The Quantum Hall Effect in quasi-1D conductors

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Abstract

The theory and experiments showing Quantum Hall effect in the quasi-one-dimensional conductors of the Bechgaard salts family are briefly reviewed. The sign reversals observed under some experimental conditions are explained within the framework of the Quantized Nesting Model. The sequence of reversals is driven by slight modifications of the geometry of the Fermi surface. It is explained why only even phases can have sign reversals and why negative phases are less stable than positive ones.

keywords: Many-body and quasiparticle theories, Hall effect, Magnetotransport, Magnetic phase transitions, Organic conductors, Bechgaard salts

1. Introduction

The organic conductors of the Bechgaard salts family \((\text{TMTSF})_2X\) where \(\text{TMTSF}\) = tetramethylenefulvalene) have remarkable properties in a magnetic field. Although these compounds are metals with a large number of carriers \(N\), they exhibit Quantum Hall Effect which normally would require a small filling factor \(\nu\).

Moreover, the Fermi surface of these systems is open, made of two almost planar sheets so that no orbital quantization is expected.\(^\text{1}\)

The quantization of Hall effect in these materials results from the magnetic field induced low temperature instability of the metallic phase versus the formation of an ordered state in which a gap opens close to the Fermi level. As a result, the ordered phase (a Spin Density Wave state) contains a much smaller number of carriers above (electrons) or below (holes) the Fermi level. These carriers form closed pockets which are quantized into Landau levels, giving rise to the Quantum Hall Effect.\(^\text{2}\)

The Bechgaard salts are strongly anisotropic systems, with a typical hierarchy of transfer integrals: \(t_a \approx 3000 K, t_b \approx 300 K, t_c \approx 10 K\). In three members of this family \((X = \text{ClO}_4, \text{PF}_6, \text{ReO}_4)\), the metallic phase is destroyed by a moderate magnetic field \(H\) applied along the \(c^*\) direction, perpendicular to the most conducting planes \((a, b)\).

The Field Induced phase consists in a series of Spin Density Wave (FISDW) subphases, separated by first order transitions.\(^\text{3}\) This field induced cascade of quantized phases results from an interplay between the nesting properties of the Fermi surface and the quantization of electronic orbits in the field: the wave vector of the SDW adjusts itself with the field so that unpaired carriers in the subphases always fill an integer number of Landau levels. As a result, the number of carriers in each subphase is quantized and so is the Hall effect: \(\sigma_{xy} = 2Ne^2/h\) (a factor 2 accounts for spin degeneracy).\(^\text{4}\) The apparition of these phases results from a new structure of the metallic phase in a field: because of the Lorentz force, the electronic motion becomes periodic and confined along the direction of the chains of high conductivity (a direction). As a result of this effective reduction of dimensionality, the metallic phase becomes unstable.\(^\text{5}\) In addition, the electrons experience a periodic motion in real space, characterized by the wave vector \(G = eHb/h, b\) being the interchain distance. Consequently, the spin susceptibility, instead of having one logarithmic divergence at \(2k_F\), exhibits a series of divergences at quantized values of the longitudinal component of the wave vector \(Q_x = 2k_F + nG\). The largest divergence signals the appearance of a SDW phase with quantized vector \(Q_y = 2k_F + NG\). These ideas have been formalized in the so-called Quantized Nesting Model which describes most of the features of the phase diagram in a magnetic field, in particular the observed Hall plateaus.\(^\text{6}\)

In this paper, we review the main features of the Quantized Nesting Model which describes the observed cascade of FISDW transitions. Then we generalize this model to explain the change in sign in the Hall plateaus which is observed in the salts \(X = \text{PF}_6, \text{ClO}_4, \text{ReO}_4\) under certain conditions.\(^\text{7}\)
2. The Quantized Nesting Model

The model which describes the FISDW starts from the effective Hamiltonian for the metallic phase:

\[ H = v_F (k_x - k_y) + t_\perp (k_y b) \]  

(1)

\( t_\perp (k_y b) \) is a periodic function which describes a warped Fermi surface. It satisfies the properties \( t_\perp (p + 2\pi) = t_\perp (p) \) and \( t_\perp (-p) = t_\perp (p) \). More specifically the following function has been chosen:

\[ t_\perp (p) = -2t_0 \cos p - 2t_0' \cos 2p \]  

