Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl16

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Version of record first published: 17 Oct 2011.

To cite this article: J. R. Cooper , L. Forro & B. Korin-hamzić (1985): Magnetoresistance of the Organic Superconductor $(TMTSF)_2ClO_4$:Kohler's Rule, Molecular Crystals and Liquid Crystals, 119:1, 121-129

To link to this article: http://dx.doi.org/10.1080/00268948508075145

Mol. Cryst. Liq. Cryst. 1985, Vol. 119, pp. 121-129
0026-8941/85/1194-0121/$15.00/0

Printed in the United States of America

MAGNETORESISTANCE OF THE ORGANIC SUPERCONDUCTOR $(TMTSF)_2ClO_4$:KOHLER'S RULE

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Abstract - We report a study of Kohler's rule for single crystals of $(TMTSF)_2ClO_4$ in the temperature range 2-22K and fields up to 7.8T for current flow parallel (a) and perpendicular (c*) to the conducting chains.

INTRODUCTION

Recent magnetoresistance measurements[1,2] on single crystals of $(TMTSF)_2ClO_4$ in the relaxed (R) state show that Kohler's rule (KR) is obeyed for $\underline{J} \| \underline{c}^*$, $\underline{H} \| \underline{b}'$ but not for $\underline{J} \| \underline{a}$, $\underline{H} \| \underline{c}^*$[3].

A textbook example[4] of KR for magnesium is shown in Figure 1. KR follows from the Boltzmann equation describing transport by electrons at the Fermi surface (FS) in the relaxation time (τ) approximation. The deflection of carriers between collisions depends on the product $H\tau$. If $\rho_o \propto \tau^{-1}$ as is usually so for metals, then the relative change in resistivity ($\Delta\rho$) over the zero field value (ρ_o) is a universal function of H/ρ_o.

The observation of KR indicates that several features of the experimental results can be understood within the framework of a classical theory based on the Boltzmann equation, as used for ordinary metals such as copper. At the present time it is not at all obvious that other properties such as anomalous optical reflectivity[5] and NMR data[6] can be accounted for in the same picture although this may in fact be possible[7]. In any case the methodology described here and in other papers[1,2,8] should be of interest and perhaps also relevant to other organic metals e.g. the BEDTTF series.

From Figure 2 it can be seen that $\rho_\perp$ is strongly increased by

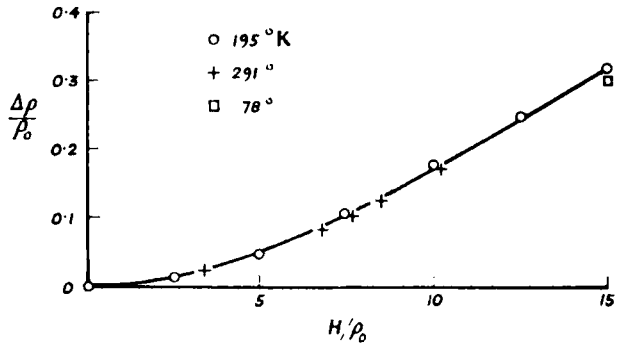


FIGURE 1 Textbook[4] example showing Kohler's rule for magnesium.

a magnetic field. Initially we thought that this was the effect we had been looking for, namely an increase in "one dimensionality" with magnetic field[9]. But we later disregarded this interpretation because it seemed to be inconsistent with the KR behaviour and a classical single particle picture. However recent theoretical work [10,11] indicates that this is not true and does indeed attribute the field induced spin density wave state to an increase in "one dimensionality" with H, but only for $\underline{H} \parallel \underline{c}^*$ and not for $\underline{H} \parallel \underline{b}''$.

In Figure 2 $\rho_{\perp}$ (T) ie. ρ_{c^*}, of R samples shows changes in slope on a log-log plot at 25K and 3.5K both of which are reproducible[2]. Although ρ_{c^*} samples can be as small as 0.3x0.2x0.1 mm^3 along $\underline{a}$, $\underline{b}''$ and $\underline{c}^*$ and are thus difficult to contact they generally do not show irreversible resistance jumps on cooling.

