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New Results for Superconductivity in κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ When an Applied Magnetic Field is Aligned Parallel to the Conducting Planes

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Using a Tunnel Diode Oscillator technique, we have measured the effect of a parallel magnetic field on the in-plane rf penetration depths in organic [α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ and κ-(BEDT-TTF)$_2$Cu(NCS)$_2$] and heavy fermion (CeCoIn$_5$) superconductors. We show that in this particular geometry, the effects due to vortex activity are minimized. The penetration depth is then governed by the density of superconducting carriers. It is shown in many experiments including rf penetration depth measurement that α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ and CeCoIn$_5$ have s-wave and d-wave pairing symmetries, respectively. The pairing symmetry of κ-(BEDT-TTF)$_2$-Cu(NCS)$_2$, however, is still an unsolved matter, showing inconsistent results. In this paper, the penetration depth of κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ is shown to be more similar to α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ than to CeCoIn$_5$, suggesting the pairing is nodeless.

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κ-(BEDT-TTF)$_2$Cu(NCS)$_2$ (ET-Cu) is an anisotropic quasi-2D organic superconductor with T_c=9.6 K. The pairing symmetry of ET-Cu as determined by, specific heat,[1] thermal conductivity,[2] and NMR[3] have consistently supported an unconventional d-wave pairing symmetry, but the penetration depth measurements have given inconsistent results, which support both conventional[4,5] and unconventional [6–9] pairings. For a better understanding, we accurately measured the penetration depth of ET-Cu with the magnetic field applied parallel to the conducting planes. Applying the magnetic field parallel to the conducting planes suppresses the vortex contribution to the penetration depth due to a lock-in effect,[10] so we can accurately measure λ_L, the penetration depth due to the density of the superconducting carriers(n_s). For the purpose of comparison, the same measurement was also conducted in two other superconductors, α-(BEDT-TTF)$_2$NH$_4$Hg(SCN)$_4$ (ET-NH$_4$) and CeCoIn$_5$, which have been known as having conventional[11] and d-wave pairing symmetries,[12] respectively. By comparing all three samples, it is coucluded that ET-Cu exhibits behavior more similar to a material with conventional pairing symmetry (no nodes).

Single crystals of ET-Cu ($0.21\times0.175\times0.04$ mm^3), ET-NH$_4$ ($1.83\times1.69\times1$ mm^3), and CeCoIn$_5$ (1mm dia., 0.1 mm thick) were used for the experiment. The samples were placed in the coils of an rf resonator with the conducting planes perpendicular to the axis of the coil. The data were taken using top loading ^{3}He-^{4}He dilution refrigerators and different magnetic field facilities at NHMFL: the 18 T superconducting magnet for ET-NH$_4$ and CeCoIn$_5$, and the 33 T resistive magnet for ET-Cu, because of its high upper critical field. The principle of operation of the TDO technique is described in Ref. 13, and its design for use in high magnetic fields can be found in Ref. 14. At a fixed temperature within the superconducting state, the change in frequency of the TDO is linear with the change of penetration depth $\Delta\lambda(H)$=-$\Gamma\Delta f(H)$. Obtaining absolute values of the penetration requires a careful calculation of the constant Γ, which depends on the coil and sample geometries and the demagnetization factor. We did not calibrate the system for obtaining absolute values, but we measured and subtracted the influence of the background by running the system both with and without the sample.

The penetration depth in superconductors is determined by two major contributions. One is the London penetration depth, $\lambda_L=\sqrt{\frac{m^*}{\mu_0 n_s e^2}}$, determined by the density of the superconducting carriers(n_s). The other term is due to the formation and motion of the vortices, λ_v. Theoretical studies have shown [15,16] that the contribution of these two factors results in an *effective* penetration $\lambda_{eff}^2=\lambda_L^2+\lambda_v^2$. In our experiment we noticed that if the sample was aligned so that the magnetic field was within 0.5° of parallel to the conducting planes, the λ_v term disappeared. The disappearance of the λ_v term is due to a lock-in effect, which is why only λ_L was measured.

In this parallel geometry, penetration depth measurements of ET-NH$_4$ were conducted at temperatures ranging from 50 to 300 mK and the results support the claim of a conventional order parameter (no nodes).[11] According to the two fluid model, the penetration depth for a s-wave superconductor

