Interplane penetration depth and coherent transport in organic superconductors

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Measurements of the interlayer penetration depth $\lambda_\perp$ have been performed on single crystals of the organic superconductors κ-$(ET)_2Cu[N(CN)_2]Br$ and κ-$(ET)_2Cu(NCS)_2$. We find that $\lambda_\perp(0) \approx 130\ \mu m$ for both materials. The normalized superfluid density $\rho_\perp = [\lambda_\perp(0)/\lambda_\perp(T)]^2$ may be fit equally well to a power law $1-\rho_\perp \sim T^n$ with $n=1.3-1.5$ or to the form $1-\rho_\perp = \alpha(T^2/T_C)/(T+T^*)$, consistent with a d-wave pairing state with impurity scattering. The data imply coherent transport between conducting planes, in agreement with recent magnetoresistive measurements [J. Singleton, P. A. Goddard, A. Ardavan, N. Harrison, S. J. Blundell, J. A. Schlueter, and A. M. Kini, Phys. Rev. Lett. **88**, 037001 (2002)] and in contrast to the copper oxides.

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I. INTRODUCTION

The κ-$(ET)_2X$ organic compounds are strongly correlated quasi-two-dimensional (quasi-2D) conductors with many similarities to the copper oxides.[1–3] The proximity to a Mott insulating phase has lead several investigators to propose a magnetic mechanism for superconductivity and a d-wave pairing state.[2–7] Although some experiments have claimed s-wave pairing,[8] penetration depth,[9–11] NMR,[12,13] thermal-conductivity,[14,15] specific-heat,[16,17] and scanning tunneling microscopy[18] (STM) experiments all provide strong support for nodal quasiparticles and d-wave pairing. In particular, high-resolution measurements of the in-plane penetration depth $\lambda_\parallel$ in both κ-$(ET)_2Cu[N(CN)_2]Br$ and κ-$(ET)_2Cu(NCS)_2$ showed power law, as opposed to exponential temperature dependence.[9] In this paper, we report measurements of the interlayer penetration depth $\lambda_\perp$ in single crystals of both compounds. Since $\lambda_\perp$ characterizes the flow of supercurrent *between* conducting planes, its temperature dependence reflects both the structure of the order parameter and the mechanism of interlayer transport, issues crucial to understanding the role of dimensionality in strongly correlated superconductors. We find that $\lambda_\perp$ and the associated superfluid density $\rho_\perp = [\lambda_\perp(0)/\lambda_\perp(T)]^2$ both exhibit power-law behavior similar to the corresponding in-plane quantities. This result implies that the interlayer transport is nearly coherent, in agreement with magnetoresistive measurements on κ-$(ET)_2Cu(NCS)_2$.[19] It highlights an important distinction between the organics and the copper oxides, which exhibit incoherent interlayer transport. How the interlayer transport is related to the mechanism of superconductivity in both organics and copper oxides is an important but still unanswered question.

II. EXPERIMENTAL

Samples of κ-$(ET)_2Cu[N(CN)_2]Br$ (T_C=11.95 K) and κ-$(ET)_2Cu(NCS)_2$ (T_C=9.8 K) in the shape of thin irregular platelets were grown using methods described earlier.[20] $\lambda_\perp$ was measured with a 12 MHz tunnel diode oscillator with frequency noise of order $\delta f/f < 10^{-9}/\sqrt{\mathrm{Hz}}$.[21] The rf magnetic field amplitude was approximately 20 mOe, which is well below Hc_1. The crystals were mounted on a moveable sapphire hot finger whose temperature could vary from 0.4–100 K. *In situ* removal of the sample from the probe coil permitted a measurement of the total rf magnetic moment at any temperature. For each sample, the rf magnetic field was applied in two orthogonal directions (in separate runs) as shown in Fig. 1. Both field orientations were parallel to the conducting planes, in turn generating both in-plane and interlayer supercurrents. These currents penetrate the sample by $\lambda_\parallel$ and $\lambda_\perp$, respectively. The oscillator frequency shift is proportional to the rf magnetic moment of the sample. For a thin rectangular slab of thickness d and width L, in which

