Sign Reversal of the Quantum Hall Number in $(TMTSF)_2PF_6$

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The quantum Hall effect is investigated in the organic conductor $(TMTSF)_2PF_6$ (TMTSF is tetramethyltetraselenafulvalene) under hydrostatic pressure. Under a pressure of $p = 9$ kbar, the sequence of the quantum Hall numbers $L = \sigma_{xy}/(-2e^2/h)$ with increasing magnetic field is regular: $L = \ldots, 3, 2, 1, \ldots$ as reported earlier. At $p = 8.5$ kbar, however, the irregular sequence $L = \ldots, 3, -2, 2, 1, \ldots$ is found. The temperature–magnetic field phase diagram is investigated in detail. We discuss our findings in the framework of recent theories of the quantum Hall effect in magnetic-field-induced spin-density waves.

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One of the most compelling results of the theory of the quantum Hall effect (QHE) is the prediction by Thouless *et al.* [1] on the Hall conductivity of a two-dimensional electron gas subjected to a periodic potential. They have shown that instead of the regular sequence $(1, 2, 3, \ldots)$ of the quantum Hall numbers $L = \sigma_{xy}/(e^2/h)$ in a periodic potential L varies wildly as a function of the Fermi energy, taking both positive and negative integer values. In the light of this result, much attention has been devoted to the organic conductors $(TMTSF)_2X$ (TMTSF is tetramethyltetraselenafulvalene, and $X = PF_6$, ClO_4, or ReO_4), the only bulk crystals showing quantum Hall effect [2], since the Hall conductivity changes sign in these materials in some cases [3].

The best-known member of the family, $(TMTSF)_2PF_6$, is quasi-one-dimensional, and has a spin-density-wave (SDW) ground state ($T_c = 12$ K) at ambient pressure. If a hydrostatic pressure of $p \sim 10$ kbar is applied, the anisotropy decreases, and the system can be considered a collection of weakly coupled conducting planes. The SDW transition is suppressed and the ground state is superconducting. Applying a magnetic field perpendicular to the planes restores the SDW instability above a critical field $B_c \approx 5$ T, and a cascade of field-induced SDW (FISDW) phases appears [4]. In a semiclassical picture, justified by a quantum mechanical treatment, the FISDW can be considered a semimetal with most electrons condensed to the SDW and with some electrons left in unnested pockets of the Fermi surface [5,6]. The position of the FISDW gap in reciprocal space and the carrier concentration are related to the wave vector of the antiferromagnetic modulation. The system minimizes its energy by adjusting the wave vector so that an integer number N of Landau levels are full. The component of the corresponding FISDW wave vector parallel to the conducting chains is

$$q_x = 2k_F - N|e|Bb/\hbar, \qquad (1)$$

where k_F is the Fermi wave number, B is the magnetic field, and b is the lattice constant perpendicular to the chains. The Hall conductivity is quantized [7]: $\sigma_{xy} = -N2e^2/h$, where the factor of 2 reflects the fact that the SDW couples spin-up and spin-down states. To be consistent with the literature, we redefine the quantum Hall number for the FISDW case as $L = \sigma_{xy}/(-2e^2/h)$, therefore we have $L = N$.

It has been found in all the three compounds ($X = ClO_4$ [3,8], PF_6 [9], and ReO_4 [10]) that under certain circumstances the Hall conductivity may change sign in a restricted range of magnetic fields. This is the "Ribault anomaly" [3]. Well-defined Hall plateaus, however, have been observed only in regions where $\sigma_{xy} < 0$, i.e., $L > 0$. No $L < 0$ Hall plateau has been described so far.

