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Surface Impedance Studies on the Electrodynamical Response of Organic Superconductors

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Abstract

We have performed detailed measurements of the surface impedance in the normal and superconducting state of κ -(BEDT-TTF)₂Cu(NCS)₂ in the millimeter wave frequency range (1 cm⁻¹ to 3 cm⁻¹), and have evaluated the complex conductivity for different crystallographic orientations. Above the transition temperature, the material behaves like a metal with a scattering rate of approximately 20 cm⁻¹. In the superconducting state the electrodynamics of both materials is in good agreement with calculations based on a BCS ground state: the penetration depth is temperature independent for $T \rightarrow 0$; while the penetration depth and the coherence length are strongly anisotropic, the superconducting energy gap shows no indications of line nodes.

1 Introduction

The nature of the superconducting ground state of (BEDT-TTF)₂Cu(NCS)₂ and (BEDT-TTF)₂Cu[N(CN)₂]Br is highly controversial. Only the strange behavior of the nuclear spin-lattice relaxation rate [1, 2], and the $H_{c2} \perp$ plane-behavior seems to be undisputed, although not explained yet. Contradictory results emerged from most of the other significant experiments, like specific heat [3, 4], magnetization [5, 6, 7], μ SR [8, 9], and microwave measurements [10, 11]. Here we want to report on our detailed studies of the surface impedance which we performed on (BEDT-TTF)₂Cu(NCS)₂ and (BEDT-TTF)₂Cu[N(CN)₂]Br in order to obtain information on the superconducting transport and the penetration depth [12].

2 Experimental

The single crystals were characterized by ac susceptibility measurements. We performed microwave cavity perturbation experiments at various frequencies [13] and measured the complex surface impedance

$$\hat{Z}_S = R_S + iX_S = \left(\frac{i\mu_0\omega}{\sigma_1 - i\sigma_2} \right)^{1/2} \quad (1)$$

as a function of temperature. The sample was placed in cylindrical TE₀₁₁ cavities in the maximum of the electric or magnetic field in such a way that either the direction parallel or perpendicular to the highly conducting planes was probed [12].

Figure 1: The temperature dependence of the in-plane resistivity of κ -(BEDT-TTF)₂Cu(NCS)₂ measured at different frequencies. $R_S^2/\mu_0\omega$ is shown for the millimeter wave data. The curves are normalized to their 15 K-value. The inset displays a typical magnetization curve of κ -(BEDT-TTF)₂Cu(NCS)₂ obtained by ac susceptibility measurements. The superconducting transition temperature $T_c = 8.6$ K is defined by a 10% change of the signal.

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3 Normal State Properties

The temperature dependence of $2R_S^2/(\mu_0\omega)$ for 35, 60 and 100 GHz and the dc resistivity along the (bc) -plane of κ -(BEDT-TTF)₂Cu(NCS)₂ is displayed in Fig. 1 where all the data were normalized to their normal state value at $T = 15$ K. Just above T_c our millimeter wave results show no significant frequency dependence within the scattering due to different sample qualities and are in accord with σ_{dc} . In the perpendicular direction $\sigma_{\perp}(T=9\text{ K}) = 4.0 (\Omega\text{cm})^{-1}$ is three orders of magnitude smaller compared to $\sigma_{\parallel}$ but basically exhibits a similar temperature dependence.

Measurements on (BEDT-TTF)₂Cu[N(CN)₂]Br in both orientations show similar results as the one for (BEDT-TTF)₂Cu(NCS)₂ as far as the absolute value and the temperature dependence is concerned [10]. The compound has a sharp superconducting transition of with ca. 0.3 K and $T_c = 11.3$ K.

Figure 2: The surface impedance $\hat{Z}_{S\parallel}$ of (BEDT-TTF)₂Cu[N(CN)₂]Br measured at a frequency of 35 GHz. (a) The measurement was done in the antinode of the electric field with $\vec{E}$ parallel to the ac -planes. (b) With $\vec{H}$ in the plane, the perpendicular response is probed.

Figure 3: Temperature dependence of the normalized components σ_1 and σ_2 of the optical conductivity at 60 GHz both (a) parallel and (b) perpendicular to the highly conducting layers of (BEDT-TTF)₂Cu(NCS)₂. The solid lines represent the results of the BCS theory with $\ell/\pi\xi_0 = 1$ and 2, respectively.

