Vortex Penetration Depth of Organic Superconductors: Evidence for Vortex Lattice Melting

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Abstract

We observe a crossover field H^* in the temperature and magnetic field dependence of the rf vortex penetration depth in $\kappa\text{-(ET)}_2\text{Cu[N(CN)}_2\text{]Br}$ for $\vec{H}_{dc} \parallel \hat{b}$ -axis. We find that H^* can be described quantitatively by the 3D Lindemann melting theory; thus, it corresponds to the melting of the vortex lattice in $\kappa\text{-(ET)}_2\text{Cu[N(CN)}_2\text{]Br}$ and lies very close to the irreversibility line. In the vortex-liquid state, we argue that the saturation of the vortex penetration depth in a magnetic field results from the finite size of the sample. Our results do not have the scaling form predicted by the Coffey-Clem model¹ in contrast to previous findings.

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Many superconductors of current interest are highly anisotropic and in the extreme type-II limit. Both cuprate high-temperature superconductors (HTSC) and many organic superconductors are in this class. In the HTSC case, theories and experiments have confirmed the existence of a line in the field-temperature phase diagram along which the vortex lattice of the mixed phase melts.²⁻⁴ Thermal fluctuations enhanced by the high T_c , large anisotropy, and quenched disorder,⁵ combine to destroy the long range order of the vortex lattice well below the nominal upper critical field line, $H_{c2}(T)$, leaving a vortex-liquid state in which the vortices move independently and the dc electrical resistance is finite. Even in Nb with low $T_c \approx 9.2$ K and cubic symmetry, several recent experiments⁶ have indicated that vortex lattice melting occurs below $H_{c2}(T)$.

In this letter, we consider whether vortex lattice melting occurs in an organic superconductor. Organic superconductors based on the ET electron-donor molecules, where ET stands for bis(ethylenedithio)-tetrathiafulvalene, have transition temperatures roughly an order of magnitude lower than the HTSC, yet they are even more strongly in the type-II limit. We report here measurements of the vortex penetration depth of the layered organic compound, $\kappa\text{-(ET)}_2\text{Cu[N(CN)}_2\text{]Br}$, that support the melting theory. At a fixed temperature, we find that the field-dependent change of the penetration depth, $\Delta\lambda(H)$, for $\vec{H}_{dc} \perp$ ac-planes increases to a shoulder at low magnetic fields, and then rises rapidly to saturation at fields (< 1 T) much below $H_{c2}(T)$. We define a crossover field $H^*(T)$ located near the shoulder and find that it does not follow $H^* \propto (1-t)^{3/2}$, as would be expected if H^* marks the crossover from a pinning dominated to a thermally activated flux-flow (TAFF) regime. We argue that the crossover at $H^*(T)$ and the saturation of $\Delta\lambda$ are strong indications of vortex lattice melting.

This new picture provides a natural explanation of the data that is more consistent with the body of recent experimental work on other layered superconductors, and is the first experimental evidence for vortex lattice melting in $\kappa\text{-(ET)}_2\text{Cu[N(CN)}_2\text{]Br}$. The Coffey-Clem (CC) theory, that treats the electrodynamics of 3D Abrikosov vortices in a periodic potential and includes the effect of thermal activation, has been employed previously to explain similar data, but will be shown to be inconsistent with our results and with earlier magnetization data⁷ on a similar organic compound.

Our measurements were performed on a single-crystal $\kappa\text{-(ET)}_2\text{Cu[N(CN)}_2\text{]Br}$ superconductor with dimensions $1.25 \times 1 \times 0.5 \text{ mm}^3$. Its transition temperature at ambient pressure is $\sim 12 \text{ K}$ and is the highest T_c of any organic superconductor reported to date. The initial discovery and crystal growth have been described elsewhere.⁸ The temperature and magnetic field dependence of the rf penetration depth were measured with the use of an ultra-stable tunnel-diode resonator⁹ operating at a frequency of $\sim 10 \text{ MHz}$. In this technique, we measure the shift in the oscillator frequency which is directly proportional to $\Delta\lambda(H,T)$. The resonator has a frequency stability of 0.02 parts per million which corresponds to a resolution of 0.2 Hz or $\sim 2 \text{ \AA}$. The rf field is always parallel to the conducting a-c planes and perpendicular to the dc magnetic field. In this case, the supercurrent induced by the rf field flows both in the plane and perpendicular to the plane so that $\lambda \propto (w\lambda_1 + t\lambda_\perp)$ where w and t are the width and the thickness of the sample respectively. We choose $\gamma \equiv \lambda_\perp/\lambda_1 \sim 29$ which is in the range of reported values $13 \leq \gamma \leq 65$ and $w/t \sim 0.5$; thus, we are measuring $\lambda_\perp$ with about 6.5% contribution from λ_1 .

