

In-plane and out-of-plane superfluid density of the layered organic superconductor κ -(BEDT-TTF)₂Cu[N(CN)₂]Br

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Abstract

The anisotropic magnetic penetration depth $\lambda_b(t)$ and $\lambda_{ac}(t)$, where $t = T/T_C$, in the layered organic superconductor κ -(BEDT-TTF)₂Cu[N(CN)₂]Br (abbreviated as κ -ET-Br) are obtained from AC susceptibility data. We find $\lambda_b(0) = 133\mu\text{m}$, consistent with earlier results and $\Delta\lambda_b(t) = \lambda_b(t) - \lambda_b(0) \propto t^2$, which is new. Also, assuming $\lambda_{ac}(0) \approx 1\mu\text{m}$, we obtain $\Delta\lambda_{ac}(t) \propto t$. The observed power laws are consistent with the ones expected in d -wave superconductivity.

The discovery of high- T_C cuprate superconductors in 1986 has been largely acknowledged to usher the scientific community into a new era in which the room temperature superconductivity and many technical applications of superconductivity might become possible. Less appreciated has been the realization of d -wave superconductivity in hole-doped high- T_C cuprates [1]. One of us has already suggested that most of newly discovered superconductors are unconventional (*i.e.* non- s -wave) and of electronic (*i.e.* non-phononic) origin [2]. Therefore, it is highly desirable to identify the underlying symmetry of the novel superconductors. In this perspective, the symmetry of the organic superconductivity in κ -(BEDT-TTF)₂X salts (abbreviated κ -ET) has been hotly debated in the last ten years. Now, it appears that the consensus is settling towards d -wave superconductivity, though the decisive proof is still missing [3].

There are striking parallels between high- T_C cuprates and κ -ET salts. First is the crystallographic (2D or layered) structure, and the second is that the superconducting state in both materials is located in the proximity to an antiferromagnetic phase revealing the dominance of the Coulomb interactions. As far as the Fermi surface is concerned in, for example, Bi2212 and κ -ET salts, the shape appears to be quite different at first sight. However, they are identical topologically if one rotates the one for the latter system by $\pi/4$ (Fig. 1) [4]. This fact naturally suggests d_{xy} -wave for κ -ET salts, while $d_{x^2-y^2}$ -wave for high- T_C cuprates. Indeed, the standard model based on Coulomb dominance and/or the antiparamagnon exchange gives d_{xy} -wave superconductivity in κ -ET salts [5–8]. Further, the T^3 dependence of the spin-lattice relaxation rate found in ¹³C NMR measurements [9], the T^2 dependence of the electronic specific heat [10], the T linear planar thermal conductivity [11] all imply the nodal structure in $\Delta(\vec{k})$ of κ -ET as in d -wave superconductor.

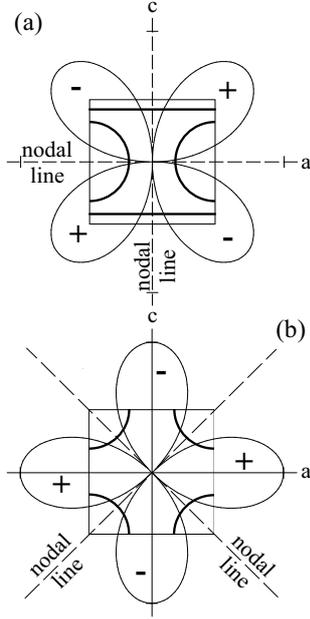


Fig. 1. Theoretically predicted 2D Fermi surface in (a) κ -ET-Br and (b) Bi2212.

