Giant Angular Dependent Nernst Effect in the Q1D Organic conductor (TMTSF)$_2$PF$_6$

Weida Wu, N.P. Ong, and P.M. Chaikin

Department of physics, Princeton University, Princeton, New Jersey, 08544

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We present a detailed study of the Nernst effect $N_{xx}$ in (TMTSF)$_2$PF$_6$ as a function of temperature, magnetic field magnitude and direction and pressure. As previously reported there is a large resonant-like structure as the magnetic field is rotated through crystallographic directions, the Lebed Magic Angles. These Nernst effect resonances strongly suggest that the transport of the system is effectively “coherent” only in crystallographic planes along or close to the applied field direction. We also present analytical and numerical calculations of the conductivity and thermoelectric tensors for (TMTSF)$_2$PF$_6$, based on a Boltzmann transport model within the semi-classical approximation. The Boltzmann transport calculation fails to describe the experiment data. We suggest that the answer may lie in field induced decoupling of the strongly correlated chains.

I. INTRODUCTION

(TMTSF)$_2$PF$_6$ is a quasi-one-dimensional electronic system, which displays various ground states ranging from triplet superconductor\textsuperscript{1,2,3} to spin-density wave(SDW) insulator, depending on pressure, temperature and magnetic field\textsuperscript{4-7}. (TMTSF)$_2$PF$_6$ consists of plate-like TMTSF molecules which stack with strong wavefunction overlap in chains. The intrachain bandwidth is $\sim$ 1eV while the inter-chain couplings give anisotropic bandwidths of 0.1eV and 0.003eV in the approximately orthogonal directions. In the ‘metallic’ phase under moderate magnetic field, a fascinating phenomenon, the so-called Lebed Magic Angle Effect (MAE) was discovered\textsuperscript{1,2} after Lebed’s initial prediction\textsuperscript{8}. The first manifestations of these MAEs were sharp resistance dips when the magnetic field was aligned at inter-chain directions in real space (lattice vectors\textsuperscript{9}). In reciprocal space a field along the magic angles induces electron motion along commensurate $\mathbf{k}$ space orbits\textsuperscript{10}. Despite many theoretical efforts to describe the magic angle effect\textsuperscript{9,11,12,13,14,15,16,17}, there is no yet satisfactory explanation. Many of the theories focus on the semiclassical motion of electron on the open Fermi surface derived from single particle band structure.

Recently, a giant Nernst effect was discovered in (TMTSF)$_2$PF$_6$. As the magnetic field was rotated toward a magic angle the Nernst signal increased then decreased toward zero, changed sign at the magic angle and continued in an inverse manner. The result is a sharp resonant-like structure\textsuperscript{18}. The magnitude of the Nernst signal at 1K is at least 3 orders larger than what we expected from simple (Drude) estimates. The sign change of the Nernst effect at the magic angles strongly suggests that the transport involved in the Nernst effect is effectively 2-dimensional at these commensurate angles. Both the sign change at the magic angles and magnitude of the signal are not yet explained, but the effect appears generic for these materials. The giant resonant Nernst voltage has recently been observed in the sister compound (TMTSF)$_2$ClO$_4$\textsuperscript{19}. Present phenomenological models for the sign change involve field induced interplane decoupling\textsuperscript{20,21,22}. Although there is some experimental evidence for this decoupling, there are not yet theoretical models which rigorously demonstrate this phenomenon. Giant Nernst signals have also been seen in high transition temperature superconducting (HT$_c$) cuprates where a model invoking superconducting vortices and 2D superconducting phase coherence has been successful\textsuperscript{20,21,22}. A similar model has been proposed for (TMTSF)$_2$PF$_6$\textsuperscript{12}. However, to apply this idea in (TMTSF)$_2$PF$_6$ is quite controversial. On one hand it naturally explains the large Nernst signal with undetectable thermopower signal, predicts a particular sign of the Nernst effect confirmed by experiments and qualitatively explains some aspects of experiments. On the other hand, it predicts a large superconducting fluctuation region in the phase diagram, which is absent in other measurements. Most of the superconducting properties in (TMTSF)$_2$PF$_6$ have been understood within a conventional mean field BCS picture. We will explore the possibility of this vortex Nernst effect with more experimental detail in a subsequent paper.

What sorely hampers progress in understanding these unusual magic angle phenomena are the lack of measurements other than charge transport. The Field Induced SDW (FISDW) and MAE have been observed primarily in the charge channel by transport measurements. The FISDW has been more thoroughly explored with magnetization, magnetocaloric effect and spin relaxation studies. Magnetic torque measurements on (TMTSF)$_2$ClO$_4$ suggest there is a thermodynamic component to the MAE\textsuperscript{20}. While a thermodynamic probe is an obvious choice for establishing the presence of unknown phases or fluctuations in (TMTSF)$_2$PF$_6$, the high pressure environment makes a measurement of specific heat or dc magnetic susceptibility impractical. Recently, the $^{77}$Se NMR spin-lattice relaxation rate measurements\textsuperscript{23} at different magnetic field orientation show no evidence for either a spin gap or a single particle gap. Furthermore, there is no evidence for an enhancement of the FISDW transition temperature. This strongly suggests that neither FISDW
ordering nor fluctuations are likely to be responsible for the MAE. The dramatic contrast between the charge channel and the spin channel at MAs suggests that spin and charge degrees of freedom may be decoupled.\textsuperscript{24,25} The thermodynamic and suggested coherent-incoherent transitions would therefore be the result of interaction and correlation effects due to subtle changes in the electronic wavefunctions and density wave susceptibilities.

Before speculating further on exotic mechanisms for the giant Nernst resonances and other MAEs in transport it is necessary to see what conventional transport theory will yield. Although Boltzmann transport calculations as a function of magnetic field magnitude and direction have been performed for resistance\textsuperscript{26}, there has been no such study for the thermoelectric transport coefficients. Such calculations are one of the main contributions of this paper.

We divide our presentation into two sections. The first section focuses on the Nernst experiments. We present a detailed study of the Nernst effect $N_{xx}$ in (TMTSF)$_2$PF$_6$ at various pressures, magnetic fields and temperatures. The second part presents both numerical and analytic calculations of Boltzmann transport in the relaxation time approximation with realistic band parameters. We then compare the calculations with our experiment data.

II. NERNST MEASUREMENT

A. Method

Figure 1 shows the experimental setup for the Nernst effect $N_{xx}$ measurement with the temperature gradient along the $a$-axis and voltage measured along $c$-axis. 3 pairs of Au wires were attached to the opposite $ab$-planes of the sample by silver paint for both resistance measurements (the end pairs) and the Nernst measurements (the middle pair). The Au wires were attached to the alloy wires (Phospher Bronze) fed through the pressure cell base. Here we used low thermal conductivity alloy wires instead of Cu wires to minimize the possible transverse temperature gradients. A miniature heater was placed on top of the sample to establish a small temperature gradient along the $a$-axis. Two thin film RuO thermometers were used to measure the temperature difference. The thermoelectric voltage is measured by a Keithley 182 Nanovoltmeter. The heater was turned on and off for several cycles for signal averaging.\textsuperscript{26} A linear-fit-extrapolation method was used to accommodate the slow drift of the baseline signal.\textsuperscript{27} The resistance was measured by a conventional 4-probe low frequency lock-in technique. The magneto-resistance $R_{zz}$ and Nernst signals $N_{xx}$ were measured simultaneously.

