

# Probing the order parameter of the layered organic superconductor $\kappa$ -(BEDT-TTF)<sub>2</sub>Cu[N(CN)<sub>2</sub>]Br by AC susceptibility measurements

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AC susceptibility measurements are reported for single crystals of the layered organic superconductor  $\kappa$ -(BEDT-TTF)<sub>2</sub>Cu[N(CN)<sub>2</sub>]Br. The value of out-of-plane penetration depth  $\lambda_b(0)$  was found to be 133  $\mu\text{m}$ , consistent with earlier results. The temperature dependence of the in-plane superfluid density, with the value of in-plane component  $\lambda_{ac}(0)$  of the order of 1  $\mu\text{m}$ , is strongly linear in  $T$ , whereas the out-of-plane superfluid density varies as  $T^2$  at low temperatures. The observed behaviour is fully consistent with the one expected for d-wave superconductivity.

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The organic superconductors containing BEDT-TTF (abbreviated ET) planes are layered materials with anisotropic physical properties. The  $\kappa$  phases i.e.  $\kappa$ -(ET)<sub>2</sub>X (X is polymerized anion) are materials in which orthogonally aligned ET dimers form 2D conducting layers sandwiched between the polymerized anion layers. One of the points of interest in the study of layered organic superconductors is the possibility that the superconducting state is something other than the conventional s-wave BCS pairing state. This idea partially arises from similarity which exists between these organic superconductors and high  $T_C$  cuprates in which the establishment of d-wave superconductivity is widely accepted and considered as the most important fact in the understanding of unconventional superconductors<sup>1,2</sup>. In addition to the similar crystallographic (2D or layered) structure, the superconducting state in both materials is situated in the proximity to an antiferromagnetic phase revealing the dominance of the Coulomb interactions. Indeed, the following experiments, in our opinion, give crucial guiding lines in favour of an unconventional superconductivity with nodes in the gap in the  $\kappa$ -(ET)<sub>2</sub>X superconductors. <sup>13</sup>C NMR measurements by Mayaffre et al. showed that the spin-lattice relaxation rate follows  $T^3$  dependence at very low temperatures. This result together with the Knight shift provided a firm evidence for a spin singlet pairing with nodes in the gap<sup>3</sup>. The low temperature specific heat measured by Nakazawa et al.<sup>4</sup> showed a power-law behaviour  $c_s(T) \propto T^2$  also consistent with nodes in the gap structure. The third one is the experiment by Belin et al.<sup>5</sup> which showed that the thermal conductivity varies linearly with temperature at  $T \ll T_C$ . Furthermore, the existence of a peak in the out-of-plane magnetoresistance<sup>6,7</sup> has been interpreted as evidence against the s-wave symmetry of the order parameter<sup>8</sup>.

The measurement of the magnetic penetration depth  $\lambda(T)$  is a useful probe of the energy-gap morphology at the Fermi surface and of the superfluid electrodynamics. Various techniques have been employed to determine the value of  $\lambda(0)$  and its temperature dependence  $\lambda(T)$  for  $\kappa$ -(ET)<sub>2</sub>X superconductors and disparate results have been obtained. Approximately, a half of the performed studies of the temperature dependence of  $\lambda$  gave evidence for non-s-wave pairing, usually in the form of a  $T$  and/or  $T^2$  behaviour for  $\lambda(T)$  at low temperatures<sup>9-12</sup>, while the other half showed exponential dependence expected for s-wave pairing<sup>13-15</sup>. Consequently, the experimental situation regarding the temperature dependence of the superfluid density in  $\kappa$ -(ET)<sub>2</sub>X superconductors is presently unclear and rather controversial<sup>16</sup>. On the other hand, some of recent theories suggest the  $d_{xy}$  superconductivity in  $\kappa$ -(ET)<sub>2</sub>X<sup>17-19</sup>.

The layered organic superconductor  $\kappa$ -(BEDT-TTF)<sub>2</sub>Cu[N(CN)<sub>2</sub>]Br (abbreviated  $\kappa$ -(ET)<sub>2</sub>Br) possesses the highest known critical temperature at ambient pressure among the anisotropic organic superconductors. In this paper we report the temperature dependence of the in-plane,  $\lambda_{ac}$ , and out-of-plane,  $\lambda_b$ , magnetic penetration depth, as well as the zero temperature value of the latter, for single-crystals of  $\kappa$ -(BEDT-TTF)<sub>2</sub>Cu[N(CN)<sub>2</sub>]Br using the AC-susceptibility technique. We have chosen this technique since it is a direct method which enables to determine  $\lambda(T)$  by detecting the diamagnetism of the single crystals. We find characteristic low-temperature dependences proportional to  $T$  for the in-plane superfluid density and  $T^2$  for the out-of-plane superfluid density, which are consistent with d symmetry of the order parameter if a cylindrical Fermi surface is taken into account.

