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Superconducting state in the layered organic superconductor κ-$(BEDT\text{-}TTF)_2Cu[N(CN)_2]Br$

M.Pinterić[a,b], M.Prester[a], S.Tomić[a], K.Maki[c], D.Schweitzer[d], I.Heinen[e], W.Strunz[e],
[a] *Institute of Physics, P.O.Box 304, HR-10000 Zagreb, Croatia*
[b] *Faculty of Civil Engineering, University of Maribor, Maribor, Slovenia*
[c] *Dept of Physics, University of Southern California, LA, CA 90089-0484, U.S.A.*
[d] *3.Physikalisches Institut, Universität Stuttgart, D-70550 Stuttgart, Germany*
[e] *Anorganisch-chemiches Institut, Universität Heidelberg, D-69120 Heidelberg, Germany*

Abstract

We summarize the main features of the magnetic field-temperature phase diagram for the superconducting state of κ-$(BEDT\text{-}TTF)_2Cu[N(CN)_2]Br$. The initial temperature dependence of the real component of ac susceptibility for field parallel and perpendicular to the ac plane are markedly different. The out-of-plane penetration depth λ_b and the related superfluid density vary at low tempeartures as T^1. Slow flux dynamics below the irreversibility line for the ac field perpendicular to the BEDT-TTF planes might be ascribed to a vortex-pair creation process. The characteristic relaxation time $\tau > 10^{-4}$ s is thermally activated with an activation energy of about 300K.

Keywords: Organic superconductors, Magnetic measurements

1. Introduction

We have measured the complex ac susceptibility ($\chi = \chi' + i\chi''$) of single crystals of the layered organic superconductor κ-$(BEDT\text{-}TTF)_2Cu[N(CN)_2]Br$. The measurements were performed in the broad temperature range between 1.5K and T_c= 11 K, and the ac field was applied either perpendicular or parallel to the conducting planes. Two regions in the field-temperature phase diagram below the irreversibility line (H_{irr}) were explored. First, in an effort to determine the magnetic field penetration depth $\lambda(T)$ and its temperature dependence we studied the vortex-free Meissner state ($H_{ac} < H_{c1}$). Namely, the experimental situation regarding the temperature dependence of the superfluid density in κ-$(BEDT\text{-}TTF)_2X$ superconductors is presently somewhat controversial [1-4]. Second, we investigated the mixed state ($H_{c1} < H_{ac} < H_{irr}$) with the aim to probe the vortex dynamics in this region. Measurements have been performed on five crystals from two different baches, rhombic platelets with large faces between 1 and 0.54 mm^2 and between 0.2 and 0.3 mm along the b axis. The complete account of the work will be given elsewhere [5, 6].

2. Experimental and results

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 χ'' component was absent. 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 field gave the same results. This is in accordance with the fact that the values $H_{c1}(T)$ corrected for demagnetization for all T < 10K are far above the earth field $H_E \sim 100$mOe [7]. The sensitivity of the system was calibrated in both geometries (ie. $H_{ac} \parallel$ plane and $H_{ac} \perp$ plane) with a piece of niobium foil whose volume and the ratio of the characteristic dimensions were close to those of the sample. $\chi(T)$ data taken at 231Hz and at H_{ac}= 14 mOe are shown in Fig.1. One can see considerable 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 T^1 law and a T^2 law below about 5K, respectively. While the anisotropy in χ' is clearly related to the anisotropy in λ, it is not clear that it can entirely be attributed to the anisotropy in the superfluid density over the whole temperature range.

In the following we will analyze the low T behaviour for $H_{ac} \parallel$ plane using the formula for a thin superconducting plate in a parallel field

$$1+\chi' = (2\lambda/D)\tanh(D/2\lambda) \quad (1)$$

where D is the sample width in the direction of the field penetration. For $H_{ac} \parallel [100]$, D is the sample width in [001] direction and $\lambda = \lambda_b$. This approach is based on the accepted values of $\lambda_b(0)$ and $\lambda_{ac}(0)$ which are about 150 μm and 1 μm, respectively [1]. Hence for the crystal in Fig.1. (width D= 0.67mm, thickness b= 0.32mm) the condition $\lambda_b/\lambda_{ac} \gg D/b$ was satisfied. This implies that the behaviour of χ' given by eq.(1) might be attributed to λ_b ie. to the penetration depth which is associated with interlayer currents. The temperature dependence of λ_b is shown in Fig.2. The full line corresponds to