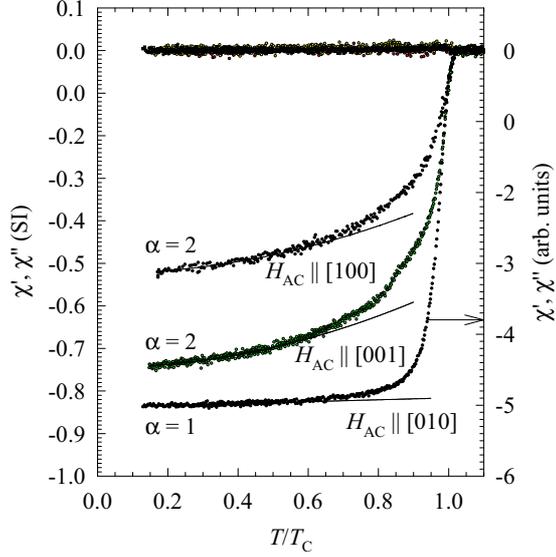


Fig. 2. Real and imaginary part of the complex susceptibility in an AC field of 14 mOe parallel to the crystal ac plane ($H_{AC} \parallel [100]$ and $H_{AC} \parallel [001]$) and perpendicular to the ac plane ($H_{AC} \parallel [010]$).

Finally, recent magnetic penetration depth measurements [12, 13] give $\Delta\lambda_{ac}(t) \propto t$ consistent with d -wave superconductivity. Indeed, for $T > 1.6$ K the data from [12] and [13] agree with each other semi-quantitatively. However, at lower temperatures the authors of [12] found $\Delta\lambda_{ac}(t) \propto t^{3/2}$, which is rather puzzling. Within a simple d -wave superconductor picture, it is expected that $\Delta\lambda_{ac}(t) \propto t$ is valid down to $t = 0$ K [14]. On the other hand, [12] and [13] disagree as to the temperature dependence of $\Delta\lambda_b(t)$. The former gives $\Delta\lambda_b(t) \propto t$ like $\Delta\lambda_{ac}(t)$, while the latter gives $\Delta\lambda_b(t) \propto t^2$. Of course, this issue has to be settled by new set of experiments.

In what follows, we review and discuss our experimental penetration depth data in the framework of a suitable theoretical model. All additional details can be found in [13].

$\chi(T)$ data, taken by AC susceptibility measurements at 231 Hz and at $H_{AC} = 14$ mOe (CryoBIND system of ACSuS/Sistemprojekt, Zagreb) are shown in Fig. 2. First we address the analysis of the low T behaviour for $H_{AC} \parallel$ plane. In that case circulating supercurrents flow within the ac planes and also across them. For the crystal in Fig. 2 (width $D[100] = 0.90$ mm and $D[001] = 0.54$ mm, thickness $b = 0.33$ mm), the former can be neglected since the condition $\lambda_b/\lambda_{ac} \gg D/b$ is easily satisfied by all estimates of the anisotropy, obtained by different techniques [15–17]. λ_b and λ_{ac} are the penetration depths associated with interlayer currents and intralayer currents, respectively. Then we can use the formula for a thin superconducting plate in a parallel field

$$1 + \chi' = \frac{2\lambda}{D} \tanh\left(\frac{D}{2\lambda}\right) \quad (1)$$

to get λ_b . D is the sample width in the direction of field penetration. The fact that the χ' response of our sample is different for $H_{AC} \parallel [100]$ and for $H_{AC} \parallel [001]$ (see Fig. 2) confirms that the field penetration along the D direction, and not the one along the b direction which

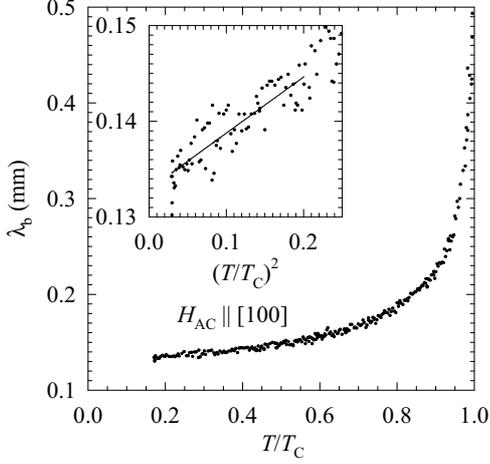


Fig. 3. Penetration depth as a function of reduced temperature for $H_{AC} \parallel [100]$. Inset: λ_b plotted versus $(T/T_C)^2$ below 5 K.

