Quantum Hall Effect in a Bulk Crystal

S. T. Hannahs,[1] J. S. Brooks,[1] W. Kang,[2] L. Y. Chiang,[3] and P. M. Chaikin[2,3]
[1]*Department of Physics, Boston University, Boston, Massachusetts 02215*
[2]*Department of Physics, Princeton University, Princeton, New Jersey 08544*
[3]*Exxon Research and Engineering Co., Route 22E, Annandale, New Jersey 08801*
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We have observed remarkably flat Hall plateaus (step ratios . . . 5:4:3:2:1) with correspondingly sharp peaks in the magnetoresistance in the Bechgaard salt $(TMTSF)_2PF_6$. This highly anisotropic conductor exhibits magnetic-field-induced spin-density-wave (FISDW) states at low temperatures and pressures above about 6 kbar. Unlike previous measurements on similar materials we find almost complete confirmation of the theoretical model for the FISDW transitions, their quantum Hall effects, and the predicted high-field semiconducting state.

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Previously the integer and fractional quantum Hall effects (QHE) have been observed unambiguously only in the single-layer quantum wells formed in silicon metal-oxide-semiconductor field-effect transistors and semiconductor heterostructures[1,2] and in a few weakly coupled layers.[3] The only bulk materials which have shown QHE-like behavior are the Bechgaard salts $(TMTSF)_2X$,[4-6] where X is the anion ClO_4, ReO_4, or PF_6 and TMTSF is tetramethyl-tetraselenafulvalene. Theoretical work has shown that there should be a QHE associated with the field-induced spin-density-wave (FISDW) states,[7] and experimental observation of the QHE[8,9] and even the fractional QHE (FQHE)[10] in these salts has been suggested. However, the interpretation has been complicated by anomalies and additional transitions which question the basic theoretical understanding of these materials.[5,6] In the present Letter we report detailed measurements of the transport properties of $(TMTSF)_2PF_6$ under pressure which show almost complete agreement with expectations for a system undergoing a series of FISDW transitions to states with filled Landau bands exhibiting the QHE. A preliminary report of these results was presented in Ref. 11. Similar experiments have been recently done by another group.[12]

The theory which has been developed over the past several years for these transitions paints a fascinating picture.[13] In zero field the materials are quasi-two-dimensional open-orbit metals. Applications of a magnetic field reduces the dimensionality by one and the resulting one-dimensional metal is unstable against the formation of a spin-density wave of wave vector $q \sim 2k_F$. The reciprocal of the magnetic length $\lambda = eHk_Fb/\hbar c$ gives another periodicity which competes with q and produces a series of field-dependent Landau bands and gaps in the vicinity of the Fermi energy. The system lowers its energy by adjusting q so that E_F sits in the largest gap for a particular field. At low field there are many filled Landau bands. Increasing the field causes the gap structure to change, q jumps, and E_F lies in the next gap with one less Landau band filled. The cascade of transitions continues until there are no filled Landau bands and E_F rests in the one large symmetric gap which remains. From the initial transition to the last E_F remains always in a gap between Landau levels, producing a Hall effect with integral conductance quanta, ne^2/h, until the final semiconducting state is reached at high field ($n=0$).

Previous studies of the FISDW's have centered on $(TMTSF)_2ClO_4$. However, instead of the progression of Hall steps n, $n-1, \ldots, 1, 0$ there were intervening phases where the Hall resistance (ρ_{xy}) changed sign,[14] there was not a transition from $n=1$ to 0 but rather apparently[10] from $n=1$ to $\frac{1}{3}$, there was no evidence for the $n=0$ state anywhere in the phase diagram, and there was a reentrance of the metallic state at high field.[15]

Another perplexing limitation of the present understanding of these materials is the observation of a series of magneto-oscillations ("rapid oscillations")[16] which appear in both transport and thermodynamic properties of the ClO_4 and ReO_4 salts. Our present observation of these oscillations in the PF_6 salt rules out many of the possible explanations.[17]

The material $(TMTSF)_2PF_6$ is distinctly different from ClO_4 and ReO_4 in two ways: It has centrosymmetric anions with no anion-ordering transition and it forms a SDW phase below 12 K at ambient pressure. Upon application of hydrostatic pressure in excess of a critical pressure P_c, the SDW phase is suppressed in favor of a metallic (and below 1 K a superconducting) phase. For pressures above P_c FISDW behavior is observed above a threshold field.[18] The initial idea of the present study was to investigate the role of the anion ordering in all aspects of the high-field phases of these materials.

