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Field Induced Spin Density Waves

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Abstract. — The Field Induced Spin Density Waves (FISDWs) found in organic conductors represent a unique series of transitions which meld the one-dimensional physics of the Peierls instability with the two-dimensional physics of the Quantum Hall Effect. This paper presents a pedagogical introduction to the FISDW's in the Bechgaard salts, along with recent experimental results on related high magnetic field phenomena.

1. Introduction

The Bechgaard salts, (TMTSF)$_2$X (where X = PF$_6$, ClO$_4$, ReO$_4$ etc.), are probably the most interesting electronic materials ever discovered. Depending on composition, temperature, pressure and magnetic field they exhibit most of the ground states and phenomena associated with interacting electrons in vastly different systems. There are competitions between metallic and insulating, magnetic and superconducting, semiconducting and semimetallic phases. The Bechgaard salts exhibit all of the electronic transport mechanisms yet discovered, metallic conductivity, sliding density wave conductivity, superconductivity and the quantum Hall effect. What is even more remarkable is that all of the above properties can be observed in one single crystal of one of the Bechgaard salts (TMTSF)$_2$PF$_6$ as temperature, pressure, and magnetic field are varied [1].

The basis for understanding the wide variety of behaviors is to be found in the strongly anisotropic bandstructure resulting from the quasi-one-dimensional chainlike crystal structure. Platelike TMTSF molecules stack face to face in a zigzag chain. The wavefunction overlap from one molecule to the next is responsible for the large bandwidth (1 eV) in the chain a direction. Neighboring chains in the b direction are also sufficiently close for Se orbital overlap and yield a bandwidth of 0.1 eV. The coupled chains form two-dimensional planes in which the electrons are delocalized. The planes are separated in the third direction by a sheet of anions. The overlaps are small and the bandwidth is down by an additional factor of ~ 30 to 0.003 eV. There is a full charge transfer of one electron per unit cell to the anion, leaving the two TMTSF molecules with half a hole on each. Were it not for a slight dimerization of the zigzag

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Fig. 1. — Log-log plot of the resistivities of some common conductors. Circles indicate the superconducting transition temperatures. Note that (TMTSF)$_2$ClO$_4$ looks most similar to Cu metal. Both are very clean, with mean free paths of many microns at low temperature. The main difference is the factor of \( \sim 1000 \) carrier density from the larger unit cell in the organic.

chain, we would expect a quarter filled TMTSF band. But the dimerization splits the TMTSF band into two bands and leaves us with a one half filled upper band as required by the charge transfer and stoichiometry.

The highly anisotropic bandwidths, \( 4t_a : 4t_b : 4t_c \approx 1 \text{ eV} : 0.1 \text{ eV} : 0.003 \text{ eV} \) imply conductivity anisotropies in the ratio \( (t_i / t_j)^2 \). The measured conductivities are: \( \sigma_a : \sigma_b : \sigma_c \approx 10^3 : 10^1 : 10^{-2} \text{ (}\Omega \cdot \text{cm})^{-1} \) [2]. The temperature dependent resistivities of several presently interesting materials is shown in Figure 1. The transition temperatures of the superconductors are indicated by the large circles terminating the low end of the resistivity curves. It is clear that the (TMTSF)$_2$ClO$_4$ salt is a good metal with resistance decreasing about three orders of magnitude on cooling from room temperature to 1.4 K. It is very similar to Copper, the main difference being that the carrier density is much less. The Bechgaard salts have about 1 carrier per 1000 cubic Angstrom unit cell, while Cu has one carrier per 1 cubic Angstrom Cu atom. This essentially accounts for the thousandfold difference in conductivity.

The low temperature phase diagram for the wonder material (TMTSF)$_2$PF$_6$ is shown in Figure 2 [3,4]. Above 12 K the system is metallic. Upon cooling at ambient pressure there is a very weakly first order transition to a Spin Density Wave (SDW) insulating phase as evidenced by antiferromagnetic resonance, NMR and muon spin rotation [5]. The SDW transition temperature can be suppressed by application of moderate pressure until at \( \sim 6 \text{ kbar} \) the metallic state is reestablished. Once the SDW is suppressed the crystal becomes superconducting at 1.2 K [6]. This material was the first organic superconductor discovered. Further increasing the pressure gradually reduces the superconducting transition temperature. Applying a small magnetic field along the least conducting direction (to induce screening currents in the highly conducting plane) kills the superconductivity above a critical field of \( \sim 500 \text{ Gauss} \) [7].
Fig. 2. — The low temperature phase diagram of (TMTSF)$_2$PF$_6$ [3, 4]. While the proximity of the SDW or antiferromagnetic insulator phase to the superconducting phase is common to many systems, the field induced SDW cascade seen above 6 kbar and ∼ 5 tesla is so far unique to the quasi-one-dimensional organic conductors [8]. Each separate phase in the cascade with increasing field corresponds to a different quantum Hall state.