Measurements of the complex AC susceptibility ( $\chi = \chi' + i\chi''$ ) were performed using an AC susceptometer (AcSuS/Sistemprojekt, Zagreb) in the broad temperature range between 1.5 K and  $T_C$ , with the AC field applied either perpendicular or parallel to the crystal ac plane. Measurements were performed for  $H_{AC} = 14$  mOe at  $f = 231$  Hz. The temperature of the sample was swept slowly (0.5 K/min typically) with the sample positioned in the upper one of the two identical pick-up coils immersed in the liquid helium bath. In order to probe the sample in the Meissner state care was taken to reduce the amplitude of the AC field ( $H_{AC}$ ) until the component  $\chi'(T)$  was independent of  $H_{AC}$  ( $H_{AC} < 42$  mOe) and the  $\chi''(T)$  component was negligible. No frequency dependence (13 Hz  $< f < 2$  kHz) was observed for  $H_{AC} < 1$  Oe. In addition no influence of the Earth's field was observed: runs performed with compensated Earth's field gave the same results. This is in accordance with the fact that the values  $H_{C1}(T)$  corrected for demagnetization for all  $T < 10$  K<sup>20</sup> are far above the Earth's field  $H_E$ . Namely, the value of the Earth's field determined in our laboratory is  $H_E \approx 360$  mOe<sup>21</sup>. The calibration of the system was performed only in the field geometry  $H_{AC} \parallel$  plane with a piece of niobium foil whose volume and ratio of the characteristic dimensions were close to those of the sample. Following this procedure the demagnetizing effect was taken into account.

Measurements have been performed on four crystals from two different batches, rhombic platelets with large faces between 0.44 and 1 mm<sup>2</sup> and between 0.25 and 0.35 mm along the  $\vec{b}$  axis. Two samples studied in the most detail exhibited qualitatively the same behaviour. Here we present data obtained on one high-quality single-crystal 0.54·0.90·0.33 mm<sup>3</sup>. Crystallographic orientation of one crystal was determined by taking one oscillation XRD pattern (Ni-filtered  $\text{Cu}_{K\alpha}$ ; Weissenberg camera) and the corresponding 0<sup>th</sup> layer-line Weissenberg pattern around  $\vec{a}$  axis. This  $\vec{a} = [100]$  axis is revealed to be parallel to the line bisecting the sharp angle (65°) of the lozenge-shaped sample, while the  $\vec{c} = [001]$  axis runs parallel to the line bisecting the obtuse angle (115°). The  $\vec{b} = [010]$  axis is aligned perpendicular to the largest facet of the crystal.

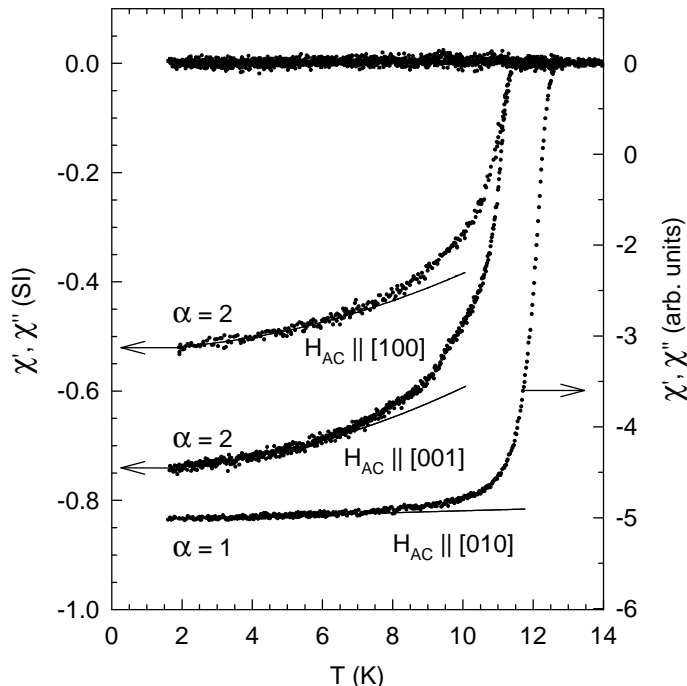


FIG. 1. Real and imaginary part of the complex susceptibility in an AC field of 14 mOe parallel to the crystal ac plane ( $H_{AC} \parallel [100]$  and  $H_{AC} \parallel [001]$ ) and perpendicular to the ac plane ( $H_{AC} \parallel [010]$ ).

