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C-axis transport in the intrinsically layered superconductor κ -(BEDT-TTF)₂Cu(NCS)₂: interplay between Josephson and quasiparticle tunneling

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Abstract

We have measured the transverse electrical transport properties of single crystals of the intrinsically layered organic superconductor κ -(BEDT-TTF)₂Cu(NCS)₂ in magnetic fields applied perpendicular to the superconducting planes. The low and intermediate field $\rho_c(T)$ curves can be fitted with a model, which takes into account the parallel conduction channels arising from intrinsic Josephson coupling and quasiparticle tunneling between the layers. At the highest measured fields, Josephson coupling between the layers is suppressed and the $\rho_c(T)$ dependence is parabolic. © 1997 Elsevier Science B.V.

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Keywords: Organic superconductor; Electrical resistivity; Mixed state; Josephson effect

1. Introduction

One of the still open questions in intrinsically layered superconductors, as, e.g., the cuprates or organic superconductors of the κ -(BEDT-TTF)₂X family [1], is the description of their perpendicular transport properties. For the high- T_c cuprates with large superconducting anisotropies $\gamma = \xi_{ab}/\xi_c$, as, e.g., Bi₂Sr₂CaCu₂O_{8+x} or oxygen reduced YBa₂Cu₃O_{7-x}, an experimental consent has been found, that the out-of-plane resistivity $\rho_c(T)$ exhibits a non-metallic, i.e. either mixed or purely semicon-

ducting behavior in the normal state [2]. This stands in contrast to their in-plane behavior, which is metallic down to low temperatures. The semiconducting $\rho_c(T)$ leads to a distinct peak around the superconducting transition temperature T_c before resistivity vanishes when temperature is further lowered. In a magnetic field H applied perpendicular to the conducting planes, the peak's height increases, and its position shifts to lower temperatures for higher fields. Several theories have been developed regarding this problem, either describing primarily the properties in the normal state and ascribing the semiconducting behavior to non-Fermi liquid behavior and/or unusual hopping processes between the conducting layers [3], or treating the normal and the superconducting properties in close connection [4,5]. The latter

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theories consider the c -axis conductivity as arising from quasiparticle tunneling processes between adjacent layers, which are subject to superconducting fluctuations. These fluctuations above the mean field T_c lead to a virtual gap, which reduces the quasiparticle density of states (DOS). The reduction of normal carriers results in a decreasing interlayer quasiparticle conductivity when the virtual gap opens. The decrease of resistivity at further lowered temperatures is explained by the onset of Josephson coupling between the superconducting layers and hence a parallel conduction channel arising from Cooper pair tunneling [4–6]. In the high- T_c cuprates, the behavior in the normal state at low temperatures, which could help to distinguish between the different types of theories, is an open question due to the experimentally inaccessible large upper critical fields $H_{c2,\perp}(T=0)$. Also measurements of the high-field $\rho_c(T)$ of the low- T_c (and low- $H_{c2,\perp}$) cuprates $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ and La-doped $\text{Bi}_2\text{Sr}_2\text{CuO}_x$ [7] only gave partial answer, since the measured fields are still in the vicinity of their upper critical fields, and a contribution of fluctuations to resistivity can hence not be ruled out.

To gain further insight into the transverse transport properties of intrinsically layered superconductors, we performed measurements on single crystals of $\kappa\text{-(BEDT-TTF)}_2\text{Cu(NCS)}_2$ and present the first convincing quantitative analysis of its c -axis resistivity.¹ The investigated compound belongs to the family of quasi two-dimensional organic superconductors $\kappa\text{-(BEDT-TTF)}_2\text{X}$ [1], which consist of alternating layers built up from the conducting organic cations BEDT-TTF and polymerized, non-conducting inorganic anions X. The in-plane conduction arises from the delocalized π -bindings of the BEDT-TTF's central carbon atoms. $\kappa\text{-(BEDT-TTF)}_2\text{-Cu(NCS)}_2$ (in the following denoted as ET-NCS) exhibits some of the cuprates' typical properties, such as short coherence lengths, short interlayer distances, and a large superconducting anisotropy $\gamma \geq 100$ [8–10]. Apart from these similarities, ET-NCS has much lower critical temperature $T_c \approx 10$ K