All of the phases discussed up to this point have been observed in other materials. The juxtaposition of superconducting and SDW or antiferromagnetic phases is now often seen in highly correlated systems such as the high temperature superconductors. However, what is new and so far unique to these materials [8] is the phase which results from further increasing the magnetic field while in the low temperature metallic state [9]. Imagine starting at 10 kbar, at 0.5 K and applying a field (along the c direction). 500 Gauss kills the superconductivity and leaves us with a “normal” metal (more about that later). At slightly more than 5 teslas we encounter another phase boundary where the resistivity sharply increases as does the Hall coefficient. Further increasing field this transition is followed by a cascade of at least 9 consecutive first order transitions to different semimetal states until at ∼ 20 teslas we enter the “final” insulating phase. Most remarkable of all, each transition takes us to a state with a well defined and sequenced (..1/4,1/3,1/2,1) quantized Hall resistance [10]. It is particularly striking since the Quantum Hall Effect (QHE) is intrinsically the property of a two-dimensional electron system and this material was the first to exhibit the QHE in a bulk three-dimensional crystal. This cascade of phase transitions is to Field Induced Spin Density Wave (FISDW) states and is the main topic of this paper.

2. One Dimensionalization in a Magnetic Field

The Fermi surface which we associate with this bandstructure is cartooned in Figure 3. It consists of two non-intersecting slightly warped sheets. The actual Fermi surface and Brillouin
Fig. 3. — Cartoon of the idealized Fermi surface of the Bechgaard salts. (The real crystal structure is triclinic not orthorhombic). The Fermi surface consists of nonintersecting sheets along the highly conducting a direction warped by $4t_b/e_F$ and $4t_c/e_F$ in the b and c directions.

Zone are more complex [11]. The crystal structure is triclinic, not orthorhombic as in the cartoon and there is overlap between one TMTSF molecule on a chain and several molecules on a neighboring chain. If the salts were truly one-dimensional electronically then the Fermi surface would consist of two parallel sheets at $\pm 2k_F$. The warping is due to the finite bandwidths in the b and c direction (the b warping is about the correct size, but the c axis is exaggerated by about a factor of 30.) A good deal of the phenomena observed in the Bechgaard salts are directly related to this Fermi surface which at first glance seems innocuous. There are no closed orbits and hence no chance for Landau quantization. The warping seems too large to allow the one-dimensional instabilities associated with the Peierls transition. For $H \parallel c$ (perpendicular to the highly conducting plane) we expect small saturating magnetoresistance along the a axis and sizable nonsaturating magnetoresistance along b and c [12].

Let us forget about the least conducting c direction for the moment and concentrate on the Fermi surface and bandsstructure in the a-b plane. The essence of the problem is that of large anisotropy and only open orbits of the Fermi surface. A dispersion relation with these properties can be written as

$$\epsilon(k_x, k_y) = -2t_a \cos k_x a - 2t_b \cos k_y b \approx \frac{h^2 k_x^2}{2m^*} - 2t_b \cos k_y b$$  \hspace{1cm} (2.1)$$

where we temporarily choose a free electron form for the dispersion along $x$ to avoid complications due to nesting and commensurability [13, 14]. We want to see the effect of a field perpendicular to the plane. A Landau-Peierls substitution $k \rightarrow i \nabla - eA/hc$, with the choice of a Landau gauge $A = (0, Hx)$ leads to:

$$\frac{\hbar^2}{2m^*} \frac{\partial^2}{\partial x^2} \Psi - 2t_b \cos(k_y \frac{\partial}{\partial y} - \frac{eHb}{hc} x) \Psi = e\Psi$$  \hspace{1cm} (2.2)$$

Since $y$ only appears in the partial derivative, the wavefunction can be factored as $\Psi(x, y) = e^{i k_y y} \psi(x)$ and defining the magnetic wavelength as $2\pi/\lambda = eHb/hc$ we have:

$$\frac{\hbar^2}{2m^*} \frac{\partial^2}{\partial x^2} \psi(x) - 2t_b \cos(k_y b - \frac{2\pi}{\lambda} x) \psi(x) = e\psi(x)$$  \hspace{1cm} (2.3)$$
Fig. 4. — Real space and momentum space quasi-classical trajectories of electrons in the presence of a magnetic field along c. In k space the motion along the open orbits periodically crosses the Brillouin zone with frequency $\omega_b$. In real space the motion is localized to $\sim 4t_b/\hbar \omega_b \propto 1/H$ chains along b. The motion is extended along the chains but there is a spatial modulation at the magnetic length $\lambda = h/c e H b = \Phi_0 / H b$ (where $\Phi_0$ is the flux quanta).

Since $k_y$ appears only in the argument of the cosine, it only serves to shift the origin of $x$ (hence the center of mass of the wavefunction, $x' \rightarrow x - \lambda b k_y / 2\pi$). Equation (3) is then a one-dimensional Schrodinger's equation for a periodic potential (in fact it is Mathieu's equation) [15]. In a magnetic field the dispersion depends on $k_z$, $\epsilon(k_z)$ rather than the zero field dispersion $\epsilon(k_x, k_y)$. Thus the magnetic field makes the system electronically one-dimensional.

