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Universal temperature dependence of the London penetration depth in κ -(ET)₂X superconductors

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Abstract

High-precision radio-frequency magnetic susceptibility measurements were performed on single crystals of fully deuterated κ -(ET)₂Cu[N(CN)₂]Br, hereafter designated as κ -(D8 ET)₂Cu[N(CN)₂]Br. This material phase separates into superconducting and antiferromagnetic regions, the degree of which depends strongly upon the cooling rate. We show that the screening fraction rise varies logarithmically with the cooling rate over nearly five decades. The average size of superconducting regions is estimated to vary from 5 to 40 μ m, depending upon cooling rate, consistent with previous infrared microscopy measurements. In the region T less than or similar to $T_c/3$, the effective magnetic penetration depth exhibits power-law behavior $\lambda(T) - \lambda(0)$ similar to T^{-n} with $n = 1.6$, independent of the cooling rate. Changes in cooling rate and the consequent phase separation evidently do not introduce the kind of disorder that would alter the exponent n in a d-wave superconductor. The exponent remains close to $n = 1.5$, reported in single crystals of κ -(ET)₂Cu[N(CN)₂]Br and κ -(ET)₂Cu(NCS)₂ [A. Carrington et al., Phys. Rev. Lett 83. 4172 (1999)]. The transition temperature fell linearly with $1 - \eta(\text{sc})$. Measurements were also made on κ -(ET)₂Cu[N(CN)₂]Cl of normal isotopic abundance in which a very small amount of superconducting phase rise $\eta(\text{sc})$ approximate to 10^{-4} developed, presumably through the strain-induced sample mounting. This material showed a power-law exponent of $n = 1.64$, independent of the cooling rate.

Disciplines

Condensed Matter Physics

Universal temperature dependence of the London penetration depth in κ -(ET)₂X superconductors

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High-precision radio-frequency magnetic susceptibility measurements were performed on single crystals of fully deuterated κ -(ET)₂Cu[N(CN)₂]Br, hereafter designated as κ -(D8 ET)₂Cu[N(CN)₂]Br. This material phase separates into superconducting and antiferromagnetic regions, the degree of which depends strongly upon the cooling rate. We show that the screening fraction η_{sc} varies logarithmically with the cooling rate over nearly five decades. The average size of superconducting regions is estimated to vary from 5 to 40 μ m, depending upon cooling rate, consistent with previous infrared microscopy measurements. In the region $T \lesssim T_c/3$, the effective magnetic penetration depth exhibits power-law behavior $\lambda(T) - \lambda(0) \sim T^n$ with $n = 1.6$, independent of the cooling rate. Changes in cooling rate and the consequent phase separation evidently do not introduce the kind of disorder that would alter the exponent n in a d -wave superconductor. The exponent remains close to $n = 1.5$, reported in single crystals of κ -(ET)₂Cu[N(CN)₂]Br and κ -(ET)₂Cu(NCS)₂ [A. Carrington *et al.*, *Phys. Rev. Lett* **83**, 4172 (1999)]. The transition temperature fell linearly with $1 - \eta_{sc}$. Measurements were also made on κ -(ET)₂Cu[N(CN)₂]Cl of normal isotopic abundance in which a very small amount of superconducting phase $\eta_{sc} \approx 10^{-4}$ developed, presumably through the strain-induced sample mounting. This material showed a power-law exponent of $n = 1.64$, independent of the cooling rate.

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I. INTRODUCTION

The temperature dependence of the London penetration depth provides important insight into the superconducting gap structure [1–3]. In conventional isotropic full gap superconductors, the London penetration depth decays exponentially in a low-temperature range, roughly at $T < T_c/3$, in which the temperature dependence of the superconducting gap, $\Delta(T)$, becomes negligible. Nonmagnetic scattering does not change that functional trend. Quasiparticle excitations in the nodal regions of clean d -wave superconductors give rise to a linear temperature dependence of $\lambda(T)$ in a similar temperature range [4]. For d -wave superconductors, nonmagnetic impurity scattering in the unitary limit leads to a finite quasiparticle density of states in the $T \rightarrow 0$ limit and to quadratic corrections to the temperature dependence, best described by the interpolation formula $\Delta\lambda(T) \equiv \lambda(T) - \lambda(0) = aT^2/(T^* + T)$ below approximately $T_c/3$ [4]. With the emergence of many other superconductors showing nonexponential temperature variation of $\lambda(T)$, e.g., iron-based superconductors, it became convenient and often insightful to use an alternative, power-law fit, $\Delta\lambda(T) = AT^n$, for the analysis of the temperature dependence of $\lambda(T)$. Usually, the fitting is done for different values of the upper limit to see whether the exponent n is robust. For YBa₂Cu₃O_{7-x}, one of the most well-studied copper

oxide superconductors, the combination of nodal quasiparticles and unitary limit scattering leads to a power-law exponent $1 \leq n \leq 2$ where the exponent increases with disorder [2]. For clean, full-gap superconductors, this approach returns $n > 4$, which is numerically nearly indistinguishable from the standard exponential variation [3]. It should be emphasized, however, that in YBCO the exponent n changes from 2 to 1 as a function of the upper fit limit. In other cases, such as iron pnictides and the present work, the exponent stays practically constant in the whole “low-temperature limit” roughly below $T_c/3$.

A pronounced variation of the superconducting gap magnitude over various sheets of the Fermi surface, as found in multiband superconductors such as MgB₂ [5], NbSe₂ [6–8], and iron-based superconductors [3], typically produces power-law behavior similar to single-gap superconductors but over a narrower temperature range, as determined by the complete opening of the smallest gap Δ_{small} . Nonmagnetic scattering has a different effect in full-gap superconductors with sign constant s^{++} and sign-changing s^\pm superconducting gaps [9,10]. Scattering averages the gap size on the Fermi surface and can lead to more exponential $\Delta\lambda(T)$ in s^{++} and complex nonmonotonic evolution with disorder in s^\pm case [11,12]. In such situations, the data is fit over a floating range [13], returning the power-law exponent n as a function of the fitting range, modeling a possible characteristic temperature-range variation.