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the calculated fit to a power-law behaviour in the temperature range $1.5K < T < 5K$

$$\lambda_b = k(T/T_c)^s + \lambda_b(0) \quad (2)$$

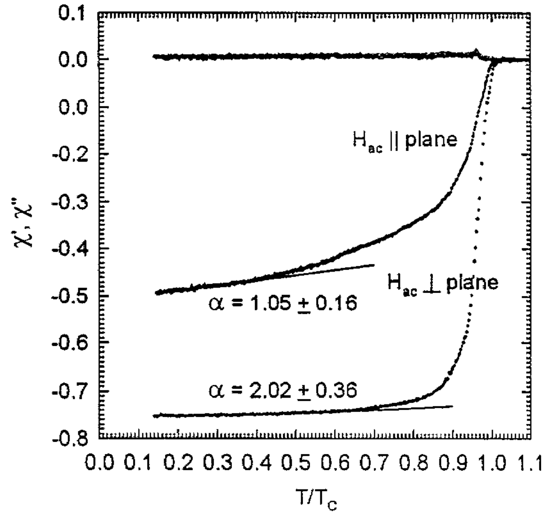

Fig. 1. Ac susceptibility data for two orientations of a single crystal. Lines present T^1 and T^2 terms in χ' for $H_{ac} \|$ plane and $H_{ac} \perp$ plane, respectively.

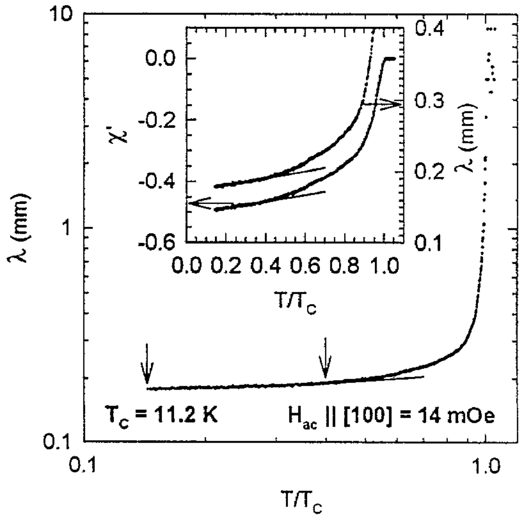

Fig. 2. Logarithm of the penetration depth vs logarithm of the reduced temperature for $H_{ac} \|$ plane. The solid line is a fit to the power-law behaviour. The inset shows T term in χ' and λ.

We get $s=1.10 \pm 0.16$ and $\lambda(0)=172.4$ μm. In the inset of Fig.2. it is shown a comparison of the temperature behaviour of χ' and λ_b. Note that initial (low T) increase in χ' is related to the increase in λ_b by $1 + \chi' = (2\lambda/D)$ (eq.(1) for $\lambda << D$), so that the linear T term is the leading term which describes the temperature dependence of both χ' and λ_b. The low-temperature data of λ_b give the first evidence for a clear linear T dependence of the out-of-plane penetration depth in κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br.

It is useful to construct the quantity $(\lambda_b(0)/\lambda_b(T))^2$ to get the information on the temperature dependence of the superfluid density ρ_{sout}

$$\rho_{sout} = (\lambda_b(0)/\lambda_b(T))^2 \quad (3)$$

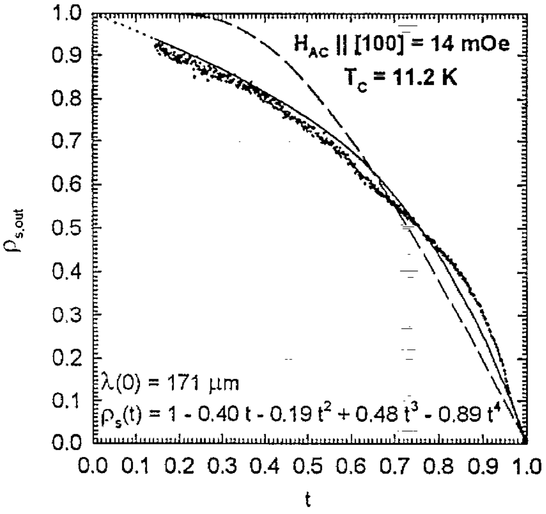

Fig. 3. The ratio $(\lambda_b(0)/\lambda_b(T))^2$ vs the reduced temperature $t=T/T_c$. The solid line is a fit to the polynomial expansion. The dashed line is the s-wave result.