(2)

Although it is essential to explain the existence of a threshold field for the cascade of FISDW, the coupling in the third direction is omitted since it is known that it does not play an important role in the sequence of subphases\([2, 11]\). This dispersion relation contains the minimum number of parameters to describe the FISDW cascade. The first harmonics \( (t_0) \) of the dispersion along the transverse direction describe the warping of the FS with a perfect nesting at wave vector \( (2k_F, \pi/b) \). The second harmonics \( (t_0') \) induces a deviation from perfect nesting which leaves a small number of carriers quantized into Landau bands. Its amplitude fixes the period of the cascade\([12,13]\). Typically in Bechgaard salts, \( t_0' \approx 10K \). This term may have two origins. One is the linearization of the dispersion relation along the \( \pi \) direction and is given by \( t_0' = -\left( \cos k_F a/4 \sin^2 k_F a \right) t_0/b \). Since the band is 3/4 filled, \( k_F a = 3\pi/4a \) and \( t_0' \) is positive. Other contributions may result directly from next nearest neighbor coupling\([13]\). The instability of the metallic phase can be described by the spin susceptibility \( \chi_0 \) whose maximum gives access to the wave vector of the ordered phase. It is given by

\[ \chi_0(Q) = \sum_n I_n(Q) \chi_0^{1D}(Q^4 - nG) \]  

(3)

This forms the variation of \( \chi_0 \) as the sum of onedimensional contributions \( \chi_0^{1D} \) shifted from the magnetic field wave vector \( G = eHb/\hbar \). The \( I_n \) depend on the zero field dispersion relation\([13]\):

\[ I_n(Q) = \left\langle e^{i(T_\perp \cdot p + Q \cdot L + np)} \right\rangle \]  

(4)

where \( T_\perp (p) = (2/\hbar \omega_c) \int_0^{\pi} t_\perp \left( p' \right) dp' \) and \( \langle ... \rangle \) is the average over \( p \). \( \omega_c = ev_F bH/2 \) is the cyclotron frequency of the open periodic motion in the metallic phase and \( \hbar \omega_c \) is the separation between Landau bands in the FISDW phases. \( \chi_0 \) has logarithmic divergences at quantized values of the wave vector and the largest divergence signal the formation of a FISDW at the corresponding wave vector \( Q_{\perp} = (Q_y, Q_z) = (2k_F + NeHb/\hbar, Q_\perp) \). When the field is varied, each of the peaks becomes in turn the absolute maximum. When \( H \) decreases \( N \) increases monotonously, \( N = 0 \rightarrow 1 \rightarrow 2 \rightarrow 3 \rightarrow 4 \rightarrow 5 \ldots \)

It is possible to go beyond the description of the SDW subphases\([13,14,15]\) to give a complete description of the SDW subphases. The divergence of the susceptibility \( \chi = \chi_0/(1 - \chi_0) \) at a quantized wave vector signals the spontaneous formation of a density wave with this wave vector. This new periodicity couples the eigenstates of the metallic phase and opens a series of gaps at quantized values of the wave vector. The total energy is maximum if the largest of these gaps stays at the Fermi level. This leads to the quantization of the Hall effect and vanishing dissipation.

The Hall conductivity can be calculated from Streda formula\([8,13]\):

\[ \sigma_{xy} = -\frac{\partial N}{\partial H} T_{\mu, Q} \]  

(5)

where \( N \) is the carrier density. The derivative is taken at fixed external parameters, in particular at fixed wave vector \( Q \). Since

\[ N = \frac{2}{4\pi^2} \frac{2\pi}{2k_F} \frac{2}{2\pi b} (Q_1 - eHb/\hbar) \]  

(6)

\[ \sigma_{xy} \] can be rewritten as:

\[ \sigma_{xy} = 2N \frac{e^2}{h} \]  

(7)

The additional factor 2 accounts for spin degeneracy. The quantization of the Hall effect has also been proven by Yakovenko by a direct derivation from the Kubo formula\([8]\).