and critical fields, e.g., for magnetic field applied perpendicular to the superconducting planes, $H_{c2,\perp} \leq 100$ kOe. These differences make ET-NCS especially suitable as a model-compound for the cuprate superconductors, since almost the complete superconducting phase diagram is accessible within experiment.

2. Experiment and results

We have measured the out-of-plane transport properties of two high-quality single crystals of ET-NCS grown in a standard electrocrystallization process [11]. The samples had sizes of about $700 \times 200 \times 25 \mu\text{m}^3$ (sample A) and $900 \times 550 \times 40 \mu\text{m}^3$ (sample B). On the top and the bottom surface of the plate-like samples, two gold contacts each (cf. Fig. 1(a)) were evaporated using a shadow mask technique. With this contact arrangement, we were able to perform measurements of the c -axis transport properties in a standard four probe setup. Due to the large resistivity anisotropy, the planes perpendicular to the current flow can be regarded as equipotential, which makes a decomposition analysis [12] unnecessary. We verified this assumption by test measurements (i) with exchanged current and voltage contacts on one side of the sample, and (ii) in a two probe setup with subsequent subtraction of the contact resistance. For resistivity measurements, a low frequency (17 Hz), low current (10 μA) lock-in technique was employed. $V(I)$ curves were taken with a DC method. The temperature was determined above 1.4 K with a carbon glass resistor, which was carefully calibrated against shifts in magnetic field. At lower temperatures, we employed an RuO_2 thermometer. Sample thermalization was ensured by the leads (gold wires) and ^4He exchange gas in the sample chamber. The use of heat conducting grease is inappropriate to these organic samples, as the induced pressure can substantially change the sample [13].

Fig. 1 depicts $\rho_c(T)$ for both crystals at different fields. The behavior is independent of the sample and the contact geometry and is found to match exactly that of another crystal reported in literature [14]. This is a clear evidence that the presented properties are intrinsic to the c -axis transport in

¹ We use the same notation as in the cuprates: the direction perpendicular to the planes will also for ET-NCS be called c -direction. In the literature, this direction is usually denoted a^* and the planes accordingly bc -planes.