The calculated fit $(\lambda_b(0)/\lambda_b(T))^2 = 1 + a\,(T/T_c) + O\,(T/T_c)^n$, where $n \geq 2$, describes rather well the experimental data as shown in Fig.3. A linear term is clearly present at low temperatures and this is qualitatively different from the s-wave BCS result also given in Fig.3. The linear term in λ_b and ρ_{sout} may arise from the presence of nodes in the superconducting energy gap as in d-wave superconductors. A complete analysis and discussion of both ρ_{sout} and ρ_{sin} in the framework of a d-wave superconductor based on the realistic Fermi surface in the κ-(BEDT-TTF)$_2$Cu[N(CN)$_2$]Br will be given in our forthcoming paper [5].

In order to probe flux dynamics, the complex ac susceptibility response has been mapped out as a function of temperature, ac field amplitude (2.8 mOe - 42 mOe) and frequency (3 Hz - 2.3 kHz), dc field (0 - 42 Oe) and field orientation (H_{ac}, H_{dc} $\|$ and $\perp$ to ac planes). The sample was measured by first cooling in zero field to low temperatures (4.2K) and then applying the field and measuring the susceptibility during the sample was slowly warmed up above T_c. The peak in the imaginary part of the susceptibility χ'' has been used to probe the flux dynamics in the range above the lower critical field H_{c1} and below the irreversibility line H_{irr}.

In the following we will show our data obtained when only the ac magnetic field was applied. Note that in Fig.4. and 5. the scale for the susceptibility is arbitrary since the calibration and correction for the demagnetizing factor has not been done. Fig.4.a. and b. show the temperature domain plots of χ' and χ'' as a function of frequency (f= 13 Hz, 231 Hz, 770 Hz, 2310 Hz) for an applied ac field of 1.4 Oe and 14 Oe perpendicular to ac planes. For H_{ac}= 1.4 Oe χ' shows a relatively sharp transition, and χ'' shows a relatively sharp peak just below T_c, both indicating that the field is not large enough to penetrate the sample except very close to T_c (Fig.4.a.). H_{ac}= 14 Oe is sufficiently large to penetrate the sample at higher temperatures, so that χ' is reduced and χ'' shows a broader peak at a lower temperature than for H_{ac}= 1.4Oe (Fig.4.b.).

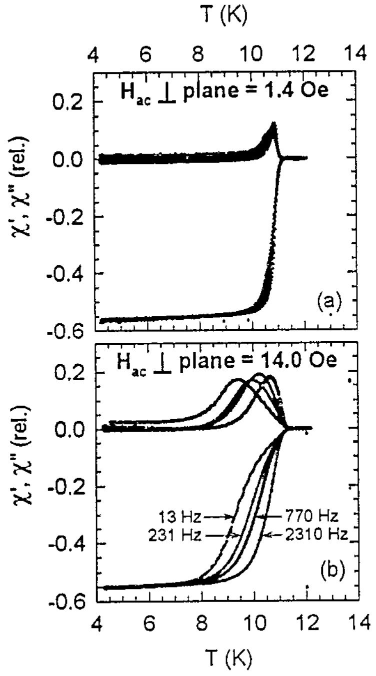

Fig.4. Complex ac susceptibility vs temperature for a number of frequencies between 13 Hz and 2.31 kHz at H_{ac}= 1.4 Oe (a) and H_{ac}= 14 Oe (b). Data were taken with $H_{ac}\perp$ plane.