We can understand this one dimensionalization from the quasi-classical motion of the electrons on the Fermi surface. The two-dimensional Fermi surface is schematically shown in Figure 4. The electron velocity $v_k = \nabla \epsilon$ is perpendicular to the Fermi surface. In the presence of a magnetic field electrons are constrained to move on constant energy surfaces. Electrons follow the equation of motion $\hbar \partial k / \partial t = e v_k \times B / c \Rightarrow$ the real space velocity $v_k \rightarrow \partial r / \partial t$ and $k$ space motion $\partial k / \partial t$ are simply rotated by 90°. There is a characteristic frequency $\omega_b \equiv (\partial k_y / \partial t) / (2\pi / b) = e v_F H b / h c$ with which the electron crosses the Brillouin zone in the b (y) direction. ($v_F$ is approximately constant at $v_F$ on the Fermi surface. The dispersion along x is often taken in linearized form as $h v_F k_x$. ) The real space electron motion is shown in Figure 4 bottom. It is limited along y and extended along x, i.e. one-dimensional. The width of the orbit (which is actually the extent of the quantum wavefunction along y [16]) is $(4t_b / \hbar \omega_b) b$ and there is a new periodicity, $G = 2\pi / \lambda = e H b / h c$, given by the magnetic length $\lambda$ along $x$ [17]. (The length is such that $\phi_0 = \lambda b H$ with $\phi_0 = h c / e$ a flux quanta. Note that for an isotropic two-dimensional electron system the magnetic length $l_H$ is such that $\phi_0 = l_H^2 H$. For 10 teslas
Fig. 5. — Susceptibility of the electronic system to a distortion of wavevector $Q$, $\chi(Q)$ [29]. The FISDW wavevector is determined by the $Q$ at the maximum of $\chi$, in this case $Q_0^*$. (Small changes in bandstructure can shift the maximum to either of the $Q_0$'s leading to changes in the sign of the quantum Hall resistance.)

The wavefunction is spread over about 60 chains in the $b$ direction and the periodicity induced on the chains in the $a$ direction is about 200 unit cells.) [18]

The effective one-dimensionalization of the electrons by the magnetic field can and does have interesting consequences. One-dimensional metals are unstable against the formation of charge and spin density waves (Peierls transitions) [19]. The effects of electron interactions become more important in one dimension, Fermi liquids give way to Luttinger liquids and other many body ground states [20, 21]. Disorder plays a drastic role and always leads to localization and insulating behavior at least for non-interacting electrons. There is no long range order and hence no finite temperature transitions in one dimension. The absence of any closed current loops would suggest that the superconducting critical field should tend to infinity for one-dimensional systems [22]. Of this rich set of proposed consequences of field induced one dimensionality, the only one of which we presently have conclusive evidence is the Field Induced Spin Density Wave (FISDW) transitions [9,17,23].

3. The FISDW Phase Diagram

The instability of the metallic state can be obtained by calculating the susceptibility to a periodic spin density modulation at the wavevector $q$. This susceptibility is shown in Figure 5. The one-dimensional character of the electronic dispersion in a magnetic field assures divergences in the susceptibility as temperature is lowered toward zero. Using a Stoner criteria we then expect a transition into an SDW state corresponding to the wavevector at which the susceptibility is maximum. The wavevectors of the maxima shift with magnetic field [17,24,25]. They always occur at $2k_F \pm n2\pi/\lambda$ along $k_a$ but the $k_b$ value varies with bandstructure and field in such a way as to yield quantized $k$ space areas.
A qualitative explanation of the FISDW's also follows from considering the possible ground states of the system (rather than the instability on cooling from the metallic state). A one-dimensional system is unstable against the formation of a density wave because a distortion of wavevector $2k_F$ exactly nests the two sides of the Fermi surface. This produces a gap at the Fermi energy. The total electronic energy is lowered by $\Delta^2 \ln \epsilon_F / \Delta$ while the distortion costs an energy of order $\kappa \Delta^2$ where $\kappa$ is an elastic constant or an exchange interaction [19]. Because of the log term the electronic energy always wins (for small $\Delta$) and the density wave is stable. For a quasi-one-dimensional dispersion, the nesting of the Fermi surface is not perfect as illustrated schematically in Figure 6a. A distortion of $2k_F$ produces a gap at only a few points on the Fermi surface. These gaps close off electron and hole pockets and the dispersion is that of a semi-metal. The electronic energy lowering is of order $\Delta^m \ln \epsilon_F / \Delta$ (with $m \geq 3$), not enough to overcome the cost of making the distortion. The metallic state of the quasi-one-dimensional system is stable. This is the situation for $(\text{TMTSF})_2\text{PF}_6$ under pressure without a magnetic field. It remains metallic to $T = 0$.