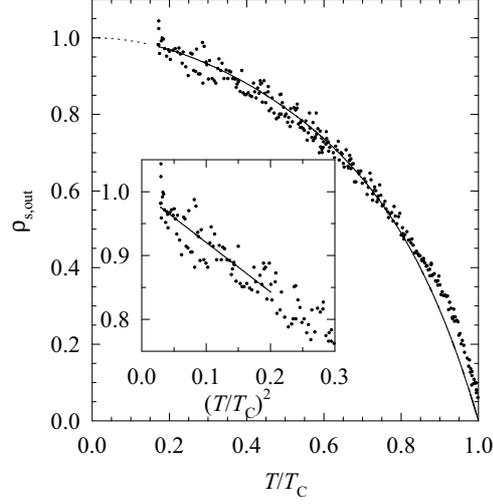


Fig. 4. Out-of-plane superfluid density $\rho_{s,out}$ as a function of reduced temperature. Full line is a fit to the theory (see Text). Inset: $\rho_{s,out}$ plotted versus $(T/T_C)^2$ below 5 K.

is associated with λ_{ac} , dominates the susceptibility of the sample.

The temperature dependence of λ_b is shown in Fig. 3. The full line corresponds to the calculated fit to the T^2 behaviour in the temperature range $1.6 \text{ K} < T < 5 \text{ K}$

$$\lambda_b = k \left(\frac{T}{T_C} \right)^2 + \lambda_0 \quad (2)$$

We get $k = 58 \pm 4 \mu\text{m}$ and $\lambda_0 = \lambda_b(0) = 132.9 \pm 0.5 \mu\text{m}$. The latter value is in a very good accordance with values for the out-of-plane penetration depth given in the literature [15].

The out-of-plane superfluid density $\rho_{s,out}$

$$\rho_{s,out} = \left(\frac{\lambda_b(0)}{\lambda_b(T)} \right)^2 \quad (3)$$

as a function of reduced temperature $t = T/T_C$ is displayed in Fig. 4. Note that the leading term which describes the low temperature behaviour of $\rho_{s,out}$ is the T^2 term (see Inset of Fig. 4).

In the perpendicular direction of the field we have determined the deviation of λ_{ac} from the minimum value at the lowest attainable temperature $T_{\min} = 1.6 \text{ K}$

$$\lambda_{ac}(T) - \lambda_{ac}(1.6 \text{ K}) = R \left[1 - \left(\frac{\chi'(T)}{\chi'(1.6 \text{ K})} \right)^{1/3} \right] \quad (4)$$

with $R = (A/\pi)^{1/2}$ where A is the area of the sample's large face. The results thus obtained from χ' are plotted in Fig. 5. Below about 5 K, there is a clear linear term with slope $3.4 \mu\text{m}$.

The observed temperature dependence of $\lambda_{ac}(T)$, when combined with the value of $\lambda_{ac}(0)$ in the widely accepted range $0.5 \mu\text{m} < \lambda_{ac}(0) < 3 \mu\text{m}$, gives

$$\rho_{s,in} = \left(\frac{\lambda_{ac}(0)}{\lambda_{ac}(T)} \right)^2 \quad (5)$$

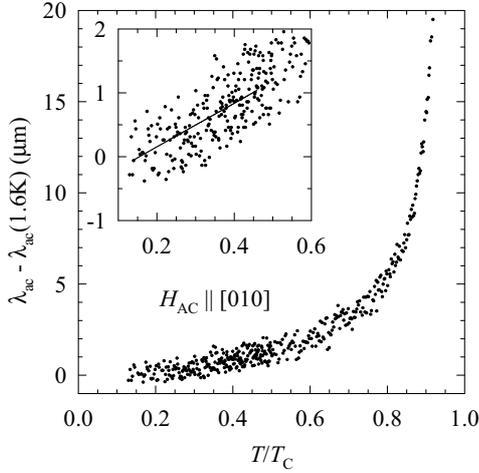


Fig. 5. Change of the penetration depth with respect to the minimum value as a function of reduced temperature for $H_{AC} \parallel [010]$. Inset: $\lambda_{ac} - \lambda_{ac}(1.6 K)$ below 5 K.

