Negative Hall plateaus and quantum Hall effect in $(TMTSF)_2PF_6$

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We present our study of negative Hall plateaus associated with the magnetic-field-induced spin-density waves (FISDW). Slightly above the the pressure necessary to stabilize superconductivity, the quantum Hall effect associated with the field-induced spin density waves is interrupted by several negative Hall plateaus and an in-between phase. Quantizations of the negative Hall plateaus suggests that these states may be identified as FISDW states with negative quantum numbers. The in-between phase occurs from splitting of a phase boundary deep inside the FISDW diagram and is reminiscent of the previously suggested arborescence.
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The $(TMTSF)_2X$ (where TMTSF is tetramethyltetraselenafulvalene and $X=PF_6$, ClO_4, . . .) family of organic conductors are notable for the first organic superconductivity, field-induced spin-density waves[1] (FISDW) and the first quantum Hall effect as a bulk crystal.[2,3] Under low temperatures, high magnetic fields, and high pressures, $(TMTSF)_2PF_6$ exhibits a series of FISDW transitions accompanied by an integral quantum Hall effect states with $\sigma_{xy}=2Ne^2/h$, where $N=1,2,3, \ldots$ is the quantum number of each FISDW phase. In the theoretical model developed over a number of years, the quantum Hall effect in $(TMTSF)_2X$ occurs as a result of orbital quantization of free carriers in the presence of spin-density wave instability.[4–6] The observation of the quantum Hall effect and the associated phase diagram have provided compelling support for the model. In contrast to the theoretical predictions, certain regions of the phase diagram in $(TMTSF)_2X$ are notable for the appearance of negative as well as positive Hall plateaus. In this paper we present our study of the negative Hall plateaus in $(TMTSF)_2PF_6$.

The dramatic sign changes in the Hall effect were initially observed in highly ordered $(TMTSF)_2ClO_4$ samples cooled slowly through its anion-ordering transition at 24 K.[7] Some of the positive Hall plateaus seen earlier in partially ordered samples were replaced by negative Hall states in the more slowly cooled samples. Subsequent experiments also showed that the Hall effect in $(TMTSF)_2PF_6$ also exhibited prominent negative Hall effect.[8,9] Instead of few negative Hall phases seen in the ClO_4 salt, the PF_6 salt exhibited many sign changes, leading to a wildly oscillatory Hall effect that alternates between positive and negative states. In spite of numerous reportings of the negative Hall effect in later studies of $(TMTSF)_2X$,[3,10,11] no coherent picture has emerged so far.

It was recently suggested that the negative Hall plateau adjacent to the $N=2$ FISDW state in $(TMTSF)_2PF_6$ may correspond to a negative quantum numbered FISDW state.[12] Performed under pressure at which only one negative dip is present, electric-field treatment of the sample has shown to produce a negative Hall plateau whose magnitude closely matched that of the positive $N=2$ plateau. This assignment of $N=-2$ quantum number to the negative Hall plateau has stimulated recent investigation of the possible origin of the negative Hall effect in $(TMTSF)_2X$.[13,14] In one approach, the effect of pressure on the band structure was modeled as higher order harmonic terms that distort the shape of the Fermi surface.[13] The additional terms in the band structure result in the appearance of FISDW with carriers of opposite signs and consequently allow for the possibility of negative quantum numbered FISDW states. In an alternate approach, the effect of umklapp scattering on the FISDW transitions was studied.[14] Presence of umklapp scattering in the electron-electron interaction alters the thermodynamics of FISDW and produces commensurate FISDW states with negative quantum numbers. Depending on the choice of parameters, both models are able to produce a monotonic sequence of FISDW or an alternating sequence of positive and negative quantum-numbered FISDW states.