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Fig. 6. — a) Distortion with $H = 0$. 1) For perfect nesting (as in one dimension) a single wavevector maps one side of the Fermi surface to the other opening a complete gap at the Fermi energy, stabilizing the distortion and the insulating phase. 2) Imperfect nesting leaves electron and/or hole pockets on a partially gapped Fermi surface. There is no longer a gap at $\epsilon_F$ and this state is not lower energy than the metal without distortion. b) No distortion, finite $H$ 1) For closed orbits area, energy and Hall resistance are quantized. 2) For the open orbit Bechgaard salts there are no interesting effects at $\epsilon_F$. c) Distortion and field. The areas and energies in the electron and hole pockets are quantized, the distortion wavevector adjusts so that $\epsilon_F$ lies between Landau levels. Since $\epsilon_F$ is completely in a gap the distortion is stabilized and concurrently we have only filled Landau levels and the quantum Hall effect.
Now let's consider the effect of a magnetic field on an undistorted metal. Figure 6b. If we have closed orbits then we have Landau quantization, discrete energy levels and the Quantum Hall effect (in a two-dimensional system). However, for open orbits there is no quantization condition and the energy dispersion remains continuous with a classical (often zero) Hall effect [26]. The quasi-one dimensional metal seems uninteresting both in its stability and lack of magnetic field effects.
If we have both a distortion and a magnetic field we regain something interesting, Figure 6c [27]. The distortion leads to closed orbits in the semi-metal pockets. These must be Landau quantized with spacing $h\omega$, $\omega = eH/m^*c$. If the Fermi energy lies between Landau levels, then we have a completely gapped system, the electronic energy gain again beats out the distortion energy and the system lowers its energy by distorting, forming an SDW. How can $e_F$ always lie in the gap? Suppose we were to change the magnetic field. The spacing of the Landau levels shifts and we might expect $e_F$ to enter a Landau level. Without a gap the SDW would collapse. However, $e_F$ remains in the gap as long as we have only filled Landau levels, i.e. the area in the $k$ space pockets must be an integral multiple of $eH/hc$ (the area which can exactly accommodate the number of states in a Landau level). The system can adjust the area of the pockets by changing the wavevector of the SDW distortion. Equivalently, the distortion wavevector dictates the top and bottom of the electron and hole pockets. As the field changes the $q_{SDW}$ changes so as to keep $e_F$ in the gap between Landau levels. At some field it may be that the energy is lowered by $q_{SDW}$ jumping to a different value so that $e_F$ sits between another set of Landau levels.

The situation where there are only completely filled landau levels is precisely the condition which gives rise to the Quantum Hall effect [28]. In a conventional two-dimensional electron gas $e_F$ sits between Landau levels only in the presence of disorder induced localized states. In the present case $e_F$ sits in the gap for intrinsic energy reasons. In fact we could not have a stable FISDW without the QHE (and conversely). As field is changed we should have first order transitions between SDW states with different quantum numbers. The ordering of the quantum occupations as field is increased depends on details of the bandstructure and the nesting [29]. In all cases the high field state should have a wide splitting between the lowest energy electron Landau level and the highest energy hole Landau level. This high field limit is an insulator. In the simplest picture as the field is lowered there is a single pocket and the quantum numbers follow $n = 0, 1, 2, 3, 4,...$ as the field is lowered.

The cartoon description given above is culled from a great deal of theoretical work by a number of groups who have converged on a "standard model" or "quantized nesting model" for the FISDW's [17, 24, 25]. The picture which emerges from these detailed calculations is illustrated in Figure 7. Starting from high field we should have the FISDW insulator state with no filled Landau levels, $N = 0$, and essentially the same transition temperature as for the zero field SDW with perfect nesting. On lowering the field we enter the $N = 1$ phase with semimetallic integer quantum Hall behavior. Further lowering the field yields the cascade of FISDW transitions to different phases, separated by first order transitions and each associated with a different Landau level filling and quantum Hall plateau. Within each phase the wavevector for the SDW changes continuously with field ($q_{FISDW} = (2k_F \pm n2\pi/\lambda, \pi/b + \epsilon)$) to maintain the complete filling of the Landau levels. Between phases the wavevector changes discontinuously with the $a$ component having a different value of $n$. In the simplest case $n = N$ and the cascade results in the stepwise decrease of $n$ as suggested in the figure. However, depending on the bandstructure, both the sequence of wavevectors $n$ and their association with $N$ (the number of filled Landau levels) may change [29].

At pressures somewhat above the critical pressure needed to suppress the ambient field SDW, both the PF₆ salt and the AsF₆ salt show the transitions as predicted by the simplest form of the quantized nesting model. The Hall resistance (at $T = 0.5$ K) and transition temperature are shown for a PF₆ sample at 11kbar in Figure 8, and Hall and longitudinal resistance are shown in Figure 9 for an AsF₆ sample at $T = 0.5$ K and 10 kbar [30].

We see the stepwise decrease in the Hall resistance following $\rho_{xy} = \hbar/2Ne^2$ as expected [31]. The absolute value of the Hall resistance in the plateaus is roughly at the quantized value given above (i.e. 13 k $\Omega/\square$ per plane) for "good" samples (where good means agreement and
Fig. 7. — According to the standard or quantized nesting model there is a cascade of FISDW transitions as field is increased. Associated with each transition the low temperature Hall resistance should show a quantized plateau. In the simplest case the plateaus, $\rho_{xy} = h/2Ne^2$, increase as $N = 0, 1, 2, 3, 4, 5$. with increasing field.