This model, with the above dispersion relation\([2,13]\), describes remarkably well the phase diagram and the QHE in \((TMTSF)_2PF_6\)\([9]\), in which there is a remarkable quantization of the Hall effect. In ref\([9]\), the transition fields \( H_n \) obey the relation \( H_n = H_f / (n + \gamma) \) with \( H_f \approx 67T \) and \( \gamma \approx 3.5 \). This is in excellent agreement with the prediction of the nesting model (fig. 1), where one finds \( evFbH_n \approx 5.8t_0' / (n + \gamma) \) with \( \gamma \approx 3.45 \). Using the parameters of the Bechgaard salts, \( evFb \approx 1.67K/T \), a value \( t_0' \approx 19.5K \) fits the observed cascade \( H_f \) depends strongly on \( t_0' \) and thus on pressure: it varies between different experiments\([8,13,12]\).

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**Figure 1**: The quantum Hall sequence in \((TMTSF)_2PF_6\)\([9]\) compared with the result of the Quantized Nesting Model, for \( t_0' = 300K, t_0'' \approx 20K \). At low field, the temperature, 150mK is too large to allow for the formation of plateaus and the Hall effect is smaller than the quantized value. Below 5T, the FISDW sequence is destroyed due to the coupling in the third direction.
The nesting model also describes very well the thermodynamic properties (magnetization, specific heat) of the FISDW phases in the PF$_6$ and ClO$_4$ salts\[4\].

3. The "Negative" phases

However, one of the most puzzling unexplained experimental results is certainly the possibility of a reversal of the Hall effect when the field varies: although most of the phases exhibit the same sign of the Hall voltage (by convention we will refer to these plateaus as the positive ones), it has been discovered by Ribault that negative plateaus may appear in (TMTSF)$_2$ClO$_4$ under certain conditions of cooling rate\[2\]. Such negative plateaus have been reproduced and also found in (TMTSF)$_2$PF$_6$, where their existence crucially depends on the pressure\[1\,\[2\,\[2\,\[2\]. One of the puzzling aspect of these negative plateaus is that most often they resemble a dip rather than a plateau and they seem less stable than the positive ones.

Quite recently, by a conditioning procedure in which current pulses depin the FISDW from lattice defects and tend to reduce hysteresis, it has been shown unambiguously that there exists at least one phase characterized by a well-formed negative plateau with a quantized value of $\sigma_{xy}$ ($N = -2$)\[1\]. In this experiment, the sequence of plateaus obtained by decreasing the field can be clearly identified with the quantum numbers $N = 1, 2, -2, 3, 4, 5, 6, 7$. Although there is only one negative plateau in this experiment, an older work has shown a sequence of phases which could be labeled by $N = 1, 2, -2, 3, 4, -5, 6, 7$. Note that the negative plateaus are labeled by even numbers only. In others salts, ClO$_4$ and ReO$_4$, there are also several negative features but it is more difficult to ascribe them a well defined quantum number\[4\,\[4\,\[2\,\[2\] (ReO$_4$: 1, 2, -2, ?). Moreover, in these two materials, the situation is complicated by the anion ordering which certainly affects the apparition of subphases\[2\]. In ClO$_4$, the existence of negative phases is also very dependent on pressure\[2\].

One very important key is to notice that at least the phase $N = -2$ extends up to the metallic phase\[4\]. This feature cannot be explained by a theory based on multiple order parameter states which would appear only at low temperature\[2\]. On the contrary, a description of the metallic phase instability should explain the existence of such phases with an appropriate form of the Fermi surface. In the next section, we show that an appropriate and very slight modification of the Fermi Surface (FS) can lead to the sequence of plateaus 1, 2, -2, 3, 4, -4, 5. Our result strengthens the validity of the Quantized Nesting Model used to describe the phase diagram of Bechgaard salts in a magnetic field.

4. Sign reversals of the Quantum Hall Effect

This change in sign in the Hall effect cannot be explained with the model using the above dispersion relation\[3\]. The reason is the following: when the field varies, the nesting vector oscillates around its zero field value, which connects the inflexion points of the Fermi surface\[3\]. The sign of the carriers is thus given by the position of the inflexion point. A simple geometric analysis shows that $sign(N) = sign(\mathbf{Q}_j - 2k_F) = sign(t'_c)$, and one sees on figs. 2a-b, that $\chi_0(\mathbf{Q}_N) > \chi_0(\mathbf{Q}_{-N})$ for all magnetic fields, so that the sign reversal could not be explained in this framework.

