Short Communication

Quantized Hall effect in the organic superconductor \((\text{TMTSF})_2\text{ClO}_4\) revisited

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Abstract. — We report Hall resistance measurements of the organic conductor \((\text{TMTSF})_2\text{ClO}_4\) at low temperatures in fields up to 12 T. We have found that the Landau filling factor of the very stable semimetallic phase above 8 T is very likely to be \(n = 1\) instead of \(n = 1/3\) as inferred from previous investigations. A comparison between different samples shows that the possible current non-uniformity can strongly affect the experimental magnitude of the Hall resistance and thus leads to a wrong determination of the Landau filling factor.

The stabilization of field-induced spin density wave phases (FISDW) in linear-chain organic conductors of the tetramethyltetraselenafulvalinium series \((\text{TMTSF})_2X\), \(X =\) monovalent anion, has been a major by-product of the superconductivity discovered ten years ago in these compounds [1].

The basic physical reason underlying the instability of a quasi 1-D conductor (with a Fermi surface open along both transverse directions) against the formation of a FISDW is the magnetic field making the electron motion along the conducting chains more one-dimensional and therefore restoring the logarithmic divergence of the \(Q\)-dependent susceptibility [2].

A FISDW state can be considered as a condensate of electron-hole pairs with opposite spins building up the local magnetic modulation of wave vector \(Q\) together with a remaining density of unpaired carriers coming from the non-nested regions of the quasi 1-D Fermi surface.

The Fermi surface of the semimetallic FISDW phase exhibits a tube-like shape parallel to the \(c^*\) crystalline direction (\(c\) is the direction of the weakest conductivity). The cross-section of this semimetallic Fermi surface by a plane perpendicular to \(c^*\) is of the order of one hundredth the area of the first Brillouin zone.

The energy spectrum of the two-dimensional low density carriers becomes quantized by the magnetic field and gives rise to a discrete comb of Landau levels (assuming the coupling along the \(c\) direction to be negligible). A particularly stable configuration of this 2-D electron gas is achieved if the Fermi level lies in-between completely filled and completely empty Landau levels.

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as the diamagnetic energy is thus minimized. Such a favorable situation can be preserved over an extended field region provided the wave vector of the modulation is allowed to vary with $H$.

This argument constitutes the essence of the standard theory for the cascade of FISDW phases which predicts a component $Q_{\parallel}$ increasing linearly with $H$ in order to maintain the Fermi level within a Landau gap with the concomitant quantization of the Hall resistance at $h/2ne^2$ (12.9 kΩ for $n = 1$) per layer. However, as $Q$ departs from the value leading to the optimum nesting vector, the energy of the unpaired carriers increases and may overcome the gain in diamagnetic energy resulting from the pinning of the Fermi level in a Landau gap. Thus the Fermi level jumps down and establishes itself at a new value corresponding to exactly $n - 1$ filled levels. There is a series of first order transitions between FISDW phases, each with quantized Hall voltages and each labeled by successive integers $n$. These effects have been observed so far in three members of the $(\text{TMTSF})_2X$ family.

In $(\text{TMTSF})_2\text{PF}_6$ under pressure the quantization of the Hall resistance corresponding to $n = 6$ to 1 has been observed by two groups with even a remarkable quantitative agreement with the theoretical value $h/2ne^2(n = 1, 2, \text{ etc.})$ [3, 4].

According to a recent study of the FISDW in $(\text{TMTSF})_2\text{ReO}_4$ under pressure Hall plateaus corresponding to the theoretical value $H/2ne^2$ with $n = 1$ and 2 have also been observed. In both compounds an ultimate transition towards the $n = 0$ phase with a gap opening over the whole Fermi surface has been observed and is accompanied by a dramatic increase of the longitudinal resistivity [5].

The first observation of quantized Hall resistance has been performed in $(\text{TMTSF})_2\text{ClO}_4$ [6, 7] and actually gave rise to the idea of a theory based on the quantization of nesting [2]. However, the behaviour of this compound is different from the others in many respects:

i) Hall resistance steps with ratios 1 : 1/3 : 1/6 were observed at 7.5, 6.3 and 5.5 T.

ii) the phase above 7.5 T is extremely stable in magnetic field and extends up to about 25 T at low temperature.

Based on the ratio of the Hall resistance values a fractional quantum Hall effect ($n = 1/3$) was suggested as a possible explanation for this very stable phase, [8].

In the present communication we report the results of a new investigation of the Hall effect performed in several $(\text{TMTSF})_2\text{ClO}_4$ single crystals. Our data show that the field dependence of the Hall resistance and the various ratios between Hall resistance plateaus are qualitatively similar in all samples. However, we have found that the magnitude of the Hall resistance depends significantly on the homogeneity of the current distribution in a given sample. We come to the conclusion that the quantum number of the very stable phase (8 to 25 T) is $n = 1$ instead of 1/3 as claimed previously.

Two pairs of Hall contacts were made by gold evaporation using a masking technique. Gold was also evaporated on all sides at the ends of the crystals to achieve a current distribution in the sample as uniform as possible. Hall resistance data were obtained after averaging Hall voltages corresponding to both directions of the magnetic field. The measurements have been conducted on two slowly cooled single crystals (average cooling rate: 10 mK/min below 40 K) at the temperature $T = 0.35$ K up to fields of 12 T. No cracks were observed on cooling down. A current of 100 and 50μA is used for samples # 1 and # 2, respectively.