In this paper we report on our study of FISDW in the region of pressure slightly above the superconducting critical pressure. The sequence of positive quantum Hall states is interrupted by a series of negative Hall plateaus and an in-between phase. The in-between phase occurs from bifurcation of a phase boundary at low temperatures and is reminiscent of the arborescence of phase diagram that had been contemplated earlier in $(TMTSF)_2ClO_4$. Our experiments reveal a sequence of even quantum-numbered negative Hall plateaus that complement the series of positive Hall plateaus. The resulting, complex sequence of FISDW transitions does not exhibit a readily recognizable pattern. A high degree of sensitivity to pressure is exhibited by the negative Hall states and the inbetween phase. At high pressures, all intervening phases disappear and the previously reported positive quantum Hall effect is recovered.[2]

The experiment was performed using a ^3He cryostat in conjunction with a superconducting magnet. A miniature pressure cell was used to pressurize the sample. The pressure was gradually increased until the pressure barely exceeded the superconducting critical pressure of 6 kbar. Electrical contacts to the samples were made in standard six-probe geometry using silver paint to attach gold wires to the sample. High quality of the samples was evident from small mixing of magnetoresistance and Hall signals. In general, Hall resistance greatly exceeded the longitudinal resistance. No electric field treatment of the sample was necessary to observe the negative Hall plateaus. Care was taken to avoid sample

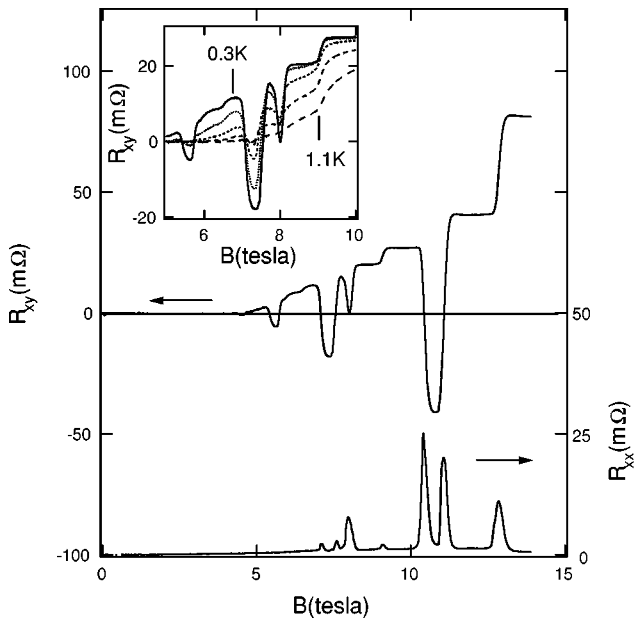

FIG. 1. Magnetoresistance and Hall effect of $(TMTSF)_2PF_6$ under 6.5 kbar of pressure at 300mK. Inset: Hall effect at 0.3, 0.5, 0.7, 0.9, and 1.1 K.

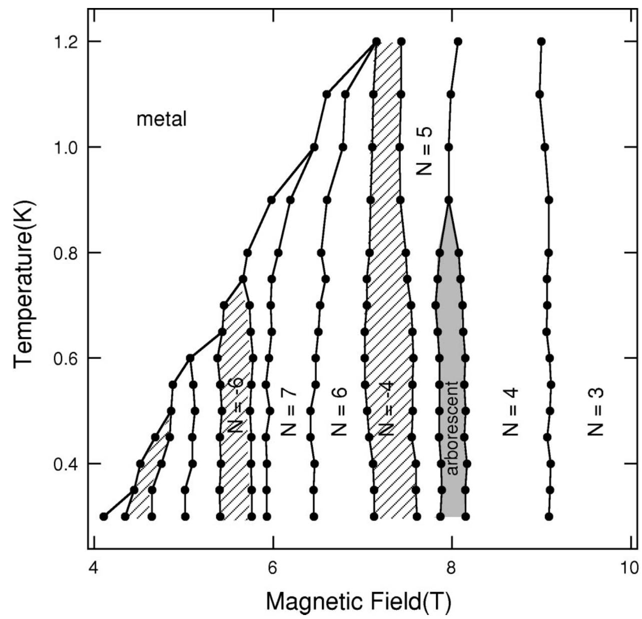

FIG. 2. Temperature-magnetic-field phase diagram of $(TMTSF)_2PF_6$ under 6.5 kbar of pressure. The gray-shaded ''arborescent'' phase corresponds to an inbetween phase that occurs from splitting of the phase boundary between the $N=4$ and 5 field-induced spin density waves. The phases that exhibit negative Hall effect are marked with diagonal lines.

heating and possible nonohmic behavior in the FISDW state. For most measurements, small excitation current, typically less than 100 μA, was used and no evidence of nonlinearity was observed in our measurements.