Early measurements of London penetration depth in the organic superconductors κ -(ET)₂Cu(NCS)₂ and

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κ -(ET)₂Cu[N(CN)₂]Br [14] showed an exponent $n \sim 3/2$ for both in-plane [14] and interplane [15] supercurrents. This observation of n well below 2 suggests a nodal superconducting gap structure. The κ -(ET)₂X organic superconductors share many common features with the copper oxides. These include extreme type-II behavior and a proximity to an antiferromagnetic Mott insulating ground state. In κ -(ET)₂Cu[N(CN)₂]Cl [16], this Mott insulating phase can be tuned by pressure or chemical substitution into superconductivity with $T_c = 12.5$ K [17–19]. Because of these similarities, the unusual exponent of London penetration depth was interpreted as evidence for d -wave pairing [20–24].

In both κ -(ET)₂Cu[N(CN)₂]Br and especially in the salt with full substitution of deuterium for hydrogen, κ -(D8 ET)₂Cu[N(CN)₂]Br, both T_c and the superconducting volume fraction depend sensitively on the rate and manner of cooling [25–44]. κ -(D8 ET)₂Cu[N(CN)₂]Br separates into an antiferromagnetic insulating phase [25] similar to κ -(ET)₂Cu[N(CN)₂]Cl [16] and a pseudogapped superconducting phase similar to κ -(ET)₂Cu[N(CN)₂]Br [45]. The origin of this nonequilibrium behavior is apparently linked to the ordering of terminal ethylene groups of the ET molecule [46–51]. Deuteration evidently moves the system closer to the superconducting/antiferromagnetic boundary and leads to electronic phase separation [45,52].

In this paper, we report high sensitivity radio-frequency magnetic susceptibility measurements of κ -(D8 ET)₂Cu[N(CN)₂]Br using a tunnel diode resonator technique [53] adopted for studying small superconducting samples [1,2]. Our motivation was to vary the degree of disorder by changing the cooling rate and then track the evolution of the temperature dependence of the effective penetration depth of the resulting composite superconductor. For a d -wave superconductor, such control of the scattering rate should lead to a suppression of the transition temperature T_c and an increase of the exponent n for increasing rates of cooling; the exponent should change from $n = 1$ in the clean case to $n = 2$ in the dirty limit [4]. For s -wave pairing, the exponent would be very large ($n \geq 4$ - mimicking exponential variation) and unchanged with disorder.

We found that despite significant changes in both T_c and the superconducting volume fraction η_{sc} , an increased cooling rate did not lead to any change in the exponent n . Interestingly, the same temperature-dependent London penetration depth with $n \sim 1.5$ was also found in the trace stress-induced superconductivity in κ -(ET)₂Cu[N(CN)₂]Cl. We found that the screening fraction in κ -(D8 ET)₂Cu[N(CN)₂]Br followed a logarithmic dependence on cooling rate over five decades, corresponding to cooling times ranging from 20 seconds to two weeks. This shows that κ -(D8 ET)₂Cu[N(CN)₂]Br does not reach true equilibrium on reasonable experimental timescales. Although these measurements did not directly probe the superconducting/antiferromagnetic phase separation, we were able to estimate the size of superconducting grains and found reasonable agreement with direct imaging measurements. The superconducting transition temperature was found to decrease monotonically with cooling rate in partial agreement with previous studies.

II. EXPERIMENTAL

Single crystals of κ -(D8 ET)₂Cu[N(CN)₂]Br were grown according to methods previously described [54]. The samples were platelets 0.2-mm thick along the b axis (normal to the conducting planes) but with irregular shapes in the a and c directions. The largest dimension was approximately 0.5 mm. The interplane electrical resistivity of the samples was measured in a four-probe configuration. Contacts to the samples were glued using DOTITE carbon paste. Resistivity measurements were performed in a *Quantum Design* PPMS.

To ensure repeatability of the data, a single cooling protocol was adopted for all measurements. The sample was first held at 100 K for several minutes after which a linear temperature ramp ($dT/dt = \text{constant}$) was applied from 100 K to 40 K. By controlling both exchange gas pressure and sample temperature, the cooling rate was varied from 0.003 K/min to 180 K/min, corresponding to cooling times ranging from two weeks down to 20 seconds. Upon reaching 40 K, the sample was cooled at 1 K/min until it reached base temperature (380 mK). With this procedure, the data were highly repeatable. The rate of cooling below 40 K had no discernible effect on the data, nor did the amount of time spent at 100 K. To test this, we varied the annealing time at 100 K from a few minutes to 12 hours and found no significant change in the data. The effort to achieve an extremely wide variation in cooling time precluded the use of a SQUID user facility to characterize the sample via susceptibility. Both the tunnel diode oscillator and the resistivity probes reveal broad transitions, as expected for a granular superconductor. This transition region, presumably involving a network of Josephson coupled grains, is outside the scope of our study. Nonetheless, our low-temperature data indicate that superconductivity within individual grains is quite similar to that of high-quality single crystals of κ -(ET)₂Cu[N(CN)₂]Br.

The susceptibility was measured with a radio frequency tunnel diode oscillator whose design has been used in many previous penetration depth studies [2,55,56]. Two samples from the same batch were examined. One was coated with a thin film of Al and one was left uncoated. The samples were mounted with silicone vacuum grease on a movable sapphire finger that allowed *in situ* removal from the rf magnetic field generated by a coil that formed part of the oscillator tank circuit.