Fig. 2 shows a typical angle dependence of the Nernst signal $N_{xx}$ in (TMTSF)$_2$PF$_6$, obtained at 1 K, 6 Tesla and 13 kbar. The magnetic field was rotated from $-40^\circ$ to $50^\circ$ with respect to the $c^*$-axis. (See Ref.\textsuperscript{18} for definitions of $c^*$, $c'$ etc.) The maximum Nernst signal is about 100 $\mu$V/K, found at approximately $3^\circ \sim 4^\circ$ off $c'$ ($\theta_c = 7^\circ$). The Nernst coefficient is of the order of 10 $\mu$V/K-T. As far as we know, this value is much larger than the Nernst effect observed in any other metal. The angular dependence of the Nernst signal agrees well with our previous thermoelectric measurement in a different geometry.\textsuperscript{18} To study the temperature, field and pressure dependence of the giant Nernst effect, we fixed the magnetic field orientation at $3^\circ$ off a magic angle ($c'$ or -$b$). First, let’s discuss the sign of the Nernst effect in (TMTSF)$_2$PF$_6$. This is very important for the vortex Nernst model.

FIG. 1: The measurement setup of Nernst effect $N_{xx}$: Three pairs of Au wires were attached to the sample along the $a$ axis on the opposite sides of the $ab$-planes of the sample. Two RuO thin film resistance thermometers were placed next to both ends of the sample to measure the temperature gradient generated by a miniature heater on the top. The middle pair of leads was used for Nernst voltage $\Delta V_z$ pickup. The other two pairs of leads were used for 4-probe interplane ($c$-axis) resistance measurement. The magneto-resistance, $R_{zz}$, was measured simultaneously with the Nernst effect.

FIG. 2: Angle dependence of the Nernst signal $N_{xx}$ and $c$-axis resistance $R_{zz}$ were measured simultaneously at 1 K, 6 Tesla and 13 kbar. The thin line data are $R_{zz}$($\theta$). The open circles "o" data are the Nernst data $N_{xx}$($\theta$). The solid line is a guide to eyes. The Nernst resonances are well aligned with the magic angles marked by the resistance dips. Here $\pm 1$ correspond to inter-chain directions $c' \pm b$.
B. The sign of the Nernst effect

In the vortex liquid phase of a type II superconductors vortices flow down the temperature gradient, $\nabla T$ and generate an electric field $E = B \times v$ transverse to the temperature gradient $-\nabla T$ according to Josephson relation\textsuperscript{28,29}. Therefore, the sign of the vortex Nernst effect is fixed by $\nabla T \times B$. In general, the Nernst effect of an electronic system can have either sign depending on details of the band structure. To determine the sign of the Nernst signal, we noted the orientation of the sample and leads, and placed an alignment mark on the base/feedthru of the pressure cell and on the cell body. We assume the alignment mark doesn’t change much on pressurization. To get the Nernst sign correct we need to know the orientation to better than 90°. We see the magic angles where we expect them to be to $\sim 15°$. We observed no orientation variation when the pressure was increased in the same pressure cell. Our measurements show that the sign of the Nernst effect is consistent with the vortex Nernst model (but certainly does not prove it).

C. Temperature dependence

![FIG. 3: Simultaneous measured temperature dependences of the c-axis resistance $R_{zz}$ (thin lines) and the Nernst signal $N_{zx}$ (filled symbols) at 8 kbar for $3°$ off $c'$. The Nernst signals rise to a maximum around 1 K, then fall exponentially and are unmeasurable below $\sim 200$ mK. The decrease of the Nernst signal is correlated with the upturn of the resistance as $T$ decreases. The upturn of $R_{zz}$ indicates the Metal-FISDW phase transition.](image)

![FIG. 4: Main panel: Temperature dependence of the Nernst signal $N_{zx}$ measured at $3°$ away from $-1$ Lebed angle at 8 kbar. The magnetic fields are: 7.5, 7, 6.5 and 6 Tesla respectively. The arrows mark the FISDW transition temperature obtained from resistance measurements $R_{zz}(T)$ (not shown for clarity) at various fields. Insert: Temperature dependence of the Nernst signal $N_{zx}$ and resistance $R_{zz}$ of $-1$ Lebed angle at 13 kbar for 7.5 Tesla magnetic field. The dash line marks the FISDW transition ($\sim 350$ mK). An enhancement of Nernst voltage was found in the FISDW phase.](image)

Interestingly, the presence of the FISDW phase affects the Nernst signal at $-1$ Lebed angle differently. As shown in Fig. 4 at 8 kbar pressure the FISDW onsets (from $R_{zz}$) coincide the onsets of a large increase of Nernst voltage at $-1$ Lebed angle at various magnetic fields. At still lower temperature the voltage at the $-1$ Lebed angle reaches a peak around $300 \sim 400$ mK then decreases quickly. The peak value is as large as $\sim 220 \mu V/K$ at 7.5 Tesla. This is further confirmed by the angle dependence of the Nernst effect at base temperature (150 mK), where there is large Nernst resonances at $\pm 1$ Lebed angles while there is none at $c'$. This behavior is consistent with our previous measurements\textsuperscript{28,29}. The effect of FISDW on $-1$ Lebed angles is further confirmed by measurements at higher pressures, where FISDW transition temperature $T_c(H)$ vary accordingly. For example, at 13 kbar the FISDW transition temperature at 7.5 Tesla is suppressed down to $\sim 350$ mK, the enhancement of the Nernst effect at $-1$ Lebed angle follows the FISDW transition accordingly as shown in the insert of Fig. 4.
The suppression of the Nernst signal at c' by the FISDW is not understood at this moment. It is probably due to the competition between the FISDW phase and 'metallic' phase. This difference seems to suggest the magic angle c' is different from -1 Lebed magic angle in a subtle way. In this paper, we limit our discussion within the “normal” state where the MAE is pronounced. We note that a full understanding of the MAE should cover the FISDW phase, where the MAE is more complicated than in the metallic phase.

D. Field dependence

![Graph showing magnetic field dependence of N_{zx} and R_{zz} measured simultaneously at 8 kbar, 1.6 K for c'. Clearly N_{zx} is non-linear with magnetic field just as observed in previous measurements. Lower panel: Ratio of N_{zx} and R_{zz} derived from the upper panel. As discussed in text, N_{zx} / R_{zz} ∝ α_{zx}. The dash line is a guide to eyes. It is clear that at low field α_{zx} is linear with field.]

FIG. 5: Upper panel: Magnetic field dependence of N_{zx} and R_{zz} measured simultaneously at 8 kbar, 1.6 K for c'. Clearly N_{zx} is non-linear with magnetic field just as observed in previous measurements. Lower panel: Ratio of N_{zx} and R_{zz} derived from the upper panel. As discussed in text, N_{zx} / R_{zz} ∝ α_{zx}. The dash line is a guide to eyes. It is clear that at low field α_{zx} is linear with field.

As seen in the temperature dependence of N_{zx} at different magnetic fields, the Nernst effect in (TMTSF)$_2$PF$_6$ is very non-linear with magnetic field. In the upper panel of Fig. 6, we show simultaneous measurements of magneto-resistance R_{zz} and the Nernst effect N_{zx} vs. magnetic field at 1.6 K, 8 kbar and 3° off c'. It is clear that the Nernst signal has a super-linear field dependence. An obvious nonlinear effect is the large magneto-resistance of (TMTSF)$_2$PF$_6$. In transport theory, the thermopower tensor S is the product of the resistivity tensor ρ and the thermoelectric tensor α.

\[
S = \rho \cdot \alpha
\]  

FIG. 6: Magnetic field dependence of N_{zx} and R_{zz} measured simultaneously at 8 kbar and 375 mK for field at 3° away from -1 Lebed angle. The upturn of the R_{zz} around 5.5 Tesla defines the threshold field of the FISDW transition. N_{zx} is highly non-linear and enhanced greatly in FISDW phase.

Therefore, S_{zx} = ρ_{zx}α_{zx} + ρ_{zy}α_{yx} + ρ_{zz}α_{zx} ≈ ρ_{zx}α_{zx}. Here we ignore the first two terms since the Hall effects are negligibly small in the metallic phase for (TMTSF)$_2$PF$_6$. To obtain α, we took the ratio of the Nernst signal N_{zx} and the resistance R_{zz} at the same field to obtain field dependence of α_{zx} ∝ N_{zx} / R_{zz} according to Eq. 1. The prefactor depends on the sample geometry. The result is shown in the lower panel of Fig. 6. Clearly α_{zx} is approximately linear with magnetic field below 5 Tesla. Therefore, the non-linearity of the Nernst signal N_{zx} in (TMTSF)$_2$PF$_6$ mainly comes from the large magneto-resistance. The linear field dependence for field along c' and -1 Lebed angle suggests that α_{zx} is probably a more fundamental quantity in the thermoelectric effect of (TMTSF)$_2$PF$_6$.