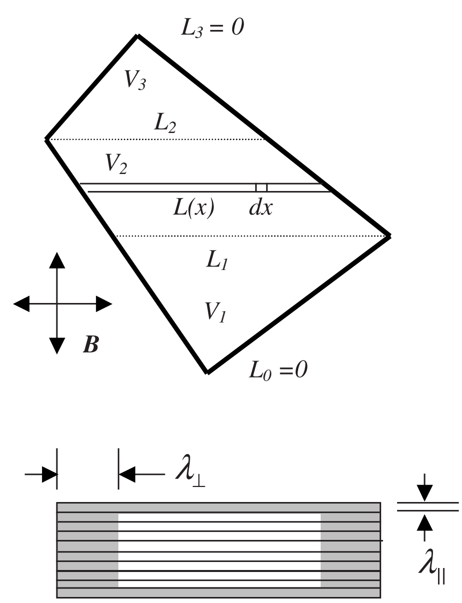

FIG. 1. (Upper) Polygon-shaped sample looking down onto conducting planes. Two different orientations of rf magnetic field B are shown. L_i and V_i refer to Eq. (3). (Lower) Penetration of in-plane and interlayer currents into sample. Horizontal lines indicate conducting planes.

TABLE I. $\lambda_\perp(0)$ for κ-$(ET)_2Cu[N(CN)_2]Br$ and κ-$(ET)_2Cu(NCS)_2$ obtained from various experiments.

Material	$\lambda_\perp(0)$ (μm)	Technique	Reference
κ-$(ET)_2Cu[N(CN)_2]Br$	130 ± 20	Tunnel diode oscillator	This work
κ-$(ET)_2Cu[N(CN)_2]Br$	100 ± 20	Tunnel diode oscillator	9
κ-$(ET)_2Cu[N(CN)_2]Br$	133	ac susceptibility	10
κ-$(ET)_2Cu(NCS)_2$	90	Josephson plasma resonance	25
κ-$(ET)_2Cu(NCS)_2$	130 ± 20	Tunnel diode oscillator	This work
κ-$(ET)_2Cu(NCS)_2$	60	Scanning SQUID probe	26
κ-$(ET)_2Cu(NCS)_2$	200	ac susceptibility	22
κ-$(ET)_2Cu(NCS)_2$	30	Surface impedance (100 GHz)	27
κ-$(ET)_2Cu(NCS)_2$	130–240	Torque magnetometry	28
κ-$(ET)_2Cu(NCS)_2$	120	Josephson plasma resonance	29

demagnetizing effects are negligible, the effective susceptibility is given by[22]

$$-\chi = 1 - \frac{2\lambda_\parallel}{d}\tanh\frac{d}{2\lambda_\parallel} - \frac{16\lambda_\perp}{\pi^2 L}\sum_{n=0}^{\infty}\frac{\tanh(q_n L/2)}{(2n+1)^2(k_n^2\lambda_\parallel^2+1)^{3/2}}, \tag{1}$$

where $k_n=(2n+1)\pi/d$ and $q_n^2=(k_n^2\lambda_\parallel^2+1)/\lambda_\perp^2$. According to this formula, the susceptibility will be dominated by $\lambda_\perp$ if the anisotropy $\gamma=\lambda_\perp/\lambda_\parallel \gg L/d$. In our case, $\gamma=\lambda_\perp/\lambda_\parallel \approx 100 \gg L/d < 10$ so this condition is well satisfied. In this limit of extreme anisotropy, the susceptibility reduces to the standard result,

$$-\chi = \left(1 - \frac{2\lambda_\perp}{L}\tanh\frac{L}{2\lambda_\perp}\right). \tag{2}$$

In fact, our samples were thin irregular polygons, as shown schematically in Fig. 1. We approximated the total rf susceptibility by assuming the slab susceptibility (2) for each section of width $L(x)$ and then integrating the result to obtain,

$$-\chi_{Polygon} = \left[1 - 8\lambda_\perp^2\sum_{i=1}^{n}\frac{V_i}{V_T}\frac{\ln\left(\dfrac{\cosh(L_i/2\lambda_\perp)}{\cosh(L_{i-1}/2\lambda_\perp)}\right)}{L_i^2 - L_{i-1}^2}\right]. \tag{3}$$