$\chi(T)$  data taken at 231 Hz and at  $H_{AC} = 14$  mOe are shown in Fig.1. The onset of superconductivity is observed to be at 12.4 K and 11.2 K for  $H_{AC} \perp$  plane and  $H_{AC} \parallel$  plane, respectively. The observed difference in  $T_C$  is somewhat

surprising, since no similar result has been reported so far. In order to eliminate spurious influences we have verified, by performing test experiments on the piece of niobium foil used for calibration, that no thermal gradient larger than 0.05 K exists along the sample holder. Therefore, the observed difference in  $T_C$  is not an experimental artifact. In addition, the second single crystal studied in the most detail has shown a smaller difference in onset temperatures of 0.8 K, nevertheless essentially the same results for either  $\lambda_b(0)$ , or low-temperature dependencies of the in-plane and the out-of-plane superfluid density have been obtained. The latter shows that the observed difference in  $T_C$  does not have an influence on the obtained final results reported in this paper. Since the very subject of onset temperatures is not the main topic of this paper, we will address this subject in our forthcoming publication. Coming back to Fig.1, note an anisotropy in the ac response for  $H_{AC}$  parallel and perpendicular to the ac planes. For the former  $\chi'(T)$  decreases rather slowly, while for the latter  $\chi'(T)$  falls sharply below  $T_C$  corresponding to screening currents flowing mostly in the ac plane. In addition, note that for  $H_{AC} \parallel$  plane and for  $H_{AC} \perp$  plane  $\chi'(T)$  obeys a power-law behaviour  $\chi'(T) \propto T^\alpha$  with the exponent  $\alpha = 2$  and  $\alpha = 1$  below about 5 K, respectively. While the anisotropy in  $\chi'$  is clearly related to the anisotropy in  $\lambda$ , it is not clear that it can entirely be attributed to the anisotropy of the superfluid density over the whole temperature range.

First we analyze 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.1 (width  $D[001] = 0.54$  mm and  $D[100] = 0.90$  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<sup>16,22,23</sup>.  $\lambda_b$  and  $\lambda_{ac}$  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  $\lambda_b$ .  $D$  is the sample width in the direction of field penetration. For  $H_{AC} \parallel [100]$  and for  $H_{AC} \parallel [001]$   $D$  is the sample width in  $[001]$  direction and in  $[100]$  direction, respectively. The fact that the  $\chi'$  response of our sample is different for  $H_{AC} \parallel [100]$  and for  $H_{AC} \parallel [001]$  confirms that the field penetration along the  $D$  direction, and not the one along the  $b$  direction which is associated to  $\lambda_{ac}$ , dominates the susceptibility of the sample. Moreover, we point out that the results obtained for both field orientations are mutually consistent. In what follows we show and analyze data for the field orientation in the  $[100]$  direction as the ratio  $D/b$  is smaller than for the  $[001]$  case.