Figure 2: a) $\chi_0(\mathbf{Q})$ in a finite magnetic field for the standard model. $t'_c = 10K, t_3 = t_4 = 0$. The best nesting vector is $\mathbf{Q}^*$. $\mathbf{Q}'$ is a degenerate secondary maximum. b) Same parameters. c) A finite $t_3 = 10K$ alters the best nesting and $\mathbf{Q}''$ is now the degenerate best nesting vector. d) A finite $t_4 = 0.2K$ lifts the degeneracy, leading to a negative quantum number.

However, the fact that the sign of the Hall sequence is fixed cannot be a general feature. The detailed structure of $\chi_0$ has to depend on the fine geometry of the FS\[3\]. If two regions of the Fermi surface exhibit almost equally good nesting properties, one can imagine that the SDW vector will oscillate between positive ($Q_\parallel > 2k_F$) and negative ($Q_\parallel < 2k_F$) regions. In this paper, we show that, by a small change in the dispersion relation, these sign reversals can be described as an equilibrium solution of the Nesting Model.

First, note that there are several maxima for each value of the quantum number and that there is a secondary maxi-
imum, noted \( Q_0 \), on the \( Q_\perp = \pi/b \) line, but for even phases only. This is because \( I_{2M+3}(\pi/b) = 0 \), a property which can be checked directly from eq. (4). This property has a simple semiclassical qualitative explanation: when \( Q_\perp = \pi/b \), the pocket of unnested carriers is be splitted into two pockets of equal size. Quantization in each of these two pockets implies an even quantization of the total number of unnested carriers. For the standard model, the absolute maximum, noted \( Q^* \), always lies outside the \( Q_\perp = \pi/b \) line[9, 13]. This is a reminiscence of the position of the zero field best nesting vector[13] (figs. 3, 4).

![Figure 3](image)

**Figure 3:** a) \( \chi_0(Q) \) in zero field for the standard model. There is a maximum corresponding to the inflexion point of the FS with \( Q_\parallel > 2k_F \). b) When \( t_3 \neq 0 \), the maximum is moved along the degenerate \( Q_\perp = \pi/b \) line. c) When \( t_4 \neq 0 \), the maximum has \( Q_\parallel < 2k_F \). To increase the effect on the figures, we have chosen large values of the parameters \( t_0' = 60K, t_3 = 20K, t_4 = 2K \).

In order to change the geometry of the Fermi surface and to change the nesting at the inflexion point, we propose that next harmonics in the dispersion relation may play a very important role. The dispersion relation is now taken as:

\[
t_\perp(p) = -2t_0\cos p - 2t_0'\cos 2p - 2t_3\cos 3p - 2t_4\cos 4p \quad (8)
\]

In Bechgaard salts, the additional harmonics exist and result directly from next nearest neighbor coupling[3]. A very slight modification of the FS induced by a third \( t_3 \) and a fourth \( t_4 \) harmonics in the transverse direction is enough to explain the existence of new phases with a change in the sign of the Hall effect. We explain now why these two terms are equally important to describe the sign reversals.

\[
I_N(\pi/b) = \langle \exp \left( \frac{4i}{\hbar \omega_c} \left[ t_0' \sin 2p - \frac{t_4}{2} \sin 4p + iNp \right] \right) \rangle \quad (9)
\]

The effect of a third harmonics \( t_3 \) in the dispersion relation is to deteriorate the nesting at the inflexion point. This is seen on figs. 4 for zero field. As a result, when the field is applied, the odd absolute maxima have still \( Q_\parallel > 2k_F \) but the even maxima can be of two different natures depending on the field: either they stand on the \( Q_\perp = \pi/b \) line (\( Q_0 \) on figs. 3, 4) or towards the zero field best nesting vector (\( Q^* \) on fig. 3). When they lie on the \( Q_\perp = \pi/b \) line, these maxima are degenerate: \( \chi_0(Q_{2M}) = \chi_0(Q_0) \). This degeneracy had already been noticed in the past[13, 14]. It would lead to a phase diagram where \(-2 \) and \( 2 \) are degenerate, which is not the case.