In this expression, V_i is the volume of the ith trapezoid or triangle (see Fig. 1) and V_T is the total sample volume. As was shown earlier, in strongly demagnetizing geometries, which would occur if the ac field were applied normal to the conducting planes, the effective length scale in Eq. (2) is quite different from the actual sample dimension.[23] For the thin samples used here, this correction was not necessary. Demagnetization was taken in account with a prefactor $1/(1-N)$, which is valid at low temperatures, where the susceptibility is close to -1. To estimate the demagnetizing factor N, we approximated the sample shape by an inscribed ellipsoid whose demagnetization factor was obtained numerically,[24]

$$N = \frac{1}{2}\int_0^\infty \frac{du}{(1+u)^{3/2}\left(1+\dfrac{Z^2}{X^2}u\right)^{1/2}\left(1+\dfrac{Z^2}{Y^2}u\right)^{1/2}}. \tag{4}$$

In this equation, Z is the sample dimension parallel to the applied field, while X and Y are dimensions orthogonal to the field. For the κ-$(ET)_2Cu[N(CN)_2]Br$ sample $N\approx .07$, while for the thicker κ-$(ET)_2Cu(NCS)_2$ sample $N\approx 0.16$. The total oscillator frequency shift is then given by $\Delta f = GV_T\chi_{Polygon}/(1-N)$, where G is a calibration factor related to the coil geometry. G was obtained by inserting a superconducting indium sphere into the coil at base temperature and measuring Δf_{indium}. $N=1/3$ for a sphere and if the sphere radius is much larger than the indium penetration depth, then $\Delta f_{indium}=3GV_{sphere}/2$, from which G was obtained. $\lambda_\perp$ was then obtained by inverting the relation $\Delta f_{sample} = GV_T\chi_{Polygon}/(1-N)$ for each temperature. A full inversion procedure was required because the factors $L_i/2\lambda_\perp$ in Eq. (3) could *not* be assumed to be $\gg 1$.

III. PENETRATION DEPTH AND SUPERFLUID DENSITY

The values of $\lambda_\perp(0)$ for both κ-$(ET)_2Cu[N(CN)_2]Br$ and κ-$(ET)_2Cu(NCS)_2$ are listed in Table I, along with other reported values. The error bars for the κ-$(ET)_2Cu[N(CN)_2]Br$ measurement are dominated by repeatability in the total frequency shift upon inserting and removing the sample *in situ* since the sample had a small volume. The errors in the κ-$(ET)_2Cu(NCS)_2$ data represent the spread of values obtained from measuring the sample in the two field orientations. As the table shows, there is a considerable disparity between the various measurements, testifying to both the difficulty of measuring $\lambda_\perp(0)$ accurately and to sample-to-sample dependence. Our previously reported measurement on a much thicker sample of κ-$(ET)_2Cu[N(CN)_2]Br$ gave $\lambda_\perp(0)=100\pm 20$ μm.[9]

In clean-limit London superconductors, the behavior of the *in-plane* penetration depth $\lambda_\parallel$ directly reflects the momentum dependence of gap function. This is true for temperatures below roughly $T_C/3$, where the gap function is essentially constant with temperature. A linear temperature