Figure 1 shows the frequencies involved in the measurement. $f_{\text{empty}} \approx 13$ MHz is the bare oscillator frequency. Insertion of the (paramagnetic) sapphire hot finger into the coil lowered the frequency by $f_{\text{empty}} - f_{\text{sapphire}} = 1231$ Hz, which was temperature independent to within 1 Hz for $T < 15$ K. Below 2 K, f_{sapphire} showed a small and highly repeatable upturn of about 1 Hz due to paramagnetic impurities. For the samples in this paper, the frequency shifts were large enough to render this effect negligible. With the sample mounted on the hot finger, the frequency shifts upward from f_{sapphire} to f_{sc} . The frequency shift due to the sample diamagnetism is therefore $f_{sc} - f_{\text{sapphire}}$. The oscillator signal at f_{sc} was amplified, heterodyned with a local oscillator, further amplified, and filtered to provide a clean intermediate frequency signal at $\Delta f_{\text{measured}} = f_{LO} - f_{sc}$. The short-term frequency noise was typically $\delta f/f \approx 10^{-9}/\sqrt{\text{Hz}}$. For the Al-plated sample, the

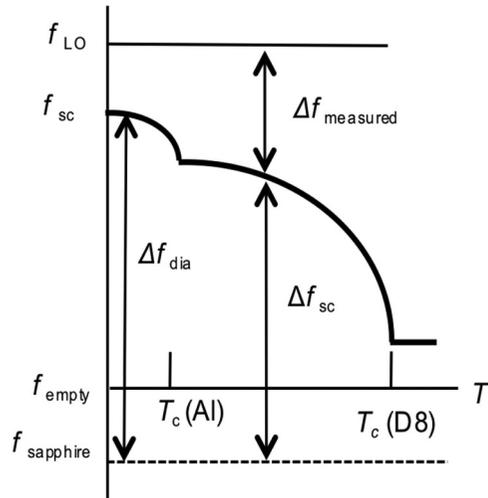


FIG. 1. Oscillator frequencies involved in the susceptibility measurement. D8 refers to κ -(D8 ET)₂Cu[N(CN)₂]Br.

onset of diamagnetism in the Al coating caused a further upward shift in frequency at the transition temperature of Al (1.2 K). Figure 2 shows the actual data for a cooling rate of 3 mK/min. The data is inverted relative to Fig. 1 since the local oscillator frequency is larger than the oscillator frequency.

III. EXPERIMENTAL RESULTS

A. Electrical resistivity

Figure 3 shows the interplane resistivity of the single crystal of κ -(D8 ET)₂Cu[N(CN)₂]Br measured for several cooling rates. The inset shows the resistive transition near T_c . At temperatures below 90 K, $\rho(T)$ becomes dependent on the cooling rate, as observed in several previous studies [26,27,43]. The dip slightly below 80 K corresponds to an equilibrium point of the transformation [31,49]. This transition has been linked to the transformation between staggered

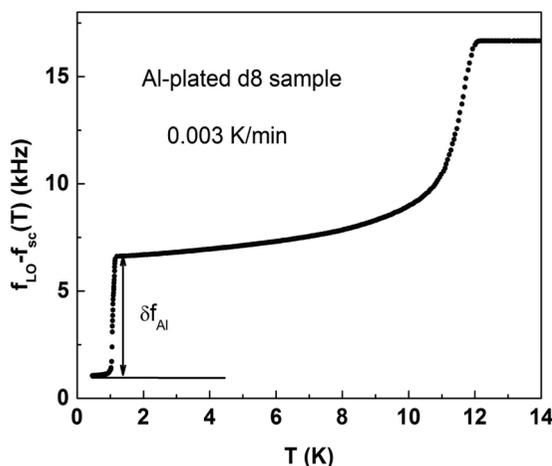


FIG. 2. Frequency shift for κ -(D8 ET)₂Cu[N(CN)₂]Br sample cooled at 3 mK/min. Δf_{Al} is due to the onset of superconductivity in the Al film coating.

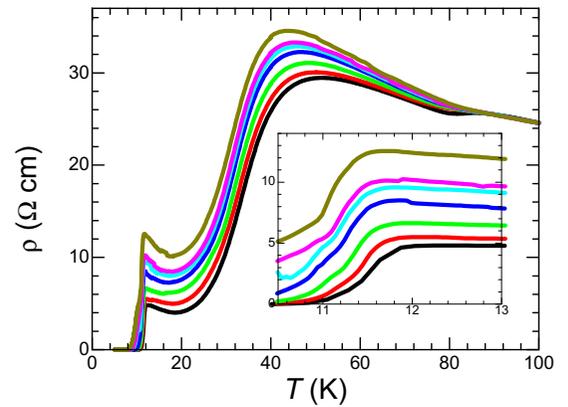


FIG. 3. Cooling rate dependence of the interplane electrical resistivity of κ -(D8 ET)₂Cu[N(CN)₂]Br. The measurements were done on cooling down from 100 K to 40 K at a rate 0.25 K/min (bottom curve), 0.5, 1, 2, 3, 4, 6 K/min (top curve). Inset shows the region near T_c .

and eclipsed conformations of the terminal ethylene groups of the ET molecule [46,48–51].

The resistivity peak near 50 K is common to both κ -(D8 ET)₂Cu[N(CN)₂]Br and κ -(ET)₂Cu[N(CN)₂]Br. In the latter material, the resistivity falls monotonically toward T_c but in κ -(D8 ET)₂Cu[N(CN)₂]Br there is a minimum near 20 K and an upturn before reaching the superconducting transition. The minimum suggests that the resistivity is determined by complex percolation between conducting and antiferromagnetically ordered insulating regions and no complete metallic path for conductivity is formed.

B. Logarithmic dependence of the screening fraction on the cooling rate

The screening fraction η_{sc} is defined to be the ratio of the susceptibility of the sample to the susceptibility of a perfectly diamagnetic sample (London penetration depth $\lambda = 0$) with the same shape and volume. Referring to Fig. 1, η_{sc} is given by the ratio of the frequency shifts:

$$\eta_{\text{sc}} = \frac{\Delta f_{\text{sc}}(T)}{\Delta f_{\text{sc}}(\lambda = 0)} \approx \frac{\Delta f_{\text{sc}}(T)}{\Delta f_{\text{dia}}}. \quad (1)$$

To obtain Δf_{dia} , one of the samples was sputter coated with 50 nm of Al. Using a focused ion beam to cross section the film, we have found that this approach yielded a film thickness uniform to better than 10% over the entire sample, including edges and corners. Once the base temperature of $T = 0.38 \text{ K} \ll T_c(\text{Al}) \approx 1.2 \text{ K}$ was reached, the penetration depth of the aluminum has essentially attained its zero-temperature value of $\lambda_{\text{Al}} \approx 50 \text{ nm}$. This value is roughly 1000 times smaller than any sample dimension, so the sample acts as a nearly perfect diamagnet. The full frequency shift at 0.38 K is labeled Δf_{dia} in Fig. 2. For $T > T_c(\text{Al}) = 1.2 \text{ K}$, the Al film is magnetically transparent since the skin depth of Al at 13 MHz is several orders of magnitude larger than the film thickness. Therefore, above $T_c(\text{Al})$ the frequency shift comes from the organic superconductor alone. Since $T_c(\text{Al}) = 1.2 \text{ K} \ll T_c(\text{D8}) = 12 \text{ K}$, the zero-temperature screening fraction of