Fig. 6 shows the field dependence of N_{zx} and R_{zz} for field along 3° off the -1 Lebed angle at 375 mK. We can see that when the (TMTSF)$_2$PF$_6$ goes into the FISDW, the resistance rises up sharply around 5.5 Tesla due to the presence of the FISDW gap. The Nernst signal also shows a sharp upturn around 5.5 T and rises up dramatically. This agrees with the observation of the enhancement of the Nernst signal at -1 Lebed angle in the temperature dependence (Fig. 6).

E. The effect of pressure

The ground state properties of the Bechgaard salts are strongly affected by hydrostatic pressure. The tempera-
ture, pressure and magnetic field (T-P-H) 3D phase diagram (Fig.1 in Ref.31) summarizes the effects of pressure on various phase transitions. For example, the threshold field of the FISDW phase progressively increases as the pressure gets higher. The superconducting transition temperature \( T_c \) is also slowly suppressed by increasing pressure. Fig.7 shows the zero field temperature dependence of resistance \( R_{zz} \) at 8 kbar, 10 kbar and 13 kbar respectively. The superconducting transition temperature decreases slowly with increasing pressure. Using the onset definition (90% of the normal state value), we found that the superconducting transition temperatures \( T_c \) are 1.02 K, 0.87 K and 0.77 K respectively. In Fig. 8 we show the comparison of the temperature dependence of the normalized Nernst signal at 3° off \( c' \) between 8 kbar, 10 kbar and 13 kbar. Qualitatively, the temperature dependence of the Nernst signal is pressure insensitive.

The angular dependence doesn’t change significantly either as we vary the pressure. Fig. 9 shows the angular dependence of \( N_{zz}(“*”) \) and \( R_{zz}(\text{thin line}) \) at (a) 8 kbar, 2.1K and 7.5 Tesla; (b) 10 kbar, 660mK and 7.5 Tesla; (c) 13 kbar, 1K and 6 Tesla. Qualitatively, no change is observed.

III. BOLTZMANN CALCULATION OF NERNST EFFECT

To gain some elementary intuition about transport processes it is often instructive to look first at a generalized Drude approximation, by which we mean a classical gas of charged particle in lowest order response to an applied set of driving fields.

A. Drude Transport

In Fig.10 we show a cartoon of the particle motion of such a charged gas. In a Drude model forces accelerate particles which then lose momentum in collisions at a rate \( 1/\tau \). The basic equation of motion is therefore \( m\dot{v}/\tau = F \), the charge per particle is \( q \), the particle density is \( n \), the current density is simply charge density times velocity \( \dot{j} = nq\dot{v} \) and \( \dot{j} = \sigma \cdot E \). In Fig.10(a), \( F = qE \) and \( \dot{v} = \frac{q\tau}{m}E, \dot{j} = \frac{nq^2\tau}{m} \cdot E \), the conductivity \( \sigma \) is given by \( \sigma = \frac{nq^2\tau}{m} \). In the presence of the magnetic
field there is a Lorentz force which deflects particles in the y direction building up charges on the upper and lower boundaries. The charges continue accumulating until the electric field they generate exactly cancels the Lorentz force, $E_y = v_x \cdot B_z$. In steady state the Hall field $E_y = \frac{2n_q}{q} \cdot B_z$ completely compensates the effect of the magnetic field, the carriers only drift in the x direction and there is no magnetoresistance. In (b) the drive is a temperature gradient. In this simple model the gas is ideal and we use the ideal gas law, $P = n k_B T$. A temperature gradient translates to a pressure gradient $\nabla P = n k_B \nabla T$, or a force per particle of $F = \frac{\nabla P}{n}$. The charged particles will flow in the x direction charging the boundaries and creating an opposing field $E_x$. The current and charging stop when $q E_x = F_x$ or $E_x = \frac{k_B}{q} \nabla_x T$.

This thermoelectric voltage is the Seebeck effect with coefficient $S_1 = \frac{k_B}{q}$. (This was a big failure of the Drude model. It overestimates the thermopower by several orders of magnitude. The reason is quite evident today. We have a degenerate electron gas (DEG) rather than an ideal classical gas. The effective number of degrees of freedom, or particles that can transport heat, is reduced by $\sim \frac{k_B T}{E_F}$ so the $S_{1\text{DEG}} \sim \frac{k_B T k_B}{E_F q}$. In this picture the electric force cancels the pressure and the particles have no velocity, $\mathbf{v} \times \mathbf{B} = 0$ and the Nernst voltage is zero. For the simplest conductors we therefore expect the thermopower to be sizeable and the Nernst effect negligible. But it is worth noting that effectively the same argument would suggest that the magnetoresistance is negligible.

In Fig 10(c) and (d) we consider the case of two oppositely charged carriers, which are otherwise identical. With an electric field along x the two carriers move at $v_x = \frac{q}{m} E_x$ in opposite directions both contributing to the electrical current and conductivity which remains $\sigma_0 = \frac{n q^2 T}{m}$. Now however, in the presence of $B_z$ both are deflected in the same direction, there is no charge accumulated on the boundaries, there is no Hall voltage, and velocities persist in both directions, $v_y = v_x \frac{q B_z \tau}{m}$, $v_x = \frac{q \tau}{m} E_x - v_y \frac{q B_z \tau}{m}$, with the solution, $v_x = \frac{q \tau}{m} E_x - \frac{q B_z \tau}{m}$, with the solution, $v_x = \frac{q \tau}{m} E_x - \frac{q B_z \tau}{m}$. In (d) the drive is a temperature gradient again producing a pressure gradient. Both types of particles move down the pressure gradient with
velocity, \( v_x = \frac{q B \nabla x T r}{m} \) there is no charge accumulation, and no field generated along \( x \) so the Seebeck coefficient is zero. In the presence of a magnetic field, the particles with the same velocity but opposite sign are separated, charges accumulate on the upper and lower boundaries until the electric field compensates the Lorentz force, \( E_y = v_z B_z = \frac{k_B \nabla z T r B_z}{m} \). The result is a Nernst voltage with coefficient \( S_{gy} = \frac{k_B}{m} \frac{|q| B_z r}{E_F} = S_1 \omega_c r \).

With the degenerate electron gas correction we should then expect \( S_{xyDEG} \sim \frac{k_B}{|q|} \frac{k_B T}{E_F} (\omega_c r) \).

| one carrier | \( \sigma \) | Hall | MR | Seebeck | Nernst |
|-------------|----------------|------|-----|---------|---------|
| \( n q \tau \) | \( m \) | \( 1 \) \( n q \) | 0 | \( \frac{k_B}{q} B \frac{T}{E_F} \) | 0 |
| ambipolar | \( n q \tau \) | \( m \) | \( (\omega_c r)^2 \) | 0 | \( \frac{k_B}{|q|} B \frac{T}{E_F} \omega_c r \) |

The extension of these results to field orientation and different ratios of the densities of the oppositely charged particles is straightforward. With \( S_1 = \frac{k_B}{q} B \frac{T}{E_F} r \), \( a = \frac{n^+ - n^-}{n^+ + n^-} \), \( f(a) = a + \frac{(\omega_c r)^2}{1 + a^2 (\omega_c r)^2} \), \( g(a) = \frac{1 - a^2}{1 + a^2 (\omega_c r)^2} \), we find \( S_1(a) = S_1 f(a), S_{xy}(a) = S_1 (\omega_c r) g(a), R_B = \frac{1}{n |q|} \frac{\Delta \rho}{\rho} f(a), \frac{\Delta \rho}{\rho} = (\omega_c r)^2 g(a) \). The Hall and Nernst voltages vary as \( \mathbf{B} \times \mathbf{E} \) and \( \mathbf{B} \times \nabla T \) respectively. \( f(a) \) and \( g(a) \) are plotted in Fig. 11. Putting in realistic parameters, we will obtain the Nernst effect in Drude model is order of 10 nV/K, linear with magnetic field. Drude picture only predicts that the Nernst effect has a simple \( \sin \theta \) dependence of magnetic field orientation and linear with magnetic field and temperature.