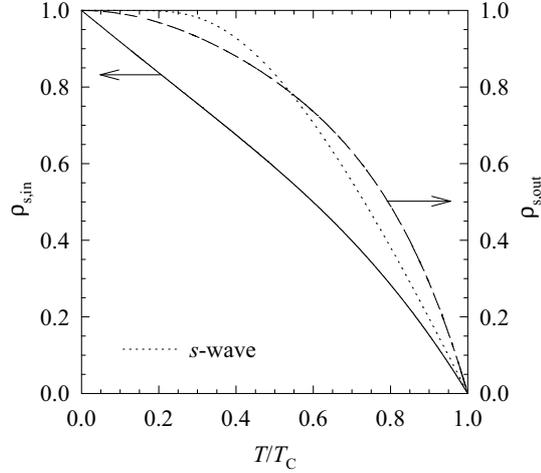


Fig. 6. In-plane superfluid density $\rho_{s,in}$ (full line) and out-of-plane superfluid density $\rho_{s,out}$ (dashed line) versus reduced temperature. The s -wave BCS result (dotted line) is shown for comparison.

which behaves linearly with T at low temperatures as expected for the d -wave superconductivity [14].

The temperature dependence of the in-plane superfluid density for the d -wave superconducting order parameter $|\Delta(\vec{k})| = \Delta f$ with $f = \sin(2\phi)$, where ϕ is the angle between the quasiparticle momentum \vec{k} and the \vec{a} axis, within the weak coupling theory is given by [14]

$$\begin{aligned} \rho_{s,in}(t) &= 1 - \frac{1}{2}(\beta\Delta) \int_0^\infty \text{Re} \left\langle \frac{x}{\sqrt{x^2 - f^2}} \right\rangle \text{sech}^2 \left(\frac{1}{2}\beta\Delta x \right) dx \\ &\approx 1 - 0.6478t - 0.276t^3 \end{aligned} \quad (6)$$

$x = E/\Delta$ where E is the quasi-particle energy, $\langle \rangle$ means average over ϕ (over the Fermi surface) and $\beta = T^{-1}$. On the other hand, the out-of-plane superfluid density is given by

$$\begin{aligned} \rho_{s,out}(t) &= \frac{\pi}{2} \frac{\Delta}{\Delta(0)} \left\langle f \tanh\left(\frac{1}{2}\beta\Delta f\right) \right\rangle \\ &\approx 1 - 0.3592t^2 \end{aligned} \quad (7)$$

Our starting assumption has been that $\rho_{s,out}(t)$ is due to the Josephson tunneling between layers, so we have used the expression for the Josephson current obtained by Ambegaokar and Baratoff [18], properly generalized for d -wave superconductor. In [13] we have addressed in detail this calculation. Here, we would like to point out that the t^2 dependence might be more consistent with the other properties of κ -ET salts than the t linear dependence found by Carrington *et al.* [12]. In what follows we elaborate this statement in more details.

It is well known that the quasi-particle motion within the conducting ac planes of κ -ET salts is described in terms of the Fermi liquid theory. This is evidenced from the observation of the de Haas-van Alphen and the Shubnikov-de Haas oscillations [19,20]. On the other hand, the out-of-plane transport appears to be not so simple for the following reasons. First, no Drude

tail is observed in the out-of-plane millimeter wave conductivity [21]. It should be noted that the similar result has been also found in the \vec{c} -axis optical conductivity in high- T_C cuprates YBCO and Bi2212 [22]. Such a behaviour is difficult to formulate in terms of the standard quasi-particle transport. Rather, a simple tunneling model across the interlayer barriers can provide the observed frequency-dependent conductivity [23]. Further, the Josephson plasmons have been observed both in κ -(BEDT-TTF)₂Cu(NCS)₂ organic superconductor [24] and in high- T_C cuprates YBCO and Bi2212 [25], implying the coherent Josephson tunneling. Finally, a similar t^2 dependence has been also observed in the out-of-plane magnetic penetration depth of YBCO [26] and Tl2201 [27].