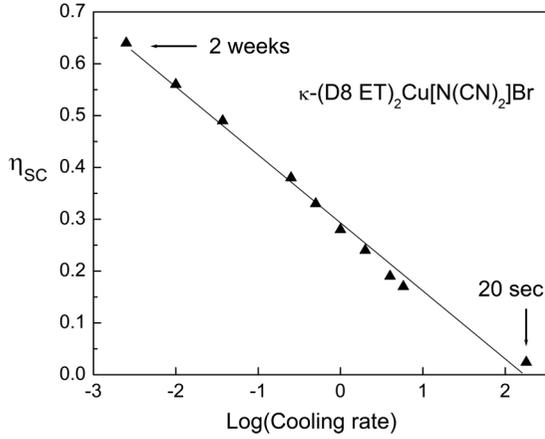


FIG. 4. Screening fraction versus log (cooling rate) for the aluminum-plated κ -(D8 ET)₂Cu[N(CN)₂]Br sample.

the κ -(D8 ET)₂Cu[N(CN)₂]Br sample was approximated by $\eta_{sc}(T = 0) \approx \Delta f_{sc}(1.2 \text{ K})/\Delta f_{dia}$.

The triangles in Fig. 4 show η_{sc} versus the logarithm of the cooling rate. The cooling time from 100 K to 40 K was varied from 20 seconds to two weeks. For the lowest rate (0.003 K/min), the screening fraction was 64%, somewhat below the theoretical maximum packing fraction of 74% for identical close-packed spheres. For the most rapid cooling rate (180 K/min), the screening fraction was 2.4%.

The logarithmic time dependence extends over nearly five decades and illustrates that κ -(D8 ET)₂Cu[N(CN)₂]Br samples never reach true equilibrium on experimentally practical timescales. Logarithmic relaxation is widely observed in disordered systems when there exists a broad distribution of barrier heights that must be overcome to achieve equilibrium [57,58]. Flux creep in superconductors is a well-established example [59,60].

C. Superconducting grain size

κ -(D8 ET)₂Cu[N(CN)₂]Br is known to phase separate into superconducting and normal domains [45,52]. Since our probe measures the bulk susceptibility, we have no direct knowledge of the size distribution or shape of the superconducting regions. However, we can perform a self-consistency check. Scanning infrared measurements indicated superconducting grain sizes of order of 10–30 μm [61], which is much larger than the zero-temperature coherence length $\xi_0 \sim 5 \text{ nm}$ in κ -(ET)₂Cu[N(CN)₂]Br. This implies that supercurrents within individual grains, rather than intergranular Josephson currents, dominate the Meissner screening [62]. We will therefore assume that below T_c the sample consists of N_g independent superconducting spheres, each of radius R_g and London penetration depth λ , surrounded by an insulating matrix. Using the expression for the susceptibility of a single superconducting sphere, the change in oscillator frequency upon inserting a collection of N_g isolated spheres into the resonator coil is given by

$$\Delta f_{sc} = \frac{GN_g V_g}{1-N} \left[1 - 3 \frac{\lambda}{R_g} \coth \frac{R_g}{\lambda} + 3 \left(\frac{\lambda}{R_g} \right)^2 \right], \quad (2)$$

where V_g is a volume of a single grain, N is the demagnetization factor, and $G = 1.16 \times 10^{-4} \text{ Hz}/\mu\text{m}^3$ is a calibration factor. The latter is determined by inserting an indium sphere ($N = 1/3$) of known volume into the oscillator coil at base temperature and measuring the frequency shift. For our simple model, the grains are also assumed to be spherical, so $V_g = 4\pi R_g^3/3$ and $N = 1/3$. There is a further complication that the London penetration depth in κ -(ET)₂X superconductors is highly anisotropic with $\lambda_{\text{interplane}}/\lambda_{\text{inplane}} \approx 100$ in κ -(ET)₂Cu[N(CN)₂]Br [14,15]; $\lambda_{\text{interplane}}$ is the penetration depth for interplane currents which can depend sensitively upon the mode of transport between planes as well as superconducting properties. To isolate $\lambda_{\text{interplane}}$, the rf magnetic field was applied perpendicular to the conducting planes so as to induce only in-plane supercurrents. This is strictly true only if the grains are magnetically isolated. Hereafter, we will refer to λ_{inplane} as simply λ .

As we will show, κ -(D8 ET)₂Cu[N(CN)₂]Br and κ -(ET)₂Cu[N(CN)₂]Br have similar transition temperatures and nearly the same thermal exponents for the penetration depth. We therefore assume that the zero-temperature penetration depth in each grain of κ -(D8 ET)₂Cu[N(CN)₂]Br is close to the zero-temperature penetration depth of uniform κ -(ET)₂Cu[N(CN)₂]Br. [Our measurements on this composite superconductor/insulator cannot yield $\lambda(T = 0)$ for individual grains.] Measurements of $\lambda(T = 0)$ on single crystals of κ -(ET)₂Cu[N(CN)₂]Br range from 0.32 to 1.1 μm [28,38,42,63–65], with some considerably larger [66]. Given the large uncertainty in this quantity, we will assume that $\lambda(T = 0) \equiv \lambda_0 = 1 \mu\text{m}$ in each superconducting grain of κ -(D8 ET)₂Cu[N(CN)₂]Br. This is still much smaller than the reported grain size of 30 μm , obtained by direct imaging of phase-separated D8-Br [52]. Therefore, at our base temperature of 0.38 K, $\lambda(0.38 \text{ K}) \ll R_g$ and

$$\Delta f_{sc}(T = 0.38 \text{ K}) \approx \frac{GN_g V_g}{1-N}. \quad (3)$$

Dividing Eq. (2) by Eq. (3), we have

$$\frac{\Delta f_{sc}(T)}{\Delta f_{sc}(T = 0.38 \text{ K})} = 1 - 3 \frac{\lambda}{R_g} \coth \frac{R_g}{\lambda} + 3 \left(\frac{\lambda}{R_g} \right)^2. \quad (4)$$

