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London penetration depth in $(BEDT\text{-}TTF)_2$ superconductors

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Abstract. Measurements of the London penetration depth, $\lambda(T)$, are reported for crystals of superconducting κ-$(ET)_2$ $Cu[N(CN)_2]Br$ (T_C = 11.6 K) and β''-$(ET)_2$-$SF_5CH_2CF_2SO_3$ (T_C = 5.2 K) at temperatures down to 0.35 K. For κ-$(ET)_2$ $Cu[N(CN)_2]Br$ the superfluid density shows the power law behavior characteristic of nodal quasiparticles with impurity scattering. The rate of change of superfluid density with temperature is higher than predicted by a weak-coupling d-wave model and higher than observed in the copper oxides. The in-plane penetration depth consistently shows a $T^{3/2}$ dependence while the interplane penetration depth varies as $T^{1.22}$. Measurements in β''-$(ET)_2$-$SF_5CH_2CF_2SO_3$ also show power law behavior consistent with nodal quasiparticles but with greater impurity scattering than in κ-$(ET)_2$ $Cu[N(CN)_2]Br$.

Key words. penetration depth - order parameter - organic superconductor - superfluid density.

1. INTRODUCTION

The symmetry of the order parameter is crucial to understanding superconductivity. The identification of an unconventional symmetry can rule out certain mechanisms for pairing. This issue has been central to high T_C superconductors where d-wave pairing is on a firm basis in the hole-doped materials and strongly suggested in the electron-doped materials. Organic superconductors have many similarities to copper oxides and it has been suggested that unconventional pairing may also hold [1]. Data from NMR [2,3], thermal conductivity [4], tunneling [5], heat capacity [6] and penetration depth [7,8] have suggested a d-wave state in the κ-$(ET)_2X$ superconductors where X = $Cu(NCS)_2$ and $Cu[N(CN)_2]Br$. In this paper we show data for both κ-$(ET)_2Cu[N(CN)_2]Br$ and β''-$(ET)_2$-$SF_5CH_2CF_2SO_3$ crystals, both of which exhibit a power law dependence of the superfluid density indicative of a d-wave pairing state with impurity scattering. Many of the results shown here were reported earlier [7,9].

1.1 Results

Penetration depth, λ(T), measurements were performed with a 12 MHz tunnel diode LC resonator used in several previous measurements [7]. Changes in the oscillator frequency were proportional to changes in the penetration depth, $G\Delta f = \Delta\lambda(T) = \lambda(T) - \lambda(T_{\min})$ through a constant G whose determination is described elsewhere [7,9]. The ac magnetic field was applied perpendicular to the conducting planes. This geometry generates in-plane screening currents and is sensitive to the in-plane penetration depth, λ_{P}(T). Several single crystals of κ and β" materials were studied. The growth procedures have all been described previously[10].

A superconducting order parameter with line nodes in momentum space exhibits a linearly increasing density of quasiparticle states. These nodal quasiparticles give rise to a normalized superfluid density that is linear in temperature,

$$\rho_s(T) = \left[\lambda_{\mathrm{P}}(0)/\lambda_{\mathrm{P}}(T)\right]^2 = \left[1+\Delta\lambda_{\mathrm{P}}/\lambda_{\mathrm{P}}(0)\right]^{-2} = 1-\alpha T/T_C \qquad (1)$$

for $T/T_c < 0.3$. For a gap function of the form $\Delta_{\max}\left(k_x^2 - k_y^2\right)$ one has $\alpha = 0.65$ with $\Delta_{\max} = 2.14 k_B T_c$. Even for a state obeying eq.(1) the penetration depth will vary as $\Delta\lambda_{\mathrm{P}}(T) \sim T + T^2 + \ldots$ Since T^2 terms can also arise from impurity scattering, it is important determine $\rho_s(T)$ in order to distinguish physically distinct sources of T^2 behavior. Resonator perturbation methods do not directly yield $\lambda_{\mathrm{P}}(0)$. To determine ρ_s it is therefore necessary to take $\lambda_{\mathrm{P}}(0)$ from some other measurement such as μSR [11].

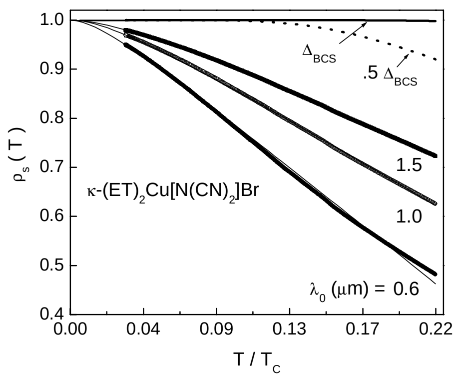

Fig. 1. Superfluid density of κ-$(ET)_2$-Cu$[N(CN)_2]$Br using three different values of λ(0) = 0.6, 1.0 , 1.5 μm. Fits are to eq.(1). Also shown: ρ_S for isotropic s-wave order parameter using Δ(BCS) = 1.76 $k_B T_C$ and Δ(BCS)/2 .