The calculated fits to our experimental data according to theory are

$$\rho_{s,\text{in}}(t) = 1 - 0.82t + 0.28t^3 - 0.64t^4 + 0.18t^5 \quad (8)$$

$$\rho_{s,\text{out}}(t) = 1 - 0.78t^2 - 0.21t^3 + 1.22t^4 - 1.23t^5 \quad (9)$$

where we assumed $\lambda_{\text{ac}}(0) = 5 \mu\text{m}$. The result is shown in Fig. 6. The linear term of $\rho_{s,\text{in}}(t)$, indicated by full line, has a slope of 0.82. In contrast, the behaviour of $\rho_{s,\text{out}}(t)$, shown as a dashed line, is much flatter and a T^2 term develops at low temperatures. Note that the shapes of both curves are qualitatively different from the s -wave BCS result. Expressions (8) and (9) are quite comparable with Eq. (6) and Eq. (7). A chosen value of $\lambda_{\text{ac}}(0)$ would indicate that the electron density (n_{el}) should be smaller by a factor of 10 than in the case of samples measured by the other authors since $\lambda_{\text{ac}}^{-2}(0) \propto n_{\text{el}}$. On the other hand, the value $\lambda_{\text{ac}}(0) \approx 1 \mu\text{m}$ gives also a t -linear dependence for $\rho_{s,\text{in}}(t)$, but with the coefficient of t -linear term being much larger than expected theoretically in the weak-coupling model. The coefficient of the leading t term in the expressions of both $\rho_{s,\text{in}}$ and $\rho_{s,\text{out}}$ depend strongly on the ratio of the superconducting transition temperature and the zero temperature superconducting order parameter. A comparison of Eq. (8) with Eq. (6), and Eq. (9) with Eq. (7), suggests that the superconducting order parameter at $T = 0$ K of κ -ET-Br is somewhat smaller than that predicted by the weak-coupling limit. Though this is somewhat surprising, a similar behaviour has been already discussed for a realistic model [6]. This might imply that the order parameter may be more complicated than the simple d -wave model predicts.

Finally, we are drawn to conclude that all recent experiments suggest the nodal structure in κ -ET-Br as expected for d -wave superconductivity. However, none of these experiments can discriminate d_{xy} -wave model from $d_{x^2-y^2}$ one. A recent STM results obtained in κ -(BEDT-TTF)₂Cu(NCS)₂ [28] suggest $d_{x^2-y^2}$ -wave symmetry, contrary to our expectation. On the other hand, d_{xy} -wave symmetry has been claimed in the same compound by Schrama *et al.* [29] from the anisotropy in the millimeter wave absorption in the presence of a magnetic field perpendicular to the conduction planes. Clearly, a more direct study of the nodal directions has to be carried out similarly as it has been done in YBCO [30–32]. In addition, the phase sensitive experiments in κ -ET salts, parallel to what has been done in high- T_C cuprates [1] will be also decisive in establishing the symmetry of the superconducting order parameter.

In summary, we have established the linear temperature dependence of the in-plane superfluid density and the T^2 dependence of the out-of-plane superfluid density below 5 K in single crystal of κ -ET-Br by using the AC susceptibility technique. The observed power laws are fully consistent with those expected in the d -wave model of superconductivity in which the bulk superconducting state is stabilized by the Josephson coherent tunneling between the superconducting layers. This observation further strengthen the evidence of d -wave superconductivity in κ -ET materials.

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