Using this equation, the data were numerically inverted to obtain λ/R_g as a function of temperature. These values were then fit, in the region $T < T_c/3$, to the form

$$\frac{\lambda(T)}{R_g} = \frac{\lambda_0}{R_g} + \frac{A}{R_g} \left(\frac{T}{T_c} \right)^n. \quad (5)$$

Table I shows λ_0/R_g along with several other parameters as a function of cooling rate for the *unplated* D8-Br sample. (Data for the Al-plated sample extended down to only 1.2 K and therefore did not provide an accurate measurement of the low-temperature behavior.) Our analysis provides only the ratio of penetration depth to grain size. Under the assumption that $\lambda_0 = 1 \mu\text{m}$, we obtain R_g . For the slowest cooling rate, this gives $R_g = 39 \mu\text{m}$, in reasonable agreement with the value of 30 μm obtained from far infrared scanning. More recent penetration-depth measurements by Milbradt *et al.* [64] give $\lambda_0 = 0.32 \mu\text{m}$ for κ -(ET)₂Cu[N(CN)₂]Br. Using this

TABLE I. Parameters for the unplated κ -(D8 ET)₂Cu[N(CN)₂]Br sample as a function of cooling rate. The estimated grain size R_g is obtained by assuming $\lambda_0 = 1 \mu\text{m}$.

Cooling rate (K/min)	T_c (K)	λ_0/R_g	R_g (μm)	A/λ_0	A/R_g (10^{-3})	n
0.037	11.52	0.0542	39.2	0.828	44.8	1.59
0.25	11.14	0.0865	23.4	0.696	60.2	1.56
0.50	11.05	0.1043	20.5	0.668	69.4	1.58
1	10.95	0.1224	18.2	0.643	78.2	1.59
2	10.85	0.1412	16.3	0.607	85.5	1.59
3	10.79	0.1581	15.4	0.561	88.3	1.57
4	10.76	0.1688	14.4	0.551	93.1	1.58
5.8	10.71	0.1863	14.1	0.560	104.2	1.61
180	10.02	0.3604	5.6	0.658	235.8	1.61

value, our estimates for the grain size R_g would be reduced by a factor of 3 from those in the table.

The fitting parameter A is proportional to the rate of change of the penetration depth in each grain with temperature. If the picture of individual grains is correct, A/λ_0 should be independent of cooling rate and grain size. The data indicates that $A/\lambda_0 = 0.64 \pm 0.19$.

D. Strain-induced superconductivity in κ -(ET)₂Cu[N(CN)₂]Cl

κ -(ET)₂Cu[N(CN)₂]Cl and κ -(ET)₂Cu[N(CN)₂]Br are isostructural. For temperatures above 50 K, the electronic and magnetic properties of the two materials are very similar. As the temperature is lowered, κ -(ET)₂Cu[N(CN)₂]Cl becomes an antiferromagnetic insulator, while κ -(ET)₂Cu[N(CN)₂]Br becomes a superconductor. With an applied pressure of $P = 300$ bar, κ -(ET)₂Cu[N(CN)₂]Cl becomes a superconductor at 13 K [67]. Since our experiments were performed in vacuum, superconductivity would not be expected to appear. However, strains due to the attachment of the sample to the sapphire stage were enough to induce a tiny amount of superconductivity [17].

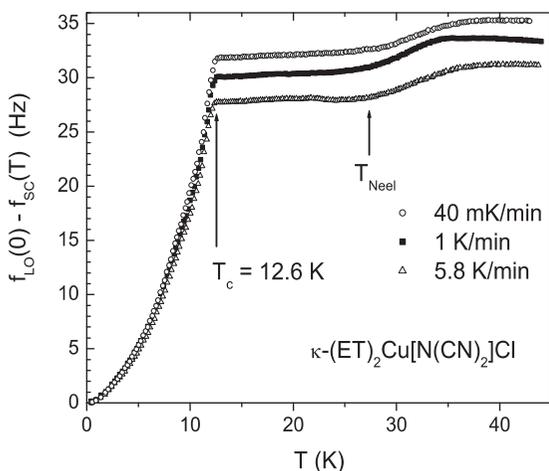


FIG. 5. Frequency shift of residual superconductivity in κ -(ET)₂Cu[N(CN)₂]Cl for three different cooling rates.

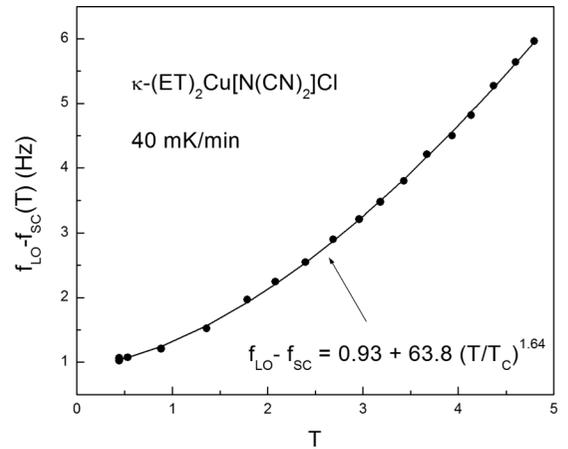


FIG. 6. Frequency shift versus temperature in the low temperature region for κ -(ET)₂Cu[N(CN)₂]Cl.