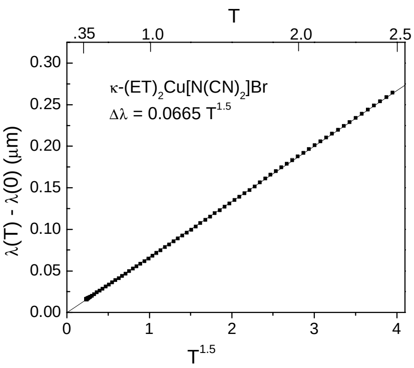

Fig. 2. Δλ(T) verus T $^{3/2}$ for κ-$(ET)_2$-Cu$[N(CN)_2]$Br .

Fig. 1 shows our measurement of ρ_s in κ-$(ET)_2$-Cu$[N(CN)_2]$Br. ρ_s plots were generated using several values of $\lambda_{\mathrm{P}}(0)$ and were fit to the form, $\rho_s(T) = 1-\alpha T^2/\left(T+T^*\right)T_C$, characteristic of a d-wave state with unitary limit impurity scattering[12]. This model has been widely used in the cuprates although a microscopic justification for unitary limit scattering is still lacking. Assuming $\lambda_{\mathrm{P}}(0)$ = 0.6 μm (a lower limit from μSR) we find α = 2.8 and $T^*/T_c = 0.05$. By comparison, α(YBCO) = 0.5 - 0.6 and $T^*/T_c < 0.01$ while α(BSCCO) = 0.7 and $T^*/T_c \approx 0.05$. A choice of $\lambda_{\mathrm{P}}(0)$ = 1.5 μm gives α = 1.6. κ-$(ET)_2$Cu$[N(CN)_2]$Br is therefore a plausible candidate for a d-wave superconductor but with a large value of α. Since $\alpha \sim \left(d\Delta/d\theta_{node}\right)^{-1}$, our results imply a gap function which increases very slowly as one moves away from the nodal direction [13]. A similar value of α was also found in recent susceptibility measurements by Pinteric et. al.[8]. Those authors suggested that the admixture of an s-wave component (d + s) would shift the k-space position of the gap nodes and give a larger value of α. To obtain α = 2.8 would require an s-wave component approximately 0.7 times the maximum d-wave gap, for which there is no obvious justification.

Fig.1 shows that an order parameter of the form s or d + is would require an extremely small s-wave component. We estimate that any finite gap on the Fermi surface would need to be less than 3% of the BCS value $\Delta_{BCS} = 1.76\, k_B T_C$. Fig. 1 shows ρ_s for both a BCS energy gap and for Δ = ½ Δ_{BCS}. In either case, ρ_s is essentially temperature independent below T/T_C = 0.1 . Our measurements for κ-$(ET)_2Cu(NCS)_2$ gave very similar results.

For all samples studied, the penetration depth obeyed a $\Delta\lambda_{\rm P} : T^{3/2}$ power law with remarkable consistency, as shown in Fig. 2. Although there is no physically motivated order parameter that would give $T^{3/2}$, it is a power law that arises from Bose excitations obeying a quadratic power law (e.g. magnons). It has also been shown that a d-wave pair fluctuation model would exhibit a $T^{3/2}$ term in the superfluid density [14]. However, we find that to fit ρ_s to a $T^{3/2}$ power law requires a very large value of $\lambda_{\rm P}(0)$ (= 6 µm) and no linear T term. Other researchers have reported a $T^{3/2}$ law for $CeCoI_5$ which was attributed to a combination of strong-coupling superconductivity and nonlocality [15]. Nonlocality is unlikely to be applicable to the organic superconductors, however. A $T^{3/2}$ power law can of course be closely approximated by a combination of T and T^2 , but the converse is not true. Neither YBCO nor BSCCO generally fit to a $T^{3/2}$ power law. We speculate that this power law may indicate a scattering rate peculiar to these materials that is highly consistent owing to the quality of the samples.

By aligning the ac magnetic field parallel to the conducting planes we measured the *interplane* penetration depth, $\lambda_\perp$. For a d-wave superconductor with coherent interlayer transport one has $1-\rho_\perp(T) : T^\beta$ with β = 1. Incoherent coupling between planes can lead to β = 2 which is widely observed in the cuprates [16]. Higher powers can also arise depending upon details of the interlayer transport and impurity scattering [17,18]. We consistently observed β=1.2-1.3 in κ-$(ET)_2$ $Cu[N(CN)_2]Br$ again giving strong support to the d-wave picture. The large difference in screening between the actual sample and a perfect diamagnet of the same dimensions also allowed us to extract $\lambda_\perp(0) \approx 85 \mu m$ directly from the change in frequency of the resonator as the sample was extracted from the coil, *in situ*. We estimate an error of 15% in this measurement [7].