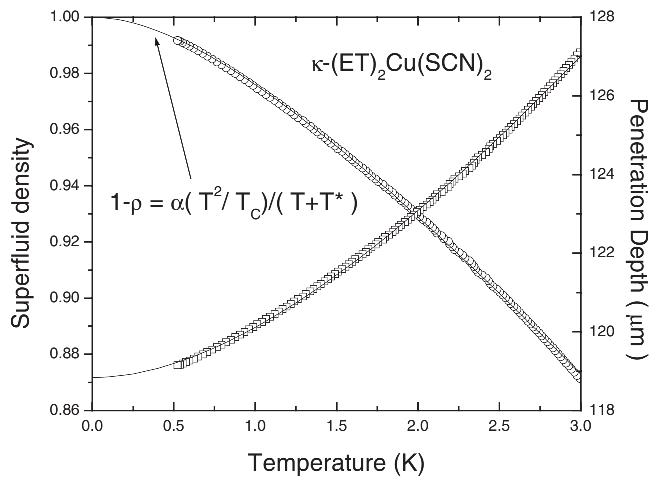

FIG. 2. $\rho_\perp$ and $\lambda_\perp$ for κ-$(ET)_2Cu(NCS)_2$ with fits to Eq. (5).

dependence of $\lambda_\parallel$ indicates an order parameter with line nodes, as widely observed in the copper oxides.[30,31] Unitary limit impurity scattering transforms the linear dependence into T^2 at the lowest temperatures. The net result is an overall dependence that is well described by the empirical form $\lambda_\parallel = 1 + AT^2/(T+T^*)$, where T^* is an impurity crossover temperature.[32] This functional form is often referred to as a "dirty d-wave" fit. Previous measurements on both κ-$(ET)_2Cu[N(CN)_2]Br$ and κ-$(ET)_2Cu(NCS)_2$ showed exactly this behavior with $T^* \approx 0.6$ K.[9] To our knowledge, there is no theoretical justification for this "dirty d-wave" form for $\lambda_\perp(T)$. Nevertheless, we find that it fits our data extremely well.

In Fig. 2 we show data for κ-$(ET)_2Cu(NCS)_2$ for one of the two field orientations measured. Fits for κ-$(ET)_2Cu[N(CN)_2]Br$ were of comparable quality. For this orientation, we found $\lambda_\perp(0) = 119$ μm. Both $\lambda_\perp$ and the normalized superfluid density $\rho_\perp = [\lambda_\perp(0)/\lambda_\perp(T)]^2$ are plotted along with fits to the dirty d-wave form for $T < 3$ K,

$$\lambda_\perp = 1 + AT^2/(T+T^*), \quad 1 - \rho_\perp = \alpha(T^2/T_C)/(T+T^*). \qquad (5)$$

Figure 3 shows the same data fit to a pure power law,

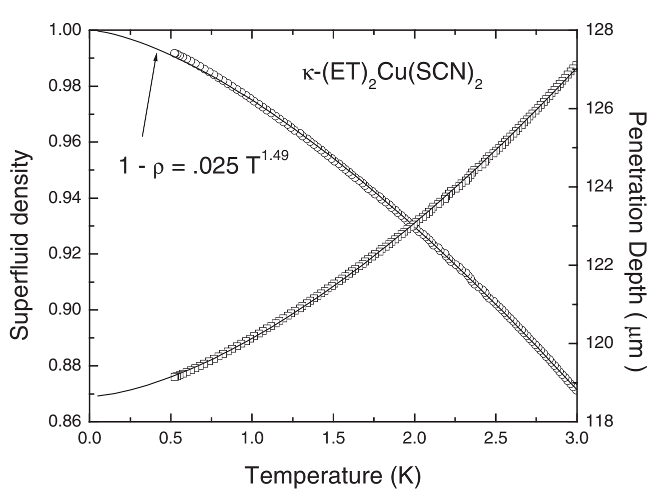

FIG. 3. $\rho_\perp$ and $\lambda_\perp$ for κ-$(ET)_2Cu(NCS)_2$ with fits to Eq. (6).

TABLE II. Parameters for fits to $1-\rho_\perp = bT^n$ and $1-\rho_\perp = \alpha(T^2/T_C)/(T+T^*)$ for both κ-$(ET)_2Cu[N(CN)_2]Br$ and κ-$(ET)_2Cu(NCS)_2$.