Figure 5 shows the oscillator frequency shift κ -(ET)₂Cu[N(CN)₂]Cl from 44 K down to 0.38 K for three different cooling rates. Near the Neel temperature of 27 K, there is a change in the skin depth as the bulk sample becomes an antiferromagnet. For the three different cooling rates shown, superconductivity sets in at 12.6 K. The total diamagnetic frequency shift is about 30 Hz, roughly 5000 times smaller than the shift for the slowly cooled κ -(D8 ET)₂Cu[N(CN)₂]Br sample which was of comparable size. Figure 5 shows that the screening fraction decreases slightly with increased cooling rate, but the change is much smaller than for the κ -(D8 ET)₂Cu[N(CN)₂]Br samples. T_c values for the three cooling rates were equal, within experimental error. Since neither the shape nor size of the superconducting regions is known, there is no reliable way to estimate changes in penetration depth from the measured frequency shifts for this sample. Nonetheless, well below T_c , the raw frequency shift provides an excellent measure of the temperature dependence of the penetration depth. As shown in Fig. 6, the frequency shift in the low-temperature region fits a power law with exponent $n = 1.64$, a value only slightly higher than those observed for κ -(D8 ET)₂Cu[N(CN)₂]Br.

The superconductivity observed in κ -(ET)₂Cu[N(CN)₂]Cl is subject to stress and highly inhomogeneous. Nevertheless, the power-law exponent is close to the values observed for the κ -(D8 ET)₂Cu[N(CN)₂]Br samples in this paper. This is an important corrective to suggestions that the power laws observed in the organics are due to a broad distribution of transition temperatures.

IV. DISCUSSION

A. Transition temperature

The superconducting transition temperature for unplated κ -(D8 ET)₂Cu[N(CN)₂]Br fell monotonically with cooling rate, as shown in Table I. While the unplated sample was required to determine the exponent at low temperatures, the Al-plated sample was more useful for studying T_c due to the wider range of cooling rates and the ability to measure the screening fraction. Figure 7 shows the frequency shift versus temperature for cooling rates ranging from 0.003 to

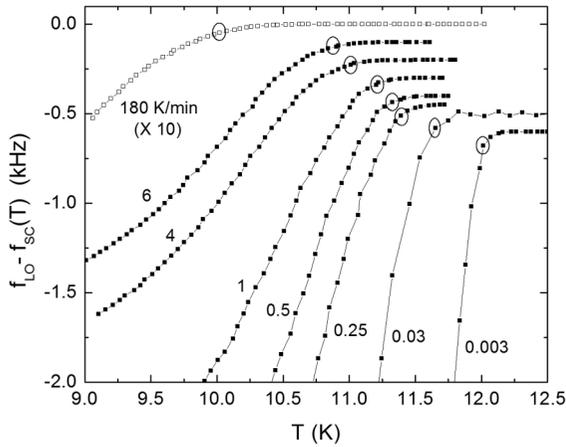


FIG. 7. Frequency shift near T_c for Al-plated κ -(D8 ET) $_2$ Cu[N(CN) $_2$]Br sample. The cooling rate varies from 0.003 to 180 K/min. T_c for each curve is circled. Data for different rates are offset vertically for clarity.

180 K/min. As the cooling rate increased, the transitions became extremely broad, making midpoint or inflection-point criteria meaningless. Instead, T_c was defined as the temperature at which the signal dropped by 1% from its normal-state value. These points are circled for each rate. The 1% criterion naturally leads to systematically higher values of T_c . For example, with κ -(ET) $_2$ Cu[N(CN) $_2$]Br cooled at a moderate rate (1.5 K/min), the midpoint criterion gave the established value of $T_c = 11.6$ K and the 1% criterion gave 12.4 K. The $T_c =$ values obtained this way are plotted in Fig. 8. For all but the 180 K/min data, T_c fell linearly with $1-\eta_{sc}$, essentially the fraction of nonsuperconducting material.

Yoneyama *et al.* [41] measured the susceptibility and transition temperature in κ -(ET) $_2$ Cu[N(CN) $_2$]Br as a function of the deuterium/hydrogen ratio, $x = [D]/[H]$, ranging from $x = 0$ to $x = 1$. They found that for slow cooling (0.2 K/min), T_c increased for $x < 0.2$ but then fell for larger x . For rapidly cooled (100 k/min) samples, T_c fell monotonically with x ,

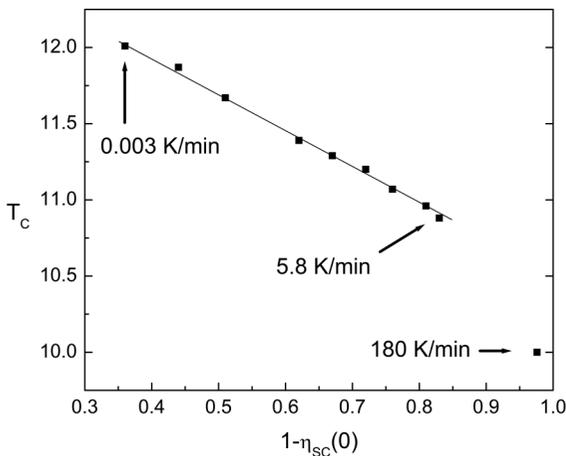


FIG. 8. Transition temperature versus $1-\eta_{sc}$ for the Al-plated sample of κ -(D8 ET) $_2$ Cu[N(CN) $_2$]Br.

reaching $T_c \approx 10$ K and a screening fraction of a few percent, similar to our own data at 180 K/min.

A possible interpretation of Fig. 8 is that $1-\eta_{sc}$ is an indirect measure of the effective scattering rate, which in turn increases with cooling rate. This may be reflected in the increase of resistivity above T_c evident in Fig. 3. The drop in T_c would then be attributable to the Abrikosov-Gor'kov (AG) formula [68], which gives, close to clean limit,

$$T_{c0} - T_c \approx A_m \rho_m T_{c0}. \quad (6)$$

Here T_{c0} is the transition temperature in the superconductor free of impurities, $\rho_m = \hbar/(2\pi k_B T_{c0} \tau_m)$ is the dimensionless scattering rate, and τ_m is the scattering time associated with the spin-flip scattering. $A_m \approx 5$ for s -wave pairing and $A_m \approx 2.5$ for d -wave pairing, assuming only pair-breaking scattering. In an s -wave superconductor, this would need to come from local moments for which there is no evidence. For d -wave pairing, both magnetic and nonmagnetic impurities are pair breaking and suppress T_c through the AG mechanism. Powell and McKenzie [69] performed a comprehensive analysis of experiments on T_c reduction and its implications for the pairing symmetry in both β -(ET) $_2$ Y and κ -(ET) $_2$ X superconductors. The available evidence strongly favors T_c reduction by nonmagnetic disorder, implying an order parameter with angular momentum greater than one, most likely d -wave [70]. Two experiments in particular illustrate this point. Analytis *et al.* [71] and Sasaki *et al.* [72] studied T_c reduction in κ -(ET) $_2$ Cu(NCS) $_2$ subject to irradiation. Transport measurements and quantum oscillation measurements were used to assess the scattering rate. Both experiments found agreement with the AG picture for low scattering rates. Therefore, unless x-ray or proton irradiation generates local moments, the reduction in T_c is consistent with a d -wave order parameter. As the irradiation level was increased beyond a certain level, corresponding to scattering rates greater than roughly 10^{12} sec^{-1} , the reduction in T_c was smaller than predicted by AG theory. One suggestion is that κ -(ET) $_2$ Cu(NCS) $_2$ has a multicomponent order parameter, although there is no independent evidence for this [71].

