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# Magnetic penetration depth of $\kappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br, determined from the reversible magnetization

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We present results of the dc magnetization of single-crystalline $\kappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br. From the slope of the linear $M(\ln H)$ dependence of the reversible magnetization, the magnetic penetration depth $\lambda(T)$ is estimated. At low temperatures, $\lambda(T)$ shows only a weak $T$ dependence, consistent with conventional pairing. For the in-plane London penetration depth we find $\lambda_L(0)=(650\pm20)$ nm. Small deviations from a pure BCS behavior indicate some tendency towards intermediate or strong-coupling and/or a somewhat reduced carrier mean free path.

Organic charge transfer salts of the (BEDT-TTF)$_2X$ family [BEDT-TTF denotes bis(ethylenedithio)tetrathiafulvalene and $X$ a monovalent anion] attract wide interest due to their exotic normal-state properties and high superconducting transition temperatures ($T_c$). To date, $\kappa$-(BEDT-TTF)$_2X$ has the highest ambient-pressure $T_c$ of 11.5 K for the anion $X=[\mathrm{Cu[N(CN)_2]Br}]^{1-}$ (Ref. 1) followed by $X=[\mathrm{Cu(NCS)_2}]^{1-}$ with $T_c$ somewhat below 10 K.[2] Orientational-dependent transport[3,4] and critical-field[5] studies reveal the strong two-dimensional electronic character of these salts following their layered crystal structure. In view of their low dimensionality along with the small charge carrier concentration (both give rise to strong Coulomb correlations), these salts have been considered as candidates for unconventional superconductivity.[6] Such a state is generally characterized by an anisotropic order parameter with a symmetry lower than the symmetry of the crystal structure. An experimental check, suitable to trace out deviations from a conventional coupling type, is provided by a careful study of the temperature dependence of the magnetic penetration depth. For the simplest unconventional states, where the energy gap vanishes at points or along lines on the Fermi surface, a power-law dependence is proposed for $\lambda(T)$ at low temperatures,[7] which contrasts the exponential behavior predicted by the Bardeen-Cooper-Schrieffer (BCS) theory.[8] For the organic superconductors $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$ and $\kappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br conflicting results have been reported for $\lambda(T)$ and related quantities: while some experiments showed conventional behavior,[9,10] others revealed deviations from the BCS predictions and were interpreted as evidence for unconventional superconductivity.[11,12] Recently, we determined the magnetic penetration depth of high-quality single crystals of $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$ from the analysis of the reversible magnetization. This method provides a most reliable estimate of $\lambda(T)$ for a type-II superconductor, since it assures a pinning-free and hence homogeneous field penetration with negligible effects of demagnetization and earth magnetic field. The resulting $\lambda(T)$ was found to be in excellent agreement with the BCS prediction.[13]

Here, we report on the analogous estimate of $\lambda(T)$ for $\kappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br. According to London's phenomenological model,[14] the reversible magnetization for a type-II superconductor with a large Ginzburg-Landau parameter, $\kappa_{\mathrm{GL}}\gg1$, is linear in the logarithm of the applied field for intermediate fields, $H_{c1}\ll H\ll H_{c2}$ ($H_{c1}$ and $H_{c2}$ are the lower and upper critical fields, respectively):

$$-4\pi M=\frac{\phi_0}{8\pi\lambda^2}x\ln\left[\frac{H_{c2}\beta}{H}\right], \quad (1)$$

where $\phi_0$ is the flux quantum and $\beta$ a constant of order unity. For an anisotropic uniaxial superconductor, the slope of the linear $M(\ln H)$ dependence is given by

$$\frac{dM}{d(\ln H)}=\frac{\phi_0}{32\pi^2\lambda_{\mathrm{eff}}^2}, \quad (2)$$

where $\lambda_{\mathrm{eff}}^2=\lambda_{\mathrm{plane}}^2$ ($H\perp$plane) and $\lambda_{\mathrm{eff}}^2=\lambda_{\mathrm{plane}}\times\lambda_{\perp\,\mathrm{plane}}$ ($H\parallel$plane).[15] Since the anisotropy between the directions perpendicular and parallel to the plane of highest conductivity (ac plane) is much larger than a small in-plane anisotropy, this system can be considered as quasiuniaxial and Eq. (2) applies.

