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Scaling in Angle Resolved Electrical Transport Measurements on \(\kappa-(\text{BEDT-TTF})_2\text{Cu(NCS)}_2\)

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Abstract. — We report on single crystal electrical transport measurements on \(\kappa-(\text{BEDT-TTF})_2\text{Cu(NCS)}_2\) in an external magnetic field applied at different angles with respect to the superconducting layers. By measuring the in-plane resistivity \(\text{versus}\) temperature we can reveal a thermally activated behavior in the vortex liquid phase and are able to extract the angular dependence of the activation energy. Voltage \(\text{versus}\) current measurements lead to angle dependent values for the critical current \(J_c\). Our data can be fitted successfully within the framework of the anisotropic effective mass model resulting in an estimate for the anisotropy parameter \(\gamma = \xi_{ab}/\xi_c \approx 40 - 110\).

1. Introduction

Anisotropic superconductivity has attracted wide interest for a long time from the theoretical as well as from the experimental point of view. Several experiments have been carried out, first on “artificially” anisotropic superconductors, \(\text{i.e.}\) multilayers of conventional superconductors [1], then on intrinsically anisotropic superconductors, \(\text{i.e.}\) high-\(T_c\) superconducting cuprates [2-4]. Several models were proposed according to the wide range of anisotropy existing both in extrinsically and intrinsically anisotropic superconductors [5].

Intrinsically anisotropic superconductivity is also observed in the organic superconductors \(\kappa-(\text{BEDT-TTF})_2X\) [6]. In this paper we will report measurements on \(\kappa-(\text{BEDT-TTF})_2\text{Cu(NCS)}_2\). This compound can be regarded as a model compound for the high-\(T_c\) superconductors, as it exhibits a similar crystal structure of alternating conducting and nonconducting layers. The conducting planes consist of the organic BEDT-TTF cations; conduction arises from the delocalized \(\pi\)-doublebindings of the BEDT-TTF’s central carbon atoms. The anorganic \(\text{Cu(NCS)}_2\) anions are polymerized and form insulating planes. As \(T_c \approx 10\) K and \(H_{c2}(0) \leq 200\) kOe, the complete superconducting phase is accessible in experiment. To date, highly contradictory values for the anisotropy parameter \(\gamma\) have been found for this compound, ranging from \(\gamma\) of the order of 10 in magnetization [7] and resistivity [8] measurements to \(\gamma > 200\)

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extracted from experiments on the magnetic torque [9], ac-susceptibility [10] and specific heat [11]. Hence, this crucial point seems still to be an open question.

In previous papers we have presented electrical transport measurements of the in-plane resistivity in magnetic fields applied perpendicular to the superconducting layers. We have shown the effect of the anisotropic superconducting properties on thermal fluctuations [12] and on the vortex dynamics in the vortex liquid state [13]. The latter can be explained in the picture of a very viscous vortex liquid with an anisotropy parameter $\gamma \approx 110 - 130$. In this paper we further investigate the anisotropic properties of $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$ by angle resolved $\rho(T)$ and $V(I)$ measurements. We compare the new data to the measurements at perpendicular fields employing different models. This also leads to an estimate for the anisotropy parameter $\gamma$.

2. Experimental Details

The measurements presented here were performed on a high-quality single crystal of $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$ grown in a standard electrocrystallization process [14]. Its size is about $125 \times 400 \times 20 \mu m^3$. On top of the platelike sample, four gold contacts were evaporated using a shadow mask technique. The two outer pads (current contacts) also cover the two outer sides of the crystals to short-circuit the superconducting planes and ensure an homogeneous current flow. The measurements were carried out in a rotatable sample holder with a relative angular resolution of 0.2°. The external magnetic field was always applied perpendicular to the current flow, but with variable angle $\Theta$ between the superconducting layers and the field direction. A standard four probe setup was used. The $\rho(T)$ characteristics were measured employing a low frequency lock-in technique, whereas for the $V(I)$ curves a pulsed dc-method was used to minimize heating effects. For the latter the voltage drop over the sample was determined with a Keithley K191 nanovoltmeter. For temperature measurements, a carbon-glass resistor was employed, which was carefully calibrated against shifts in the presence of a magnetic field.