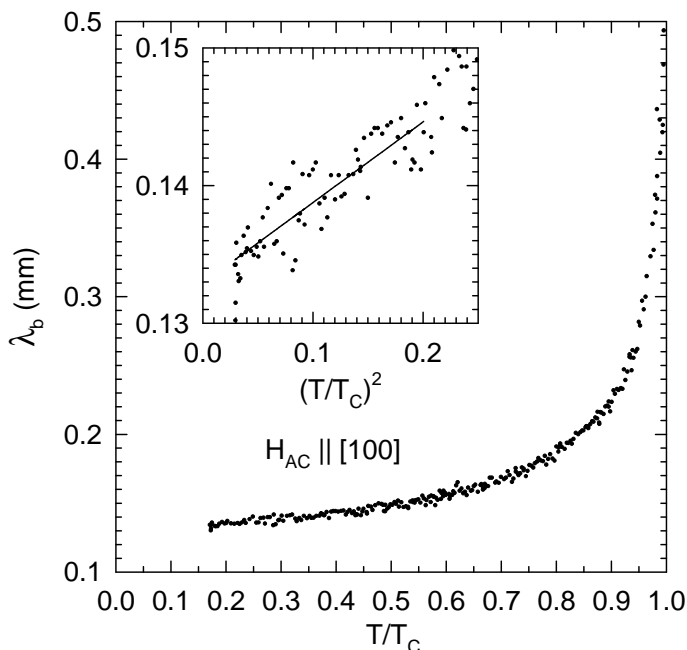


FIG. 2. Penetration depth as a function of reduced temperature for  $H_{AC} \parallel [100]$ . Inset:  $\lambda_b$  plotted versus  $(T/T_C)^2$  in the low temperature region  $1.6 \text{ K} < T < 5 \text{ K}$ .

The temperature dependence of  $\lambda_b$  is shown in Fig.2. The temperature dependence of  $\lambda_b$  is well described by the  $T^2$  law at temperatures below about 5 K i.e.  $(T/T_C)^2 < 0.2$  (see Inset of Fig.2). 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<sup>16</sup>. Note that initial (low  $T$ ) increase in  $\chi'$  is related to the increase in  $\lambda_b$  by  $1 + \chi' = 2\lambda/D$  (Eq.(1) for  $\lambda \ll D$ ), so that the quadratic  $T$  term is the leading term which describes the temperature dependence of both  $\chi'$  and  $\lambda_b$ .

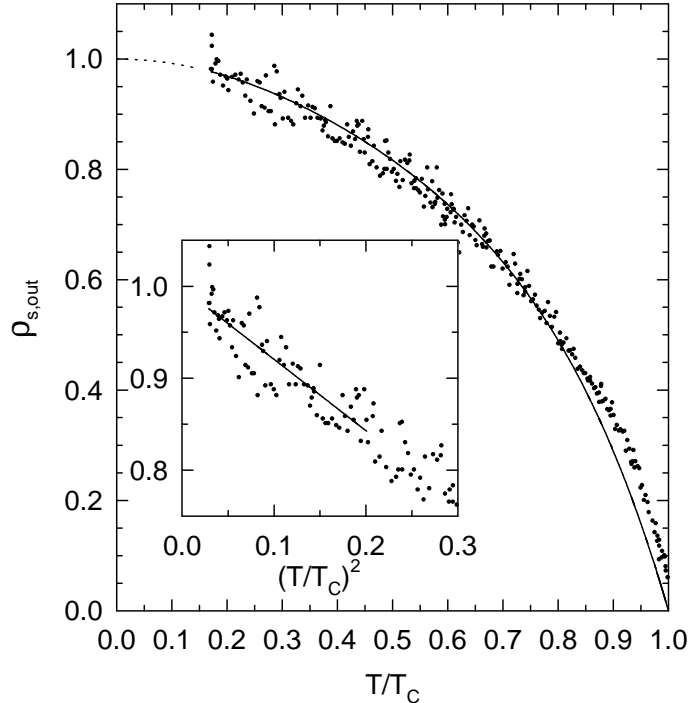


FIG. 3. Out-of-plane superfluid density  $\rho_{s,out}$  as a function of reduced temperature. Full line is a fit to the theory (see Text). Inset:  $\rho_{s,out}$  plotted versus  $(T/T_C)^2$  in the temperature range  $1.6 \text{ K} < T < 5 \text{ K}$ .

It is useful to construct the quantity  $(\lambda_b(0)/\lambda_b(T))^2$  to get information on the temperature dependence of the out-of-plane superfluid density  $\rho_{s,out}$  and on the symmetry of the superconducting state

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

The out-of-plane superfluid density  $\rho_{s,out}$  as a function of reduced temperature  $t = T/T_C$  is displayed in Fig.3. Note that the leading term which describes the low temperature behaviour of the out-of-plane superfluid density  $\rho_{s,out}$  is the  $T^2$  term (see Inset of Fig.3).