The $\kappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br single crystals were grown by an electrocrystallization method as described elsewhere.[4] A platelike single crystal with dimensions of $1.2\times1.8\times0.6$ mm$^3$, and a weight of 2.48 mg was selected. The magnetization measurements were carried out with a commercial superconducting quantum interference device (SQUID) magnetometer, where fields up to 5 T were applied perpendicular to the ac plane. Figure 1 shows the temperature dependence of the magnetization near the superconducting transition, measured upon cooling in a field of $H=20$ G. An extrapolation of the linear $M(T)$ behavior slightly below $T_c$ back to $M=0$ yields a nucleation temperature of 11.5 K$\pm$0.1 K. Figure 2 shows typical isotherms of the magnetization on a logarithmic field scale. A diamagnetic core contribution of $\chi^{\mathrm{core}}=-4.06\times10^{-4}$ emu/mol, determined from normal state $M(T)$ measurements at various fixed fields, had been subtracted. At each temperature, the magnetization was measured upon increasing and decreasing fields to

 

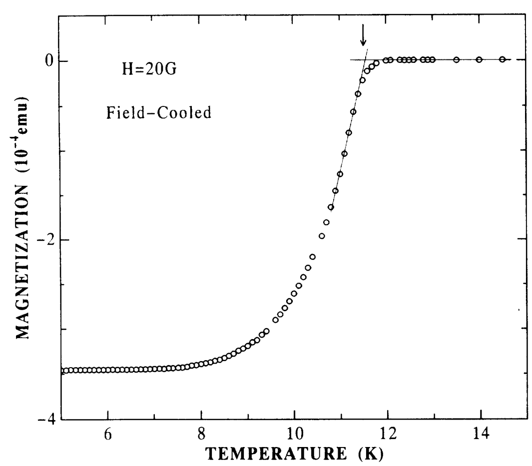


FIG. 1. Magnetization of $\kappa$-(BEDT-TTF)$_2$[N(CN)$_2$]Br measured upon cooling in a field of $H = 20$ G (H$\perp$$a$-$c$ plane).

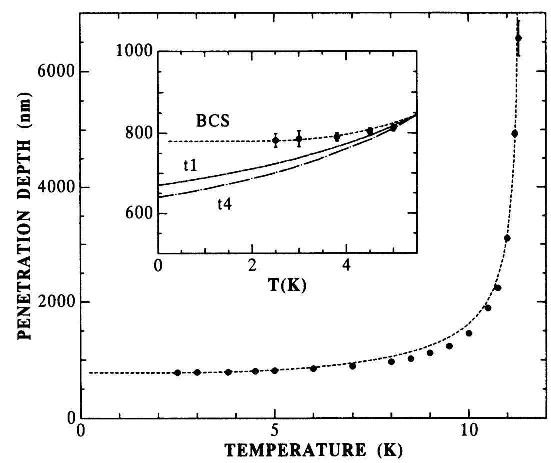


FIG. 3. In-plane penetration depth of $\kappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br. The dashed line represents a BCS temperature dependence. Inset: low-temperature data along with model curves for a BCS (- - -) superconductor and two triplet states, $t_1$ and $t_4$, taken from Ref. 12.

check for reversibility. For a wide field and temperature range, we found an entirely reversible $M(T)$ behavior indicating the formation of a pinning-free homogeneous vortex lattice. In accordance with the London model, the magnetization for intermediate fields follows the linear $M(\ln H)$ dependence. Employing the slopes, $dM/d(\ln H)$, and using Eq. (2), we can determine the in-plane penetration depth $\lambda_{ac}$. The resulting $T$ dependence is shown in Fig. 3. At low temperatures, i.e., for 2.5 K $\leq T \leq 5.5$ K (cf. inset), the data reveal only a weak variation upon decreasing temperature with the tendency to saturate for $T \rightarrow 0$. As indicated by the short-dashed line, this behavior is consistent with the exponential $T$ dependence expected for a BCS superconductor.[16] In contrast, for an unconventional order parameter with gap zeros along certain symmetry directions a stronger $T$ dependence (power law) of $\lambda(T)$ can be expected, due to the finite number of excited states close to the gap zeros.[7] Also shown in the inset of Fig. 3 are model calculations for the in-plane penetration depth used by Le *et al.*[12] to analyze their low-temperature muon-spin-rotation ($\mu$SR) data. The authors compare different unconventional coupling types, proposed by Hasegawa and Fukuyama for a superconducting state in a quasi-two-dimensional system.[6] The two curves shown correspond to the spin-triplet states $t_1$ and $t_4$ of Ref. 12, which represent the models with the weakest and strongest $T$ dependences, respectively. In order to compare with our data, the curves were normalized at $T = 5.5$ K, where the slopes come close to that of the BCS curve. Figure 3 clearly demonstrates that our experimental data are incompatible with any of these unconventional pairing schemes.