Fig. 8. — Data for (TMTSF)$_2$PF$_6$ at 11 kbar pressure showing the phase diagram and the Hall resistance at 0.5 K [3,4].
Fig. 9. — (TMTSF)$_2$AsF$_6$ shows similar behavior to PF$_6$. Here the longitudinal resistance is seen to decrease with increasing field in the region of the Hall plateaus and to exhibit peaks in the region of the transitions between FISDW-QHE states. Above 18 tesla the sample enters the $N = 0$ state which is a SDW insulator [3, 4].

presumably that the current paths are uniformly distributed between the conducting planes that make up the crystal).

The differences from conventional integer quantum Hall Effect in single layer two-dimensional electron systems (e.g. GaAs) are the factor of 2 in the denominator (from the spin degeneracy resulting from the spin pairing in the SDW state) [31], the absence of any linear region between the plateaus (from the first order nature of the transitions between the different $N$ states, or equivalently, the fact that the system can energetically never find itself with $\epsilon_F$ located in a Landau level), and the fact that the plateaus do not sit on a line $\rho_{xy} = H/n_e e$ (with $n_e$ the fixed electron density) which extrapolates to zero at zero field (from the fact that the effective carrier concentration is changing with field as the nesting changes and the pockets shrink toward zero at high field.)

In the conventional two-dimensional electron system (2DES) QHE the longitudinal resistance $\rho_{xx}$ goes toward zero in the Hall plateaus and has local maxima between the plateaus. A similar behavior is seen in Figure 10. In fact $\rho_{xx}$ decreases with decreasing temperature in the plateaus (approximately exponentially) and increases exponentially in the $N = 0$ phase [32]. There are several other interesting differences between this system and the 2DES. The closed orbits which quantize into the Landau levels are created by the SDW distortion and its resulting SDW gap. The gap is relatively small and the magnetic fields present are of comparable magnitude. We can therefore have magnetic breakdown through these gaps and the result is tunneling between Landau levels. The Landau levels become Landau bands. Depending on the particular bandstructure and $q_{SDW}$ we have an intricate set of bands and gaps. An example is shown in Figure 11 which shows the calculated gaps about $\epsilon_F$ as the field is varied and different states in the cascade are present [33].
Fig. 10. — Temperature dependence of the longitudinal and Hall resistance of (TMTSF)$_2$AsF$_6$. The Hall resistance monotonically increases to its quantized value as temperature is lowered through the FISDW transition. For the $N=0$ SDW insulating state the resistance increases due to the opening of the SDW gap and the absence of any filled Landau levels. For $N \neq 0$ states the resistance rises just below $T_{SDW}$ from the loss of carriers but then decreases as the dissipationless transport associated with the QHE takes over at lower temperatures.

4. Tilted Magnetic Fields and Lebed Resonances

So far the FISDW has been described as the instability of a quasi-one-dimensional metal in a two-dimensional space. (Experimentally the phase diagram basically scales only with the field perpendicular to the a-b plane when the field is tilted.) The real electronic system is of course three-dimensional and here we consider some of the effects of the three-dimensionality. If the bandwidth in the third direction is larger than the SDW gap, or the spacing of the Landau levels (these numbers are comparable) then the bands will overlap and the FISDW and the associated QHE will disappear. The best measurements and calculations agree that $4t_\epsilon \approx 0.003 \ eV \sim 30 \ K$, easily enough to kill the two-dimensional effect. The clever solution that nature has found and applied in these salts, is to choose a wavevector of $\pi/c$ along c, i.e. to alternate the SDW in neighboring planes. Just as the wavevector $(2k_F, \pi/b)$ leads to perfect nesting and complete gapping of the Fermi surface for the dispersion relation $\epsilon(k) = \hbar v_F k_x - 2t_b \cos k_y b$, $(2k_F, \pi/b, \pi/c)$ does the same for $\epsilon(k) = \hbar v_F k_x - 2t_b \cos k_y b - 2t_b \cos k_z c$. If the $x$ dispersion is quadratic, the nesting is imperfect and what remains of the dispersion is $t_{\text{eff}} \sim t_b^2/\epsilon_\text{f}$ and $t_{\text{ceff}} \sim t_c^2/\epsilon_\text{f} \sim 10^{-5} \ eV \sim 0.1 \ K$. For a strictly two-dimensional system any finite magnetic field
Fig. 11. — The gap structure in the FISDW states are shown as a function of magnetic field [33]. The dark regions are the gaps which separate the white Landau bands. Note that the largest gap is centered around $\epsilon_F$.

would produce an FISDW if we cooled to low enough temperature [17]. A finite $t_{\text{eff}}$ mandates a threshold field below which we cannot attain an FISDW. Experimentally this has been found to be when $\hbar\omega_{\text{crit}} \sim t_{\text{eff}} \sim 0.1$ K as expected also from consideration of the QHE and Landau quantization [34].