In Fig. 1 we present magnetoresistance and Hall effect from a high quality $(TMTSF)_2PF_6$ sample under 6.5 kbar of pressure. Above the initial FISDW transition at 4 tesla, approximately 17 transitions are observed at 300 mK. The transitions between FISDW subphases appear either as peaks in the magnetoresistance or jumps in the Hall effect. The Hall effect is distinguished by a series of plateaus that are interrupted by a number of sign reversals. At least four distinct negative Hall states are found. In general, these negative dips are much narrower than the neighboring positive plateaus and are accompanied by a doublet of magnetoresistance peaks. In addition to the negative Hall states, a shallow dip in the Hall effect is found at 8 tesla. The temperature dependence of the Hall plateaus between 5 and 10 tesla is illustrated in the inset of Fig. 1. While the magnitudes of the positive and the negative Hall plateaus decrease monotonically with increasing temperature, the dip in the Hall effect at 8 tesla initially becomes shallower as the temperature is raised. Above 0.9 K, the dip disappears and is displaced by the adjacent phase at lower magnetic fields.

The quantum number of the various FISDW states can be determined from the ratios of the Hall plateaus. The first four positive Hall plateaus can be clearly identified as the $N=1$, 2, 3, and 4 FISDW subphases. The narrow Hall plateau between the shallow dip and the second negative dip near 8 tesla is identified as the $N=5$ phase. The two Hall plateaus below the second negative Hall dip correspond to the $N=6$ and 7 subphases. Thus, we find a full sequence of $N=1, 2, 3, \ldots$ positive Hall plateaus. Turning to the negative Hall plateaus, the height of the largest negative Hall plateau at 11 tesla is within 1% of the Hall resistance of the adjacent $N=2$ plateau and we assign N $= -2$ as the quantum number for the state. The second-largest negative Hall plateau at 7.5 tesla is similarly identified as the $N=-4$ FISDW state. Based on the trend of the first two negative plateaus, the next two negative Hall phases occurring at 5.8 and 4.2 tesla are assigned as the $N=-6$ and -8 states, giving rise to the sequence of $N=-2$, -4, -6, and -8 negative Hall plateaus.

In Fig. 2 we present a temperature-magnetic-field phase diagram constructed from our extensive magnetotransport studies. The quantum number for each phase is derived from the ratios of the Hall plateau as discussed above. The FISDW states are separated from the metallic state by a boundary that rises roughly linearly with the magnetic field. Though differing in the sign of the Hall effect, the FISDW subphases with negative Hall plateaus are nearly indistinguishable from other states within the phase diagram. More interestingly, the phase boundary between $N=4$ and 5 states splits and defines an inbetween phase below a tricritical point at 0.9 K. This inbetween phase occurs as a shallow dip in the Hall effect at 8 tesla and displaces the $N=5$ phase at lower temperatures. This splitting of the phase boundary is suggestive of ''arborescence'' or a treelike splitting of the FISDW states that had been studied earlier in $(TMTSF)_2ClO_4$ (Refs. 15–17). While a multiple splitting of the FISDW phases into subphases and sub-subphases had been suggested, we observe only one phase that clearly demonstrates such splitting inside the FISDW phase diagram.