Material	n	α	T^* (K)	Reference
κ-$(ET)_2Cu[N(CN)_2]Br$	$1.5 \pm .05$	0.72	3.6 ± 0.1	This work
κ-$(ET)_2Cu[N(CN)_2]Br$	1.2 ± 0.1	0.42	0.5	9
κ-$(ET)_2Cu[N(CN)_2]Br$	2			10
κ-$(ET)_2Cu(NCS)_2$	1.4 ± 0.1	0.73	1.9 ± 0.1	This work

$$\lambda_\perp = \lambda_\perp(0) + aT^{1.58}, \quad 1 - \rho_\perp = bT^{1.49}. \qquad (6)$$

In Ref. 9 it was shown that the data for $\rho_\parallel$ could also be fit to both forms with essentially the same degree of precision.

Table II summarizes the results for $\rho_\perp$. For κ-$(ET)_2Cu[N(CN)_2]Br$, the error came from uncertainty in our determination of $\lambda_\perp(0)$ described earlier. Nonetheless, the final parameters were largely insensitive to this quantity. For κ-$(ET)_2Cu(NCS)_2$, the errors came predominantly from differences in the two field orientations measured since the demagnetization factors were larger. Overall, we found that $\rho_\perp$ data for both samples could be accurately fit to power laws with relatively small exponents $n = 1.5 \pm 0.05$ ($\kappa-(ET)_2Cu[N(CN)_2]Br$) and $n = 1.4 \pm 0.1$ [κ-$(ET)_2Cu(NCS)_2$]. Alternatively, choosing the dirty d-wave expression, we found $T^* = 3.6$ K for κ-$(ET)_2Cu[N(CN)_2]Br$ and $T^* = 1.9$ K for κ-$(ET)_2Cu(NCS)_2$. These values should be compared to our earlier values of $T^* = 0.6$ K for $\rho_\parallel$.[9] Printeric *et al.*[10] also measured $\rho_\perp$ in κ-$(ET)_2Cu[N(CN)_2]Br$ and reported an exponent $n \approx 2$.[26] Their data do not go below $T = 1.6$ K and shows rather more scatter than our own.

IV. DISCUSSION

A model for $\rho_\perp(T)$ requires assumptions about the transport channels that participate in the supercurrent. These channels may include wave-function overlap, impurity scattering, phonon-assisted hopping, and resonant tunneling processes. In one limit, the system is considered a stack of superconductor-insulator-superconductor (SIS) junctions, in which interlayer transport occurs through incoherent processes such as impurity or bosonic scattering. Graf. *et al.* studied this "interlayer diffusion" model for both s- and d-wave pairings.[33] A standard relationship between $\lambda_\perp(0)$ and the critical current $J_c^\perp(0)$ holds, independent of the pairing symmetry,

$$\lambda_\perp(0) = \sqrt{\frac{\hbar}{2e\mu_0 d J_c(0)}} \quad (SI). \qquad (7)$$

For the organics, the spacing between conducting layers $d = 1.5$ nm. Using the values in Table II, we obtain $J_c(0) \approx 10^3$ Amp/cm^2 for both materials. Within this model, the normal-state interlayer resistivity $\rho_n^\perp$ is related to the maximum energy gap Δ_0 and $\lambda_\perp(0)$,[33]

$$\frac{4\pi^2}{\hbar c^2}\frac{\Delta_0 \lambda_\perp^2}{\rho_n^\perp} = \begin{cases} 1 & (s \text{ wave}) \\ R_d \geq 1 & (d \text{ wave}). \end{cases} \tag{8}$$

If the scattering is isotropic then $R_d \to \infty$ and so $\lambda_\perp \to \infty$, indicating the lack of Josephson screening currents due to averaging over the d-wave order parameter in momentum space. Josephson screening does appear if the scattering is anisotropic, in which case R_d is finite. Recent specific-heat measurements yield a d-wave gap $\Delta_0 = 2.14\beta k_B T_C$ with a strong-coupling enhancements of $\beta = 1.73$ [κ-$(ET)_2Cu[N(CN)_2]Br$] and $\beta = 1.45$ [κ-$(ET)_2Cu(NCS)_2$].[17] Taking these values, together with $\rho_n^\perp \approx 1\ \Omega-\text{cm}$,[34] we obtain $R_d \approx 30$. This value would imply a d-wave state with isotropic interlayer scattering, but again, this conclusion is based on an incoherent transport model. We note that Eqs. (7) and (8) lead to a generalized Ambegaokar-Baratoff relation,[35]

$$\frac{2e}{\pi \Delta_0}\rho_n^\perp J_c^\perp d = \begin{cases} 1 & (s \text{ wave}) \\ R_d^{-1} & (d \text{ wave}). \end{cases} \tag{9}$$

