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Power-law dependence of the ab-plane penetration depth in $Nd_{1.85}Ce_{0.15}CuO_{4-y}$

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It is shown that measurements of the low-temperature ab-plane penetration depth, $\lambda_{ab}(T)$, of the electron-doped superconductor $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ may be affected by the paramagnetism arising from the Nd^{3+} ions. Making a correction based on the static susceptibility measured for single crystals alters the limiting temperature dependence of $\lambda_{ab}(T)-\lambda_{ab}(0)$ from an exponential behavior, typical of s-wave pairing, to a T^1 or T^2 behavior associated with d-wave pairing. It is argued that this correction is probably still appropriate at microwave frequencies. Thus the widely held belief that the electron-doped cuprate superconductors are s wave is still not established and further experiments are proposed. [S0163-1829(96)52130-2]

The limiting low-temperature (T) dependence of the superconducting penetration depth (λ) of high-T_c oxides is an important initial indicator of nodes in the energy gap and the possibility of unconventional (d-wave) pairing. Although there was much early evidence[1–3] for power-law behavior in $\lambda(T)-\lambda(0)$ of yttrium barium copper oxide (YBCO) rather than the exponential decay expected for an s-wave superconductor with a constant gap, the subject remained controversial until the microwave studies of Hardy and colleagues on single crystals of YBCO.[4] That work and subsequent investigations of untwinned YBCO crystals[5] and $Bi_2Sr_2CaCu_2O_8$ crystals[6,7] have all shown the limiting T^1 behavior in $\lambda_{ab}(T)-\lambda_{ab}(0)$ that is expected for a superconducting order parameter with $d_{x^2-y^2}$ symmetry and line nodes in the gap function $\Delta(\mathbf{k})$. Recent measurements of $\lambda_{ab}(T)$ of magnetically aligned $HgBa_2Ca_2Cu_3O_{8+\delta}$ powders[8] also fit the d-wave picture.

More stringent tests[9,10] using tunneling techniques which are sensitive to the phase of the order parameter have generally favored d-wave pairing. However, it is widely believed that the electron-doped cuprate superconductor $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ (NCCO), with a maximum critical temperature of ca. 24 K, is s wave. The main experimental evidence for this is the studies of $\lambda_{ab}(T)$ for thin films and single crystals at microwave frequencies.[11–13] Although there is no reason to doubt the quality of the measurements or the materials studied, in this article it is shown that when corrections for the paramagnetic Nd^{3+} ions are made to the published microwave data[11,12] then $\lambda_{ab}(T)-\lambda_{ab}(0)$ approximately follows a T^1 law with about the magnitude expected for d-wave pairing. However, comparison with results for Zn-doped YBCO (Ref. 14) shows that a T^2 law arising from a d-wave superconductor with a small amount of pair breaking may be a more appropriate description. The crucial question of the frequency dependence of the paramagnetism is briefly discussed and further experiments are suggested.

There is considerable evidence in the literature that Nd is trivalent in NCCO with a $4f^3$ ionic configuration. Neutron scattering work shows that the lowest crystal field level is a magnetic doublet separated by approximately 140 K from the next highest quartet[15] and so the free ion magnetic moment of $3.6\mu_B$ is reduced because of crystal field splitting. The magnetic susceptibility (χ) of a single crystal of NCCO has been measured from 4 to 300 K.[16] There is considerable anisotropy in χ because of crystal field effects. χ is larger for H perpendicular to the c axis (the geometry used in the microwave work [11,12]). From 6 to at least 80 K it can be fitted by a Curie-Weiss law $\chi_\perp=\chi_0+C/(T+\Theta)$. The fitting parameters are $C=71\times10^{-2}$ emu K/mole Nd, corresponding to an effective moment of $2.4\mu_B$ per Nd^{3+} ion, $\chi_0=0.5\ 10^{-2}$ emu/mole Nd and the antiferromagnetic interaction term $\Theta=4.0$ K. Taking the density of NCCO to be 7.35 gm/cm^3, and using cgs units, the above value of $\chi_\perp$ corresponds to a magnetic permeability $\mu=1+4\pi\chi=1+0.295/(T+4.0)$.