3. Results and Discussion

3.1. Resistivity versus Temperature Measurements. — In this section we will concentrate on the temperature dependence of the resistivity with respect to the angle between the magnetic field and the superconducting layers. Measurements were performed at different external fields. Figure 1 gives an example of the data at $H = 12$ kOe. In this Arrhenius representation it can be clearly noticed, that the resistivity is thermally activated at all angles. As reported previously [13] for the case of a magnetic field applied perpendicular to the superconducting planes ($\Theta = 90^\circ$), this behavior can be understood in the picture of thermally activated flux flow (TAFF). The resistivity can then be described as $\rho(T) = \rho_0 \exp(-U(H,T)/k_B T)$, where $U(H,T)$ denotes the activation energy and $k_B$ the Boltzmann constant. In reference [13] we were able to show, that the activation energy $U(H,T)$ can be ascribed to energy barriers arising from plastic deformations in the vortex liquid, i.e. the creation of double kinks in the three dimensional flux lines. It then follows $U(H,T) = U_{pl} = U_0 (1 - T/T_C)$, where $T_C$ denotes the transition temperature, and $U_0 \propto H^{-1/2}$. We will now analyze the angular dependent data in the same way as the data at $\Theta = 90^\circ$. For this purpose, we employ linear fits to the apparently linear parts of the curves and derive the angular dependent activation energy. These are plotted in Figure 2 for $H = 12$ kOe and another field investigated ($H = 450$ Oe). It turns out, that the activation energy increases slowly from its perpendicular ($\Theta = 90^\circ$) value and has a rather sharp peak at $\Theta = 0$. To understand this behavior, we further analyze these data employing two models designed to describe anisotropic, or layered superconductors,
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Fig. 1. — Arrhenius-plot of the resistivity versus temperature characteristics. Solid lines: in a magnetic field $H = 12$ kOe applied at different angles $\Theta$ with respect to the superconducting planes. The angle between the field and the planes increases from left to right. For comparison, also the curve for $H \approx 0$ is plotted (dashed line).

Fig. 2. — Angular dependence of the activation energy for $H = 12$ kOe and $H = 450$ Oe, respectively.

respectively. The first model was proposed by Kes et al. [15] for layered superconductors: it predicts a breakdown of the concept of the usual 3D flux line structures when the magnetic field is applied parallel to the superconducting planes. For an arbitrarily oriented field only the perpendicular component of the magnetic field should be relevant for the formation of the flux-line-structure. This idea leads to a scaling formula for all field and angular dependent quantities $Q$:

$$Q(H, \Theta) = Q(H \sin \Theta, \Theta = 90^\circ),$$

i.e. the behavior at different angles and fields of a certain quantity $Q$ is completely described by its dependence on field directed perpendicular to the planes.

The second model we will employ is the anisotropic effective mass model, which follows from the Lawrence-Donnach equations [16]. This model is further generalized by the scaling approach proposed by Blatter et al. [17]. Here again the magnetic field is scaled with an angular dependent scaling function $\varepsilon(\Theta)$,

$$Q(H, \Theta) = Q(H \varepsilon(\Theta), \Theta = 90^\circ).$$
Fig. 3. — Activation energy dependence on the magnetic field for fields applied perpendicular to the layers (closed symbols) and at different angles Θ with respect to the planes (open symbols). The magnetic field for the angular dependent values is scaled according to the models explained in the text. The lines are guides to the eye. The bar represents the error due to the experimental uncertainty in angle and field.

with

$$\varepsilon(\Theta) = \sqrt{1/\gamma^2 * \cos^2 \Theta + \sin^2 \Theta},$$

(3)

where γ denotes the anisotropy parameter. γ = \xi_{ab}/\xi_{c} = \sqrt{m_c/m_{ab}}. \xi_{ab}, \xi_{c} and m_{ab}, m_{c} are the in-plane and out-of-plane coherence lengths and effective masses, respectively. It can easily be seen, that the first model follows formally from the second by setting γ to infinity, as already pointed out by Blatter et al. [17]. This can be understood within the framework of the anisotropic effective mass model as neglecting the interlayer coupling, i.e. setting the effective mass m_{c} for motions perpendicular to the layers to infinity.

In Figure 3 we plot the activation energy found in measurements with the magnetic field applied perpendicular to the planes and additionally the angle resolved activation energies at 12 kOe, for which the magnetic field is scaled for different values of γ following equations (1, 2, 3). It turns out, that the overall fitting quality is quite convincing. The fits for different γ differ considerably only at low fields (i.e. Θ → 0), whereas at higher fields (Θ → 90°) the \cos^2 \Theta term in the scaling function (3) becomes negligible and both models result in approximately the same behavior. In the regime, where the field is directed almost parallel to the layers, the best fit can be achieved with a value of γ ≈ 70. Values for γ in the range 40 ≤ γ ≤ 110 still result in fits, which lie well within the error resulting from our experimental resolution in angle and in field (for low applied fields). Furthermore, the model of Kes et al. (or γ = ∞) does not seem to be appropriate to describe our data for Θ → 0. At the lower field, \( H = 450 \text{ Oe} \), similar results could be achieved in an analogous analysis (not shown here).