![FIG. 11: Drude picture in general. Here \( a = \frac{n^+ - n^-}{n^+ + n^-} \) is the net charge density of mobile charge carriers.](image)

**B. Boltzmann Transport and Q1D Fermi Surface**

Simple Drude calculations are not capable of handling the highly anisotropic nature of the Bechgaard salts nor the angular orientation of the field relative to the lattice vectors. The simplest treatment which includes the bandstructure comes from a steady state Boltzmann equation. It has previously been shown that Boltzmann transport, appropriately modified to follow electron trajectories over many Brillouin zones (or equivalently many Umklapp scatterings) can give magic angle effect in \( R_{zz} \) Moreover, this model is in qualitative agreement with the measurements in the \((TMTSF)_2ClO_4\) salt. Although, Boltzmann transport and the Osada model have not been successful for \((TMTSF)_2PF_6\), they are still the only reasonable single particle treatment available. We therefore performed both numerical and analytical calculations using a Boltzmann Transport formulation based on the Tight Binding Approximation band structure within the single Relaxation Time Approximation. For simplicity, the triclinic crystal structure of \((TMTSF)_2X\) is taken as orthorhombic.

\[
\epsilon = -2 t_a \cos k_x a - 2 t_b \cos k_y b - 2 t_c \cos k_z c
\]

For \((TMTSF)_2X\), \( t_a \gg t_b \gg t_c \). Often people linearize the \( k_x \) dispersion for simplicity. One would obtain the so-called linearized dispersion:

\[
\epsilon - \epsilon_f = \pm h v_f (k_x + \frac{1}{t_f}) - 2 t_b \cos (k_y b) - 2 t_c \cos (k_z c)
\]

In this approximation, the Fermi velocity \( v_f \) (or the density of states on Fermi surface \( N_f(\epsilon_f) = 1/h v_f \)) is a constant for a given energy. Many of the angular magnetoresistance oscillations (AMRO), e.g. the Danner-Kang-Chaikin oscillation (\( ac \)-rotation)\(^{32}\), the third angle effect (\( ab \)-rotation)\(^{32}\) and the combination of them\(^{32}\) can be understood within this approximation. There is excellent agreements between experiment and theory, especially for \((TMTSF)_2ClO_4\). However, for a linearized dispersion relation the Hall effect is zero (\( \sigma_{xy} = \sigma_{xz} = 0 \)). It is not surprising the Nernst effect is also zero (\( S_{xy} = S_{xz} = 0 \)) in this approximation. In order to calculate the Nernst effect, we have to use the full dispersion or a nonlinear approximation in either numerical computation or in an analytic calculation.

In general transport theory, we consider both an electric current \( \mathbf{J} \) and a thermal current \( \mathbf{J}_T \) in response to an electric field \( \mathbf{E} \), a magnetic field \( \mathbf{B} \) and a temperature gradient \( -\nabla T \):

\[
\begin{aligned}
\mathbf{J} &= \sigma \cdot \mathbf{E} + \alpha \cdot (-\nabla T) \\
\mathbf{J}_T &= T \alpha' \cdot \mathbf{E} + \kappa \cdot (-\nabla T)
\end{aligned}
\]

Here \( \sigma \) is the electric conductivity tensor, \( \alpha \) and \( \alpha' \) the thermoelectric tensor and \( \kappa \) is the thermal conductivity tensor. Here \( \alpha_{ij}(\mathbf{H}) = \alpha_{ij}'(\mathbf{H}) \) according to Onsager relation\(^{34,35}\). For free electron gas, these coefficients can be obtained by applying the relaxation time
approximation\(^{36}\)

\[
\begin{align*}
\sigma &= \frac{e^2 T}{4\pi^3} \int dS_k \frac{v \langle v \rangle}{\hbar} \\
\alpha &= \frac{1}{eT} \frac{\pi^2}{3} (k_B T)^2 \frac{\partial \sigma(\varepsilon)}{\partial \varepsilon}
\end{align*}
\]

Here we assume an energy and momentum independent relaxation time \(\tau\). The velocity average is defined as:

\[
\langle v(k(t)) \rangle = \int_{-\infty}^{0} dt \frac{e^{\varepsilon t}}{T} v(k(t)), \text{ where } k(t) \text{ is the semi-classical motion of electrons on Fermi Surface in the presence of a magnetic field.}
\]

Eqs. (4) are the starting point of our calculation. From the general transport equations (Eqs. [4]), by setting \(J = 0\) as the boundary condition in thermoelectric measurements, one would find Eq. (11) The Nernst signal \(N_{xx}\) is an off-diagonal element of thermoelectric power tensor \(S\).

Now let’s focus on a quasi-1D (Q1D) system, (TMTSF)\(_2\)X. The Fermi surface of Q1D consists of two slightly warped sheets. In order to evaluate the Fermi surface averaging velocity \(\langle v \rangle\) we have to calculate the motion of electrons on Fermi Surface in the presence of the magnetic field. Here the \(a\)-axis \((k_x)\) is the best conductivity direction, so the Fermi surface is approximately normal to the \(k_x\) axis. Therefore, \(k_x\) is a function of \(k_y\), \(k_z\) and \(\varepsilon\) from the dispersion relation (Eq. 2), i.e.

\[k_x = k_x(k_y,k_z;\varepsilon)\]

Therefore, \(k_z\) is not an independent variable for the semi-classical motion of electrons, given that the motion of the election is confined to the Fermi surface in the presence of the magnetic field \(B\). The Equations of motion can be reduced to:

\[
\begin{align*}
\frac{dk_y}{dt} &= \frac{e}{\hbar} (v_x B_x - v_x B_z) \\
\frac{dk_z}{dt} &= \frac{e}{\hbar} (v_x B_y - v_y B_x)
\end{align*}
\]

By solving these two equations of motion Eq. 7 we can evaluate \(v(t) = v(k(t))\). This is what we need for the conductivity tensor \(\sigma\). From Eqs. 5 the thermoelectric coefficient tensor \(\alpha\) is proportional to the energy derivative of the conductivity tensor \(\sigma(\varepsilon)\) at the Fermi energy.

\[
\begin{align*}
\sigma_{ij} &= \frac{e^2 T}{4\pi^3} \int dS_k \frac{dk_y dk_z}{h|v_x|} v_i \langle v_j \rangle \\
\alpha_{ij} &= \frac{k_B^2 e^2 T}{12\pi^2} \frac{\partial}{\partial \varepsilon} \int dS_k \frac{dk_y dk_z}{h|v_x|} v_i \langle v_j \rangle
\end{align*}
\]

If we could find an analytic form of \(\sigma(\varepsilon)\), evaluating \(\alpha\) would be straightforward. In our analytic calculation, we derive an approximate analytic form for \(\sigma(\varepsilon)\), and we obtain \(\alpha\) by taking differentiation. On the other hand, it is straightforward to evaluate both \(\sigma\) and \(\alpha\) numerically. Here \(\alpha\) can be calculated by taking the energy derivative of each term in the integral. (Eq. 5)

C. Numerical Calculation

From Eq. 5 we can obtain:

\[
\alpha = \frac{k_B^2 e^2 T}{12\pi^2} \int dS_k \frac{dk_y dk_z}{h} \left[ \frac{\partial}{\partial \varepsilon} \left( \frac{1}{|v_x|} \right) v_i \langle v_j \rangle + \frac{1}{|v_x|} \frac{\partial v_i}{\partial \varepsilon} \langle v_j \rangle + \frac{1}{|v_x|} \frac{\partial v_i}{\partial \varepsilon} \langle v_j \rangle \right]
\]

