This is a distinct advantage over the situation in which these two quantities are obtained by different groups using different samples and techniques. In addition, no new physical model is required to obtain $\lambda(T)$ from the data, unlike the case with techniques such as $\mu$SR or reversible magnetization. Our method has been tested on single crystals of YBCO, BSCCO and PCCO and compared with values of $\lambda(T)$ obtained from other techniques.

It is first worth 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 $f_0 = 1/\sqrt{LC}$. When a superconducting sample is inserted into the resonator, the inductance $L$ decreases due to a decrease of the magnetic field energy $W_m = LI^2/2c^2$ as a result of Meissner expulsion. 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 $\Delta f = f(T) - f(0)$ given by,

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

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. For the standard "Meissner" configurations 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 AC field is normal to the face of a platelet, $R \approx w/2d$.

The measurement process is sketched in Fig. [1]. The superconducting sample is inserted into the resonator, resulting in a change in frequency $\Delta f_0$. For typical HTSC samples $\Delta f_0 10^4$ Hz. In principle, if $R$ were known precisely then one could use Eq. (1) together with the measured $\Delta f_0$ to determine $\lambda(0)$. Unfortunately, there are several sources of error. First, the accuracy with which $\Delta f_0$ can be determined is limited by repeatability. Extracting and inserting the sample in situ typically leads...
to an error of $\delta f_0 \approx 10$ Hz out of a total $\Delta f \approx 10^4$. This “static” uncertainty is shown by the gray band in Fig. 1. According to Eq. (1), the difference between the perfect diamagnet and sample with finite $\lambda$ is only $f_0(1 - \lambda/R) \approx 30$ Hz for a typical YBCO crystal where $R \geq 50$ $\mu$m and $\lambda(0) = 0.15$ $\mu$m which is quite comparable to the static uncertainty, $\delta f_0 \approx 10$ Hz. Furthermore, extracting and inserting the sample gives the value of $\Delta f_0$ already reduced by finite $\lambda(0)$. Other methods of estimation of $\Delta f_0$ such as measuring a ball made of a conventional superconductor or replicating an HTSC sample using low-$T_c$ materials result even in greater uncertainty, because in addition to an inevitable “static” uncertainty related to differences between real sample and the substitute. Furthermore, realistic samples are irregular and so have dimensions which are uncertain to much more than $\lambda(T)$. They may also have large demagnetizing effects. Finally Eq. (1) itself involves approximations for $R$ that adds further error. 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 $\lambda$ can be measured is much higher. This is illustrated in Fig. 1 by the change $\Delta f$ 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 $\lambda(Al)$ 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 $\lambda(T < T_c(Al)) = \lambda(Al) + \exp(-t/\lambda(Al)) [\lambda(HTSC) - \lambda(Al)]$. Above $T_c(Al)$ the penetration depth is $\lambda(T > T_c(Al)) = t + \lambda(HTSC)$. Converting the frequency change $\Delta f = f(T > T_c(Al)) - f(T < T_c(Al))$ to a change in the effective penetration depth, $\Delta \lambda$, using Eq. (2), we obtain:

$$\Delta \lambda = \lambda(Al) + \frac{\Delta f}{1 - \exp(-t/\lambda(Al))}$$

The errors in this method arise from uncertainties in the film thickness $t$, the resonator calibration constant and $\lambda(Al)$. Literature values for the effective penetration depth of aluminum films, $\lambda(Al) \approx \lambda_L(Al)\sqrt{\xi(0)/\ell}$ range from 400 to 600 $\AA$. Here, the BCS coherence length $\xi(0) \approx 16000$ $\AA$, the mean free path, $\ell \approx 1000$ $\AA$ and the London penetration depth $\lambda_L(Al) \approx 160$ $\AA$. We choose the commonly accepted value, $\lambda(Al) \approx 500 \pm 100$ $\AA$. The Al film was 800 ± 50 $\AA$ thick. Uncertainty in the calibration constant gives an additional error of about 10 $\AA$ giving a total error of approximately ±150 $\AA$. 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. Although it is clearly desirable to improve the accuracy, an error of 150 $\AA$ still results in only a 1% deviation of $\rho_s$ over 20 K range for YBCO. A somewhat similar measurement technique was used earlier to determine $\lambda$ in heavy fermion compounds. In that experiment, a flux trapped or screened by a thin Cd layer was used, but owing to the much reduced sensitivity of SQUID magnetization measurements, it is not suitable for high-$T_c$ materials.$\lambda(HTSC)$ was measured in three different superconductors: YBa$_2$Cu$_3$O$_{7-\delta}$ (YBCO), Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$...
FIG. 3: Penetration depth in single crystal YBCO calculated from Eq. (2). Inset: Temperature range in which Al becomes normal.