Lebed [35] suggested that there was a way around the three dimensionality and the zero temperature threshold field. Up to this point we have treated the case when the magnetic field is aligned along the c axis. If the field is tilted in the b-c plane, the electronic orbits sweep across the Fermi surface as illustrated in Figure 12. The $k$ space equation of motion is simply $\hbar \frac{dk}{dt} = e \mathbf{v} \times \mathbf{B} \approx e\mathbf{v}_F \times \mathbf{B}$ so that there are now two natural frequencies, one for crossing the Brillouin zone in the direction, $\Omega_b = \frac{k_b}{(2\pi/\hbar)} = e\mathbf{v}_F bB \cos \theta/\hbar$ and one for the direction, $\Omega_c = \frac{k_c}{(2\pi/c)} = e\mathbf{v}_F cB \sin \theta/\hbar$. At certain angles, $\tan \theta = pb/qc$, with $p$ and $q$ integers, these frequencies are rationally related, $\Omega_c/\Omega_b = p/q$. For these particular angles, the electron orbits retrace their motion and the trajectory is a line, for other angles the electron orbits do not retrace and end up covering the entire Fermi surface. Lebed argued that the commensurate motion at the "magic" angles couples the dispersion in the two open orbit directions and reduces the three-dimensional dispersion to one-dimensional. At these angles the threshold
field would be reduced to zero (at zero temperature). The analogous effect, the increase of the transition temperature of the FISDW at the Lebed angles has been seen [36].

A few years after Lebed’s prediction of the commensurability effect on the threshold field, came the idea that the angular resonance on the Fermi surface should also have drastic effects on the transport properties [37]. Experimentally, these were first observed in the ClO₄ salt a axis resistance [38]. Later they were seen along both a and c directions in both the ClO₄ and PF₆ salts when they are in the metallic state at low temperature [39]. It is fairly easy to understand why transport properties are changed at the Lebed magic angles: the repeating orbits can avoid high scattering regions of the Fermi surface and/or provide different averages over the velocities. The one or two dimensionality of the dispersion has drastic effects on the nature of both impurity and electron-electron scattering. To date there are ~ 10 different models/explanations for the magic angle resonances, none of which has satisfactorily explained all of the data [38, 40, 41]. This is partly because the effects differ from one salt to the other. There are especially sharp and unusual structures found in the PF₆ salt under pressure: Figure 13.

Although the basic idea for the Lebed resonances comes from k space, Fermi surface arguments Figure 12, the fact that k space vectors are perpendicular to real space vectors, and that the velocity is perpendicular to the field, translates to the fact that the “magic” angles have a real space interpretation. They are simply the angles at which the field is oriented in the direction between the actual molecules in different chains (Fig. 12d). This observation led to
one of the most intriguing ideas on the origin of the magic angle effects, at least for the PF$_6$ salt. The resistivities along the most and least conducting directions are shown for this salt in Figure 13. The suggestion was that the salt is marginally a Fermi liquid [41]. It is presumed that the planes would be in a non-Fermi liquid (NFL) state if they were uncoupled. The electronic coupling along the c axis, $t_c$, is (presumably) just barely enough to lead to coherent transport along c and consequently the destruction of the 2 dimensional NFL state in favor of a conventional 3 dimensional Fermi liquid. In this scenario a small field along b reduces $t_{c_{\text{eff}}}$ below the critical value and leaves a 2D NFL. In such a 2D system all magnetotransport must depend only on the field perpendicular to the 2D plane and might be in the form of power
Fig. 14. — The power law dependence on field perpendicular to a-b plane \((H \cos(\theta))\) is illustrated by log-log plot of c axis magnetoresistance for sample rotations taken at different fields. The same curve results when field sweeps are taken at fixed angles. The deviations occur at the Lebed angles. Same parameters as in Figure 13.

laws, \((H \cos(\theta))^\alpha\). At the magic angles \(t_{\text{cell}}\) is not as reduced and the resistance drops. Measurements show that aside from the magic angles the magnetoresistances vary \(\rho_{xx} \propto (H \cos(\theta))^{0.5}\) and \(\rho_{zz} \propto (H \cos(\theta))^{1.2 \text{ to } 1.5}\) when either \(H\) is varied at constant \(\theta\) or \(\theta\) is varied at constant \(H\) [42, 43], Figure 14. From this point of view the “normal” metal state of (TMTSF)_2PF_6 is a non Fermi liquid in the presence of a small field along \(b\).

The deceptively simple Fermi surface, which at first glance yields no hint of interesting behavior in a magnetic field, also produces large angular dependent magnetoresistance for rotations in the a-c plane [44] and most recently in the a-b plane [45]. So far these resonances have been readily explainable, at least in the ClO_4 salt in terms of conventional Boltzmann transport, with a coherent Fermi surface [46]. They have been able to provide useful information about the bandstructure parameters much as has been done previously with closed orbit metals.