An example of Kohler plots for ρ_{c^*} is shown in Figure 3. These are newer results for a crystal from another preparation batch, with the more usual morphology, the largest face being an (001) plane, in the R state (6 hours from 42 to 4.2K). The corresponding results for ρ_{c^*}(T) and the quenched (Q) state are also given in these Proceedings[2]. It is reassuring to see that KR is again obeyed. $\Delta\rho/\rho_0$ is about a factor of 5 smaller than before[1], but this only

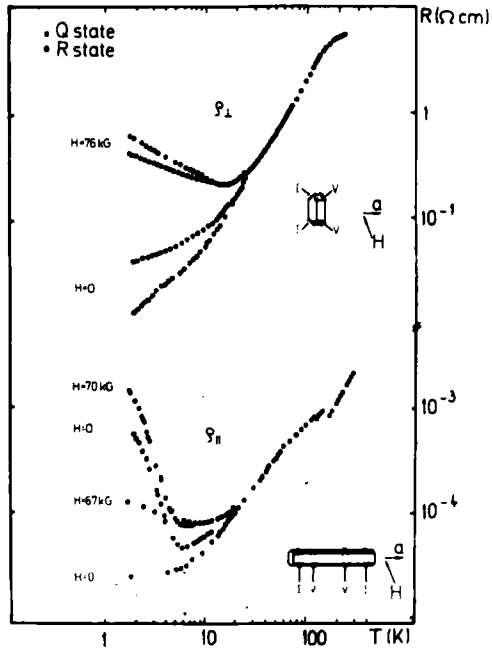


FIGURE 2 Overall view of resistivity data for $\rho_\perp$ ($\underline{c}^*$) and $\rho_\parallel$ ($\underline{a}$) for $(TMTSF)_2ClO_4$ from ref. 1.

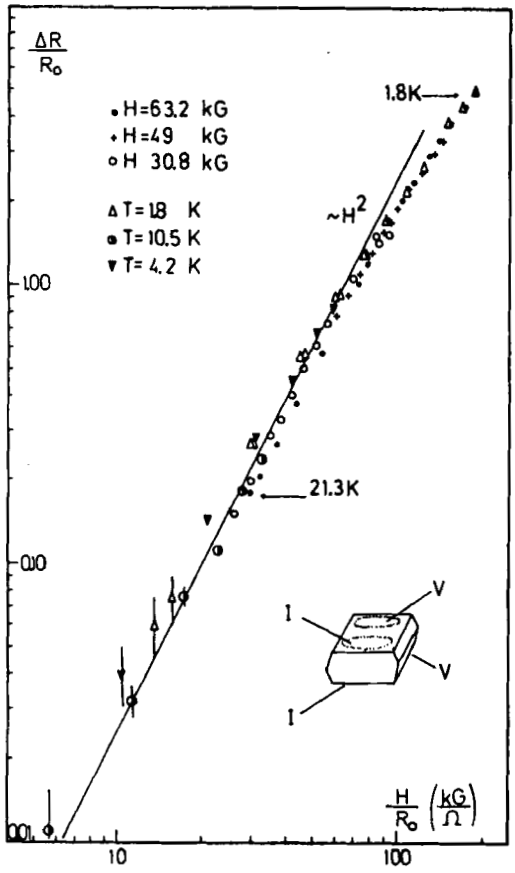
corresponds to τ being 2.2 times smaller and indeed agrees with the ratio of the measured values of $\sigma_\perp(2)$.

CALCULATION OF $\sigma_{ij}(H)$

Following the observation of KR we made analytical calculations of the conductivity tensor for an open tight binding FS using the standard formula[12]:

$$\sigma_{ij} = -\frac{e^2}{4\pi^3} \int \frac{ds}{\hbar|V_k|} \int_0^\infty V_i(0)V_j(t)\, e^{-t/\tau} dt \qquad (1)$$

Here $\int ds$ is an integral over the FS which can be transformed to one over $k_\perp$. V_k is the FS velocity, $\hbar$ Planck's constant and e the



FIGUPE 3 Kohler plots for ρ_{c^*} for a crystal of $(TMTSF)_2ClO_4$ in the relaxed state, for $\underline{H} \parallel \underline{b}$. A sketch of the sample is shown, the largest face being an 001 plane.

electron charge. $V_k(t)$ is determined from the time dependence of $\underline{k}$ caused by the Lorentz force. Initially we assumed orthorhombic symmetry, but later[1] in an attempt to account for $\rho_{aa}(H)$ via a non zero value of $\sigma_{ab^*}(0)$, we took $a<b = 70^\circ$ while the other two triclinic angles were still taken to be 90°. The physical insight given by these calculations complements recent numerical work[13] taking the full triclinic symmetry into account. It may be important to use an accurate a band structure as possible[1,4] because $\sigma_{ab^*}(0)$

seems[1] to be sensitive to b* zone boundary effects. The main result is that for $\underline{H} \| \underline{b}'$

$$\sigma_{c^*c^*} = \sigma_{c^*c^*}(0)\,[1 - \Omega^2 \tau^2] \qquad (2)$$

Equations[1,8] for the other components of σ will not be repeated here.