Figure 1 shows $\Delta\lambda(H,T) \equiv \lambda(H,T) - \lambda(0,T)$ for $\vec{H}_{dc} \perp$ ac-planes at various temperatures below T_c . All the data were taken after zero field cooling to avoid trapped flux; however, no hysteresis was observed at any temperature upon increasing the field to 1 T and then returning the field to zero. The overall behavior of $\Delta\lambda$ is similar at all temperatures. The lower critical field, $H_{c1}^\perp$, in our sample is very small; thus, there is no signature of crossing H_{c1} in our data.

Using the Ginzburg-Landau expression, we estimate $\mu_0 H_{c1}^\perp \approx \frac{\Phi_0}{4\pi\lambda_\perp^2} \ln \kappa \approx 7 \times 10^{-7}$ T where

$\kappa \equiv \lambda/\xi \approx 400$ and $\lambda_\perp = 38 \mu\text{m}$ (Ref. 10); thus, we are always in the mixed state. In practice, it is very difficult to measure $H_{c1}^\perp$ because of the huge demagnetization effect.

In Fig. 2a, we plot $\Delta\lambda(H)$ and $\partial(\Delta\lambda)/\partial H$ vs. H at $T = 8$ K. We define H^* to be the position of the peak in $\partial(\Delta\lambda)/\partial H$ and $\Delta\lambda^*(T) \equiv \Delta\lambda(H^*,T)$. The resultant $H^*(T)$ is plotted vs. $1-t$, where $t \equiv T/T_c$, on a logarithmic scale in Fig. 2b. The dashed line corresponds to $H^* \sim (1-t)^{3/2}$ as expected for the TAFF model, and clearly, H^* does not follow the 3/2 power law of the TAFF model, especially close to T_c . We now consider whether H^* represents the melting of the vortex lattice. Experiments on HTSC find that the vortex lattice melting line $B^m(T)$ coincides with or lies very close to the so-called irreversibility line observed in dc-magnetization measurements. Small angle neutron scattering experiments³ on $\text{Bi}_{2.15}\text{Sr}_{1.95}\text{CaCu}_2\text{O}_{8+x}$ have provided direct observation of the melting of the vortex lattice; this melting transition is found to be first order.⁴ In Fig. 2b, the squares are the values of $H_{ir}(T)$ determined from dc-magnetization measurements by Lang, et al.¹¹ Within experimental error, the data for $H_{ir}(T)$ coincide with our results for $H^*(T)$. We therefore associate $H^*(T)$ in κ -

(ET)₂Cu[N(CN)₂]Br with the melting of the vortex lattice and give the following justifications.

1) For κ -(ET)₂Cu[N(CN)₂]Br, thermal fluctuations are important. The figure of merit for fluctuation effects is given by the Ginzburg number $G = A(T_c \lambda^2 / \gamma \xi)^2 = AG'$ where A is a constant, λ is the London penetration depth and γ is the anisotropy. For κ -(ET)₂Cu[N(CN)₂]Br, $\xi_{\perp}(0) \approx 5 \text{ \AA}$, $\xi_{\parallel}(0) \approx 37 \text{ \AA}$, $\xi \equiv (\xi_{\perp}(0) \xi_{\parallel}^2(0))^{1/3}$, $\lambda \equiv (\lambda_{\perp}(0) \lambda_{\parallel}^2(0))^{1/3} \approx 12 \text{ \mu m}$, and $\gamma = 29$, we find $G'_{\text{organic}} \sim 10^{-3}$, which lies between YBCO ($G'_{\text{YBCO}} \sim 10^{-5}$) and BSCCO ($G'_{\text{BSCCO}} \sim 0.1$); note that $G'_{\text{organic}} \sim 10^{10} G'_{\text{Nb}}$. However, the thermal energy ($\sim k_B T$) for κ -(ET)₂Cu[N(CN)₂]Br is about one order of magnitude lower than the high- T_c superconductors. One may ask if the effect of thermal fluctuations is strong enough to destroy the vortex lattice. The answer is affirmative because the vortex pinning strength in organic superconductors is six to nine orders of magnitude weaker than in YBCO.¹² 2) κ -(ET)₂Cu[N(CN)₂]Br is strongly anisotropic ($\gamma = 29$) due to its layered structure. 3) $\kappa \equiv \lambda/\xi \approx 400$ so that κ -(ET)₂Cu[N(CN)₂]Br is an extreme type-II superconductor. Thus, the flux lines are very elastic and can wander more easily. 4) Quenched disorder also helps to destroy the long range order of the vortex lattice. Disorder plays a very important role in the superconductivity of κ -(ET)₂Cu[N(CN)₂]Br. Kwok, et al.¹³ observed a reduction in T_c by about 10% from the disorder induced when they quenched their sample from room to helium temperature in less than five minutes.