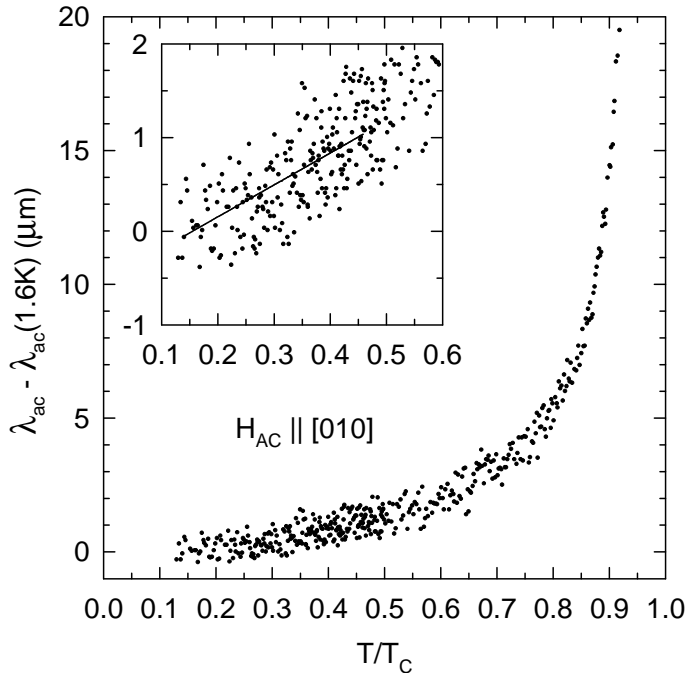


FIG. 4. Change of the penetration depth with respect to the minimum value as a function of reduced temperature for  $H_{AC} \parallel [010]$ . Inset:  $\lambda_{ac} - \lambda_{ac}(1.6 \text{ K})$  in the low temperature region  $1.6 \text{ K} < T < 5 \text{ K}$  is clearly linear in temperature with a slope of  $3.4 \mu\text{m}$ .

In the perpendicular direction of the field, it is much more difficult to obtain the absolute value of the penetration depth,  $\lambda_{ac}$ . Namely, a large demagnetization factor for this orientation is very sensitive to the sample's geometry and cannot be evaluated accurately enough. Therefore, we adopt the following approach to extract  $(\lambda_{ac}(0)/\lambda_{ac}(T))^2$  in order to get the temperature dependence of the in-plane superfluid density. We follow the method of Kanoda et al.<sup>9</sup> and determine the deviation of  $\lambda_{ac}$  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  $\chi'$  are plotted in Fig.4. Below about 5 K, there is a clear linear term with slope  $3.4 \mu\text{m}$  (see Inset of Fig.4).

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

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

which behaves linearly with  $T$  at low temperatures as expected for the d-wave superconductivity<sup>24</sup>. The temperature dependence of the in-plane superfluid density for the d-wave superconducting order parameter  $|\Delta(\vec{k})| = \Delta f$  with  $f = \sin(2\varphi)$ , where  $\varphi$  is the angle between the quasiparticle momentum  $\vec{k}$  and the  $\vec{a}$  axis, within the weak coupling theory is given by<sup>24</sup>

$$\begin{aligned} \rho_{s,in}(t) &= 1 - \frac{1}{2}(\beta\Delta) \int_0^\infty \Re \left\langle \frac{x}{\sqrt{x^2 - f^2}} \right\rangle \text{sech}^2 \left( \frac{1}{2}\beta\Delta x \right) dx \\ &\approx 1 - 2(\ln 2)(\beta\Delta)^{-1} - \frac{9}{4}\zeta(3)(\beta\Delta)^{-3} \\ &\approx 1 - 0.6478t - 0.276t^3 \end{aligned} \quad (6)$$

$x = E/\Delta$  where  $E$  is the quasi-particle energy,  $\langle \rangle$  means average over  $\varphi$  (over the Fermi surface) and  $\beta = T^{-1}$ . Here we used  $2e^{\beta\Delta x} (1 + e^{\beta\Delta x})^{-2} = \frac{1}{2}\text{sech}^2 \left( \frac{1}{2}\beta\Delta x \right)$  and  $\Re \left\langle \frac{x}{\sqrt{x^2 - f^2}} \right\rangle = \frac{2}{\pi}xK(x)$  for  $x \leq 1$ , where  $K(x)$  is the

complete elliptic integral. In the last step we used the weak-coupling result for the d-wave order parameter at  $T = 0$  K,  $\Delta(0) = 2.14 T_C$ . On the other hand, the out-of-plane superfluid density is given by

$$\begin{aligned} \rho_{s,out}(t) &= \frac{\pi}{2} \frac{\Delta}{\Delta(0)} \left\langle f \tanh\left(\frac{1}{2}\beta\Delta f\right) \right\rangle \\ &\approx \left(1 - \frac{\pi^2}{6}(\beta\Delta)^{-2} - \frac{7\pi^4}{120}(\beta\Delta)^{-4}\right) \left(1 - \frac{3}{2}\zeta(3)(\beta\Delta)^{-3}\right) \\ &\approx 1 - 0.3592t^2 \end{aligned} \quad (7)$$