Note here \(\alpha \propto T\) if we assume \(\tau\) is T independence, which is a good approximation at low temperature. It is straightforward to evaluate the first two terms. Since:

\[
\frac{\partial v_i}{\partial \varepsilon} = \hbar M^{-1} \frac{\partial k_i}{\partial \varepsilon}
\]

Here we define the inverse mass tensor \(M^{-1}\) as:

\[
(M^{-1})_{ij} = \frac{1}{h} \frac{\partial v_i}{\partial k_j} = \frac{1}{h^2} \frac{\partial^2 \varepsilon(k)}{\partial k_i \partial k_j}
\]

Here \(k_y\) and \(k_z\) are independent variables, so the energy dependence of the first two terms only comes from \(k_x\), which is a function of \(\varepsilon\) (see Eq. 6). Then, we will have:

\[
\frac{\partial \varepsilon}{\partial \varepsilon} = \frac{\partial k_x}{\partial \varepsilon} \frac{\partial k_x}{\partial \varepsilon} = \frac{1}{hv_x} \frac{\partial}{\partial k_x}
\]

Therefore:

\[
\frac{\partial}{\partial \varepsilon} \left( \frac{1}{|v_x|} \right) = \frac{-m_{xx}^{-1}}{v_x^3} \text{ and } \frac{\partial v_i}{\partial \varepsilon} = \delta_{i,x} \frac{m_{xx}^{-1}}{v_x}. \text{ For the (c) term, we exchange the differentiation and averaging(integral), i.e. } \frac{\partial}{\partial \varepsilon} \frac{\partial (\varepsilon)}{\partial \varepsilon} = \left[ \frac{\partial v_i}{\partial \varepsilon} \right]. \text{ Putting the (a), (b) and (c) terms together, we obtain an expression for the thermoelectric coefficient tensor } \alpha:
\]

\[
\alpha = \alpha^{(+)} + \alpha^{(-)}
\]

where \(\alpha^{(\pm)}\) is defined as:

\[
\alpha_{ij}^{(\pm)} = \frac{k_B^2 e^2 T}{12\pi^2} \frac{\partial}{\partial \varepsilon} \left[ \int dS_k dk_z (\pm) \left[ \frac{-m_{xx}^{-1}}{v_x^3} v_i \langle v_j \rangle + \frac{m_{xx}^{-1}}{v_x^3} \delta_{i,x} \langle v_j \rangle + \frac{v_j}{v_x} \langle \frac{\partial v_i}{\partial \varepsilon} \rangle \right] \right]
\]

Now we need to find \(\frac{\partial v_i}{\partial \varepsilon}(t)\), which involves the motion of an electron in the magnetic field. \(\frac{\partial v_i}{\partial \varepsilon}(t)\) depends on the value of the Fermi Energy not only through \(k_x\), but also through \(k_y\) and \(k_z\), because \(k_y\) and \(k_z\) are also functions of \(t\) when the electron is moving on the Fermi surface in the magnetic field. These functions depend on energy \(\varepsilon\) and the “initial” condition \((k_y^0, k_z^0)\), i.e.

\[
\begin{align*}
\begin{cases}
\dot{k}_y(t) = \dot{k}_y(t;\varepsilon, k_y^0, k_z^0) \\
\dot{k}_z(t) = \dot{k}_z(t;\varepsilon, k_y^0, k_z^0)
\end{cases}
\end{align*}
\]
which are satisfied by motion. However, we can find the differential equations directly if we don’t know the solution of the Equations of motion. Combining Eq.14 with Eq.9, we can obtain numerical solutions of \( \partial k_y / \partial \varepsilon \) and \( \partial k_z / \partial \varepsilon \) which are independent of \( \varepsilon \) at \( t = 0 \), the “initial” conditions for \( \partial k_y / \partial \varepsilon (t) \) and \( \partial k_z / \partial \varepsilon (t) \) are \( \partial k_y / \partial \varepsilon (0) = \partial k_z / \partial \varepsilon (0) = 0 \).

In summary, to calculate both \( \sigma \) and \( \alpha \), we numerically solve two sets of equations of motions, Eqs. 7 and Eqs. 14. To treat the differential equations in Eqs. 7 and 14, we use a 4th order Runge-Kutta Method. Then we numerically integrate \( \langle v \rangle \) and \( \langle \partial v / \partial \varepsilon \rangle \) and evaluate both \( \sigma \) and \( \alpha \) by Fermi surface integrals. Here we use the band parameters \( (t_a = 0.25 eV, t_b = 0.024 eV \) and \( t_c = 0.008 eV) \) from tight binding approximations, realistic lattice parameters \( (a=3.49 \ A, b=7.7 \ A \) and \( c=13.264 \ A) \) and a scattering time \( \tau = 4.26 \times 10^{-12} \text{sec} \) from previous studies by Danner et al. Here we use \( B=8 \) Tesla, and \( T=1 \) K, which are comparable with experiment conditions. The combination of the parameters produce \( \omega_c \tau = eB\tau / m_e = 6 \) for an isotropic free electron gas. We use a \( 20 \times 20 \) grid on the Fermi surface in the calculations.

We can’t derive the energy dependence of \( k_y \) and \( k_z \) directly if we don’t know the solution of the Equations of motion. From (Eq.6), we will find: 

\[
\begin{align*}
\frac{d}{dt} \frac{\partial k_y}{\partial \varepsilon} &= \frac{\partial k_y}{\partial \varepsilon} = \frac{e}{\hbar} \left( \frac{\partial v_x}{\partial \varepsilon} B_y - \frac{\partial v_z}{\partial \varepsilon} B_z \right) \\
\frac{d}{dt} \frac{\partial k_z}{\partial \varepsilon} &= \frac{\partial k_z}{\partial \varepsilon} = \frac{e}{\hbar} \left( \frac{\partial v_x}{\partial \varepsilon} B_y - \frac{\partial v_z}{\partial \varepsilon} B_z \right)
\end{align*}
\]

From (Eq.6), we will find:

\[
\frac{\partial k_y (t)}{\partial \varepsilon} = \frac{1}{\hbar v_x (t)} \left[ 1 - \hbar v_y (t) \frac{\partial k_y (t)}{\partial \varepsilon} - \hbar v_z (t) \frac{\partial k_z (t)}{\partial \varepsilon} \right]
\]

Combining Eq.14 with Eq.9 we can obtain numerical solutions of \( \partial k_y / \partial \varepsilon (t) \). Since \( k_y, k_z \) are independent of \( \varepsilon \) at \( t = 0 \), the “initial” conditions for \( \partial k_y / \partial \varepsilon (t) \) and \( \partial k_z / \partial \varepsilon (t) \) are \( \partial k_y / \partial \varepsilon (0) = \partial k_z / \partial \varepsilon (0) = 0 \).

In summary, to calculate both \( \sigma \) and \( \alpha \), we numerically solve two sets of equations of motions, Eqs. 7 and Eqs. 14. To treat the differential equations in Eqs. 7 and 14, we use a 4th order Runge-Kutta Method. Then we numerically integrate \( \langle v \rangle \) and \( \langle \partial v / \partial \varepsilon \rangle \) and evaluate both \( \sigma \) and \( \alpha \) by Fermi surface integrals. Here we use the band parameters \( (t_a = 0.25 eV, t_b = 0.024 eV \) and \( t_c = 0.008 eV) \) from tight binding approximations, realistic lattice parameters \( (a=3.49 \ A, b=7.7 \ A \) and \( c=13.264 \ A) \) and a scattering time \( \tau = 4.26 \times 10^{-12} \text{sec} \) from previous studies by Danner et al. Here we use \( B=8 \) Tesla, and \( T=1 \) K, which are comparable with experiment conditions. The combination of the parameters produce \( \omega_c \tau = eB\tau / m_e = 6 \) for an isotropic free electron gas. We use a \( 20 \times 20 \) grid on the Fermi surface in the calculations.
ρ tensor (σ⁻¹)

FIG. 13: Resistivity tensor ρ = σ⁻¹. Here we just perform a simple matrix inverse operation.