The superconducting-layer spacing of κ -(ET)₂Cu[N(CN)₂]Br is $s = 15 \text{ \AA}$ and, using the BCS expression $\xi_{\perp}(T) = 0.74 \xi_{\perp}(0) / [1 - t]^{1/2}$ with $\xi_{\perp}(0) = 5 \text{ \AA}$, we expect a 3D-2D crossover, when $\xi_{\perp}(T_{2D}) = s$, which occurs at $T_{2D} \approx 0.93 T_c = 11.2 \text{ K}$. Thus, κ -(ET)₂Cu[N(CN)₂]Br

behaves as a quasi-2D superconductor over nearly the entire temperature range below T_c . However, this does not mean that the vortex structure in the mixed phase is two dimensional. When a magnetic field is applied perpendicular to the layers in layered superconductors, an individual vortex is composed of a stack of pancake vortices in the layers connected between layers by Josephson vortices.¹⁴ In weak fields, the interplanar interaction (tilt modulus) of the stack is larger than the intraplane interaction (shear modulus); hence, these pancake vortices align themselves to form a 3D flux line. In the high-field regime, the planar interaction between the pancake vortices dominates and the pancake vortices do not arrange in a straight line, but form a zig-zag pattern. Thus, there is a change of vortex structure from a 3D pancake flux line to 2D pancake vortices with little or no alignment. This dimensional change in vortex pinning has been observed in the BSCCO superconductor.³ Using the same criterion, we find that the crossover field is roughly $B_{2D} \approx \Phi_0 / (s\gamma)^2 = 1.1$ T for $\kappa\text{-(ET)}_2\text{Cu[N(CN)}_2\text{]Br}$, where we take $\gamma = 29$. Because our fields are less than 1 T, we are always in the 3D vortex limit, in which case, the melting transition can be described by the Lindemann criterion. If $H^*(T)$ is indeed the melting line, we should be able to fit it to the 3D melting function based on the Lindemann criterion,¹⁵

$$B_m^{3D} \approx B_0 \frac{1}{t^2 \lambda_{\perp}^4(t)} \quad (1)$$

where

$$B_0 = \frac{1}{[4(\sqrt{2} - 1) + 1] \pi \mu_0^2} \left(\frac{\Phi_0^5 c_L^4 \gamma^2}{(k_B T_c)^2} \right)$$

and c_L is the Lindemann number which is a constant ~ 0.2 . We take $\lambda_{\perp}^2(t) = \lambda_{\perp}^2(0) / \sqrt{1 - t^n}$ and allow n and γ to vary. The fit of Eq. 1 to $H^*(T)$, which is shown as the solid line in Fig. 2b, yields $n = 5.62$ and given the range of reported values for the in-plane penetration depth $0.67 \mu\text{m} \leq \lambda_{\parallel}(0) \leq 1.5 \mu\text{m}$, the anisotropy is $13 \leq \gamma \leq 65$. For the simple two-fluid model, $n = 4$ and Lee, et al.³ and R. Cubitt, et al.³ obtained $n = 3.3$ for BSCCO. The value of γ determined from microwave measurements is ≈ 25 .¹⁰ The anisotropy is very sensitive to the mosaic spread of the sample and other imperfections and as a result, can be a factor of 2 or more higher. Thus, the fitting parameters are in reasonable agreement with other experiments. Since $B_m^{3D} \propto 1/\gamma^2 \lambda_{\parallel}^4$, the large in-plane $\lambda_{\parallel}(0)$ and anisotropy of κ -(ET)₂Cu[N(CN)₂]Br push the melting line far below H_{c2} . This makes it plausible to associate $H^*(T)$ with flux lattice melting for the organic superconductors.