Here  $1 - 3\zeta(3)(\beta\Delta)^{-3}$  comes from  $\Delta/\Delta(0)$ . Also, we have assumed that  $\rho_{s,out}(t)$  is due to the Josephson tunneling between layers and used the expression for the Josephson current obtained by Ambegaokar and Baratoff<sup>25</sup>, properly generalized for d-wave superconductor. We would like to point out that this is the first time that this model is used to calculate  $\rho_{s,out}(t)$  in d-wave superconductors, although the Josephson tunneling between layers in high- $T_C$  cuprates has been established experimentally<sup>26</sup>. The related Josephson plasmons have been observed both in high- $T_C$  cuprates<sup>27</sup> and in  $\kappa$ -(BEDT-TTF)<sub>2</sub>Cu(NCS)<sub>2</sub> organic superconductor<sup>28</sup>. The latter authors have estimated the out-of-plane penetration depth  $\lambda_{out}(0) = 120 \mu\text{m}$  which is very close to the value we found in the the  $\kappa$ -(BEDT-TTF)<sub>2</sub>Cu[N(CN)<sub>2</sub>]Br system.

In what follows we address the issue of calculating  $\rho_{s,out}(t)$  for a particular layered compound in some detail, since there appears to be a great confusion in the literature about what are the appropriate starting assumptions in this calculation. Actually, there have been 3 different approaches to calculate  $\rho_{s,out}(t)$  in high  $T_C$  cuprates in literature, depending on the relation between the in-plane electron momentum  $\vec{k}$  and  $\vec{k}'$  across the barrier: (a)  $\vec{k} = \vec{k}'$ , the in-plane momentum is conserved, (b)  $\vec{k} \neq \vec{k}'$ , the in-plane momentum is completely randomized and (c)  $\vec{k} \parallel \vec{k}'$ , only direction, but not magnitude of the in-plane momentum, is conserved. We consider that neither the case (a), nor (b) can be properly viewed as the tunneling model, since (a), which might be called the anisotropic 3D model, is the usual tight-binding model and (b), which might be viewed as the limiting case of the tunneling model, is dominated by impurity scattering between the layers (the incoherent limit). Moreover, these two approaches give the expressions different from Ambegaokar-Baratoff model<sup>25</sup> of Josephson tunneling.

The simplest result for d-wave superconductors within the approach (a) is given by Radtke et al.<sup>29</sup>. As one expects this model gives  $\rho_{s,out} \propto t$ , that is the same behaviour as found for  $\rho_{s,in}(t)$  at low temperatures. Therefore, this model does not describe our present result. The model considered by Xiang and Wheatley<sup>30</sup> is more elaborated version of this approach, where they inserted an extra  $\vec{k}, \vec{k}'$  dependence in the scattering matrix. Their model gives  $t^5$  dependence for  $\rho_{s,out}(t)$ , which also does not describe our result. In the approach (b),  $\vec{k}$  and  $\vec{k}'$  are totally uncorrelated, i. e. only incoherent scattering processes are taken into account. The simplest version of this approach was elaborated by Graf et al.<sup>31</sup>. Their result looks fine for s-wave superconductors, but gives  $\rho_{s,out} = 0$  for d-wave superconductors. Indeed, Hirschfeld et al.<sup>32</sup> have tried to save this unphysical situation by introducing, in our opinion, a rather artificial  $\varphi$  and  $\varphi'$  dependence in the scattering matrix and found  $\rho_{s,out}(t) \propto t^3$ . Here  $\varphi$  and  $\varphi'$  are the angles  $\vec{k}$  and  $\vec{k}'$  make with the  $\vec{a}$  axis, respectively. In our calculation the model (c) is used. We assumed  $\varphi = \varphi'$ , that is the specular transmission as mentioned in Ambegaokar and Baratoff original paper, and more clearly characterized by Hirschfeld et al.<sup>32</sup>. Naturally, in this limit the tunneling should be coherent, since we do not introduce the impurity scattering, and we obtain Eq.(7) as given above. We stress that the model (c) is the only approach in which Josephson coupling is present in unconventional superconductors like d-wave superconductors considered in this paper. Therefore, the coherent tunneling or the specular transmission is our crucial assumption. It is rather gratifying that our model appears to describe  $\rho_{s,out}(t)$  of both  $\kappa$ -(ET)<sub>2</sub>X salts and high  $T_C$  cuprates like YBCO and Tl2201 as discussed later in text.