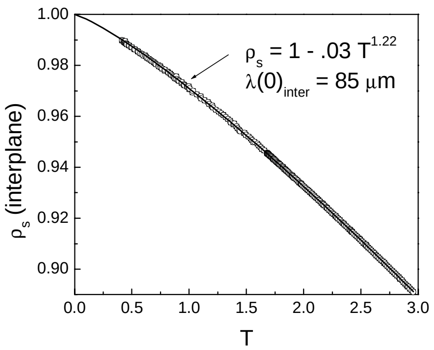

Fig. 3. Interplane superfluid density versus temperature in κ-$(ET)_2$-$Cu[N(CN)_2]Br$.

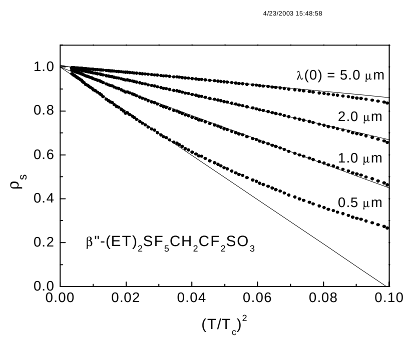

Fig 4. In-plane superfluid density in all organic β"-$(ET)_2SF_5CH_2CF_2SO_3$ versus $(T/T_C)^2$, for several different values of λ(0).

More recently we reported penetration depth measurements in single crystals of the superconductor β"-$(ET)_2SF_5CH_2CF_2SO_3$ (Tc = 5.2 K). This material has both organic cations and anions. It has been suggested that pairing occurs through charge fluctuations and the order parameter should have d_{xy} symmetry [19]. Our measurements average over in-plane directions and would not distinguish d_{xy} from $d_{x^2-y^2}$ symmetry. We found that $\Delta\lambda_{\rm P} : T^3$ over a large temperature range, but consistently crossing over to a lower power law exponent at the lowest temperatures obtainable. In Fig. 4 we plot the in-plane superfluid density for a range of plausible $\lambda_P(0)$ values. For all curves, the asymptotic dependence is $\rho_s = 1 - cT^2$. A T^2 power law can arise from a spin triplet, p-wave state but we know of no measurement that suggests triplet pairing in this material [20]. Assuming singlet pairing, a T^2 power law would imply a d-wave state with $T^*/T_c > 0.5$ and thus very strong scattering. It could also imply a very inhomogeneous sample with a spread of transition temperatures. However, the latter case is unlikely in that the values we observe for $d\lambda/dT$ are extremely consistent, within 10% from sample to sample.

Since the penetration depth is determined by the quasiparticle energy, power law behavior can indicate gap nodes but not the phase of the order parameter directly. It is possible to indirectly detect the phase through penetration depth measurements. For sample surfaces perpendicular to nodal directions, quasiparticles in a d-wave superconductor suffer a sign change in the pair potential upon specular reflection. Barash et. al. have shown that this leads to a singular density of surface Andreev bound states at zero quasiparticle energy and a corresponding paramagnetic term $\lambda \sim 1/T$ at very low temperatures [21]. The competition between the T and T^2 terms from nodal quasiparticles and the 1/T term from Andreev bound states gives a minimum in λ_P below $T_{\min} = g(\theta)\sqrt{\xi/\lambda}$ where $g \leq 1$ is a measure of amount of crystalline surface normal to the nodal direction and ξ is the coherence length[21]. Recently, Carrington et. al.[22] verified these predictions in YBCO crystals where $T_{\min} \sim 10K$. A similar 1/T term was observed earlier in irradiated films of YBCO where surfaces favorable for bound states were purposely created [23]. For κ-$(ET)_2Cu[N(CN)_2]Br$, this theory predicts $T_{\min} \approx 0.1\,K$. The observation of $\lambda \sim 1/T$ would provide a definitive test of d-wave pairing in the organic superconductors.

In conclusion, we find that the κ-$(ET)_2Cu[N(CN)_2]Br$ data is consistent with a d-wave order parameter in the presence of impurity scattering but with a larger value of $d\rho_S/dT$ than predicted in a weak-coupling model. All samples consistently showed a near perfect fit to $\Delta\lambda \sim T^{1.5}$. β''-$(ET)_2$ superconductors exhibited $\rho_s = 1 - cT^2$ behavior, again consistent with nodal quasiparticles but with much greater impurity scattering.

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