Although it is a little rough, it catches all the main features. We also used a 40 × 40 grid for some points and did not find a significant difference.

D. Analytic Calculation

The basis of our analytic approximation scheme is finding the proper correction to the linear dispersion approximation Eq. 3. We expanded v_x to next order to include the effect of non-linearity of the dispersion, i.e. \( v_x = v_f + \delta v_f \). This approach is basically the semi-classical version of Lebed’s quantum approach. It is straightforward to find: \( v_f = \frac{2t_a a}{\hbar} \sin k_f a \) here \( \cos k_f a = -\frac{\varepsilon}{2t_a} \) and

\[
\frac{\delta v_f}{v_f} = \frac{\cos k_f a}{\sin^2 k_f a} \left[ \frac{t_b}{t_a} \cos(k_f b) + \frac{t_c}{t_a} \cos(k_f c) \right] + ...
\]

Defining \( \beta \equiv -\frac{\cos k_f a}{\sin^2 k_f a} \frac{t_b}{t_a} \) and \( \gamma \equiv -\frac{\cos k_f a}{\sin^2 k_f a} \frac{t_c}{t_a} \), then we have:

\[
\begin{align*}
v_x &\approx v_f \left[ 1 - \beta \cos(k_f b) - \gamma \cos(k_f c) \right] \\
v_y &\approx \frac{2t_b b}{\hbar} \sin k_b b \\
v_y &\approx \frac{2t_c c}{\hbar} \sin k_c c
\end{align*}
\]  (15)

By substituting \( v_x \) in the equations of motion Eqs. 4, one can obtain analytic expressions of \( k_y(t) \) and \( k_z(t) \) and evaluate velocity averages \( \langle \psi \rangle \) for \( \sigma(\varepsilon) \). Then it is straightforward to obtain \( \alpha \) from Eq. 5. Details of the analytic calculation are presented in the appendix. As shown in Fig. 12 Fig. 13 Fig. 14 and Fig. 15 our analytic calculation reproduces the main features of the numerical calculation. In some cases, the results from different methods overlap. Therefore, we believe our calculations describe the main behavior of the Nernst signal in Boltzmann transport within the tight binding approximation.
taking the product of \( \rho \alpha \), we got the thermoelectric power tensor confirming our numerical results. Once we obtained our motivation for performing the analytic calculation to \( \alpha \) of tensor angle. We also confirmed other AMROs, e.g. \( \tau \) small hump at 1st Lebed angle: \( c+b \), which is about 30°in our approximation. This is one test that our calculations reproduce the angular dependence of \( \sigma \) in the matrix form. Here \( \theta \) is defined respect to the \( c \)-axis. Therefore \( \theta = 90^\circ \) corresponds to the magnetic aligning at the \( b \)-axis. As shown in the insert graph of \( \sigma_{zz} \), there is a small hump at 1st Lebed angle: \( c+b \), which is about 30°in our approximation. This is one test that our calculations reproduce the angular dependence of \( \sigma_{zz} \) calculated by Osada et al.\(^\text{39} \). By increasing the scattering time \( \tau \), or magnetic field, we can clearly resolve a peak at this angle. We also confirmed other AMROs, e.g. \( ac \)-rotation and reproduce the Danner oscillations\(^\text{22} \).

However, as far as we know, there is no calculation of tensor \( \alpha \) in the literature for comparison. This was our motivation for performing the analytic calculation to confirm our numerical results. Once we obtained \( \sigma \) and \( \alpha \), we got the thermoelectric power tensor \( S \) (Fig. 15) by taking the product of \( \rho = \sigma^{-1} \) (Fig. 14) and \( \alpha \) (Fig. 14).

\[
\alpha \sim \frac{\partial \sigma}{\partial \epsilon} \mu.
\]

E. Results

Fig. 12 shows the calculated angular dependence of the conductivity tensor \( \sigma \). The graphs are arranged in the pattern of the tensor elements \( \sigma_{ij} \) in the matrix form. For this case, \( \theta \) is defined respect to the \( c \)-axis, or magnetic field, we can clearly resolve a peak at this angle. We also confirmed other AMROs, e.g. \( ac \)-rotation and reproduce the Danner oscillations\(^\text{22} \).

However, as far as we know, there is no calculation of tensor \( \alpha \) in the literature for comparison. This was our motivation for performing the analytic calculation to confirm our numerical results. Once we obtained \( \sigma \) and \( \alpha \), we got the thermoelectric power tensor \( S \) (Fig. 15) by taking the product of \( \rho = \sigma^{-1} \) (Fig. 14) and \( \alpha \) (Fig. 14).

It is clear that the quality of numerical calculation of \( \sigma \) is much better than that of \( \alpha \). For \( \sigma \) most curves are very smooth and only minor oscillations are observed. For \( \alpha \) most curves are smooth, except \( \alpha_{yz} \) and \( \alpha_{zy} \). \( \alpha_{yz} \) has some spiky features for \( \theta \) close to \( c \); while \( \alpha_{zy} \) has some spiky features for \( \theta \) close to \( b \). This is because the energy derivative of the velocities is very sensitive to the location on the Fermi surface \( (k_y, k_z) \). Finite size grid integration could also generate artificial spikes if the integrand oscillates a lot.

By varying the grid size and the integration cut off limit, these artificial features can be suppressed, but at the expense of much more computation time. Since we are only interested in the general behaviors and magnitudes for a given set of parameters, we will use these non-perfect calculation results to compare with experiments, while keeping in mind that sharp features might be artificial. Also, our analytic results will help us to find out the physical features.

The Nernst signal \( S_{xx} \) corresponds to the experimental results discussed in the previous section. It is clear that
its angular dependence is sinθ-like, which agrees with the simple Drude model. The maximum magnitude is about 1 μV/K, which is 2 orders smaller than what we found in (TMTSF)2PF6 as shown in Fig. 15. This result is very different in shape from our observation, missing the resonances at magic angles and it gives the wrong temperature dependence (S ∝ T in Boltzmann Transport). Therefore, we conclude that the single particle picture is not able to describe the Nernst effect observed in (TMTSF)2PF6.

It is interesting to note that the Nernst signal Szz has a similar angular dependence as the experimental Nernst effect Szz, showing a peak near c and a sign change. However, the geometry is completely opposite and the value is about 8 orders of magnitude too small. Experimentally we couldn’t detect a sizable Szz, though we did not optimize the experimental setup for that measurement. Comparing the off-diagonal elements of S (i.e. the Nernst effect), we find that the elements in the upper triangle, Szxy, Szxz and Szxx are much smaller that those in the lower triangle, Sxy, Sxx and Syy. More surprisingly, the conjugate elements don’t have the same angular dependence.

Does this violate the Onsager relation? The answer is no. The Onsager relation only states constraint on σ and α (defined in Eqs. 4):

\[
\begin{align*}
\sigma_{ij}(H) &= \sigma_{ji}(-H) \\
\alpha_{ij}(H) &= \alpha_{ji}(-H)
\end{align*}
\]

By symmetry, we know that \(\sigma_{ij} (\alpha_{ij})\) is an odd function of magnetic field H for \(i \neq j\).

\[
\begin{align*}
\sigma_{ij}(H) &= -\sigma_{ij}(-H) \\
\alpha_{ij}(H) &= -\alpha_{ij}(-H)
\end{align*}
\]

Together with the Onsager relation Eq. 16, we can find:

\[
\begin{align*}
\sigma_{ij}(H) &= -\sigma_{ji}(H) \\
\alpha_{ij}(H) &= -\alpha_{ji}(H)
\end{align*}
\]

This is exactly what we see in the calculations (except \(\sigma_{yz} (\alpha_{yz})\) and \(\sigma_{zy} (\alpha_{zy})\), which are not real Hall effects since the magnetic field is in the plane). However, the thermopower tensor S is the product of \(\rho = \sigma^{-1}\) and α.
In general one should not expect $S_{ij} = -S_{ji}$. This is only true when we consider an isotropic system, where $\rho_{ii} = \rho_0$ and $\rho_{ij} = \rho_H$ for $i \neq j$. In an anisotropic system like (TMTSF)$_2$X, $\rho_{xx} \ll \rho_{yy} \ll \rho_{zz}$. If we ignore the Hall effect, we will have $S_{xy} \sim \rho_{xx} \alpha_{xy} \ll \rho_{yy} \alpha_{yx} \sim S_{yx}$. Nevertheless, the Nernst effect $S_{ij} (i \neq j)$ is an odd function of $H$ and Seebeck effect $S_{ii}$ is an even function of $H$ as long as there is an inversion symmetry.