5. The Anomalous behavior of (TMTSF)_2ClO_4

Although the PF_6 salt of TMTSF seems an ideal example of the simplest form of the standard model for the FISDW’s, the ClO_4 salt presents a much more complex behavior. To date the ClO_4 salt is more studied than the PF_6 for both FISDW’s and for superconductivity even though both phenomena were discovered first in the PF_6. ClO_4 is an ambient pressure superconductor and FISDW system while PF_6 requires a pressure of greater than 6 kbar. This makes it much harder to mount, align, and rotate the PF_6 samples and makes most thermodynamic and elastic properties almost impossible to measure. All of these measurements have been done on ClO_4 [47, 48].

ClO_4 has an anion ordering transition at 24K which leads to a doubling of the unit cell in the \(b\) direction [49]. It was well known that the low temperature properties of this salt, its resistance, its superconducting transition temperature etc. depended strongly on how rapidly the sample was cooled through this anion ordering transition. However, the extreme sensitivity of the quantum Hall steps, especially the change in sign or “Ribault” anomaly [50], was unexpected. For many years the only Hall data was from ClO_4 and masked the acceptance of
Fig. 15. — (TMTSF)$_2$PF$_6$ exhibits "negative" Hall steps at pressures below about 9 kbar [51], similar to what has been observed earlier in (TMTSF)$_2$ClO$_4$ by Ribault et al. [50]. They have been attributed to a specific warping of the Fermi surface, $\rho_0 = H/2e^2$.

The lack of a simple staircase for the Hall resistance is only one of many anomalous behaviors of the FISDW in ClO$_4$. Although experiments have been carried out to very high fields (50 T at ~ 1 K) the $N = 0$ insulating state has yet to be found. At low temperature there is a very wide field region from 7.5 to 27 Tesla where the Hall resistance is flat and the longitudinal resistance is clearly semi-metallic. Beyond 27 T there is a first order transition to a state with very large oscillations in resistance, Hall effect and thermodynamic properties [53]. For many years this was considered a reentrant metallic phase, but the most recent results suggest that the phase diagram (Fig. 16) at ambient pressure is yet more complicated with some FISDW phases enclosed in other phases [54].
Fig. 16. — The most recent version of the phase diagram of (TMTSF)\textsubscript{2}ClO\textsubscript{4} shows significant deviations from that of the PF\textsubscript{6} salt including the absence of the $N = 0$ insulating phase and the presence of two transitions as the sample is cooled in fields near 25 tesla.

It seems that many of the complications of ClO\textsubscript{4} and its differences with PF\textsubscript{6} can be associated with the anion ordering. In particular, it was found that above 4.5 kbar the anion ordering is suppressed and the phase diagram vs. $H$ comes to resemble that of PF\textsubscript{6} with a well defined $N = 0$ phase at high field [3, 4].

There is yet another strikingly strange phenomena associated with the Bechgaard salts. Despite the open orbit nature of the Fermi surface, there are “fast” oscillations observed in magnetotransport which greatly resemble Shubnikov de Haas (SdH) oscillations (normally associated with closed orbits). They are seen in transport in the metallic and FISDW phases and in thermodynamics (magnetization, specific heat, magnetocaloric effect, sound velocity) in the FISDW of the ClO\textsubscript{4} salt [55]. They are seen at a slightly different frequency in the PF\textsubscript{6} salt but only in the SDW phase and the low N FISDW phases in transport [56]. The Fermi surface of ClO\textsubscript{4} is schematized in Figure 17 where the effect of the anion ordering is seen to have halved the Brillouin zone and doubled the Fermi surface. The area associated with the fast oscillations is about 6 percent of the Brillouin zone similar to the shaded area in the figure. (The slow oscillations are just the FISDW’s themselves, their frequency is associated with the unnested area of the Fermi surface, about 0.6 percent of the area of the Brillouin zone.) At high magnetic fields it might be expected that magnetic breakdown would allow electrons to tunnel across the anion ordering gap at the zone edge (A). Since both Fermi surfaces near A are electron like (i.e. the trajectories are both up) the breakdown does not result in a closed orbit. However, since there is a phase difference between an electron on path i and the same electron on path ii, the breakdown results in the possibility of quantum interference between two trajectories. This would produce a field dependent oscillation in transport properties but not in thermodynamics. It is likely that this is the explanation for the resistance oscillations in ClO\textsubscript{4} in the metallic state [57]. However, it has also been pointed out that the interference from the magnetic breakdown can also have sizable effects on the electron impurity and electron-
electron interactions which can effect both transport and thermodynamics. Several models to explain the fast oscillations are found in reference [58].

The observation of thermodynamic oscillations in the FISDW state in ClO$_4$ suggest that the anion ordering and in particular the doubling of the FS as in Figure 17 has an interesting effect on the nature of the FISDW gaps and phase transitions. Several authors have looked at the problem of treating the anion ordering, the opening of the SDW gap and the magnetic breakdown on an equal footing. Depending on the treatment and the parameters, the condensation energy either shows a small oscillation at the fast oscillation frequency or varies so strongly to cause an oscillating reduction of $T_{\text{FISDW}}$ from its maximum to zero [59]. Experimentally there are certainly fast oscillations in thermodynamic quantities and in transport inside the FISDW phases, but as yet there is no evidence of an oscillating phase boundary [54].