Figure 8 also suggests that electronic phase separation plays a role in determining T_c . Haddad *et al.* [70] proposed that, for slow cooling, the insulating regions in κ -(D8 ET) $_2$ Cu[N(CN) $_2$]Br act as extended scattering centers that lower T_c through the AG mechanism. For more rapid cooling, they propose that the sample consists of superconducting islands connected to each other through Josephson coupling. The increased cooling rate lowers the superconducting fraction, weakens the Josephson coupling, and lowers T_c . Figure 8 shows that T_c does fall more rapidly for cooling times of less than 5.8 hours. This may indicate the onset of a regime dominated by Josephson coupling but we have insufficient data to test this hypothesis.

B. Power-law exponent of London penetration depth

The last column in Table I shows the power-law exponent n , which is plotted versus the log of the cooling rate in Fig. 9. Despite changes in cooling rate by nearly four orders of magnitude and changes in the screening fraction by a factor of 30, the thermal exponent remains constant within

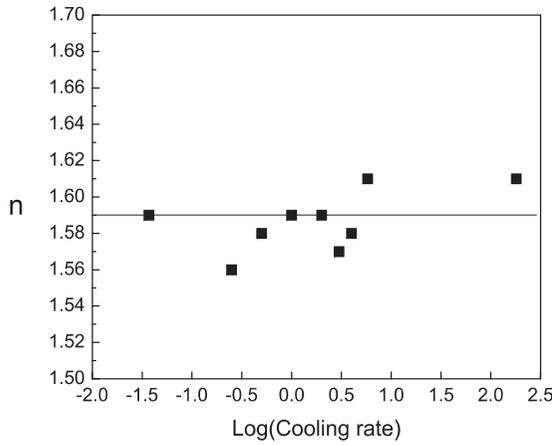


FIG. 9. Dependence of the exponent n of the power-law function describing temperature-dependent London penetration depth, $\lambda = \lambda_0 + AT^n$ on the logarithm of the cooling rate for κ -(D8 ET)₂Cu[N(CN)₂]Br.

error bars, with $n = 1.59 \pm 0.03$. The assumption of spherical grains used in Eq. (2) is not critical. Since $\lambda \ll R_g$ over the temperature range in question, the frequency shift is a linear function of the penetration depth, as it would be for rectangular- or cylindrical-shaped grains:

$$\Delta f_{sc} \approx GN_g V_g \left[1 - 3 \frac{\lambda(T)}{R_g} \right] = a + b\lambda. \quad (7)$$

The exponent can therefore be obtained directly from fits to the raw frequency shift versus temperature, as shown in Fig. 10.

The value $n = 1.59$ is close to the value of $n = 1.5$ first reported for both the in-plane [14] and out-of-plane [15] penetration depths in both κ -(ET)₂Cu[N(CN)₂]Br and κ -(ET)₂Cu(NCS)₂. If we adopt a (d -wave + impurity) model for κ -(D8 ET)₂Cu[N(CN)₂]Br, it appears that rapid cooling and the ensuing AF/SC phase separation does not generate the type of unitary limit scattering that is widely observed in the copper oxides. Otherwise, the exponent would increase with cooling rate. The increased cooling rate does

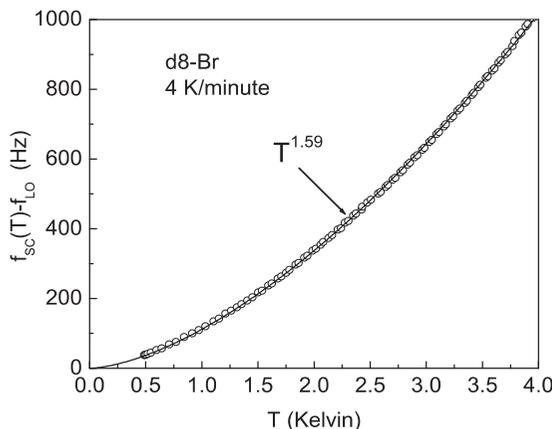


FIG. 10. Raw data: frequency shift versus temperature along with power-law fit for unplated κ -(D8 ET)₂Cu[N(CN)₂]Br sample.

lead to large changes in the resistivity (Fig. 3), a reduction in T_c (Fig. 8), and considerable rounding of the superconducting transition, all indicators of increased scattering. Measurements in κ -(ET)₂Cu[N(CN)₂]Br show an increase in λ_0 [42] and a decrease in the Dingle temperature of quantum oscillations [35]. Together, these experiments indicate that more rapid cooling directly increases the electronic scattering rate. The fact that $n \approx 1.6$ is insensitive to the cooling rate would seem to imply either a peculiar kind of impurity scattering or that this exponent is intrinsic to the κ -(ET)₂X superconductors. This situation is unlike either the copper oxides [2] or the iron-based superconductors [9,11,12,73–75], where the exponent evolves in predictable ways with doping or impurity content. The difference in response to cooling rate between T_c and the power-law exponent forces us look for alternative explanations.