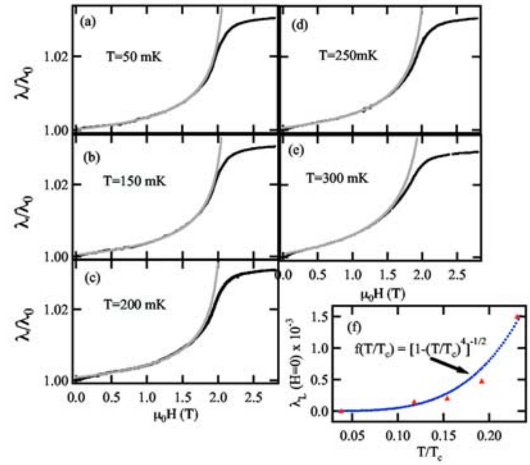

Fig. 1. (a)-(e) λ measurements in ET-NH_4 with fits to BCS theory. (Gray lines: the fit by Eq. 1). (f) Zero-field penetration verses T, fit by Eq. 2.

as a function of magnetic field is,

$$\lambda_L(H) = \frac{\lambda_L(H=0)}{\left[1 - \frac{H}{H_{c2}}\right]^{1/2}}. \tag{1}$$

As is seen in Fig. 1(a)-(e), Eq. 1 fits the data well, proving that the penetration depth we measured is determined by the density of superconducting carriers(n_s). ¿From the fits with Eq. 1, the zero-field penetration depths, λ_L(H=0), were found for each temperature, and in Fig. 1(f) these values were fitted to an equation, which determines the temperature dependence of penetration depth again according to the two fluid model,

$$\lambda(T) = \frac{\lambda(T=0)}{[1-(T/T_c)^4]^{1/2}}. \tag{2}$$

The fact that Eq. 2 fits the data well in Fig. 1(f) supports the s-wave pairing symmetry of ET-NH_4.

The same measurements were performed on $CeCoIn_5$, a $d_{x^2-y^2}$ superconductor. Below T=250 mK, there is a clear linear dependence of the penetration depth on the magnetic field, which supports the d-wave pairing symmetry. The details have been discussed elsewhere.[17]

In Fig. 2, the penetration depths of the three different samples, ET-NH_4, $CeCoIn_5$, and ET-Cu, are plotted using normalized scales. The penetration depths of ET-NH_4 and ET-Cu are consistently small as compared to

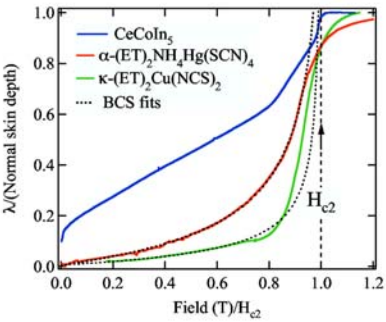

Fig. 2. Penetration depths of ET-NH_4, ET-Cu, and $CeCoIn_5$ on a normalized scale. Black-dots: the fit by Eq. 1.

$CeCoIn_5$. Also, the ET-Cu data has a slight curvature even though it does not fit Eq.1 perfectly. This penetration depth data suggests that ET-Cu has a nodeless character which is more consistent with s-wave pairing symmetry.

In conclusion, we have reported on the electronic pairing symmetry of ET-Cu. Using ET-NH_4 and $CeCoIn_5$ our as a baseline, our penetration depth measurements show that ET-Cu has a nodeless order parameter. We acknowledge the DOE #ER46214 and the NSF #DMR-SGER-0331272.

REFERENCES

1. Y. Nakazawa and K. Kanoda, *Physica (Amsterdam)* **282C**, 1897 (1997).
2. S. Belin, K. Behnia, and A. Deluzet, *Phys. Rev. Lett.* **81**, 4728 (1998).
3. S. Lefebvre et al., *Phys. Rev. Lett.* **85**, 5420 (2000).
4. D. R. Harshman et al., *Phys. Rev. Lett.* **64**, 1293 (1990).
5. M. Lang et al., *Phys. Rev. Lett.* **69**, 1443 (1992).
6. A. Carrington at al., *Phys. Rev. Lett.* **83**, 4172 (1999).
7. K. Kanoda et al., *Phys. Rev. Lett.* **65**, 1271 (1990).
8. L. P. Le et al., *Phys. Rev. Lett.* **68**, 1923 (1992).
9. D. Achikir et al., *Phys. Rev. B* **47**, 11595 (1993).
10. P. A. Mansky et al., *Phys. Rev. B* **50**, 15929 (1994).
11. Y. Nakazawa et al., *Phys. Rev. B* **52**, 12890 (1995).
12. R. Movshovich, et al., *Phys. Rev. Lett.* **86**, 5152 (2001).
13. C. T. Van Degrift, *Rev. Sci. Instrum.* **46**, 599 (1975).
14. T. Coffey et al., *Rev. Sci. Instrum.* **71**, 4600 (2000).
15. M. W. Coffey and J. R. Clem, *Phys. Rev. Lett* **67**, 386 (1991).
16. E. H. Brandt, *Phys. Rev. Lett.* **67**, 2219 (1991).
17. C. Martin et al., *Phys. Rev. B* **71**, 020503 (2005).