We used a miniature BeCu pressure clamp with a fluorocarbon fluid for pressures of 8 to 12 kbar. A loss of 2–3 kbar is expected on cooling to 4.2 K. Samples were mounted in a conventional six-probe configuration for longitudinal and ρ_{xy} measurements. Electrical contact to the samples was made with gold wires and silver paint. Experiments were carried out at the Francis Bitter National Magnet Laboratory.

We show in Fig. 1(a) low-temperature data for the magnetoresistance (ρ_{xx}) of $(TMTSF)_2PF_6$ at 10.5 kbar

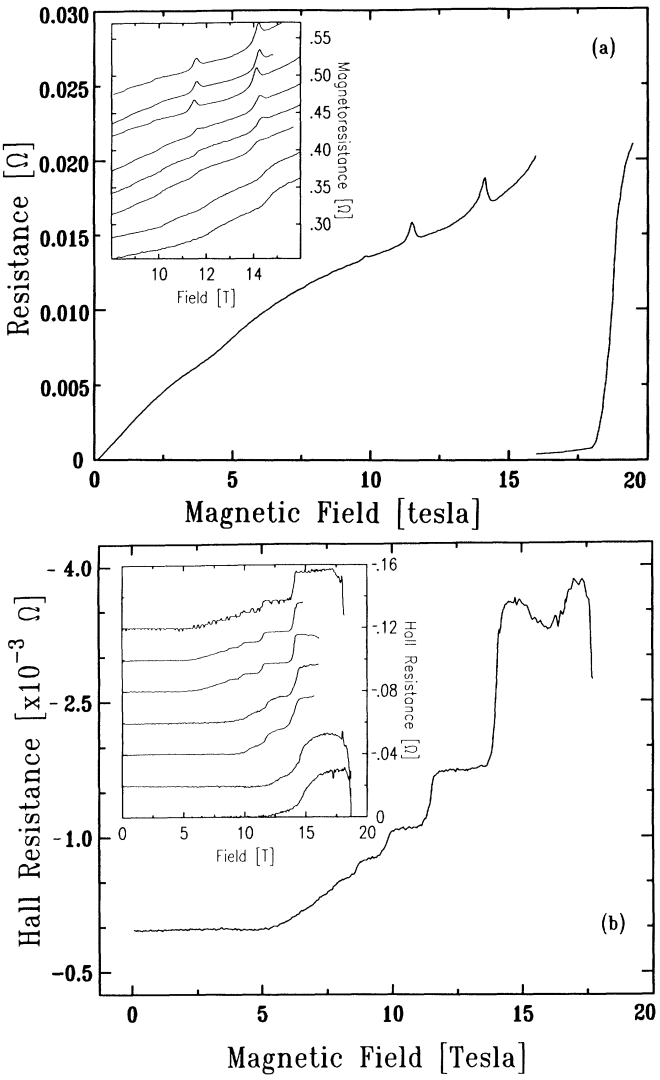

FIG. 1. Magnetoresistance and Hall resistance of $(TMTSF)_2PF_6$ vs magnetic field for different temperatures. (a) Original trace of mixed magnetoresistance and Hall signal at 0.1 K. Scale above 16 T is reduced by a factor of 50. Inset: Temperature evolution of the corrected magnetoresistance for (top to bottom) 0.1, 0.3, 0.33, 0.62, 1.01, 1.1, 1.5, and 2.0 K. (b) Hall signal at 0.1 K. Uncertainties above 15 T originate from errors in subtraction of digitized signals with low resolution and the rapidly increasing magnetoresistance terms. Inset: Similar traces for 0.1, 0.1, 0.33, 1.01, 1.1, 1.5, and 2.0 K. Note the negative sign of the Hall signal.