This has two effects on measurements of λ_{ab}. First the usual solution of London's equation shows that λ_{ab} is *reduced* by a factor $\sqrt{\mu}$, in a similar way to the skin depth of a magnetic conductor. Namely, in the usual notation, the standard equations $\mathbf{J}=ne\mathbf{v}$, $m\,d\mathbf{v}/dt=e\mathbf{E}$, $\mathbf{B}=\mu\mathbf{H}$, c curl$\mathbf{E}=-\partial B/\partial t$ and curl$\mathbf{H}=4\pi\mathbf{J}/c$, lead to $\lambda^2(T)=\lambda_{\rm sf}^2(T)/\mu$ where $\lambda_{\rm sf}$ is the intrinsic penetration depth that is related to the superfluid density.

Second, the extra permeability of that part of the crystal or epitaxial film penetrated by the microwave field gives a T-dependent frequency shift because the resonant frequency of the cavity is determined by the volume integral of $\mu H^2/8\pi$. For H parallel to the ab plane and in the limit where $2\lambda_{ab}\ll d$ (the film thickness) this effect will simply *increase* the apparent value of λ by a factor μ. Thus in the large d limit, the raw data for $\lambda_{ab}(T)$ need to be divided by a factor $\sqrt{\mu}$ in order to show up the true T dependence of the superfluid density. For smaller values of d the correction has to be made in two stages using the formula $M/M_{\rm max}=\{1-x\tanh(1/x)\}$, where $x=2\lambda/d$, M is the diamagnetic moment of the film and $M_{\rm max}$ the maximum possible value corresponding to $\lambda\ll d$. First $1-M/M_{\rm max}$ (as determined from the measured frequency shift) has to be divided by a factor μ and then the value of λ calculated from the new value of $M/M_{\rm max}$ has to be multiplied by $\sqrt{\mu}$ to obtain the quantity of interest $\lambda_{\rm sf}$.

The effect of these corrections is shown in Fig. 1. In fact the appropriate value of the molecular field parameter Θ is probably 1.2 K, as indicated by neutron scattering experiments[15] in the superconducting state and the actual ordering temperature (1.2 K) seen in the specific heat[17] instead of 4 K obtained above by fitting $\chi_\perp$ measured in a field of 1

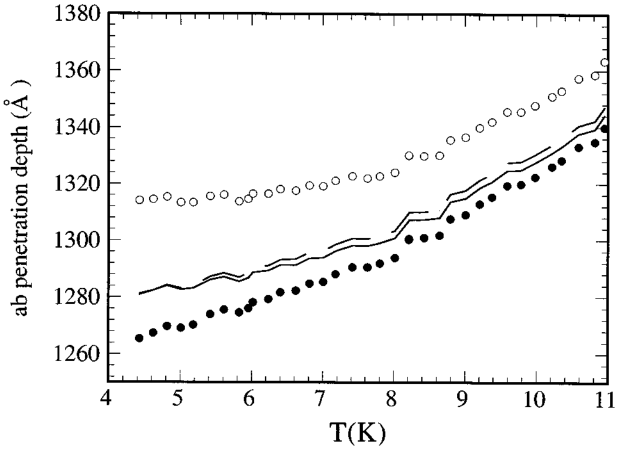

FIG. 1. Open symbols, values of the superconducting penetration depth $\lambda_{ab}(T)$ for a 5000 Å ab-plane film of $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ as derived from the measured frequency shift in Fig. 1 of Ref. 11 without making any correction for paramagnetism. As discussed in Refs. 11 and 12 these data give an excellent fit to the Bardeen-Cooper-Schrieffer theory for an s-wave superconductor. Solid circles, ''best'' values corrected for the paramagnetic effects of the Nd^{3+} ions, taking $\Theta=1.2$ K and using the full formula for $M/M_{\max}$ (see text). Solid line, same correction but with $\Theta=4.0$ K. Dashed line, correction made using the thick film formula and $\Theta=1.2$ K, i.e., simply dividing the measured values by $\sqrt{\mu}$.

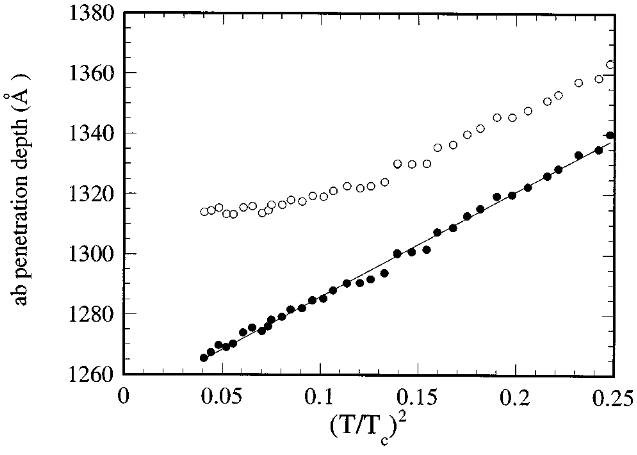

FIG. 2. Plot of $\lambda_{ab}(T)$ for $Nd_{1.85}Ce_{0.15}CuO_{4-y}$ versus $(T/T_c)^2$ where $T_c=22$ K. Measured and corrected values are shown by open and filled circles, respectively.