C. Type of disorder in systems with ethylene ordering transformation

In the first scenario, we consider the possibility that variation of T_c with cooling is not caused by the pair-breaking action of disorder and the persistent $n = 1.6$ exponent is determined by the superfluid response in relatively clean grains of d -wave superconductors with a pseudogap [45]. Here, pseudogap refers to the decrease in spin susceptibility below 50 K observed via electron spin resonance measurements in κ -(ET)₂Cu[N(CN)₂]Br. Rapid cooling of κ -(ET)₂Cu[N(CN)₂]Br is known to affect this pseudogap-related decrease of susceptibility, possibly by reducing the density of states [33]. If so, then rapid cooling would likely reduce T_c as well, irrespective of the amount of disorder.

It is worthwhile to recall the situation in β -(ET)₂I₃ in which the superconducting state is also a strong function of pressure and annealing history. β -(ET)₂I₃ has two superconducting phases. The β_L phase ($T_c = 1.5$ K) shows an incommensurate superstructure [76] with modulation of ethylene group conformations and triiodide I₃ group displacements [51]. The β_H phase ($T_c = 7.5$ K) is stabilized by pressure [77] and the superstructure is suppressed [78]. However, the superconducting state with $T_c = 7.5$ K can be obtained by annealing the crystal at temperatures in the 100 K to 120 K range [79,80], resulting in the formation of a metastable state with $T_c \sim 2$ K. Here the superstructure changes its wave vector from the state in crystals with usual cooling protocol of approximately 1 K/min [79].

X-ray diffraction studies in the closely related compound κ -(ET)₂Cu[N(CN)₂]I have shown that the glass transition is accompanied by the formation of a highly disordered superstructure [81,82]. If a similar scenario works for κ -(D8 ET)₂Cu[N(CN)₂]Br, short-range correlations would form extended defects, which would have a very different effect on scattering than point defects. However, x-ray studies on κ -(D8 ET)₂Cu[N(CN)₂]Br are less conclusive. No new periodic modulations that depend on cooling protocol are found and, even with moderately (4 K/min) slow cooling, 97% of the molecules still adopt the eclipsed configuration at low temperatures [50].

Despite notable differences in the shape of the Fermi surface, both β and κ phase organic superconductors exhibit

“dimerization” of the BEDT-TTF molecule and half-filled energy bands [20]. As with β -(ET)₂I₃, it seems likely that the ultimate source of the cooling rate dependence is the equilibration of ethylene groups into ordered positions in the superstructures. Senoussi *et al.* performed a detailed examination of the glass transition and superconducting fraction in κ -(D8 ET)₂Cu[N(CN)₂]Br samples, using SQUID susceptometry [43]. The final superconducting fractions they observed were similar to our own. They reported stretched exponential relaxation but their cooling procedures were rather different from those used here. They did not report values for the thermal exponent of the penetration depth, which was the primary motivation for our experiment.

D. Quantum critical scenario

Since the first report [14] of an $n = 1.5$ power law in the organics, this same exponent has been observed in the heavy-fermion superconductors CeCoIn₅ [83,84] and Ce₂PdIn₈ [85] and in the iron-based superconductor BaFe₂(As_{0.7}P_{0.3})₂ [85,86]. In the latter, the $n = 1.5$ power law appears at a doping level coincident with a peak in λ_0 and an antiferromagnetic quantum critical point beneath the superconducting dome [56,86].

Nomoto and Ikeda calculated an $n = 1.5$ power-law exponent coming from renormalization of the superfluid density by quantum critical fluctuations in a d -wave superconductor [87]. They predict that the power law should return to $n = 1$ at the lowest temperatures. This may resolve the disagreement between our results and recent microwave measurements by Milbradt *et al.* [64] who reported a linear T dependence of the superfluid density in κ -(ET)₂Cu[N(CN)₂]Br at temperatures below ours. She *et al.* [88] also showed that quantum critical fluctuations will produce logarithmic corrections to the linear T penetration depth in nodal superconductors. These corrections may lead to an effective power law whose origin is entirely distinct from impurity scattering. However, the phase transition between antiferromagnetic and superconducting states in κ -phase superconductors is strongly first order [45], which makes a quantum criticality scenario unlikely. And, contrary to the quantum critical scenario, no mass enhancement has been reported in heat-capacity measurements [36]. Moreover, in the two compounds where quantum criticality and an $n = 1.5$ exponent occur (CeCoIn₅ [89] and BaFe₂(As_{1-x}P_x) [11]), the exponent still changes with disorder.

Whether or not quantum criticality plays a role, the power-law behavior adds to the already strong case for d -wave pairing in both κ -(ET)₂Cu[N(CN)₂]Br and in

κ -(ET)₂Cu(NCS)₂ [14,20,21,90–101]. Fewer tests of order-parameter symmetry have been reported in the deuterated versions of these compounds but scanning tunneling measurements have shown evidence for d -wave pairing in κ -(ET)₂Cu[N(CN)₂]Br with varying amounts of deuteration [102].

V. CONCLUSIONS

In conclusion, rf susceptibility measurements have been performed on κ -(D8 ET)₂Cu[N(CN)₂]Br crystals. The screening fraction falls logarithmically with cooling rate over nearly five decades. By modeling the electronically phase-separated material as a collection of independent spheres we estimate a sphere diameter in reasonable agreement with scanning infrared measurements. The transition temperature falls linearly with screening fraction over a wide range of cooling rates but then drops sharply for quenched samples. The effective penetration depth obeys a power law with exponent $n = 1.6$ independent of cooling rate and close to the value for the undeuterated compound κ -(ET)₂Cu[N(CN)₂]Br. The appearance of nearly the same power-law exponent in κ -(ET)₂Cu[N(CN)₂]Br, κ -(ET)₂Cu(NCS)₂, highly strained κ -(ET)₂Cu[N(CN)₂]Cl, and κ -(D8 ET)₂Cu[N(CN)₂]Br independent of cooling rate and screening fraction suggests an intrinsic origin to this exponent. While a d -wave model with impurity scattering provides a perfectly satisfactory fit to all the low-temperature data, it is difficult to imagine how the degree of impurity scattering remains nearly independent of every parameter we have been able to adjust. It is important to stress that the interplay of antiferromagnetism and superconductivity is the common thread in all the materials exhibiting this unusual power-law behavior. In any case, the persistent power law variation together with a continuously reduced transition temperature as a function of cooling rate strengthens the case for d -wave pairing symmetry in the κ -(ET)₂X organic superconductors.

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