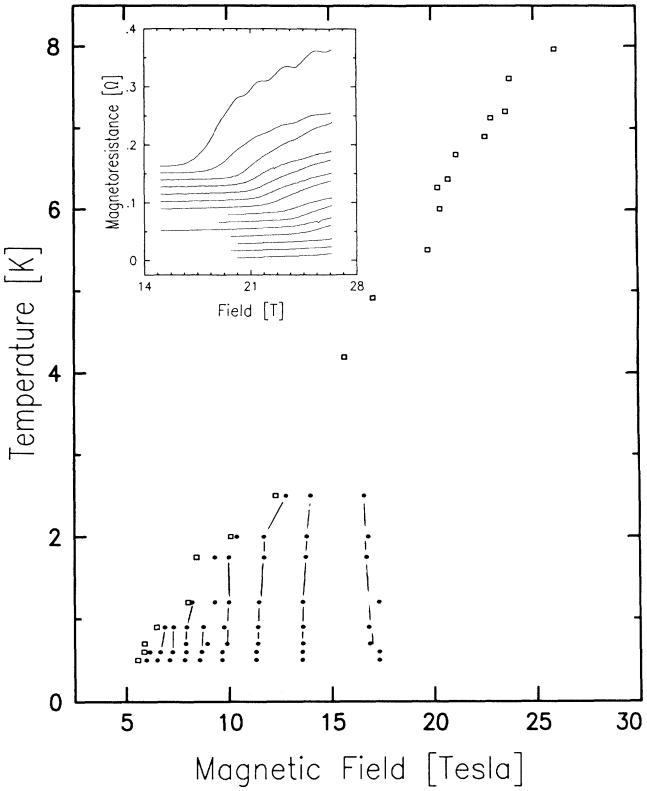

FIG. 2. *B-T* phase diagram produced from the positions of the peaks in the magnetoresistance. Threshold field and phase boundary are represented by squares. Inset: Magnetoresistance measurements used to determine the phase boundary at high temperature.

for fields up to 20 T. The ρ_{xx} data show the threshold field, and remarkably sharp structure at the onset of each FISDW phase. The inset shows the temperature evolution of the peak structure. The Hall signal, obtained by subtraction, is shown in Fig. 1(b). The data show a negligible ρ_{xy} in the metallic state at low field, a sharp upturn in ρ_{xy} at the threshold field, followed by a series of steps for each FISDW phase, and finally a large fluctuation in the Hall signal above 18 T. ρ_{xx} increases by 2 orders of magnitude and a subtraction to obtain the Hall signal is difficult. From many measurements on many samples we believe that ρ_{xy} is close to zero in this phase. The inset in Fig. 1(b) shows the temperature evolution of the shapes and magnitudes of the Hall plateaus.

A phase diagram is shown in Fig. 2. The inset shows the threshold region at high temperatures. Also evident are the rapid oscillations which become most apparent above the final transition. These oscillations are largest near 2 K.

In Fig. 3 we show field and temperature dependences. Figure 3(a) refers to the plot of the inverse Hall voltage versus step index. The data indicate the integer ratio of the steps. Deviations from integer behavior in the high-index limit arise from the closer proximity of these transitions to the threshold field (i.e., the steps have not yet reached their low-temperature limit). Figure 3(b) refers to the index of each transition versus $1/H$. Some hysteresis in the transition positions was observed for increasing and decreasing field sweeps. This hysteresis gives a 3% change in the intercept and a 0.3% change in the slope in this figure. Figure 3(c) shows the temperature dependence of the Hall steps, and Fig. 3(d) shows the corresponding temperature dependence of the ρ_{xx}