There have also been interesting suggestions about the coexistence of several order parameters both well into a particular FISDW phase and certainly near the transition line between two FISDW phases [60]. It is a particularly intriguing question since we associate a different quantum Hall state with each different FISDW nesting vector. If we have order parameters for both $N = 2$ and $N = 3$ coexisting, what will the Hall resistance be? Theoretically the answer involves the winding number of the phase of the total order parameter [61]. In the case of the coexistence of two order parameters the one with the bigger amplitude wins, but there again are subtleties regarding the existence of a gap and the magnitude of the longitudinal resistance as the amplitudes of the order parameters pass one another with increasing field.

Experimentally the thermodynamics of the transition are measured only for the ClO$_4$ salt. They are a fascinating story on their own [47,48]. On entering the FISDW phase from the normal metal the transitions are second order. Some of the low field phases are reentrant. Below 6 T at low temperature, the transitions between adjacent FISDW phases are continuous with a region of coexistence of order parameters. The magnetocaloric effect shows two thermodynamic transitions (when $\Delta_1$ and $\Delta_2$ separately go to zero [48]) associated with each Hall transition (when $\Delta_1$ crosses $\Delta_2$). The two transitions above 6 tesla are strongly first order and hysteretic.
6. Conclusions

The "standard" or quantized nesting model of the FISDW has been very successful in describing many of the features seen in the Bechgaard salts at high magnetic field. The phase boundary, the quantum Hall effect, much of the thermodynamics are semi quantitatively in agreement with experiments especially on the PF$_6$ salt. How successful these one electron models will be in understanding the more complex behavior of the ClO$_4$ salt. with its complication of anion ordering remains to be seen. However, there appear to be some hints that the simple picture we have painted may not be the whole story. Experiments, in hand with theory, say that interactions may be important, especially in the presence of the magnetic fields relevant to this study. The magnetic fields tend to reduce the dimensionality for perpendicular electron motion (that is the essence of the FISDW) and this should enhance interaction effects [21,41]. Whether the metallic state remains a Fermi liquid and whether the nesting ideas remain relevant awaits further theoretical and experimental work.

Developments in related research areas also present challenges to our studies of the FISDW state. The distinction between integer and fractional QHE has been altered by the "discovery" of composite Fermions [62]. It is reasonable to ask whether such objects can be found (theoretically or experimentally) in the Bechgaard salts and whether we can distinguish between the integer and fractional QHE. Along these lines it has not escaped the attention of people working on the QHE that the Bechgaard salts present a rather unique opportunity to study the effects of coupling of the QHE layers [63]. The question is the nature of interplane transport (c axis conductivity in the QHE state). Since the system is gapped everywhere but at the edges [64], the transport must be from a circulating surface sheets whose properties change dramatically if the inplane state is integer or fractional QHE.

Returning to the inplane transport the nature of the transition between QHE steps is also quite interesting. Is there a low temperature Quantum phase transition between extended and localized states as in conventional QHE systems? Such a possibility is suggested by the resistance peaks between Hall steps seen in Figures 9 and 12 and by the multiple order parameter models [61].

As of yet there are many models but no consensus for understanding the origin of both the "fast oscillations" and the Lebed magic angles. Both single particle and electron interaction effects can account for many of the observations. The advent of easier pressure apparatuses along with better angle control and lower temperature in high magnetic fields should allow us to answer many of these outstanding questions experimentally. The physics of the anisotropic quasi-one, quasi-two-dimensional electron systems should prove at least as fascinating as the strictly two-dimensional electron system long studied in GaAs heterostructures.

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this field and particularly influenced my early work in thermoelectricity. In the several times that I met and hosted Igor I was equally impressed by his intelligence and his humanity. He was a rare person who will be greatly missed.

References


[18] Flux quantization provides an interesting interpretation of the interchain pairing suggested by Yakovenko V.M., *Europhys. Lett.* 3 (1987) 1041. If we are in the FISDW state with $q_x = 2k_F \pm N \pi$ then electrons are paired with holes $N$ chains away. The area defined by the wavelength along $x$, $2\pi/|N\pi$ times the interchain distance. $N_b$ contains one flux quantum. i.e. for $N = 5$ the wavefunction along $x$ is modulated by $2\pi/(5eHb/\hbar c)$ and the pairing is across 5 chains with a spacing $5b$. The flux enclosed is $\Phi = H \times 2\pi/(5eHb/\hbar c) \times 5b = hc/e = \Phi_0$. This construction results from a discussion with V. M. Yakovenko.


[27] The diagram in Figure 6c and the discussion here are merely meant as schematic. The actual nesting is probably closer to the ideal nesting vector $(2k_F, \pi)$ than the $(2k_F, 0)$ shown. Moreover, the nesting may leave both electron and hole pockets as here schematized or either electron or hole pockets depending on details of the bandstructure.