3.2. Voltage versus Current Measurements. — We now turn to angle resolved measurements of \( V(I) \) characteristics and analyze the critical current determined from these curves in the same context as before. We measured \( V(I) \) curves at different external magnetic fields (12 kOe, 48 kOe and 96 kOe) and temperatures (4.4 K, 5.0 K, 6.0 K and 7.0 K). Typical curves at 4.4 K and 48 kOe are displayed for some angles in Figure 4. From these curves, we extract the angle dependent critical current. We do not employ a voltage criterion, but take the intercept of a linear fit to the high-current parts of the curves with the x-axis [18].
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**Fig. 4.** — Voltage \textit{versus} current curves at 4.4 K and 48 kOe for different angles of the applied magnetic field.

**Fig. 5.** — Angular dependence of the critical current at \( T = 4.4 \) K.

This decision was motivated by the fact that the curves are rather rounded, seemingly due to the thermally activated flux flow. Accordingly, the determination of the critical current by a voltage criterion would result in too low values. The resulting values at \( T = 4.4 \) K are shown in Figure 5. Their dependence on angle is rather similar to the behavior of the activation energy described above. Again, the critical current increases slowly from its \( \Theta = 90^\circ \) value and has a rather sharp peak at \( \Theta = 0^\circ \). This general feature also turns out at the other temperatures investigated (not shown here).

We now compare the angle resolved critical currents to the values found for \( H \) applied perpendicular to the superconducting planes. Figure 6 shows the critical currents at \( \Theta = 90^\circ \) and \( T = 4.4 \) K for different fields, derived in the same way as described above. Additionally, we plot the angle dependent values at 12 kOe, 48 kOe and 96 kOe with scaled values of the magnetic field for different \( \gamma \). Again, the overall fitting quality is quite convincing. But as in the previous section — due to the scatter in the data — it can not unequivocally be decided, which \( \gamma \) results in the best fit. Nevertheless, values of \( \gamma \) lower than about 40 seem to be excluded. But in contrary to the \( \rho(T) \) measurements, the model of Kes \textit{et al.} seems not entirely inappropriate to describe the data, also taking into account the accuracy in the determination of the critical currents employing the method described above.
3.3. DISCUSSION. — In this section we want to compare our results to the values for \( \gamma \) and to angle resolved measurements reported in literature. First, compared to the values given by magnetization [7] and resistivity [8] measurements, where values of \( \gamma \approx 10 - 20 \) are reported, our lower limit of 40 seems to be too large. This may be due to the fact, that for example in resistivity measurements, \( \gamma \) is usually determined from the anisotropy in the upper critical magnetic field \( H_{c2} \) perpendicular and parallel to the layers. Here, errors may arise from a) the method employed to determine \( H_{c2} \) from the measured curves b) the accuracy, which can be achieved to align the magnetic field parallel to the superconducting layers and c) the fact that \( H_{c2,c}(0) \) is Pauli limited to about 190 kOe \( (H_{\text{pauli}} = 18.4 \text{ kOe/K}\times T_c \ [19]) \). In contrary, our method does not test the upper critical field and does not depend on a perfect alignment in the parallel position, but involves several values close to \( \Theta = 0 \).

In fact, angle resolved measurements of the magnetic torque [9] and ac-susceptibility [10] reveal much larger values of \( \gamma \). Farrell et al. [9] find a value of \( \gamma \approx 200 \), Mansky et al. [10] \( \gamma = 160 - 350 \). These values are not far from our values found for the angle resolved \( \rho(T) \) measurements, and fit perfectly into our \( J_{c}(\Theta) \) dependence. Accordingly, we believe that it is not the measuring method (resistivity, magnetization, ac-susceptibility etc.) which is decisive for the value found for \( \gamma \), but instead, whether the angle dependence of the measured value is taken into account. The more reliable values seem to result from angle dependent measurements.

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**Fig. 6.** — Critical current dependence on the magnetic field at \( T = 4.4 \text{ K} \) for fields applied perpendicular to the layers (closed symbols) and at different angles \( \Theta \) with respect to the planes (open symbols). The magnetic field for the angular dependent values is scaled according to the models explained in the text. The solid line for the perpendicular values is a guide to the eye.
4. Conclusion

In summary, we have presented angle resolved measurements of $\rho(T)$ and $V(I)$ characteristics in order to determine the anisotropic properties of $\kappa$-(BEDT-TTF)$_2$Cu(NCS)$_2$. The $\rho(T)$ measurements reveal activated behavior at low temperatures; the angular dependence of the activation energy can be fitted by scaling the magnetic field according to the anisotropic effective mass model. The same property was found for the angle dependence of the critical currents. Both methods result in a lower limit for $\gamma$ of about 40. From $\rho(T; H, \Theta)$ measurements an upper limit of $\gamma \approx 110$ could be estimated, whereas the $V(I; T, H, \Theta)$ characteristics do not agree with such an estimate.

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[18] A more accurate way to determine the critical currents was to fit the $V(I)$ characteristics to the appropriate theoretical models. The results of fits in the TAFF picture do not differ considerably from the values determined by the simple method described above. They will be presented elsewhere: Pasquier C. and Friemel S., in preparation.