In Fig. 3 we present the temperature dependence of the Hall effect in various FISDW phases. While differing in sign, both positive and negative Hall phases are found to exhibit a rather similar temperature dependence. The magnitude of the Hall effect in both positive and negative phases increase with decreasing temperature. As the temperature is further decreased the Hall effect reaches saturation, with the

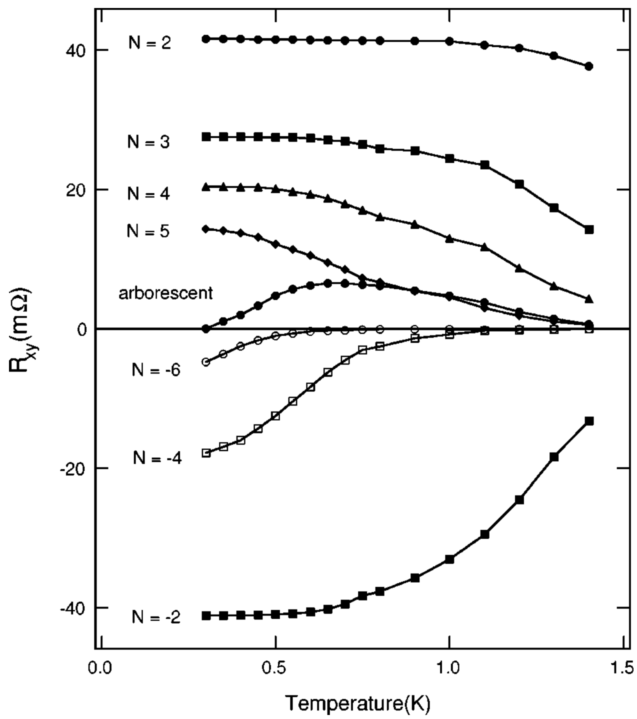

FIG. 3. Temperature dependence of Hall effect for various field-induced spin-density wave phases. The Hall resistance was measured at the middle of the plateau in each FISDW phase.

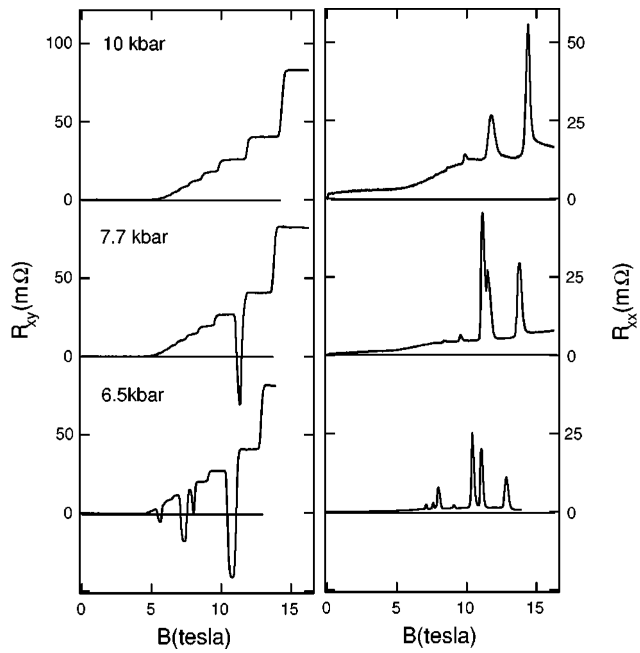

FIG. 4. Evolution of magnetoresistance and Hall effect of $(TMTSF)_2PF_6$ under pressure at 300mK.

saturation point being a function of the magnetic field. The Hall effect of the inbetween phase is accompanied by a strikingly different temperature dependence. Temperature sweep at 8 tesla cuts across two FISDW phases as can be seen in Fig. 2. Above 1 K the Hall effect mimics the data from the adjacent $N=5$ phase. Upon entering the in-between phase below 1 K, the Hall effect rapidly decreases to zero as temperature is lowered.

In Fig. 4 we compare the magnetoresistance and Hall effect under different pressures. While the sample under 6.5 kbar of pressure exhibits a sequence of negative Hall plateaus and the in-between phase, a slight increase in pressure to 7.7 kbar removes all but the strongest negative Hall plateau at the $N=-2$ phase. At 10 kbar no negative Hall plateau is found and only the sequence of positive Hall plateaus is found. Similar evolution of the phase diagram is also reflected in the magnetoresistance. The split peaks in magnetoresistance observed at 6.5 kbar has begun to merge into one at 7.7 kbar of pressure, and the splitting has completely disappeared at 10 kbar of pressure.