We have found that this degeneracy can be removed by the addition of a fourth harmonics of amplitude \( t_4 \). On the \( Q_\perp = \pi/b \) line, the \( I_N \) are given by:

\[
I_N(\pi/b) = \langle \exp \left( \frac{4i}{\hbar \omega_c} \left[ t_0' \sin 2p - \frac{t_4}{2} \sin 4p + iNp \right] \right) \rangle \quad (9)
\]

![Figure 4](image)

**Figure 4:** \( x_{max}(H) \) for \( t_3 = t_4 = 0 \) and for \( t_3 \neq 0, t_4 \neq 0 \).

![Figure 5](image)

**Figure 5:** Hall voltage versus field for a) \( t_0' = 10K, t_3 = t_4 = 0 \) b) \( t_3 = 7K, t_4 = 0.025K \), obtained from \( \chi_0(Q) \) at \( T = 0.5K \).
By changing $N$ into $-N$ and $p$ into $p + \pi/2$, one has:

$$I_{-N}(\pi/b) = (-1)^{N/2} \exp\left\{ \frac{4i}{\hbar \omega_c} (t'_4 \sin 2p + \frac{t_4}{2} \sin 4p + iNp) \right\}$$

(10)

One immediately sees that if $N$ is odd, $I_N = 0$ as stated above. If $N$ is even, the degeneracy is broken by a non-zero $t_4$. When $\text{sign}(t_4) = \text{sign}(t'_4)$, $I_{-N}^2 > I_N^2$, so that $\chi_0(Q_{-N}) > \chi_0(Q_N)$ and a phase with negative even $N$ is favored for some values of the field.

Fig. 4 shows the evolution of the maximum of the susceptibility with the field. It is made of different sheets corresponding to the different nesting vectors. It is seen that for the standard model ($t_3 = t_4 = 0$), all the SDW phases will have a positive $N$, and that for some finite $t_3$ and $t_4$, negative phases appear and the sequence 1, 2, -2, 3, 4, -4, 5 is found, as observed experimentally.

Fig. 5 shows the variation of the Hall voltage with the field. It is qualitatively similar to the experimental ones. At low $T$, a sequence of fine quantized structures may be resolved and that an alternance of many subphases with positive and negative quantum numbers can appear. It is in very good agreement with the complex structure observed ten years ago, near the threshold field, see fig. 6.

Figure 6: The Hall voltage in $(TMTSF)_2PF_6$ under pressure, from [3]. It is qualitatively well described by the result of fig. 4.

We believe that the experimentally observed high sensitivity of the sequence of sign reversal to external pressure is due to the sensitivity of the parameters of the dispersion relation to pressure. We emphasize that although the metallic electron gas has a metallic behavior with a large Fermi energy, of the order of the $eV$, the cascade of SDW subphases is driven by extremely small energy scales, of the order of a few Kelvins. The orders of magnitude of these two additional harmonics are non incompatible with the estimations of a refined microscopic model [13].

Finally it is worth noticing that positive and negative phases are almost degenerate because the energy scale $t_4$ is certainly very small, of the order of $1K$. The relative energy difference between these two phases is very small, of order $t_4^2/\omega_c$. This explains which the negative phases are always very sensitive to external parameters like pressure or probably anion ordering.

Fig. 7 shows the evolution of the QHE sequence with the parameter $t_3$. One sees that the even negative phases appear when $t_3$ increases.

5. Conclusion

The Quantized Nesting Model explains very well the quantization of the Hall effect observed in Bechgaard salts. The observed structure is extremely sensitive to the details of the dispersion relation and the nesting of the Fermi surface. We have explained the ten-years-old puzzle of the observed sign reversals of the Quantum Hall effect in the cascade of FISDW phases of Quasi-1D organic conductors. They can be described with a slight modification of the dispersion relation of the metallic phase. We have been able to...
reproduce the observed sequence of "negative" phases with an even quantum number, to understand why they are very sensitive to pressure and why it is more difficult to measure a well defined plateau. Our result shows that the electronic properties of the Bechgaard salts are extremely sensitive to very small changes in the geometry of the FS and that the standard model and its variations still continues to describe very well the observed phase diagram of these salts in a magnetic field.

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