For an s-wave superconductor, the Ambegaokar-Baratoff model leads to the temperature dependence,

$$\left(\frac{\lambda_\perp(0)}{\lambda_\perp(T)}\right)^2 = \rho_\perp = \frac{\Delta(T)}{\Delta_0}\tanh\frac{\Delta(T)}{2k_B T}. \tag{10}$$

Our data cannot be fit to this form, even approximately. However, this picture was generalized by Maki and Haas to a d-wave gap function.[36] They found that $1-\rho_\perp \sim T^2$, in reasonable agreement with microwave data on underdoped Bi-2212,[37] susceptibility measurements on aligned powders of $HgBa_2Ca_2Cu_3O_{8+\delta}$,[38] and measurements on κ-$(ET)_2Cu[N(CN)_2]Br$ by Printeric *et al.*[10] Using a somewhat different model for diffusive interlayer transport in a d-wave superconductor, Hirschfeld *et al.* predicted $1-\rho_\perp \sim T^3$.[39] Xiang and Wheatley[40] showed that anisotropy in the transfer integrals can lead to $1-\rho_\perp \sim T^5$, which was observed in aligned powders of $HgBa_2CuO_{4+\delta}$.[38] Atkinson and Carbotte calculated the $\rho_\parallel$ and $\rho_\perp$ for proximity-coupled layers superconductor-normal-superconductor for both s- and d-wave pairing.[41] They show that the superfluid density develops *upward* curvature below some characteristic temperature, where the proximity coupling sets in. Upward curvature in the superfluid density appears to be a general characteristic of proximity coupling and was observed previously in Mg-coated MgB_2.[42] However, Figs. 2 and 3 show no upward curvature so the proximity model for $\rho_\perp$ is not applicable to the organics. The fact that our data accurately obey $1-\rho_\perp \sim T^n$ with $n < 1.5$ appears to rule out all of the above models.

Radtke *et al.* included several contributions to the transport in their calculation of $\rho_\perp$ for a d-wave superconductor.[43] In the limit of purely coherent transport (wave-function overlap), they showed that $\rho_\perp$, $\rho_\parallel$ should have the same temperature dependence. Sheehy *et al.* developed an alternative theory for $\rho_\perp$ in a d-wave superconductor.[44] In their model, applied to underdoped yttrium-barium-copper-oxide (YBCO), nodal quasiparticles with energies beyond a doping-dependent scale E_C are given reduced weight in determining the superfluid density. Depending upon the relative

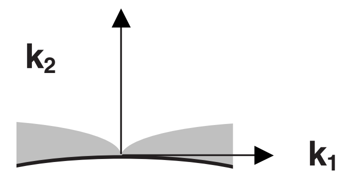

FIG. 4. Wave-vector components near a nodal point on the Fermi surface in a d-wave superconductor.

magnitudes of $k_B T$, Δ_0, and E_C, the power law for $\rho_\perp$ may vary from T to T^3.[44] Qualitatively, the various regimes may be visualized using the quasiparticle wave-vector components $\mathbf{k_1}$, $\mathbf{k_2}$ defined parallel and perpendicular to the Fermi surface at a nodal point, as shown in Fig. 4.