Fig.5.a. and b. show χ' and χ'' plots as in Fig.4. but obtained for a parallel field orientation ie. H_{ac} || plane. An anisotropy in the complex ac magnetic susceptibility response function for two field orientations is clearly visible. The anisotropy ratio is even larger than it appears in the figure since for H_{ac} ⊥ plane the internal magnetic field is larger than the applied field due to the rather large demagnetizating factor in this geometry. From these measurements we have determined the temperature of the peak in the dissipative component of the ac susceptibility χ'' as a function of the ac field amplitude and frequency.

In the following we show and analyze only our data observed for the field orientation perpendicular to the ac planes. The phase diagram H_{ac} vs T is drawn in Fig.6. Note that the χ'' peak temperatures are at field values much below the irreversibility line obtained with dc magnetization measurements as well as ac susceptibility measurements [7]. Here we point out that for the case of a small ac field and a larger dc field and for the highest frequency used f= 2.31 kHz our data for the χ'' peak temperature versus field coincide closely with the irreversibility line [6]. This indicates that the origin for absorption might be different for these two cases. We briefly recall that two possible sources of ac losses, pinning and relaxation, might be present. In the critical-state model no vortex motion is allowed, and the peak in χ'' versus the temperature occurs at the point where the ac magnetic-field flux just penetrates to the center of a sample. However, the obtained results are not only amplitude but also frequency dependent, contrary to the prediction of this model. This fact reveals that the relaxation processes are also active and have to be taken into account. In the following we attempt to explain the ac susceptibility response as the result of thermally activated vortex-pair creation processes or by using a vortex-glass theory.

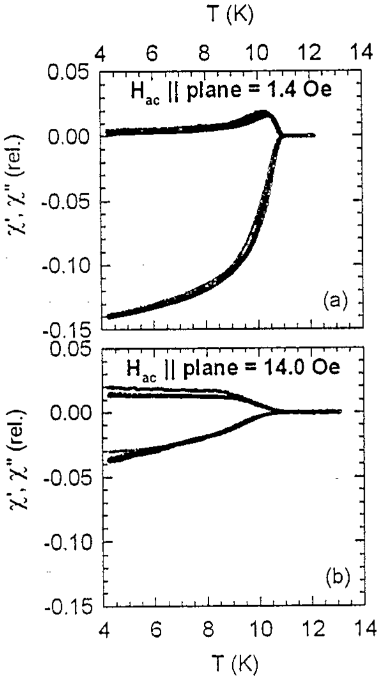

Fig.5. Complex ac susceptibility vs temperature for a number of frequencies between 13 Hz and 770 Hz at H_{ac}= 1.4 Oe (a) and H_{ac}= 14 Oe (b). Data were taken with H_{ac}|| plane.

Fig.4.b. shows a representative temperature domain plot of χ'' for the selected choise of frequencies. Note that the width of the χ'' peak increases and its position shifts to lower temperature when the frequency f of the applied ac field is decreased. The position of the χ'' maximum gives a measure for the dynamic effect. Namely, when the relaxation time τ of the system becomes larger than $1/2\pi f$, the system ceases to be able to follow the ac field. The initial increase of χ'' indicates a more viscous response as T lowers, while the subsequent decrease might be interpreted as a consequence of more active pinning sites which impedes the vortex response. In the temperature range below the maximum the dependence of χ'' on frequency obeys the power-law with exponent $\alpha < 0.5$ indicating a broad distribution of metastable states [6]. The temperature dependence of the characteristic relaxation time for ac field amplitude H_{ac}= 33.6 Oe is shown in Fig.7.a. The solid line is

the best fit to an Arrhenius behaviour with the activation energy $\Delta = 270K$ and the attempt time $\tau_0 = 4.68 \cdot 10^{-17}$ sec. We note that the energy scale of the order of a few hundred kelvin corresponds rather well to an energy needed for the creation of a pancake vortex-pair. An alternate approach assumes critical slowing down which leads to a complete freezing of the response at enough low temperatures $T<T_G$. (Fig.7.b.). The solid line is a fit to $\tau = \tau_0 ((T/T_G) - 1)^{-6.5}$. The activation energy and the attempt time as a function of the ac field amplitude are shown in Fig.8. Note that after the initial strong change, both parameters saturates at the constant value for $H_{ac} > 14$ Oe.