Tesla to a Curie-Weiss law. In other words, the Nd-Nd interactions are probably factor of 3 weaker in the superconducting state than in the normal state but the effective moment is unchanged. As shown in Fig. 1 this alters the effect of the paramagnetic correction slightly below 8 K.

The experimental data in Fig. 1 are taken from Ref. 11. The effect of the finite film thickness (5000 Å) has also been included in the best estimate of $\lambda_{ab}(T)$ shown by the solid circles. When the paramagnetic correction is included there is a marked linear term with an initial slope of 8.6 Å/K. For comparison, untwinned YBCO has a linear term of 4.7 Å/K for the a direction (where the Cu-O chains are not important) and a $\lambda(0)$ value of 1600 Å.[5] If the normalized initial slope $\lambda(0)^{-1}d\lambda/dT$ for YBCO is simply scaled by the T_c ratio (22 K vs 92 K) then an even larger linear term, 15 Å/K, would be expected for NCCO. One possible reason for this discrepancy is that the ratio of the superconducting gap to T_c is a factor of 2 larger for NCCO than for YBCO, however this seems to be unlikely. As shown in Fig. 2, the corrected data also give a good fit to a T^2 law over a wider range of T/T_c. It is known that the addition of only 0.3% Zn to YBCO alters the behavior of $\lambda_{ab}(T)$ from a T to a T^2 law (Ref. 14) and there could easily be similar effects in the present NCCO sample which modify the behavior of $\lambda_{ab}(T)$ but only give (say) a 1% reduction in T_c.

A crucial assumption in the preceding analysis is that the susceptibility of the NCCO crystals at 9 GHz is essentially the same as the static value. For noninteracting Nd^{3+} spins this will only be true if the spin lattice relaxation rate $1/T_1$, is much larger than 9 GHz.[18] Estimates of $1/T_1$ derived from electron spin resonance (ESR) data for insulating Nd compounds[19] are several orders of magnitude smaller than 9 GHz. Estimates based on ESR studies of Gd-doped $YBa_2Cu_3O_7$, where the relaxation is dominated by the Korringa conduction electron mechanism,[20] also seem to be too small (by a factor of 100 at 10 K). So at first sight the above correction is not appropriate. However, the neutron results[15] show that antiferromagnetic Nd-Nd interactions split the ground state Kramers doublet of the Nd^{3+} ions in NCCO by 0.3 meV (i.e., 70 GHz) for temperatures from 2 to 10 K, well above the ordering temperature of 1.2 K.[17] Therefore in low applied fields the imaginary part of the susceptibility $\chi''(\omega)$ (i.e., the energy absorption) will be peaked at around 70 GHz and it will probably be small at 9 GHz. It then follows from the Kramers-Kronig relations[18] that the real part of the uniform susceptibility will not be frequency dependent up to 9 GHz and the correction to $\lambda_{ab}(T)$ discussed here *will* be valid. This is quite a subtle point and needs to be verified by further experiments; for example, those suggested below. It also implies that the paramagnetic effects discussed here cannot be checked simply by making microwave measurements of $\chi(T)$ in the normal state above T_c.

One simple test would be to measure $\lambda_{ab}(T)$ for NCCO crystals in the other geometry where the microwave field is along the c axis as was done recently for $Bi_2Sr_2CaCu_2O_8$,[6,7] because in this case the static susceptibility from the Nd^{3+} ions[16] is approximately a factor of 3 smaller. Because of their smaller static susceptibilities[16] the corresponding electron doped Pr and Sm compounds would also be suitable candidates for microwave studies.

In conclusion more microwave experiments or tunneling studies are needed to probe the superconducting properties of the electron-doped high-T_c oxides. There is still a strong possibility that the measured values of $\lambda_{ab}(T)$ for NCCO are *not* consistent with a standard s-wave picture and that the electron-doped compounds are also d-wave superconductors perhaps with some pair breaking.

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