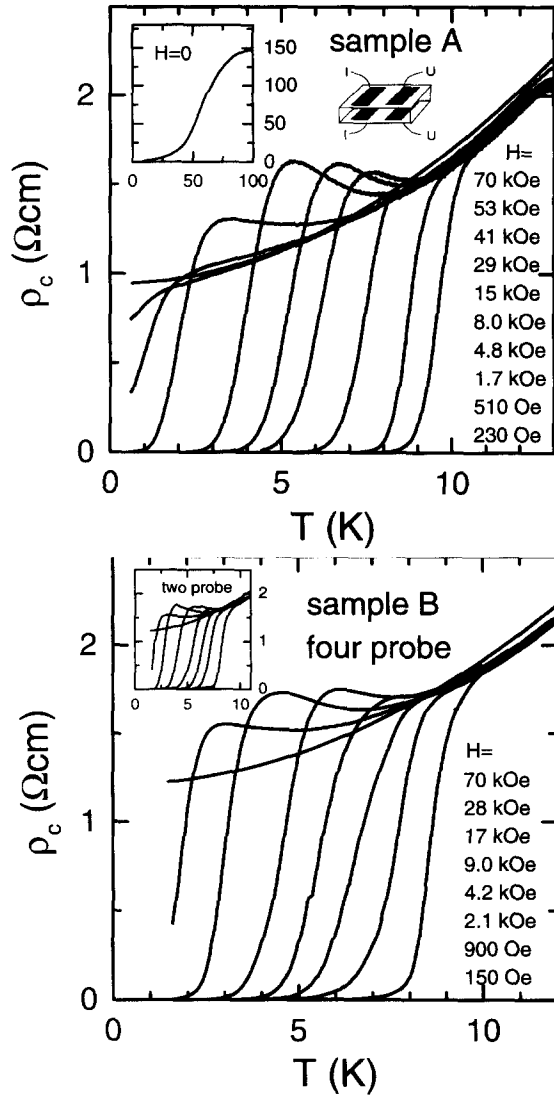


Fig. 1. Out of plane resistivity ρ_c versus temperature. (a) Sample A. Inset: metallic dependence of resistivity at higher temperatures. (b) Sample B. Inset: in a two probe setup with subsequent subtraction of the contact resistance.

ET-NCS. Let us shortly outline the main issues of the measurements: In the normal state, $\rho_c(T)$ is metallic. In the superconducting state, three field regimes can be distinguished: (i) At low fields ($H < 1.7$ kOe), resistivity decreases monotonically with decreasing temperature. (ii) At intermediate fields (1.7 kOe $< H < 45$ kOe), the curves show a distinct peak. Its position shifts to lower temperatures with

increasing field. Up to fields of about 15 kOe, the peak height simultaneously increases with increasing field. As field is further increased, the peak height decreases again. (iii) At the highest measured fields ($H > 45$ kOe), the peak disappears completely. At 70 kOe even the decrease of $\rho_c(T)$ at the low temperature end of the curve is suppressed.

In the following we develop a phenomenological model, which fits the c -axis resistivity of ET-NCS at low and intermediate fields. At high fields, our picture still predicts the measured resistivity qualitatively. The basic idea is, as in Refs. [4,5], to describe the c -axis conductivity σ_c as the sum of a contribution σ_{Jos} due to Josephson tunneling and a contribution σ_{qp} from quasiparticle tunneling between adjacent layers, $\sigma_c = \sigma_{\text{Jos}} + \sigma_{\text{qp}}$. Accordingly, we model the investigated crystal as an array of intrinsic Josephson junctions [15], each following the resistively shunted junction (RSJ) model [16]. The individual junction's length is given by the layer distance $d = 15$, whereas the junction area depends primarily on the vortex density induced by the external magnetic field, since the vortices limit the maximum size of phase coherent areas. The Josephson contribution to conductivity σ_{Jos} is calculated from the average junction conductance g_{jct} as $\sigma_{\text{Jos}} = mdg_{\text{jct}}$, where m denotes the density of junctions per plane, i.e. the number of junctions per unit area. The conductance g_{jct} of each junction follows from the RSJ model and subtraction of the shunt conductance g_n [5],

$$g_{\text{jct}} = g_n \{ I_0^2(\gamma(T)/2) - 1 \}, \quad (1)$$

where I_0 denotes the modified Bessel function and $\gamma(T)$ the Josephson coupling parameter in the Ambegaokar–Baratoff (AB) model [17],

$$\gamma(T) = \frac{\pi \hbar \Delta(T) g_n}{2 e^2 k_B T} \tanh\left(\frac{\Delta(T)}{2 k_B T}\right). \quad (2)$$

Here $\Delta(T)$ is the superconducting gap. The shunt conductance is calculated from the average critical current $i_c(T)$ of each junction in the $T \rightarrow 0$ limit and the relation $i_c(0) = \pi \Delta(0) g_n / 2e$ [17]. To determine $i_c(T)$ we additionally performed series of $V(I)$ measurements at fixed field for different temperatures, and employed $i_c(T) = J_c(T)/m$, with $J_c(T)$ the sample critical current density. $J_c(T, B)$ was calcu-