The regime of coherent transport corresponds to both $\mathbf{k_1}$ and $\mathbf{k_2}$ preserved during interlayer hopping. In an impurity-free d-wave superconductors, this would lead to $1-\rho_\perp \sim T$. The intermediate regime corresponds to the conservation of $\mathbf{k_1}$ but not of $\mathbf{k_2}$. This situation leads to $1-\rho_\perp \sim T^2$. When neither component is preserved, one has $1-\rho_\perp \sim T^3$. Hoessini *et al.* reported $1-\rho_\perp \sim T^{2-2.5}$ in samples of extremely underdoped YBCO.[45] This result signified the presence of nodal quasiparticles and incoherent transport between layers. For the κ-$(ET)_2X$ organics studied here, $1-\rho_\perp \sim T^{1.3-1.5}$, which implies the existence of nodal quasiparticles but—in contrast to the copper oxides—an interlayer transport mechanism that is close to *coherent*, despite strong anisotropy ($\lambda_\perp / \lambda_\parallel > 100$).

Probably the clearest indication of coherent interlayer transport in the normal state is the observation of a peak in the angular dependence of the magnetoresistance[46] of κ-$(ET)_2Cu(NCS)_2$ by Singleton *et al.*[19] Those authors noted that coherence is maintained despite an extremely small transfer integral $t_\perp \approx 0.04$ meV, which leads to a violation of the usual condition for coherent transport $t_\perp > \hbar/\tau$, where τ is the in-plane scattering time. In addition, the interlayer resistivity $\rho_n^\perp \approx 1\ \Omega-\text{cm}$ is many orders of magnitude higher than that of ordinary metals, although its temperature dependence is metallic. Therefore, one would naively expect these materials to display incoherent transport, And, in fact, infrared measurements by McGuire *et al.* of the interlayer conductivity in κ-$(ET)_2Cu[N(CN)_2]Br$ exhibited no Drude peak.[47] The authors concluded that the transport was indeed incoherent and speculated that the lower-frequency measurements were dominated by defects that formed interlayer short circuits. However, it should be noted that the electronic behavior of κ-$(ET)_2Cu[N(CN)_2]Br$ [but not κ-$(ET)_2Cu(NCS)_2$] is highly sensitive to the rate of cooling. Rapid cooling through 80 K leads to partial phase separation,[48–50] which may account for some of the discrepancies between infrared and low-frequency measurements. Finally, recent work by Gutman and Maslov provided new insight into the conduction mechanisms at work.[51,52] They noted that in κ-$(ET)_2Cu(NCS)_2$, Sr_2RuO_4, and several other quasi-2D metals, the interlayer resistivity versus temperature passes through a maximum, below which it shows metallic temperature dependence. In κ-$(ET)_2Cu(NCS)_2$, this maximum occurs near 80 K. In their model, there are two parallel interlayer conductance channels. The first, via normal interlayer hopping, leads to bandlike metallic contribution. The second consists of phonon-assisted tunneling through resonant defects located between conducting planes. This channel gives an insulatorlike temperature dependence. The resistivity maximum represents a competition between the two processes. Their model also predicts a non-Drude frequency dependence in some limits, though it is not clear whether it can reconcile the infrared data with the magnetoresistance and penetration depth measurements.

V. SUMMARY

We have measured the interlayer penetration depth $\lambda_\perp$ in both κ-$(ET)_2Cu[N(CN)_2]Br$ and κ-$(ET)_2Cu(NCS)_2$, two closely related quasi-2D organic superconductors. We find that $\lambda_\perp(0) \sim 130$ μm for both materials. For $T/T_C < 0.3$, the temperature dependence of the interlayer superfluid density $\rho_\perp$ may be fit equally well to a power law $1-\rho_\perp \sim T^{1.3-1.5}$ or to the form $1-\rho_\perp = \alpha(T^2/T_C)/(T+T^*)$ widely used for the in-plane superfluid density in a d-wave superconductor with impurity scattering. Our observations imply that the energy gap is nodal, consistent with d-wave pairing. The relatively low power-law exponent ($n=1.3$–1.5) shows that the interlayer transport is close to coherent, in agreement with magnetoresistance measurements. The appearance of coherent transport differs from the case of copper oxides, in which a power-law exponent of $n=2$–2.5 has been taken as evidence for incoherent transport. This finding may be relevant to theories, in which interlayer coupling and two dimensionality play a central role in determining the superconducting transition temperature.[53,54]

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