Figure 1 displays the field dependence of the Hall resistance for the two pairs of contacts in sample # 1 of thickness $t = 75$ μm. The Hall voltage is negative (electron-like) at high fields and becomes large only above the threshold field of $\approx 4$ T. The sign reversal which is observed between 6 and 7 T is the signature of a well relaxed sample [9]. The magnitude of the Hall resistance at the largest plateau amounts to 12.9 ± 0.5 and 16.3 ± 0.5 kΩ/molecular layer for contacts a and b respectively. Both experimental values are very close to the theory $h/2e^2 = 12.9$ kΩ per layer.
Fig. 1. — Hall resistance at 0.35 K vs. magnetic field along $c^*$ direction for two pairs (a), (b) of opposite contacts on sample #1. The sample dimension is $5.75 \times 0.38 \times 0.075$ mm$^3$. The quantized value $\hbar/2e^2$ (12.9 kΩ) per molecular layer is marked on the right for contact (a).

Fig. 2. — Hall resistance at 0.35 K of sample #2. Experimental conditions are identical as sample #1. The sample dimension is $4.5 \times 0.38 \times 0.225$ mm$^3$. $\hbar/2e^2$ per molecular layer corresponds to 3.8 μV for this sample.
for a phase $n = 1$. Another sample (# 2) with a thickness $t = 225 \mu m$ has given quite a similar field dependence of the Hall resistance by very different values for its magnitude, figure 2. For instance the experimental value of the Hall resistance at the largest plateau is significantly smaller than the value $h/2e^2$. These two examples have shown that one must be extremely careful about the indexation of Hall plateaus with a given quantum number.

![Graph](image)

**Fig. 3.** — Low field hole-like data in the metallic state of sample # 1.

There is no doubt that the main problem lies in the uniformity of the current distribution throughout the whole sample cross section. We guess that better conditions, as far as a homogeneous current flow is concerned, are met for thin and long samples. In this respect sample # 1 is about 3 times thinner than sample # 2. However, another mean to ascertain the current is indeed flowing homogeneously through the sample is to perform a measurement of the Hall voltage in the metallic phase, below 4 T, when both the density and the sign of the carriers are known.

Data for the low field Hall voltage of sample # 1 are displayed in figure 3. The sign is positive in agreement with hole carriers and the magnitude of the linearly field dependent Hall voltage with $R_H = 4.6 \times 10^{-9} m^3/C$ corresponds remarkably well to the theoretical expression of the Hall constant for a quarter-filled hole tight binding band $R_H = k_Fa/N e \tan k_Fa$ with $k_Fa = \pi/4$. Using $N$ corresponding to one hole per unit cell (694.4 Å$^3$) we obtain $R_H = +3.4 \times 10^{-9} m^3/C$. The deviation from the theoretical value seems to come from an overestimation of the sample thickness since the current is hardly perfectly homogeneous in this highly anisotropic metallic state. However, the current will be more homogeneous in FISDW states as the longitudinal conductivity decreases rapidly and the conductivity anisotropy is not as large as the one of the metallic state. The low field Hall data of sample # 2 do not agree with the theory by a factor 2 at minimum. We are thus led to the conclusion that a proper indexation of the Hall plateaus cannot be trusted unless the measurement of the low field Hall voltage is performed on the same sample. Another recent publication of Hall effect data in (TMTSF)$_2$ClO$_4$ do also lead to an $n = 1$ very stable state although the authors of that work have claimed the state resembles the 1/3 fractional quantized Hall effect [10].

To summarize, the reinvestigation of the Hall resistance in the FISDW phases of (TMTSF)$_2$ ClO$_4$ has shown that the indexation of the Hall plateaus cannot be based on the only knowledge of the experimental Hall resistance and sample thickness. The current flow may be highly non-
uniform in these very anisotropic conductors. Therefore a good check for the current uniformity is the experimental value of the Hall constant in the metallic phase ($H < 4$ T). We have reached the conclusion that the very stable FISDW phase between 8 and 25 T must be labeled $n = 1$ instead of $1/3$ as claimed previously [8]. The $n = 1$ indexation of the stable phase does not affect the relation between the magnetic field dependent specific heat and the phase diagram $T_c(H)$ which are model independent [11].

The comparison of Hall data between FISDW phases of different (TMTSF)$_2$X compounds shows that the behaviour of (TMTSF)$_2$PF$_6$ is in excellent agreement with the predictions of the quantized nesting model. At high fields (TMTSF)$_2$ReO$_4$ is also in agreement with the standard theory. However, the data of (TMTSF)$_2$ClO$_4$ reveal an absence of integer plateaus ($n = 2, 4, ...$), the extreme stability of the phase $n = 1$ and the apparent absence of the $n = 0$ phase. It becomes a crucial experiment to investigate whether the reentrance of the non-distorted phase in (TMTSF)$_2$ClO$_4$ at very high fields is linked to the existence of unpaired carriers in the $n = 1$ phase or/and whether the reentrance is also present in materials such as (TMTSF)$_2$PF$_6$ where the $n = 0$ phase has been seen to become stable at high fields.

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References


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