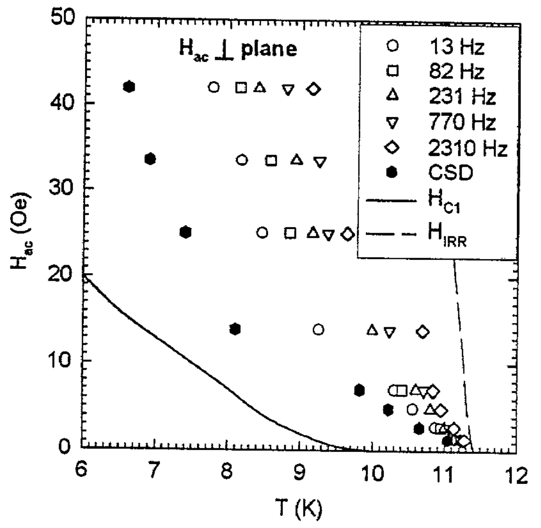

Fig.6. Loss (χ") peak lines at a number of frequenices for the ac field orientation perpendicular to the BEDT-TTF planes.

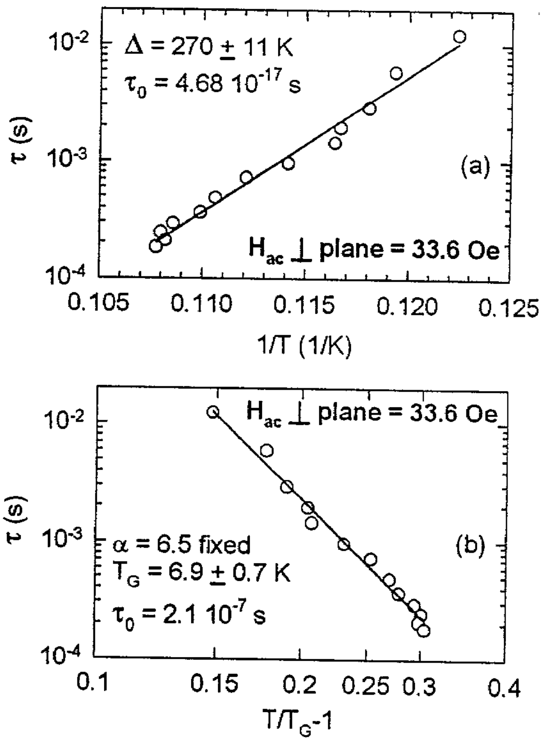

Fig.7. (a) Characteristic relaxation time vs inverse temperature. Full line shows a fit to Arrhenius form. (b) Same data analyzed within the vortex glass theory. Full line is a fit to theory.

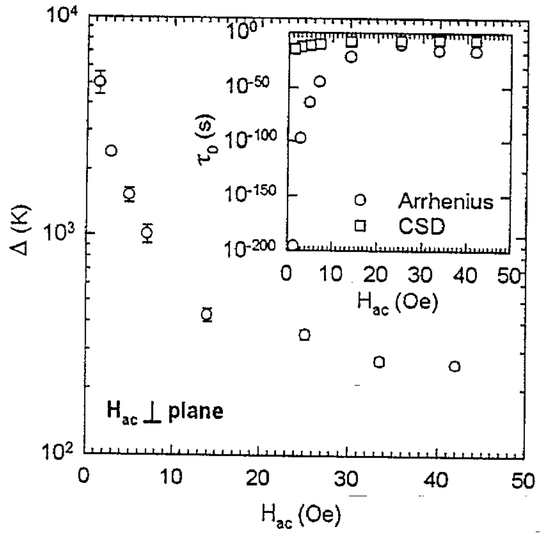

Fig.8. Activation energy vs ac field amplitude. Inset shows attempt time vs ac field amplitude for gradual slowing down (circles) and critical slowing down (squares) behaviour.

3. Conclusion

The low temperature linear dependence of the out-of-plane superfluid density offers a new piece of evidence for the d-wave nature of the SC state. Dynamically determined boundaries in the H_{ac} -T phase diagram are frequency dependent. For an investigated range of ac amplitudes and in our frequency window the response of vortex-pairs gradually slows down with temperature. Studies at lower frequencies are needed to check the existence of a real glass transition.

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