At somewhat higher temperatures, $\lambda(T)$ shows small deviations from a pure BCS behavior. This becomes particularly clear when the quantity $[\lambda(0)/\lambda(T)]^2$ is plotted against reduced temperature, $t = T/T_c$, as displayed in Fig. 4. The parameters $T_c(0) = 11.4$ K and $\lambda(0) = 780$ nm were determined as follows: close to $T_c$ $\lambda(T)$ follows

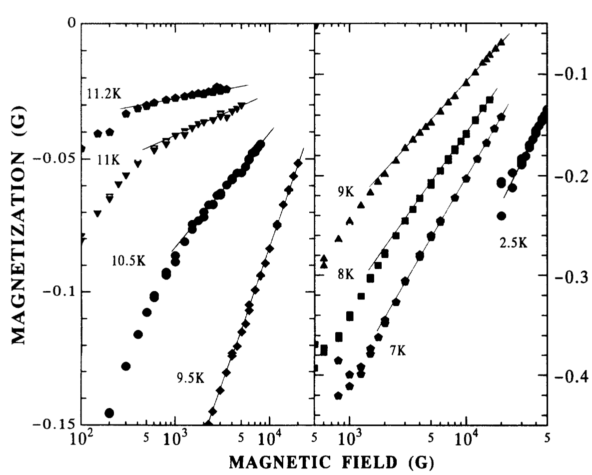


FIG. 2. Isothermal magnetization curves vs $\ln H$ (H$\perp$$a$-$c$ plane). Straight lines are guides to the eye. A factor of 801 was used to transform the magnetization data in units of Gauss.

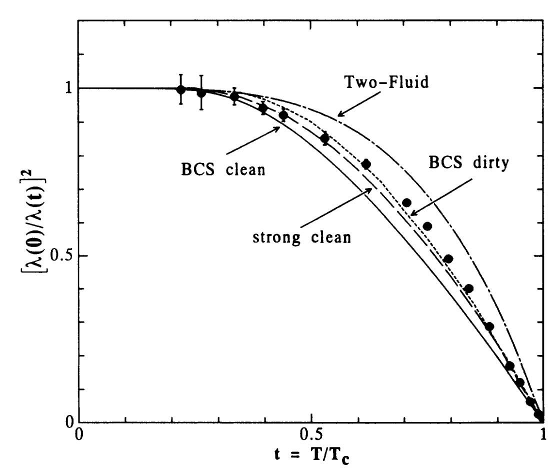


FIG. 4. Normalized penetration depth vs reduced temperature. Curves correspond to BCS prediction for clean (——) and dirty (- - -) limit, the two-fluid model (—·—·—·) and a model calculation for a strong-coupling superconductor (— — —) taken from Ref. 19.

the general and model-independent Ginzburg-Landau behavior, i.e., $\lambda^{-2} \propto (T - T_c)$, which fixes $T_c$. $\lambda(0)$ was obtained by an extrapolation to $T=0$ (cf. the inset of Fig. 3). Also shown are theoretical curves for a BCS superconductor in the clean[16] and dirty[17] limits and the empirical two-fluid expression, $[\lambda(0)/\lambda(t)]^2 = (1-t^4)$,[18] as well as a model curve for a strong-coupling superconductor, implying an energy gap $2\Delta_0 = 3.88 \times k_B T_c$.[19] Both a reduction of the carrier mean free path as well as an enhanced coupling strength cause a flattening of $\lambda(T)$ down to low temperatures and could, therefore, account for the deviations from the BCS clean-limit curve. A somewhat reduced carrier mean free path for the present salt is consistent with the fact that, so far, no quantum oscillations have been observed even for the extreme parameters $T=0.5$ K and $B \leq 28$ T,[20] while for $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$, clear oscillations show up above $B=10$ T at this temperature.[21] However, since both salts show a residual-resistivity ratio of comparable size,[20] it appears most likely that the deviations from the BCS behavior are mainly due to a somewhat enhanced coupling strength. In fact, some tendency towards intermediate or strong coupling has been inferred from specific-heat measurements, yielding a discontinuity at $T_c$ of $\Delta C = (2 \pm 0.5) \times \gamma T_c$ ($\gamma$ is the Sommerfeld coefficient).[22] However, a similar value was reported for $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$,[23] where our $\lambda(T)$ data gave no indication for deviations from weak coupling.[13]