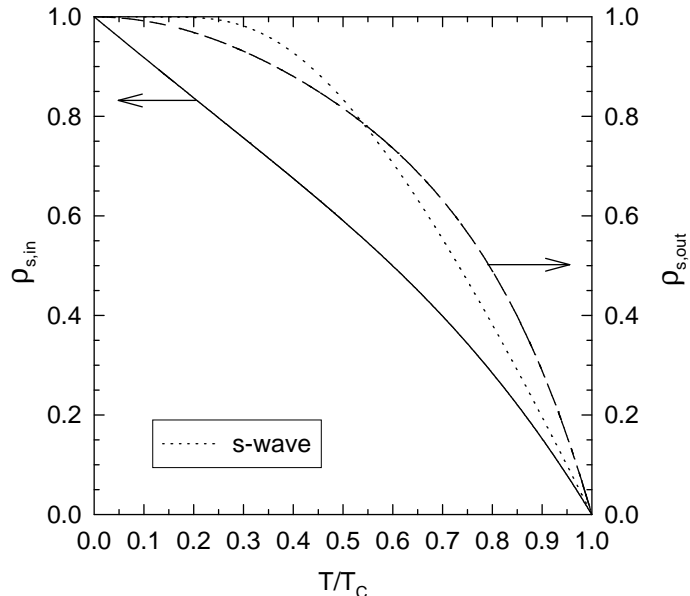


FIG. 5. In-plane superfluid density  $\rho_{s,in}$  (full line) and out-of-plane superfluid density  $\rho_{s,out}$  (dashed line) versus reduced temperature.  $\rho_{s,out}$  is qualitatively different from the behavior seen in the ac plane. The s-wave BCS result (dotted line) is shown for comparison.

The calculated fits to our experimental data according to theory are

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

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

where we assumed  $\lambda_{ac}(0) = 5 \mu\text{m}$ . The result is shown in Fig.5. The linear term of  $\rho_{s,in}(t)$ , indicated by full line, has a slope of 0.82. In contrast, the behaviour of  $\rho_{s,out}(t)$ , 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). First, we note that the coefficient of the  $t^2$  term in expression (9) for the out-of-plane superconducting density is very close to the result found by Kanoda et al.<sup>9</sup> for  $\kappa$ -(BEDT-TTF)<sub>2</sub>Cu(NCS)<sub>2</sub> single crystals. Second, a chosen value of  $\lambda_{ac}(0)$  would indicate that the electron density ( $n_{el}$ ) should be smaller by a factor of 10 than in the samples measured by the other authors since  $\lambda_{ac}^{-2}(0) \propto n_{el}$ . On the other hand, the value  $\lambda_{ac}(0) \approx 1 \mu\text{m}$  gives also a  $t$ -linear dependence for  $\rho_{s,in}(t)$ , but with the coefficient of  $t$ -linear term much larger than expected theoretically in the weak-coupling model. The coefficient of the leading  $t$  term in the expressions of both in-plane and out-of-plane superconducting densities 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  $\kappa$ -(BEDT-TTF)<sub>2</sub>Cu[N(CN)<sub>2</sub>]Br is somewhat smaller than that predicted by the weak-coupling limit. Though this is somewhat surprising, a similar behaviour is already discussed for a realistic model<sup>18</sup>. This might imply that the order parameter may be more complicated than the simple  $d_{xy}$  model predicts.