4 Superconducting State Transport

Using Eq. 1, both the real and imaginary part of the conductivity $\hat{\sigma} = \sigma_1 + i\sigma_2$ in the superconducting state can be evaluated and compared with the results of calculations based on BCS theory. Since the Mattis-Bardeen formulae are only correct in the dirty limit which may not be appropriate in the case of these organic superconductors, we went beyond this assumption and performed calculations similar to Chang and Scalapino [14]. The temperature dependences of R_S and X_S for (BEDT-TTF)₂Cu[N(CN)₂]Br at low temperatures are displayed in Fig. 2 normalized to the normal state sur-

face resistance R_n at 13 K. In both directions our results can be well described by using the parameters $\ell/\pi\xi_0 = 2.5$, or 4, respectively, and a slightly larger gap value $2\Delta/k_B T_c = 5$. This is in accord with results from dc magnetization experiments [7] which indicated strong or medium coupling effects in $(\text{BEDT-TTF})_2\text{Cu}[\text{N}(\text{CN})_2]\text{Br}$. The parameters obtained by our analysis are listed in Table I.

In Fig. 3 the complex conductivity parallel and perpendicular to the planes of $(\text{BEDT-TTF})_2\text{Cu}(\text{NCS})_2$ is displayed as a function of temperature. We restricted ourselves to the weak coupling limit $\Delta = 1.76k_B T_c$ and fitted our results for various values of the only remaining parameter $\ell/\pi\xi_0$. The best fit to our data at 60 GHz was obtained with $\ell_{\parallel}/\pi\xi_{0\parallel} = 1$; i.e. $\omega\tau_{\parallel} = 0.18$ at $f = 60$ GHz and the relaxation rate $1/2\pi\tau_{\parallel} = 15 \text{ cm}^{-1}$ at 10 K. The values for the perpendicular direction are $\ell_{\perp}/\pi\xi_{0\perp} = 2$ which yields $\ell_{\perp} = 30 \text{ \AA}$, using $\xi_{0\perp} = 5 \text{ \AA}$. It is obvious from Figs. 3 that the normalized behavior of the electrodynamical response is highly similar in both orientations, despite the fact that the normal state values differ by three orders of magnitude between the two configurations. The similarity between the different crystallographic orientations suggests isotropic pairing. The dominant feature of the σ_1 -curve is the peak below the superconducting transition temperature which finds a natural explanation in the case-II coherence factor. Both of our results are in full agreement with a BCS ground state.

Figure 4: The penetration depth $\lambda_{\parallel}$ in $(\text{BEDT-TTF})_2\text{Cu}[\text{N}(\text{CN})_2]\text{Br}$ as a function of temperature. The solid circles show μSR data [9] and the open boxes results from measurements of the magnetization [7]. The dashed line is the BCS weak coupling limit, the solid line represents the local regime, and the dotted line gives the results of the two fluid model which are close the strong coupling limit.

Figure 5: Temperature dependence of the penetration depth of $(\text{BEDT-TTF})_2\text{Cu}(\text{NCS})_2$ measured at 35 GHz (pluses) and 60 GHz (open diamonds) in comparison with results from surface impedance experiments of Achkir et al. [10] (solid line), and with μSR experiments of Harshman et al. [8] (full boxes) and Le et al. [9] (solid circles). The experiments probe $\lambda_{\parallel}$, except for the open diamonds where $\lambda_{\perp}$ corresponds to the right axis. The inset demonstrates the agreement with the predictions of the BCS theory where the dashed line represents the London regime and the solid line the local regime.