In summary, we numerically and analytically calculated the thermopower tensor $S$ by evaluating both the conductivity tensor $\sigma$ and thermoelectric coefficient tensor $\alpha$. The numerical results agree well with the analytic approximation. This gives us confidence on the reliability of our calculations. It is clear that Boltzmann transport within a single particle picture is not consistent with our observation in (TMTSF)$_2$PF$_6$. Therefore, correlation effects due to the strong $e-e$ interaction should be considered in understanding the giant Nernst effect found in (TMTSF)$_2$PF$_6$. However, we note that the angular dependence of $S_{xx}$ fits the data in (TMTSF)$_2$ClO$_4$ very well, though there is a factor of 10 or so difference in magnitude. Our results are not limited to the Bechgaard salts (TMTSF)$_2$X. For any Q1D system with open Fermi surface, all the transport coefficients can be calculated using our results based on Boltzmann transport in a tight binding model. Our original results should prove useful for further investigations.

IV. DISCUSSION

The giant value of the Nernst effect and the Nernst resonances at Magic angles are not understood and it appears difficult to explain them in conventional Fermi Liquid models as illustrated by comparing our experiments and Boltzmann transport calculations. An exotic feature is that the Nernst signal changes its sign sharply at magic angles, with 3 “resonances” within $70^\circ$ in our measurements. As we know, the sign of the transverse electric field is determined by the cross product: $\nabla T \times B$. Of course, the physics really involves $E = v \times B$. Since the temperature gradient is fixed, the only quantity that could possibly change its sign is the component of $B$. As the magnetic field passes through a magic angle, the only component of magnetic field that could change sign relative to a magic angle is the one that is perpendicular to the direction of the magic angle. Therefore, we have to conclude that the Nernst signal in (TMTSF)$_2$PF$_6$ comes from $v \times B_{\perp}$. This means that whatever is moving is confined in the Magic angle plane. The main idea that underlies our interpretation of the Nernst resonances is that the transport is only coherent at the planes which are “parallel” (or close to parallel) to the magnetic field. In other words, the coherent electronic motion is controlled by the orientation of the magnetic field relative to the planes defined by the conducting chains and the interchain directions.

The nature of the coherence is not clear at this moment. One possibility is quasi-particle coherence. For example we may have a Field Induced Inter-
Field Induced Interchain/Interplane Decoupling picture.

Fig. 18: Field Induced Interchain/Interplane Decoupling

1. \( B_\perp > B^* \), \( t_{\text{eff}} = 0 \), chains are decoupled, Nernst signal is zero \( e_N = 0 \);
2. \( B_\perp < B^* \), \( t_{\text{eff}} \neq 0 \), chains are coupled, \( e_N > 0 \);
3. \( B_\perp = 0 \), \( t_{\text{eff}} \neq 0 \), chains are coupled, \( e_N = 0 \);
4. \( B_\perp < B^* \), \( t_{\text{eff}} \neq 0 \), chains are coupled, \( e_N < 0 \);
5. \( B_\perp > B^* \), \( t_{\text{eff}} = 0 \), chains are decoupled, Nernst signal is zero \( e_N = 0 \).

Fig. 18 shows the basic idea of this picture. (TMTSF)_2PF_6 is a Q1D system, which consists of conduction chains along the \( a \)-axis. The chains are weakly coupled with each other. When the magnetic field \( B \) is far from a magic angle, e.g. \( c' \), the inter-chain coherent coupling in this direction is effectively suppressed by the large normal field component, i.e., \( t_{\text{eff}} = 0 \) when \( B_\perp > B^* \) (here \( B^* \) is a cross-over magnetic field scale).

In other words, there is no coherent transport in the \( ac \)-plane for sufficient perpendicular field. (Fig. 18) When \( B \) is parallel to the \( c' \) direction, the coherent coupling in the \( c \)-direction is restored and a coherent \( ac \)-plane is formed. (Fig. 18) If we tilt the magnetic field slightly upward, there is a small component \( B_\perp \) of \( B \) normal to this plane. Here \( \delta \theta \) is small enough that \( B_\perp < B^* \). So the plane is still coherent. In the presence of temperature gradient \(-\nabla T\), there is a transverse electric field \( E \), the Nernst effect. When we tilt the field slightly to the other side of the magic angle, everything is the same except the sign of \( B_\perp \) reverses. Therefore, the sign of the Nernst signal is reversed. (Fig. 18) This scenario is then repeated at the other magic angles as in Fig. 17.

In fact, the idea of field induced decoupling is not new. Strong et al. \(^{20}\) considered the isolated conducting planes, \( ab \)-planes, of (TMTSF)_2PF_6 as 2D Non-Fermi-Liquid due to the strong \( e-e \) interactions. The possible non Fermi Liquid ground state of ”isolated” (TMTSF)_2PF_6 chains is supported by transport, optical, \(^{24}\) and thermal transport measurements. The effect of field induced coherent coupling/decoupling is supported by temperature and angular dependence magneto-resistance studies.\(^{45}\) However, this theory is not universally accepted due to the unknown nature of the non Fermi Liquid state and the lack of a detailed model.

Another possibility is the 2 dimensional superconducting phase coherence proposed by Ong et al.\(^{20}\) Ong points out that normal quasi-particles usually give a thermopower much larger than the Nernst signal whereas we have a large Nernst signal with undetectably small thermopower. On the contrary, vortex flow naturally produces an electric field that is predominantly transverse. This is generally true for most conventional systems, as well as HTc cuprates.\(^{20}\) Implicit in this model is the ability of the magnetic field to destroy phase coherence in the planes to which it is normal. It is natural that the vortices penetrating perpendicular planes destroy superconductivity. This is similar to the decoupling model discussed above and has the consequent “resonances” at magic angles.

V. CONCLUSION

We present a detailed study of the Nernst effect \( N_{zx} \) in (TMTSF)_2PF_6 as a function of temperature, pressure and magnetic field magnitude and direction. The data agree well with our previous measurements.\(^{15}\) We have calculated Boltzmann transport coefficients by both numerical and analytic methods with realistic band parameters within the single relaxation time approximation. The two calculations agree with either very well, but fail to describe the experimental data. The large magnitude, resonant-like angular field dependence and the non-linear field and temperature dependence cannot be understood within the semi-classical approximation of Boltzmann transport. The sign change of the Nernst effect at magic angles strongly suggests that the transport is effectively 2-dimensional in lattice planes parallel or close to the orientation of magnetic field. The nature of the coherence is not clear at this moment. We suggest that the answer may lie in field induced decoupling of the strongly correlated chains.