Therefore, the observed temperature dependences of  $\rho_{s,in}$  and  $\rho_{s,out}$  perfectly agree with the behaviour expected in the framework of the  $d_{xy}$  model. The  $T$  dependence of  $\rho_{s,in}$  at low temperatures implies linear nodes at the Fermi surface. These linear nodes also give rise to both the  $T^3$  dependence of the spin-lattice relaxation rate  $T_1^{-1}$  and the  $T^2$  dependence of the specific heat at low temperatures consistent with the observation in the  $\kappa$ -(BEDT-TTF)<sub>2</sub>Cu[N(CN)<sub>2</sub>]Br superconductor<sup>3,4</sup>. In addition, the thermal conductivity within the ac plane should behave linearly in  $T$  for  $T < T_C$ <sup>33,34</sup> as observed by Belin et al.<sup>5</sup>. The presence of linear nodes in  $\Delta(\vec{k})$  indicates that the order parameter in the  $\kappa$ -(ET)<sub>2</sub>X salts might be very similar to the d-wave order parameter in high- $T_C$  cuprates. We recall that  $T$ -linear dependence of  $\rho_{s,in}(t)$  in YBCO is one of the crucial experiments which indicates d-wave superconductivity in high- $T_C$  cuprates<sup>35</sup>. Further,  $T^2$  behaviour in  $\rho_{s,out}(t)$  has been only recently also established experimentally in high  $T_C$  - cuprate YBCO<sup>36</sup>. In addition,  $\rho_{s,out}(t)$  appears to display  $T^2$  behaviour also in Tl2201 system<sup>37</sup>, consistent with our theoretical model. On the other hand the  $T$  linear dependence of  $\rho_{s,out}(t)$  is observed in Bi2212<sup>38</sup>, what is rather puzzling.

Now, we address the possibility that the interlayer tunneling (ILT) model<sup>39</sup>, suggested to explain the superconductivity in the high- $T_C$  cuprates, might be also a relevant theory of superconductivity in layered organic superconductors.

The basic assumption of the ILT model is that the transport of carriers between layers is incoherent in the normal state, while the coherent interlayer transport is allowed by Cooper pairs in the superconducting state. It is exactly the latter process which creates superconductivity in contrast to the model we use in which superconductivity first arises by pairing correlation within each layer. While the ILT model appears to explain well the superconductivity in La-214 cuprate high- $T_C$  superconductors, the recent high-precision experimental determination of the out-of-plane penetration depth for Hg-1201 and Tl-2201 systems gave the values of  $\lambda_{out}$  in disagreement with  $\lambda_{ILT}$  predicted by the ILT model<sup>40</sup>. As far as  $\kappa$ -(ET)<sub>2</sub>X superconductors are concerned, the ILT model is less likely to be relevant as the mechanism of superconductivity for the following reasons. First, the experimental observation of de Haas van Alphen oscillations<sup>41</sup>, also predicted by the band calculations<sup>42</sup>, clearly shows that the normal state is the traditional Fermi liquid state. Second, taking into account the value  $\lambda_{out}(0) = 133 \mu\text{m}$ , the Josephson coupling energy appears to be thousand times smaller than the condensation energy, which is at variance with the basic prediction of the ILT model. Moreover, this result implies  $T_C = 0.37 \text{ K}$  within the ILT model, which is 30 times lower than actually observed ( $T_C = 12 \text{ K}$ ).

Finally, the standard model based on Coulomb dominance and/or the antiparamagnon exchange gives  $d_{xy}$ -wave superconductivity in  $\kappa$ -(ET)<sub>2</sub>X salts<sup>17–19</sup>. However, a startling result within the standard model is reported in ref. 43. For example, Fig. 3 in ref. 43 shows that the superconductivity is not d-wave, but g-wave with extra nodal lines running along the diagonal directions, as well. It appears to us that this strange result is due to the wrong choice of the relative sign between two order parameters attached to two distinct Fermi surfaces. When this error is corrected, the result in ref. 43 agrees with those in refs. 17–19. In spite of this (sign) problem, ref. 43 stressed correctly the importance of the interband scattering in organic superconductors. Therefore, we conclude that most of available approaches in the framework of the standard model expects d-wave superconductivity in  $\kappa$ -(ET)<sub>2</sub>X salts.

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  $\kappa$ -(BEDT-TTF)<sub>2</sub>Cu[N(CN)<sub>2</sub>]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 should further strengthen the possibility of d-wave superconductivity in  $\kappa$ -(ET)<sub>2</sub>X materials. In this circumstance the phase sensitive experiments in  $\kappa$ -(ET)<sub>2</sub>X salts, as the ones done in high- $T_C$  cuprates<sup>1</sup>, are highly desirable.

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