The main result of the present study is that the magnetic penetration depth determined from the reversible magnetization shows only a weak variation in the low-temperature range down to the lowest temperature of our experiment of $T=2.5$ K ($t=0.22$). This result is in clear contrast to $\lambda(T)$ evaluated from complex susceptibility measurements[11] and low-temperature $\mu$SR studies.[12] As we have already pointed out,[13,25] we think that the deviations from the BCS behavior in these experiments originate in flux-pinning-related phenomena: at $T=2.5$ K for example, our experiment reveals an irreversible magnetization for fields $H < H_{\mathrm{irr}}(2.5\ \mathrm{K}) = 25$ kG (cf. Fig. 2). At lower fields, pinning of vortices becomes increasingly important and prevents the formation of a homogeneous vortex structure in this strongly anisotropic superconductor. Hence, for measurements carried out at lower fields and lower temperatures, pinning-induced inhomogeneities of the local-field distribution can be expected. This is also true for a field-cooled experiment, as long as the field is far below the irreversible field, $H_{\mathrm{irr}}$. In light of this, the low-temperature data of the $\mu$SR study of Ref. 12, performed at $H=3$ kG, appear questionable. Pinning related phenomena might also have affected the complex susceptibility experiments of Ref. 11: for platelike crystals (as used there), demagnetization for $H \perp$plane strongly reduces the effective lower critical field, $H_{c1}^{\mathrm{eff}} = (1-N) \times H_{c1}$ (demagnetization factor, $N \approx 1$, for this configuration). Since for both salts $X = [\mathrm{Cu(NCS)_2}]^{1-}$ and $X = [\mathrm{Cu[N(CN)_2]Br}]^{1-}$, $H_{c1\perp}$ is of the order of 20 G,[13] both $H_{c1\perp}^{\mathrm{eff}}$ and $H_{c1\parallel}^{\mathrm{eff}}$ (where $H_{c1\parallel}$ is of the order of a few Gauss and $N < 1$) became comparable to the earth magnetic field, $H_E$ (not compensated in their experiments). It is well known that in an ac-susceptibility experiment under these conditions, the frozen-in remnants of $H_E$ flux can strongly affect the estimate of both the temperature dependence and the magnitude of $\lambda(T)$.[25,26]

Assuming an exponential (BCS) $T$ dependence of $\lambda(T \to 0)$ we find $\lambda(0) = (780 \pm 20)$ nm. According to a recent work by Hao and Clem,[24] this value is related to the London penetration depth by $\lambda_L(0) = \sqrt{\alpha} \times \lambda(0)$, where the correction factor $\alpha$ accounts for the core contribution neglected in the London expression, Eq. (2) ($\alpha = 0.7$ for the field range used in our experiment). The resulting in-plane London penetration depth $\lambda_{ac}(0) = (650 \pm 20)$ nm is of comparable size to the in-plane penetration depth $\lambda_{bc}(0) = (535 \pm 20)$ nm found for $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$.[13]

Using clean-limit formulas[27] and employing $\lambda_L(0)$ and the Ginzburg-Landau coherence length $\xi_{\mathrm{GL}} = (3.4 \pm 0.4)$ nm,[28] we can estimate the Ginzburg-Landau parameter $\kappa = 0.96 \times \lambda_L \times \xi_0^{-1} = 136 \pm 20$, where the BCS coherence length $\xi_0$ is related to $\xi_{\mathrm{GL}}$ by $\xi_{\mathrm{GL}}(0) = 0.74 \times \xi_0$. For the lower critical field $H_{c1}$ and the thermodynamical critical field $H_c$, defined as $H_{c1} = \phi_0 \times \ln\kappa \times [4\pi\lambda_L^2(0)]^{-1}$ and $H_c = \sqrt{24} \times H_{c1} \times \kappa \times (\pi \ln\kappa)^{-1}$, respectively, we find $H_{c1}(0) = (19 \pm 2)$ G and $H_c = (820 \pm 200)$ G.

In conclusion, the magnetic in-plane penetration depth of single-crystalline $\kappa$-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br determined from the reversible magnetization shows only a weak $T$ dependence at low temperatures, consistent with a conventional order parameter. The same conclusion has been drawn from the analogous analysis for $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$, where an excellent agreement with the BCS prediction was found over the whole temperature range investigated.[13] These results exclude the unconventional pairing schemes proposed by Kanoda *et al.*[11] and Le *et al.*[12] From our results, there is no need to invoke any unconventional pairing schemes to explain superconductivity in these materials. The small deviations from a clean-limit BCS behavior in the intermediate temperature range for the present salt point to some tendency towards intermediate or strong coupling and/or a somewhat reduced carrier mean free path.

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[27]A somewhat reduced carrier mean free path, $l$, for the present salt compared to $X=$ [$Cu(NCS)_2$]$^{1-}$ [where $\xi_{GL}/l \approx 0.1$ (Ref. 13)] still satisfies $\xi_{GL}/l < 1$.

[28]For an $s$-wave superconductor, the initial slope of the upper critical field, $H'_{c2}$, is related to the Ginzburg-Landau coherence length by $H'_{c2} = \phi_0 \times (2\pi)^{-1} \times \xi_{GL}^{-2} \times T_c^{-1}$. We find $\xi_{GL} = (3.4 \pm 0.4)$ nm in agreement with Ref. 5. These values are consistent with $\xi_{GL}$ of about 40 nm as derived from the analysis of resistive transitions measured in various fields taking fluctuation effects into account (Ref. 4).