Our study of $(TMTSF)_2PF_6$ shows that there are at least four distinct FISDW phases that exhibit the negative Hall effect in the vicinity of the superconducting critical pressure. The Hall effect at these pressures consists of $N=1,2,3,4,\ldots$ positively quantized Hall plateaus as well as the sequence of $N=-2$, -4, -6, and -8 negatively quantized Hall plateaus. The evolution of the negative Hall states with temperature, as shown in the Hall effect and the phase diagram, suggests that these states are rather similar to other FISDW states. These results provide a strong support for the theoretical predictions that the negative Hall plateaus correspond to FISDW with even and negative quantum numbers.[13,14] The observed pressure dependence of the Hall effect is qualitatively consistent with changes in either the band structure[13] or the amplitude of umklapp scattering.[14] Our experiment shows that addition of approximately ~ 3 kbar of pressure is sufficient to completely alter the Hall effect. It remains to be seen whether such a small change in the pressure produces large enough modification of parameters in these models to explain the observed changes in the Hall effect.

Both theoretical models predict that some of the positive quantum numbered FISDW states are suppressed due to the appearance of negative Hall plateaus.[13,14] The experimentally observed sequence of FISDW states at low temperatures is as follows: $N=0$, 1, 2, -2, 3, 4, the inbetween phase, 5, -4, 6, 7, 8, -6, ... No obvious trend or pattern is evident in the sequence of observed transitions. While some FISDW phases are slightly displaced by the negative Hall plateaus and the inbetween phases, no suppression of positive quantum-numbered states are found. One possible source of the discrepancy may lie in the difference between the orthorhombic symmetry employed in the model band structure and the triclinic symmetry found in the real crystal. Additional refinement of the quantized nesting model may be necessary to resolve the difference from experimentally observed to the theoretically predicted phases.

In addition, the appearance of the inbetween phase below 1 K suggests of a more interesting possibility. The inbetween phase derives from the splitting of the phase boundary between $N=4$ and 5 FISDW subphases. Unlike other FISDW phases, the Hall effect in the inbetween phase rapidly decreases to zero as the temperature is lowered, and the magnetoresistance never fully develops into a minimum. At pressures slightly below 6.5 kbar, the Hall effect in the arborescent phase actually becomes negative at low temperatures. At higher pressures, it quickly disappears. The bifurcation of the phase boundary, giving rise to a FISDW subphase deep inside the phase diagram, is reminiscent of the ''arborescence'' or the treelike splitting of FISDW phases that has been reported in $(TMTSF)_2ClO_4$.[15] In our experiment, only one phase is observed with no evidence of additional transitions.

The theoretical study on arborescence has suggested possible mechanisms for arborescence as a fractional quantization of the Landau levels from a generalized nesting condition,[15] multiple order parameter FISDW,[16] and many-order parameters and higher-order energy gaps.[17] However, subsequent experiments have discounted the presence of arborescence in the ClO_4 salt,[18] and the applicability of these works to the inbetween phase seen in the PF_6 salt is unclear at this time.

While our paper supports the notion of sign-reversed Hall states being negative, even quantum numbered FISDW states, there still remains a question over the negative Hall states in $(TMTSF)_2ClO_4$. The negative Hall dips in the ClO_4 salt do not exhibit the quantization observed in the PF_6 salt, and the strongest negative Hall state in the ClO_4 salt is considerably wider and stronger than the negative Hall plateaus in the PF_6 salt. Since the ClO_4 salt is complicated by the presence of anion ordering, whether the sign reversals of the Hall effect in both salts occur from the same mechanism remains an open question.

In summary, we have identified the series of FISDW states that produce sign reversals of the Hall effect in $(TMTSF)_2PF_6$. At pressures slightly above the superconducting critical pressure, the negative Hall plateaus can be identified as even quantum-numbered FISDW states that are interspersed about the phase diagram. In addition, an inbetween phase reminiscent of previously contemplated arborescence is found. Both the negative Hall plateaus and the inbetween phase exhibit sensitivity to pressure. While a good qualitative agreement can be found with the modified quantized nesting models, the sequence of FISDW transitions and the origin of the inbetween phase require additional investigation.

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