Using the simplest approximation that τ is independent of $\underline{k}$ and of the direction of $\underline{J}$ (i.e. the direction in which the FS is displaced from equilibrium)

$$\frac{\sigma_{c^*c^*}}{\sigma_{aa}} = \frac{\overline{V_{c^*}^2}}{\overline{V_a^2}} = \left[\frac{t_\perp c}{t_\parallel}\right]^2 \frac{c^2}{a^2} \qquad (3a)$$

$$\sigma_{aa} = \frac{2}{\pi}\frac{e^2}{\hbar c}\frac{V_a}{b}\tau \qquad (3b)$$

where[3] a=3.63Å, b=7.68Å and c=13.3 Å.

If τ is indeed a constant than the relative change in $\sigma_{c^*c^*}$ depends on[1]

$$\Omega\tau = \frac{e\,c\,H}{\hbar\,c'}\,V_a\tau = \frac{ecH\,\ell_\parallel}{\hbar c'} \qquad (4)$$

where c' is the velocity of light. Note that in this simple approximation the anisotropy is also temperature independent.

SAMPLE GEOMETRY

In dealing with such anisotropic conductors the concept of the equivalent isotropic sample[15] (EIS) used during early work on TTF-TCNQ[16], is often convenient. It provides a simple way of thinking about the current distribution in the anisotropic sample. The EIS is generated by a coordinate transformation according to the formulae[15] $\ell_i' = \ell_i \sqrt{\sigma/\sigma_i}$, $\sigma = \sqrt[3]{\sigma_1\,\sigma_2\,\sigma_3}$, where ℓ_i is the sample dimension along the i-th principal axis of σ. Examples of EIS are shown in Figure 4 for the typical crystal shapes used in our work for ρ_{c^*} and ρ_a with typical anisotropy ratios 500:10:0.2 $(\Omega cm)^{-1}$

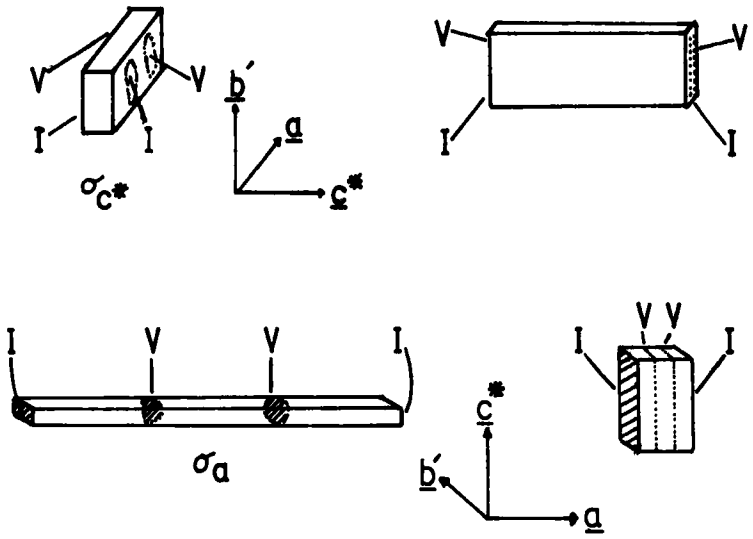


FIGURE 4 Comparison of the shapes of the equivalent isotropic samples, on the right, with those of the crystals, on the left, for σ_{c^*} (upper) and σ_a (lower). The samples are drawn to scale for dimensions a,b,c of 0.3x0.2x0.1 mm^3 for σ_{c^*} and 3x0.2x0.1 mm^3 for σ_a, and conductivity ratios of 2500:50:1.*

It is clear that for ρ_{c^*} samples the direction of $\underline{J}$, i.e. the geometry, is better defined than it is for ρ_a (or for that matter ρ_b.) Strictly speaking the concept of an EIS is not valid in a magnetic field because of the Hall field, and ideally the current-voltage distribution should be worked out "ab initio" in the plane $\perp$ to H. However it is reasonable to assume that the EIS concept holds until the Hall field (Lorentz force) becomes an appreciable fraction of the applied electric field. In practice we also checked experimentally that for ρ_{c^*} samples with current contacts in the $\underline{a}$ direction, there was no detectable voltage at low T and high fields which also confirms that the EIS is long and thin.