We can now address the saturation of $\Delta\lambda$ at fields $H^* < H \ll H_{c2}$. In the melting scenario, the flux lines form a lattice below $H^*(T)$ so that $\Delta\lambda$ represents the change in the effective penetration depth, λ_{eff} , arising from the bending of the vortices. Above $H^*(T)$, where the flux lines are in the liquid state, each flux line moves independently leading to a finite electrical resistance. The corresponding rf skin depth δ_{liq} will be much larger than λ_{eff} although still less than the normal-state skin depth. Once δ_{liq} is greater than or equal to half the sample thickness, $w/2$, the magnetic energy stored in the coil will not change with field. As a result, we observe little frequency shift with increasing magnetic field, and $\Delta\lambda$ appears to saturate. While our technique measures only the relative change of the frequency, we can roughly calibrate the coil to give a measure of the actual $\Delta\lambda$ values. The largest frequency shift

observed is $\Delta f_{\text{sat}} \cong 27$ kHz at $T = 5$ K. Assuming the rf field to be uniform over the surface of the sample, we equate the fractional change in frequency to the fractional change in the filling of the coil, $\Delta f_{\text{sat}}/f_0 \approx A_s \Delta \lambda_{\text{sat}}/V_{\text{coil}}$ giving $\Delta \lambda_{\text{sat}} \approx 0.06$ mm. The factor of two accounts for both sides of the sample. The total $\lambda_{\text{sat}}(H, 5\text{K}) = \lambda(5\text{ K}) + \Delta \lambda_{\text{sat}} = 0.10$ mm which is about a half of $w/2$ where $\lambda_{\perp}(T = 5\text{ K}) \approx 0.04$ mm (see Ref. 10). The conversion factor is then ≈ 2.2 $\mu\text{m}/\text{Hz}$.

If we now set $\lambda(H > H_{\text{sat}}, T) = w/2$ at all temperatures, we can extract the actual penetration depth from the expression $\lambda(H = 0, T) = \lambda_{\perp}(T = 0) + [w/2 - \Delta \lambda(H, T)]/2$. Using the above geometrical factor of 2.2 $\mu\text{m}/\text{Hz}$ and assuming $\lambda_{\perp}(T = 0) = 38$ μm (Ref. 10), we can determine the absolute value of $\lambda(H = 0, T)$; we plot this in Fig. 3. One can test the prediction of Eq. 1, that $\lambda_{\perp} = B_0^{1/4} \frac{1}{(t^2 B_m^{3D})^{1/4}}$ using H^* and $\lambda(H = 0, T)$ as extracted from the data.

The solid line is a guide to the eyes; clearly, $\lambda(H = 0, T)$ is consistent with Eq. 1, lending further support for vortex lattice melting in $\kappa\text{-(ET)}_2\text{Cu[N(CN)}_2\text{]Br}$. We can also estimate the anisotropy from the slope of Fig. 3, $B_0^{1/4}$; the value of γ obtained is ≈ 23 which is consistent with γ from our fit of H^* to Eq. 1.

Finally, we comment on the purported scaling behavior of the vortex penetration depth. Sridhar, et al.¹⁶ argued that $\Delta \lambda(T, H)/\Delta \lambda^*(T)$ is a universal function of H/H^* where $\Delta \lambda^*(T)$ is a temperature-dependent flux-flow skin depth, and they demonstrated that this form is contained within the CC theory. Within the CC theory, the scaling form $\Delta \lambda(T, H)/\Delta \lambda^*(T) = f(\omega \tau_o, H/H^*)$ is valid only for $T \ll T_c$, where the normal skin depth is much greater than the London penetration depth. In Fig. 4, we test this scaling relation at seven temperatures. While Sridhar, et al. found this scaling to hold for $\kappa\text{-(ET)}_2\text{Cu(NCS)}_2$ in the range $5\text{ K} < T \leq T_c$ ($\approx 8.9\text{ K}$), our

data do not collapse into a single curve over similar temperature range. Sridhar, et al. chose H^* and $\Delta\lambda^*$ near the saturation value, and because their data do not give a clear indication of the crossover fields, reasonable data collapsing is possible. Mansky, et al.⁷ also observed the shoulder features similar to ours in $\kappa\text{-(ET)}_2\text{Cu(NCS)}_2$ using ac-magnetization measurements. Perhaps the data of Sridhar, et al. do not show clear evidence of the melting line due to stronger pinning by defects. While the CC theory is able to explain the electrodynamics of 3D Abrikosov vortices, it does not include the possibility of flux-lattice melting; therefore, it is not applicable close to the melting line.