lated from the critical currents, the latter extracted as the intercepts of the tangent to the inflection point of each $V(I)$ curve with the current axis. (See also Figs. 2 and 3 for $J_c(T)$ at some fields.) The parallel quasiparticle resistivity $\rho_{qp} = \sigma_{qp}^{-1}$ is modeled as the sum of a contribution due to scattering processes, which was in in-plane measurements shown to lead to a parabolic $\rho(T)$ dependence due to electron–electron scattering [18], and a tunneling contribution. The choice of a parabolic contribution to the quasiparticle resistivity also in the superconducting state is justified by the fact that the in-plane resistivity does not vanish at the mean field T_c [19], and accordingly the in-plane quasiparticle contribution can not be regarded as short-circuited by the superconducting channel. The quasiparticle tunneling contribution is taken constant in the normal state. In the superconducting state, the quasiparticle tunneling re-

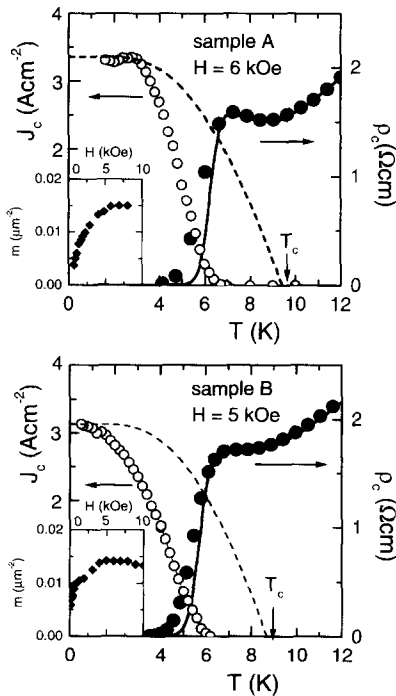


Fig. 2. (a) Resistivity (solid circles) and critical current density (open circles) versus temperature at $H = 6$ kOe for sample A. The solid line is a fit according to the model developed in the main text. The dashed line depicts the calculated critical current density in the Ambegaokar–Baratoff model. Inset: dependence of the density of junctions m on magnetic field. (b) The corresponding data for sample B at $H = 5$ kOe.

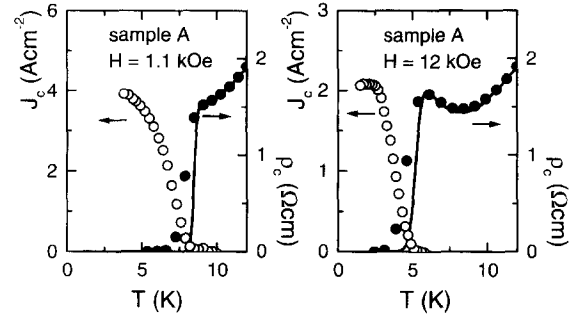


Fig. 3. Resistivity, fit to resistivity and critical current density for sample A at $H = 1.1$ kOe (left panel) and $H = 12$ kOe (right panel), respectively. Symbols as in Fig. 2.

sistivity increases with decreasing T as $\propto \exp(\Delta(T)/k_B T)$ [5], since the number of quasiparticles decreases with the establishment of the superconducting gap due to the formation of Cooper pairs. Because the two processes can be regarded as independent [20], the total quasiparticle resistivity is written as

$$\rho_{qp} = \sigma_{qp}^{-1} = A + BT^2 + \rho_{tl} \exp(\Delta(T)/k_B T), \quad (3)$$

where A , B , and ρ_{tl} are field independent in the limits of this model. To fit the data, we use the BCS temperature dependence of $\Delta(T)$ and $\Delta(0) = 1.76 k_B T_c$. The field dependent critical temperature is adjusted to the values found in a fluctuation analysis of the in-plane resistivity of a comparable sample [19]. The parameters A and B are derived from a fit of the resistivity in the normal state to $A = 0.73 \Omega \text{ cm}$ and $B = 7.6 \times 10^{-3} \Omega \text{ cm K}^{-2}$ (sample A) and $A = 1.14 \Omega \text{ cm}$ and $B = 7.1 \times 10^{-3} \Omega \text{ cm K}^{-2}$ (sample B), respectively. The tunnel resistivity ρ_{tl} is determined universal for all fields to $\rho_{tl} = 0.087 \Omega \text{ cm}$ (sample A) and $\rho_{tl} = 0.050 \Omega \text{ cm}$ (sample B). The only field dependent parameter is the junction density m .