COMPARISON WITH EXPERIMENTAL RESULTS FOR $\rho_{c^*}(H)$ AND $\rho_a(H)$

According to the above argument $\perp$ is $\parallel$ to c^* in a magnetic field for the ρ_{c^*} sample. It turns out that the offdiagonal (Hall) components of σ are negligible so that $\Delta\rho_\perp/\rho_\perp = \Omega^2\tau^2$. For the two samples studied in detail (ref.1 and Figure 3) at 2K and 1T, $\Delta\rho/\rho = 1$ and 0.2 respectively. From equation 4 this implies that $\ell_{\parallel} = 1400$ a or 620 a at 2K. These values of $\ell_{\parallel}$ are in good agreement with those estimated from the magnitude of $\rho_{\parallel}(2)$ on samples from the same batch, namely $\ell_{\parallel} = 300\text{-}3000$ a (17) for residual resistance ratios ranging from 200-2000. The corresponding τ values (taking $t_{\parallel} = 0.3$eV) at 2K are 2.10^{-12} and $0.89.10^{-12}$ secs in good agreement with those found[13] from σ_{bb} at 0.5K. Furthermore the ratio of these values of τ is nearly the same as that of the measured values of $\sigma_\perp(2)$, (100 and 55 $(\Omega cm)^{-1}$). Knowing τ and $\sigma_\perp$ at 2K and using equations 3 leads to $t_\perp^c = 1.1 \pm 0.1$ meV[17] in both cases, in good agreement with theoretical estimates[18]. Values of $t_\perp^c$ in the Q state are also within the above limits but a similar procedure for the PF_6 salt leads to $t_\perp^c = 0.16$ meV[2,17].

As reported previously[1,19] KR is not obeyed for R samples with $\underline{H} \parallel \underline{c}^*$ and $\underline{J} \parallel \underline{a}$. There are also difficulties in accounting for the magnitude of the observed effect. Nevertheless there are three points which favour some kind of single particle explanation (i) in the limited metallic region available (5.5 to 12K) the Q sample does obey KR (ii) in fixed fields $\Delta\rho_{\parallel}/\rho_{\parallel}$ for the R samples[1,19] goes as ρ_o^{-2} (iii) at high fields and higher temperatures the results for the R sample tend to the Kohler line of the Q sample.

The differences between the R and Q states lead us to believe that magnetic breakdown across the superlattice gap associated with anion ordering may be important[1].

If one draws the superlattice gaps on the original FS without halving the Brillouin zone, then it is clear that for $\underline{H} \parallel \underline{c}^*$ the

Lorentz force will drive electrons "through" the gaps. These gaps are not effective when $\underline{H} \| \underline{b}'$ which is why ρ_{c^*} obeys KR. However this interpretation now needs more experimental support. It does not seem to be consistent with the result[13] that ρ_b follows an H^2 law at 0.5K. For $\underline{H} \| \underline{b}'$ and $\underline{J} \| \underline{a}$ the shape of the equivalent isotropic sample in Figure 4 indicates that the direction of $\underline{J}$ may not be very well defined unless the current contacts are extremely uniform and thus that geometrical effects could be troublesome. Note also that we did not use end contacts for $\underline{J} \| \underline{a}$.

In summary we believe that several features of the magnetoresistance of single crystals of $(TMTSF)_2ClO_4$ can be understood in terms of the classical picture. However many of the details are still not understood, for example the resistance anomaly at 3.5K. The geometry $\underline{J} \| \underline{c}^*$, $\underline{H} \| \underline{b}'$ is a particularly good one. In the region where Kohler's rule is obeyed, it can be used to estimate the mean free path along the chains, the relaxation time τ, the interchain overlap integral t_1^c, and the average mean free path along $\underline{c}$. Deviations from Kohler's rule might be used to look for the coherent-diffusive transition[19] in the transverse conductivity[2], changes in band structure with magnetic field (magnetic breakdown) or temperature (band narrowing), changes in τ with magnetic field or pretransitional fluctuations.

ACKNOWLEDGEMENTS

We are grateful to our coworkers in Zagreb M.Miljak and K. Biljaković for their efficient co-operation in the experiments as well as many discussions. Discussions with S.Barišić, P.Chaikin, D.Jérome, K.Murata and T.Timusk are also gratefully acknowledged as well as with K. Bechgaard and A.Moradpour who also prepared the single crystals used in this work.

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