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VII. APPENDIX

Define \( \phi_0 = k_y b \) and \( \phi_c = k_z c \). In the presence of a magnetic field in the \( bc \)-plane \( B = (0, B \sin \theta, B \cos \theta) \), define \( \omega_0^b \equiv \omega_0 \cos \theta \) and \( \omega_0^c \equiv \omega_0 \sin \theta \), here \( \hbar \omega_0 = |e| \nu_f B b \) and \( \hbar \omega_c = |e| \nu_f B c \). \( e < 0 \) is the electron charge.
Then the equation of motion become:

\[
\begin{align*}
\frac{d\phi_b}{dt} &= \omega_b^0 (1 - \beta \cos \phi_b - \gamma \cos \phi_c) \\
\frac{d\phi_c}{dt} &= -\omega_c^0 (1 - \beta \cos \phi_b - \gamma \cos \phi_c)
\end{align*}
\]  

(19)

The exact solutions are hard to obtain. However, since \( \beta, \gamma \ll 1 \), the solutions can be approximated to 1st order by an iterative method:

\[
\begin{align*}
\phi_b(t) &= \phi_b^0 + \omega_b^0 t - \beta \sin(\phi_b^0 + \omega_b^0 t) - \gamma \frac{\omega_b^0}{\omega_c^0} \sin(\phi_c^0 - \omega_c^0 t) \\
\phi_c(t) &= \phi_c^0 - \omega_c^0 t + \beta \frac{\omega_c^0}{\omega_b^0} \sin(\phi_b^0 + \omega_b^0 t) - \gamma \sin(\phi_c^0 - \omega_c^0 t)
\end{align*}
\]  

(20)

Note here \( \frac{\omega_b^0}{\omega_c^0} \) diverges as \( \theta \to 0 \), and \( \frac{\omega_c^0}{\omega_b^0} \) diverges as \( \theta \to 90^\circ \). So this solution is only good at \( 0 \leq \theta < 90^\circ \).

One has to be aware that as \( \mathbf{B} \) approaches \( \mathbf{b} \) or \( \mathbf{c} \), this solution may not give the correct result. With the help of Jacobi’s expansions, it is straightforward to evaluate \( \mathbf{v}(t) \) (Eq.15) and \( \mathbf{v} \). Then we can obtain analytic expression for \( \sigma \). After tedious but straightforward calculation, we obtain the conductivity tensor \( \sigma \). Here we assume \( \gamma \ll \beta \ll 1 \), and only keep the leading terms in \( \beta, \gamma, \beta^2 \) and \( \beta \gamma \). For simplicity, we also use the anti-symmetric property of \( \sigma \). It is clear from Fig.12 that the analytic calculation reproduces the numerical results very well. Due to the limitation of our expansion, not every minor detail was reproduced. For example, in this 1st order approximation, \( \sigma_{xx} \) is independence of angle in analytic form, while numerically it shows a very weak angle dependence. The good agreement between different calculations give us confidence about the reliability of the calculations. Once we know the analytic form of the conductivity tensor \( \sigma \), we can take its energy derivative to obtain \( \alpha \).
\[
\begin{align*}
\alpha_{xx} &= -\frac{2\pi k_B^2 T \alpha_{\tau a}}{3\hbar^2 b c} \left\{ (1 - \beta J_1(\beta) - \gamma J_1(\gamma)) + 3[\beta J_1(\beta) + \beta^2 J_1'(\beta) + \gamma J_1(\gamma) + \gamma^2 J_1'(\gamma)] \right\} \\
\alpha_{yx} &= \frac{4\pi k_B^2 T \alpha_{\tau a}}{3\hbar^2 c} \frac{t_a^2 \omega^0_\tau[1 + 2(\omega^0_\tau)^2]}{t_a^2 [1 + (\omega^0_\tau)^2]^2} \\
\alpha_{xy} &= -\alpha_{yx} \\
\alpha_{zz} &= \frac{2\pi k_B^2 T \alpha_{\tau a}}{3\hbar^2 b} \left\{ \gamma \omega^0_\tau \left[ \frac{1 + (\omega^0_\tau)^2}{1 + (\omega^0_\tau)^2} - \frac{\gamma^3}{4} \cdot \frac{2 \omega^0_\tau}{1 + (\omega^0_\tau)^2} \cdot \left[ J_1(\gamma) \cdot \frac{5 + 7(\omega^0_\tau)^2}{1 + (\omega^0_\tau)^2} + 3 \gamma J_1'(\gamma) \right] - \frac{\gamma^2}{2} \right\} \\
&= \beta J_1 \left( \frac{\omega^0_\tau}{\omega^0_c} \right) + \gamma J_1 \left( \frac{\omega^0_\tau}{\omega^0_b} \right) \left[ \frac{1}{1 + (\omega^0_\tau - \omega^0_\tau)^2} + \frac{1}{1 + (\omega^0_\tau + \omega^0_\tau)^2} \right] \\
\alpha_{yy} &= \frac{\pi k_B^2 T \alpha_{\tau a}}{3\hbar^2 a} \left\{ 1 + 3(\omega^0_\tau)^2 \right\} \\
\alpha_{yz} &= \frac{\pi k_B^2 T \alpha_{\tau a}}{3\hbar^2 a} \left\{ \frac{1}{1 + (\omega^0_\tau)^2} \frac{\gamma J_1(\gamma)}{1 + (\omega^0_\tau - \omega^0_\tau)^2} + \frac{7 + 11 \cdot (\omega^0_\tau)^2}{1 + (\omega^0_\tau)^2} \right\} \\
&= \frac{1}{1 + (\omega^0_\tau - \omega^0_\tau)^2} + \frac{1}{1 + (\omega^0_\tau + \omega^0_\tau)^2} \\
\alpha_{zy} &= \frac{\pi k_B^2 T \alpha_{\tau a}}{3\hbar^2 a} \left\{ 1 + 3(\omega^0_\tau)^2 \right\} \\
\alpha_{xx} &= \frac{2\pi k_B^2 T \alpha_{\tau a}}{3\hbar^2 b} \left\{ \gamma \omega^0_\tau \left[ \frac{1 + (\omega^0_\tau)^2}{1 + (\omega^0_\tau)^2} - \frac{\gamma^3}{4} \cdot \frac{2 \omega^0_\tau}{1 + (\omega^0_\tau)^2} \cdot \left[ J_1(\gamma) \cdot \frac{5 + 7(\omega^0_\tau)^2}{1 + (\omega^0_\tau)^2} + 3 \gamma J_1'(\gamma) \right] - \frac{\gamma^2}{2} \right\} \\
&= \beta J_1 \left( \frac{\omega^0_\tau}{\omega^0_c} \right) + \gamma J_1 \left( \frac{\omega^0_\tau}{\omega^0_b} \right) \left[ \frac{1}{1 + (\omega^0_\tau - \omega^0_\tau)^2} + \frac{1}{1 + (\omega^0_\tau + \omega^0_\tau)^2} \right] \\
&= \frac{1}{1 + (\omega^0_\tau - \omega^0_\tau)^2} + \frac{1}{1 + (\omega^0_\tau + \omega^0_\tau)^2} \\
+ \beta J_1 \left( \frac{\omega^0_\tau}{\omega^0_c} \right) \left[ \frac{2 + 3(\omega^0_\tau)^2}{1 + (\omega^0_\tau - \omega^0_\tau)^2} - \frac{2 + 3(\omega^0_\tau + \omega^0_\tau)^2}{1 + (\omega^0_\tau + \omega^0_\tau)^2} \right] \\
&= \frac{1}{1 + (\omega^0_\tau - \omega^0_\tau)^2} + \frac{1}{1 + (\omega^0_\tau + \omega^0_\tau)^2} \\
&= \frac{1}{1 + (\omega^0_\tau - \omega^0_\tau)^2} + \frac{1}{1 + (\omega^0_\tau + \omega^0_\tau)^2} \\
\end{align*}
\]

Once we have \( \sigma \) and \( \alpha \), we can calculate \( S = \sigma^{-1} \cdot \alpha \). The analytic form of \( S \) would be too long to write down here since every term in \( S \) is the sum of three products of two matrix elements. We just numerically calculate the matrix product. Here we only present the angular dependence of all quantities in order to compare with experiment. In principle one could obtain temperature and field dependence with these formulae.

\* Current address: Department of Physics, University of Texas, Austin, Texas 78712

\[ \text{I. J. Lee, M. J. Naughton, G. M. Danner, and P. M. Chaikin, Phys. Rev. Lett. 78, 3555 (1997).} \]