In summary, we have measured the temperature and magnetic field dependence of the effective penetration depth in the mixed state of $\kappa\text{-(ET)}_2\text{Cu[N(CN)}_2\text{]B}$ for $\vec{H}_{dc} \perp$ ac-planes and have shown that the crossover field H^* is quantitatively consistent with the 3D Lindemann melting theory. Our result is significant because it is the first experimental evidence for vortex lattice melting in the ET family of organic superconductors. We do not find the scaling behavior as reported previously¹⁶ and suggest that the Coffey-Clem theory must be modified to account for pancake vortices and the melting of the vortex lattice in layered superconductors.

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References

1. M. W. Coffey and J. R. Clem, Phys. Rev. Lett. **67**, 386 (1991).
2. D. J. Bishop, et al., Science **255**, 165 (1992).
3. R. Cubitt, et al., Nature **365**, 407 (1993).
 S. L. Lee, et al., Phys. Rev. Lett. **71**, 3862 (1993).
 K. Harada, et al., Phys. Rev. Lett. **71**, 3371 (1993).
4. E. Zeldov, et al., Nature **375**, 373 (1995).
5. D. S. Fisher, et al., Phys. Rev. B **43**, 130 (1991).
6. J. W. Lynn, et al., Phys. Rev. Lett. **72**, 3413 (1994).
 M. F. Schmidt, et al., Phys. Rev. Lett. **70**, 2162 (1993).
7. P. A. Mansky, et al., Phys. Rev. B **50**, 15929 (1994).
8. A. M. Kini, et al., Inorg. Chem. **29**, 2555 (1990).
9. C. Van DeGrift, Rev. Sci. Instrum. **46**, 599 (1975).
10. M. Dressel, et al., Phys. Rev. B **50**, 13603 (1994).
11. M. Lang, et al., Phys. Rev. B **49**, 15227 (1994).
12. D-H. Wu and S. Sridhar, Phys. Rev. Lett. **65**, 2074 (1990). They found that the pinning constants for YBCO are $\kappa_p^{\parallel} \approx 2 \times 10^5 \text{ N/m}^2$ and $\kappa_p^{\perp} \approx 2 \times 10^4 \text{ N/m}^2$. In Ref. 7, the pinning constants for organic superconductors are $\kappa_p^{\parallel} \approx 10^4 \text{ N/m}^2$ and $\kappa_p^{\perp} \approx 5 \times 10^{-2} \text{ N/m}^2$.
13. W. K. Kwok, et al., Phys. Rev. B **42**, 8686 (1990).
14. G. Blatter, et al, Rev. Mod. Phys. **66**, 1125 (1994).
15. A. Houghton, et al., Phys. Rev. B **40**, 6763 (1990).
16. S. Sridhar, et al., Phys. Rev. Lett. **68**, 2220 (1992).

Figure Captions

Fig. 1. A plot of $\Delta\lambda(H, T)$ vs. H of $\kappa\text{-(ET)}_2\text{Cu[N(CN)}_2\text{]Br}$ ($T_c \sim 12$ K) for $\vec{H}_{dc} \perp$ planes at various temperatures 4.2, 4.6, 5, 5.5, 6, 6.5, 7, 7.5, 8, 9, 9.5, 10, 10.5, 11, 11.5, 13 K.

Fig. 2. (a) Determination of $\mu_0 H^*(T)$ from the peak position of $d(\Delta\lambda)/dH$. (b) Open circle is a plot of $\mu_0 H^*$ at several temperatures below T_c . Open square is $B_{ir}(T)$ from Ref. 11. The dashed line is a fit of $\mu_0 H^* \propto (1-t)^{3/2}$ to the data (open circles). The solid line is a fit to the 3D melting function (Eq. 1 in text).

Fig. 3. A plot of $\Delta\lambda(T, H = 0)$ vs. $(t^2 \mu_0 H^*)^{-1/4}$, see text for detail.

Fig. 4. A plot of $\Delta\lambda(T, H)/\Delta\lambda^*(T)$ vs. H/H^* for several temperatures below T_c .

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Fig. 1

Fig. 2

Fig. 3

Fig. 4