5 Penetration Depth

The penetration depth for $(\text{BEDT-TTF})_2\text{Cu}[\text{N}(\text{CN})_2]\text{Br}$ as a function of the reduced temperature T/T_c is shown in Fig. 4, in comparison with other experimental results. The measured penetration depth of $(\text{BEDT-TTF})_2\text{Cu}(\text{NCS})_2$ in the two directions is displayed in Fig. 5 in comparison with published results. With the exception of [9] and [10], there is a good agreement between all the measurements. Even with the extended temperature range down to 0.8 K and the increased accuracy of $\delta\lambda/\lambda = \pm 0.007$, no indication of a temperature dependence can be found below $0.3 T_c$. In the inset of Fig. 5, the measured results of the normalized penetration depth $\lambda(T)$ of $(\text{BEDT-TTF})_2\text{Cu}(\text{NCS})_2$ are compared with theoretical predictions of the weak coupling BCS theory, the resolution of our measurement does not allow us to distinguish between different coupling limits. However, we can conclude that in both directions $\lambda(T)$ is well described by assuming a singlet ground state. They conclude $(\text{BEDT-TTF})_2\text{Cu}(\text{NCS})_2$ is in the weak coupling limit while $(\text{BEDT-TTF})_2\text{Cu}[\text{N}(\text{CN})_2]\text{Br}$ seems to be medium coupling, but for both compounds perfect agreement with s-wave pairing was found.

Table 1: Electrodynamical properties of $(\text{BEDT-TTF})_2\text{Cu}(\text{NCS})_2$ measured at 60 GHz, and $(\text{BEDT-TTF})_2\text{Cu}[\text{N}(\text{CN})_2]\text{Br}$ measured at 35 GHz with the current flowing either parallel or perpendicular to the highly conducting plane

$\kappa\text{-(BEDT-TTF)}_2X$ $X =$		T_c [K]	$\sigma_n(T = 300 \text{ K})$ [(Ωcm) $^{-1}$]	$\sigma_n(T > T_c)$ [(Ωcm) $^{-1}$]	δ [μm]	λ [μm]	ℓ [$\text{\AA}$]	ξ [$\text{\AA}$]	$1/(2\pi\tau)$ [cm^{-1}]	2Δ [cm^{-1}]
$\text{Cu}(\text{NCS})_2$	$\parallel$	8.3	20	3.7×10^3	3.4	1.4	150	70	15	21
	$\perp$	8.2	2.0×10^{-3}	4.0	100	40	30	5	5	21
$\text{Cu}[\text{N}(\text{CN})_2]\text{Br}$	$\parallel$	11.3	23	4.0×10^3	4.2	1.5	290	37	30	40
	$\perp$	11.3	0.5	6.4	110	38	50	6	5	40

6 Discussion

In the temperature region above T_c (but $T < 100 \text{ K}$) both materials behave like metals with a scattering rate of approximately 20 cm^{-1} when probed with microwave frequencies, but seems to depend on the measurement frequency. From this we infer that the simple Drude model does not apply.

Since we observe the same temperature dependence for $\sigma_1(T)$ both parallel and perpendicular to the planes, we deduce that the divergency in the optical response of the system is identical at various orientations. We conclude that the ratio $\Delta/\hbar\omega$ is a fundamental constant of the superconducting phase and thus the gap is isotropic for all three crystallographic axes. The coherence peak in σ_1 below T_c rules out a superconducting ground state which is significantly different from singlet pairing. For $\kappa\text{-(BEDT-TTF)}_2\text{Cu}(\text{NCS})_2$ we find $2\Delta = 3.53k_B T_c = 20 \text{ cm}^{-1}$, while a larger gap is obtained in $\kappa\text{-(BEDT-TTF)}_2\text{Cu}[\text{N}(\text{CN})_2]\text{Br}$: $2\Delta = 5k_B T_c = 40 \text{ cm}^{-1}$. As predicted by the theory, the maximum of σ_1/σ_n smoothes out as the photon energy becomes comparable to the single particle gap.

The temperature dependence of the penetration depth is a direct indication of the pairing mechanism; $\lambda(T)$ of the two organic superconductors $\kappa\text{-(BEDT-TTF)}_2\text{Cu}(\text{NCS})_2$ and $\kappa\text{-(BEDT-TTF)}_2\text{Cu}[\text{N}(\text{CN})_2]\text{Br}$ shows a flat behavior for low temperatures in both direction parallel and perpendicular to the planes. This $\lim_{T \rightarrow 0} d\lambda(T)/dT = 0$ is in agreement with the BCS prediction and conventional s-wave pairing.

We conclude that our surface impedance measurements on both $\kappa\text{-(BEDT-TTF)}_2\text{Cu}(\text{NCS})_2$ and $\kappa\text{-(BEDT-TTF)}_2\text{Cu}[\text{N}(\text{CN})_2]\text{Br}$ can be described in all respects by the BCS theory. A more detailed discussion can be found in [12].

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