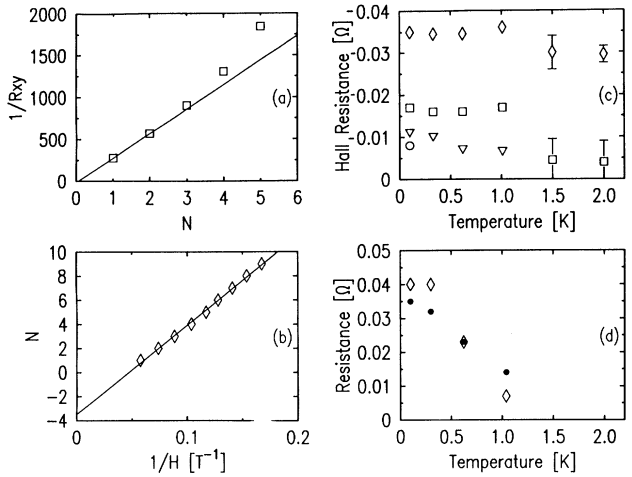

FIG. 3. (a) Plot of $1/R_{xy}$ vs FISDW phase index at low temperatures. The solid line is a linear fit to the $i=1$ and 2 values. (b) Plot of FISDW transition index vs inverse field at low temperatures. The line is a fit to all points. Note the negative intercept of -3.5. (c) Temperature dependence of the $i=1$ and 2 Hall plateaus: diamonds, $i=1$; squares, $i=2$; triangles, $i=3$; and circle, $i=4$. (d) Temperature dependence of the magnetoresistance peaks for $i=1$ (diamonds) and 2 (circles).

transition peaks.

At this point a comparison with the integer quantum Hall effect is clear: (1) We observe eight transitions above threshold, five of which yield measurable Hall signals in consecutive integer ratio and four of which are remarkably flat. (2) The peaks in ρ_{xx} are coincident with the onset of each new Hall step. The fact that the QHE *alone* will not explain all of the data is evident from the temperature dependence of the Hall steps and the negative intercept of the $1/H$ plot in Fig. 3(b). Both of these discrepancies are related to the existence of the FISDW's which make the gap, Fermi surface, and number of carriers temperature and field dependent.

It is also worth comparing this new data with previous experiments on the Bechgaard salts. The phase diagram is similar to that found before for $(TMTSF)_2ClO_4$ [15] and $(TMTSF)_2PF_6$ [19–21] but with important differences. The quasi de Haas–van Alphen frequency associated with the FISDW states is about 74 T at 10.5 kbar in contrast to 31 T in ClO_4. The FISDW subphases are much less temperature dependent than in the ClO_4 salt and there is no indication of reentrance in either the low-field [22] or high-field regions. There are no intervening phases between the consecutive integer phases, no sign reversals in ρ_{xy}, and no fractional phase after the $n=1$ phase. For the first time a transition is seen from the $n=1$ to an FISDW state with an activated resistance which can be identified by its Hall coefficient and field and temperature dependence at the *long missing* $n=0$ state.

The rapid oscillations seen here at high field and previously in the ReO_4 and ClO_4 salts are not yet understood. They have been interpreted as due to the effects of anion ordering, magnetic breakdown through the anion-ordering gap, or the existence of edge states. [17] The fact that we observe them in the PF_6 salt where anion ordering has never been observed and is not expected (since the anions are centrosymmetric) effectively rules out any of the interpretations relying on anion ordering. Rapid oscillations have also been seen in the PF_6 salt at ambient pressure in the non-field-induced SDW state. [23] Virtually all of the models of the rapid oscillations predict that their frequency is proportional to the b-axis bandwidth t_b. The quasifrequency of the FISDW's, on the other hand, is proportional to the area of the unnested part of the Fermi surface which for a simple band structure varies at t_b^2/E_F. Our values of the FISDW and rapid frequencies from four separate measurements (all at room-temperature pressures of about 10 kbar) are 72.6, 263; 64.9, 262; 74.1, 283; and 60.7, 291 T, respectively. Thus at least in this small range any correlations are not evident.

The above observations are based on investigation of nearly half a dozen samples. A variable in our measurements has been the *absolute* value of the Hall plateaus. Here, for $n=1$, we expect a value of $h/2e^2$ [corresponding to a value of 12.9 (kΩ/□)/layer for a spin degeneracy of two due to the SDW formation [7]]. To compare with our results we simply model the system as a parallel electric circuit so that the measured resistances are layer resistances divided by the number of layers. There can be considerable uncertainty in estimating the number of layers in a sample, and in the presence of sample cracks, and imperfect contacts, we further expect nonideal current paths to occur within the sample. However, we consistently find a value of 6.4 ± 0.8 (kΩ/□)/layer, a factor of 2 less than $h/2e^2$ and similar to the value found in ClO_4.