(BSCCO), and the electron-doped Pr$_{1.85}$Ce$_{0.15}$CuO$_{4-\delta}$ (PCCO). YBCO crystals were grown in yttria stabilized zirconia crucibles as described, and annealed to achieve maximal $T_c \approx 93$ K. BSCCO samples where grown using a floating zone process and had $T_c \approx 89.5$ K. Single crystals of PCCO were grown using directional solidification technique and annealed in argon to achieve $T_c \approx 22.5$ K. 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 an Inficon XTC 2 with 6 MHz gold quartz crystal and later directly measured using SEM edge imaging of a broken sample.

The measurement technique utilized a 10 MHz tunnel diode oscillator whose specifications have been reported previously. Samples were mounted on a moveable 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 magnetic field 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. Since supercurrents for the $H$||c orientation flow along both $a$ and $b$ axes, we obtain an average of $\lambda_a$ and $\lambda_b$. Two crystals, plated in separate evaporation runs, were measured. The first is shown in Fig. 3. Note that contribution due to Al film is subtracted using Eq. (3) and therefore the $\Delta\lambda(T)$ curve begins at negative values. Thus, at $T = T_c(Al)$, $\lambda(HTSC)(T_c(Al))$ is obtained. Linear extrapolation to $T = 0$ yields $\lambda(YBCO) \approx 1460$ Å. This value should be compared to values obtained from $\mu$SR, $1405 \pm 92$ Å, magnetic neutron reflectometry, $1400 \pm 100$ Å, and infrared spectroscopy, 1440 Å.

Since $T_c(Al)$ is quite low and the Al plating in its normal state is transparent to 10 MHz RF, it is possible to monitor $d\lambda(T)/dT$ of YBCO for $T > T_c(Al)$. This is an important check on the method since it is conceivable that the Al coating might change the surface properties of the cuprate enough to alter its penetration depth. The slope $d\lambda/dT \approx 5.1$ Å/K. This slope is somewhat larger than the value of 4 Å/K reported previously, but is in agreement with our recent measurements conducted on unplated samples in the $H$||ab configuration. The second YBCO sample, shown in Fig. 4, gave $\lambda(0) \approx 1460$ Å and $d\lambda/dT \approx 5.10$ Å/K, both within the estimated error with the first sample.

The Inset to Fig. 3 shows details of the penetration depth variation warming the sample above $T_c(Al)$. The measured $T_c(Al) \approx 1.69$ K is significantly larger than the bulk value $T_c(Al) \approx 1$ K. This increase could be due to proximity effects, but could also be caused by disorder and altered chemical composition of aluminum film.

Figure 4 summarizes the measurements for all three cuprates. For BSCCO-2212 crystal we obtained $\lambda(BSCCO) \approx 2690$ Å, which can be compared to data from: reversible magnetization, $\lambda \approx 2100$ Å; $\mu$SR, $\lambda \approx 1800$ Å; and lower critical field measurements, $\lambda \approx 2700$ Å. It is clear that a fairly large disagreement still exists over the value of $\lambda(0)$ in this material. 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 $\mu$SR measurements. To within our current precision, it appears that the Al plating has no effect on the electrodynamics of 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 \(\lambda(T)\) varies quadratically with temperature, indicative of a nodal order parameter in the presence of impurity scattering. This is shown in the figure with \(d\lambda/dT \approx 4.38 \text{ Å}/K^2\). We find \(\lambda(0) \approx 2790 \text{ Å}\). The only published value was obtained from measurements of \(H_{c1} \approx \Phi_0/[4\pi\lambda(0)^2] \ln \kappa\) which gave \(\lambda(0) \approx 1000 \text{ Å}\). It is difficult to reliably determine \(H_{c1}\) in thin crystals owing to demagnetization and pinning surface barrier effects. Our approach is arguably more reliable since no DC fields or vortices are involved and we have obtained close agreement with other methods in YBCO and BSCCO.

In conclusion, we have developed a new technique to measure \(\lambda(T)\) in high-\(T_c\) superconductors. We obtained \(\lambda(0) = 1390\) and 1460 Å for two YBCO crystals, \(\lambda(0) = 2890 \text{ Å}\) for BSCCO, and \(\lambda(0) = 2790 \text{ Å}\) for PCCO. All values are determined with the \(\pm 150 \text{ Å}\) accuracy. The plating has no discernable effect on the underlying temperature dependence of \(\lambda(T)\). The accuracy of the method is limited principally by uncertainties in the Al film thickness.

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