Fig. 2 depicts the results of the fits for sample A and B at two different fields, whereas Fig. 3 displays the data at two further fields for sample A. It is found, that the $\rho_c(T)$ dependence is well reproduced down to temperatures below the peak. In the insets of Fig. 2, we plot the field dependent fitting parameter – the junction density m . As expected, m increases monotonically with field, since the field induced vortices limit the junction size. Below the peak down to the disappearance of resistivity, the

model underestimates the measured values. It is clear that this is due to the neglected vortex contribution to resistivity: diffusive motion of (pancake-) vortices results in phase slips between adjacent layers and hence in additional dissipation [21]. The activation energies of these thermally activated processes correspond directly to the values found in in-plane measurements [8,22]. In Figs. 2 and 3 we additionally plot the measured critical current densities, and in Fig. 2 also the theoretical predictions following from the AB model. Together with the fitted $\rho_c(T)$ curves this leads to a coherent picture of the c -axis transport in ET-NCS: At temperatures above the critical temperature, the interlayer resistivity is due to quasiparticle scattering processes and tunneling. Slightly below T_c , $\rho_c(T)$ is still dominated by the quasiparticle contribution. Here the resistivity increases with decreasing temperature due to the loss of normal carriers. At further lower temperatures, about at the position of the peak in $\rho_c(T)$, effective Josephson coupling between the layers sets in, characterized by a non-vanishing Josephson critical current density. Below the peak, in the Josephson coupled regime, resistivity is non-zero due to thermally activated vortex motion. It should also be noted that the measured critical currents are always lower than expected from the AB model. This reduction of J_c is also caused by the presence of fluctuating vortices [6], which give rise to an average phase difference of the superconducting wave function between adjacent layers. The lower critical currents compared to the AB model were also reported for Josephson plasma resonance measurements in this compound [23].

The curves at high fields can not be fitted successfully: although the model predicts the suppression of the peak for $H \gg H_{c2}$ (where $\Delta = 0$ at all T and $\rho_c(T)$ is parabolic), its shift to lower temperature at fields $12 \text{ kOe} < H < H_{c2}$ can not be modeled correctly with reasonable values of m . This is due to the enhancement of quasiparticle tunneling by vortices, which reduce the superconducting gap locally (in the vortex cores), or uniformly (at very high vortex densities). Taking into account the hence lowered quasiparticle resistivity, the faster shift of the peak can qualitatively be understood. Another effect, which is neglected in the model, is the ordinary positive magnetoresistance, which becomes more important at higher fields.

3. Conclusion

In summary, the behavior of c -axis transport in ET-NCS can be described as the interplay between quasiparticle tunneling and scattering, Josephson tunneling, and dissipative vortex motion. It was shown that $\rho_c(T)$ curves at low and intermediate fields can be fitted with a model, which takes into account the lowered quasiparticle tunneling contribution to conductivity due to the reduction of the DOS as the superconducting state develops, and interlayer Josephson coupling. The behavior at high fields can qualitatively be understood, taking additionally into account the further reduction of the quasiparticle resistivity. Here, a quantitative description should also include vortex-related effects on the quasiparticle tunneling. At the highest measured fields, $\rho_c(T)$ is parabolic, as predicted by the model for $H \gg H_{c2}$. For $T \rightarrow 0$, the out-of-plane resistivity is finite and metallic.

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