The temperature dependence of ρ_{xx} and ρ_{xy} should differ in the present case from that found for the conventional QHE. At low temperature the ρ_{xy} should be flat and ρ_{xx} should drop toward zero. At temperatures above 1 K we expect substantial changes in the resistances due to excitations above the SDW gap and to the temperature dependence of the gap itself. The SDW gap vanishes in a second-order transition as the phase boundary is approached with increasing temperature. The behavior of ρ_{xy} is as expected, but the behavior of ρ_{xx} is complicated by the presence of a background term (whose origin is not yet clear) on top of which a more conventional behavior is found. It is possible that the temperature dependence of the ρ_{xx} in $(TMTSF)_2PF_6$ is intrinsically different from that in semiconductor materials. In the standard model E_F is pinned energetically in a gap and the quantum number changes via a first-order transition to a state with E_F in the next gap. Thus unlike the conventional QHE there is no need for the system to sample extended states in going from levels n to $n-1$.

We finish our discussion by considering the possibility

of reentrance from a final SDW state above the last transition back into a 1D metallic state, as is the case for $(TMTSF)_2ClO_4$.[15] In the present data for $(TMTSF)_2PF_6$ ρ_{xx} increases very rapidly on leaving the $n=1$ state at the field where the $n=0$ transition is expected from the $1/H$ plot [Fig. 3(b)] and is thermally activated in this state. ρ_{xy} is near zero in this phase and heating reveals that a phase boundary must be crossed to enter the metallic state. These observations constitute our main evidence for the existence of the $n=0$ state. In contrast, for $(TMTSF)_2ClO_4$ ρ_{xx} does not rise significantly after the $n=1$ plateau, and ρ_{xy} increases to a larger value. The material remains a semimetal and has been suggested as being in the FQHE state. At much higher field the reentrant transition is reached. *Then* ρ_{xy} collapses, and ρ_{xx} increases rapidly, and is thermally activated. Theoretical work on the very-high-field phases of these materials is relatively new.[24] An estimate for the reentrance field is the frequency of the FISDW transitions. For $(TMTSF)_2ClO_4$ it is 31 T (28 T actual) and for $(TMTSF)_2PF_6$ it would be around 65 to 90 T for the pressures we have studied. In Fig. 2, it is clear that to 27 T there is no evidence for reentrance. Thus $(TMTSF)_2PF_6$ either has no reentrant phase or it will occur at fields unattainable with dc magnets.

Why are the present results so much cleaner and different from previous results? It it tempting to attribute the differences from $(TMTSF)_2ClO_4$ to the presence of anion ordering there and its absence in $(TMTSF)_2PF_6$. However, previous experiments on $(TMTSF)_2PF_6$[19-21] under pressure have indicated some of the more complicated behavior, such as sign reversals in ρ_{xy}, seen in $(TMTSF)_2ClO_4$. Thus the extraordinary effects observed in these materials may be very dependent on detailed subtleties of the band structure and scattering.

In conclusion, we elaborated on the first reported measurements on a bulk material which indicate an unambiguous integer quantum Hall effect. The QHE is no longer the exclusive domain of two-dimensional semiconductor devices. We have also found evidence for the complete cascade of field-induced spin-density-wave transitions expected from theory and have observed the previously unseen semiconducting state indicating the theoretical high-field limit. Thus it appears that the standard model for these transitions is experimentally well confirmed at least for one material and at one pressure. It remains to explain why other pressures and very similar materials show marked deviation from the standard model. The observation of rapid oscillations in the PF_6 salt rules out an explanation of this additional phenomena in terms of anion ordering and leaves it as the one remaining mystery in this material at this pressure.

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[1]K. von Klitzing, G. Dorde, and M. Pepper, Phys. Rev. Lett. **45**, 494 (1980).

[2]D. C. Tsui, H. L. Stormer, and A. C. Gossard, Phys. Rev. B **25**, 1408 (1982).

[3]H. L. Stormer *et al.*, Phys. Rev. Lett. **56**, 85 (1986).

[4]D. Jerome and H. J. Schultz, Adv. Phys. **31**, 299 (1982).

[5]See review articles by M. Ribault, P. M. Chaikin, G. Montambaux, and M. Herritier, in *Low Dimensional Conductors and Superconductors,* edited by D. Jerome and L. G. Caron, NATO Advanced Study Institute, Ser. B, Vol. 55 (Plenum, New York, 1987).

[6]See articles by K. Yamaji, P. Lederer, F. Pesty, X. Yan, G. Montambaux, P. M. Chaikin, P. Garoche, J. Brooks, and M. J. Naughton, in *Proceedings of the International Conference on Science and Technology of Synthetic Metals,* edited by A. J. Heeger [Mol. Cryst. Liq. Cryst. **27B** (1989)].

[7]D. Poilblanc *et al.*, Phys. Rev. Lett. **58**, 270 (1987).

[8]M. Ribault *et al.*, J. Phys. (Paris), Lett. **44**, L953 (1983).

[9]P. M. Chaikin *et al.*, Phys. Rev. Lett. **51**, 2333 (1983).

[10]R. V. Chamberlin *et al.*, Phys. Rev. Lett. **60**, 1189 (1988).

[11]S. T. Hannahs *et al.*, Bull. Am. Phys. Soc. **34**, 740 (1989).

[12]J. R. Cooper *et al.*, preceding Letter, Phys. Rev. Lett. **63**, 1984 (1989).

[13]L. P. Gor'kov and A. G. Ledel, J. Phys. (Paris), Lett. **45**, L433 (1984); P. M. Chaikin, Phys. Rev. B **31**, 4770 (1985); G. Montambaux, M. Heritier, and P. Lederer, J. Phys. (Paris), Lett. **45**, L533 (1984); M. Heritier, G. Montambaux, and P. Lederer, J. Phys. (Paris), Lett. **45**, L943 (1984); K. Yamaji, J. Phys. Soc. Jpn. **54**, 1034 (1985); M. Ya. Azbel, Per Bak, and P. M. Chaikin, Phys. Lett. A **117**, 92 (1986); K. Maki, Phys. Rev. B **33**, 4826 (1986).

[14]M. Ribault, Mol. Cryst. Liq. Cryst. **119**, 91 (1985).

[15]M. J. Naughton *et al.*, Phys. Rev. Lett. **61**, 621 (1988).

[16]H. Schwenk *et al.*, Phys. Rev. Lett. **56**, 667 (1986); T. Osada, N. Miura, and G. Saito, Solid State Commun. **60**, 441 (1986); Physica (Amsterdam) **143B**, 403 (1986); J. P. Ulmet *et al.*, Physica (Amsterdam) **143B**, 400 (1986); X. Yan *et al.*, Phys. Rev. B **36**, 1799 (1987).

[17]K. Yamaji, Physica (Amsterdam) **143B+C**, 439 (1986); J. Phys. Soc. Jpn. **55**, 1424 (1986); S. A. Brazovskii and V. M. Yakovenko, Pis'ma Zh. Eksp. Teor. Fiz. **43**, 102 (1986) [JETP Lett. **43**, 134 (1986)]; M. Ya. Azbel and P. M. Chaikin, Phys. Rev. Lett. **59**, 582 (1987); T. Osada and N. Muira, Solid State Commun. **69**, 1169 (1989).

[18]J. F. Kwak, J. E. Schirber, R. L. Greene, and E. M. Engler, Phys. Rev. Lett. **46**, 1296 (1981); Mol. Cryst. Liq. Cryst. **79**, 121 (1981).

[19]J. F. Kwak *et al.*, Phys. Rev. Lett. **56**, 972 (1986).

[20]M. Ribault *et al.*, Physica (Amsterdam) **143B**, 393 (1986).

[21]L Brossard *et al.*, Physica (Amsterdam) **143B**, 406 (1986).

[22]G. Montambaux *et al.*, Phys. Rev. B **39**, 885 (1989).

[23]J. P. Ulmet, A. Khmou, P. Auban, and L. Bachere, Solid State Commun. **58**, 753 (1986).

[24]V. M. Yakovenko, Zh. Eksp. Teor. Fiz. **93**, 627 (1987) [Sov. Phys. JETP **66**, 355 (1987)].