In this Letter we show unambiguous evidence for the existence of a quantized "negative" Hall conductivity with $\sigma_{xy} = -2 \times (-2e^2/h)$ in $(TMTSF)_2PF_6$ in the pressure range $p \leq 8.5$ kbar. The phase with negative Hall number appears between two positive-Hall-number phases with $L = 2$ and 3. The transitions $L = 3 \to -2$ and $-2 \to 3$ are strongly hysteretic as a function of magnetic field and temperature, and exhibit a complex fine structure. We show, however, that if the FISDW's are depinned by applying a high current every time after the magnetic field is changed, the hysteresis and the fine structure disappear, indicating that there are no additional phases at the transition to or from the negative-L phase. We investigate the temperature–magnetic field phase diagram and show the existence of a transition line between the metallic and $L = -2$ FISDW phases. This is a serious constraint on the interpretation, and we conclude that there is no need to invoke multiple-order-parameter FISDW states to describe our findings. If the pressure is increased to 9 kbar, the $L = -2$ phase disappears.

We investigated two single crystals of $(TMTSF)_2PF_6$. The hydrostatic pressure was produced by a Cu-Be pressure clamp. Pressure values throughout this Letter are nominal values applied at room temperature; the low-temperature values may be lower by up to 2 kbar due to pressure loss upon cooldown. We find the critical

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pressure for suppressing the SDW state without magnetic field at about 8 kbar. The samples were visually aligned with the a-b face perpendicular to the magnetic field. Most of the measurements have been performed on sample 2 in a 12-T solenoid with a ^{3}He refrigerator. Some control measurements on sample 1 were performed in a 20-T solenoid equipped with a dilution refrigerator.

To prepare the electrode contacts, four gold stripes were evaporated on both a-c^* faces of the crystal as indicated in the inset of Fig. 1(a). This arrangement allowed us to measure the four components of the resistance tensor in the conducting planes, R_{ij}, where $i, j = x$ refers to the direction parallel to the conducting chains (a direction) and y to the second best conducting (b') direction. To measure R_{xx} and R_{yx}, the current flows from contacts 1 and 8 to 4 and 5 and the voltage is measured on contacts 2 and 3 as well as on 2 and 7, and we define the resistance tensor components as $R_{xx} \equiv V_{23}/I_{18,45}$, $R_{yx} \equiv V_{72}/I_{18,45}$. Similarly, $R_{yy} \equiv V_{45}/I_{23,67}$ and $R_{xy} \equiv V_{14}/I_{23,67}$. In this Letter we discuss results obtained in high magnetic fields where $|R_{ii}| \ll |R_{xy}|$. In this region, taking the symmetric (for R_{ii}) or antisymmetric (for R_{xy}) averages over the two signs of the magnetic field gives a negligible correction to the components of the resistance tensor.

Figure 1(a) shows the Hall resistance as a function of magnetic field in $p = 9$ kbar and $T = 150$ mK. Under these conditions, FISDW's appear above $B_c = 5.1$ T. Well-defined Hall plateaus develop in high fields with resistance ratios $1 : \frac{1}{2} : \frac{1}{3} : \ldots$ corresponding to the FISDW phases $N = 1, 2, 3, \ldots$, respectively. If the pressure is reduced from 9 to 8.3 kbar, a new feature appears in the Hall resistance: The resistance becomes negative in a narrow field range between the $N = 2$ and 3 phases [Fig. 1(b)]. This result is reproduced on sample 2 with a nominal pressure of 8.5 kbar [Fig. 1(c)]. As indicated in the figure, the Hall resistance in this region is very close to the value corresponding to $L = -2$.

Figures 1(d) and 1(e) show the longitudinal resistances R_{xx} and R_{yy} under the same conditions as in Fig. 1(c). Both quantities exhibit qualitatively the same behavior: Above the critical field of FISDW formation, the resistance first increases reflecting the decrease of carrier concentration, then decreases in high fields because of the increasing carrier scattering time in the quantum Hall region. The transition from one phase to another is marked by peaks in the diagonal resistances probably because a mixture of domains of the two phases is present [11]. Notably, there are peaks at the $L = 3 \to -2$ and $-2 \to 2$ transitions, and between these peaks the diagonal resistances are low. This is exactly the behavior expected for a new FISDW phase showing QHE.

Figures 2(a) and 2(b) display the neighborhood of the $L = -2$ phase on an inflated field scale. The transitions to or from this phase are strongly hysteretic and have a

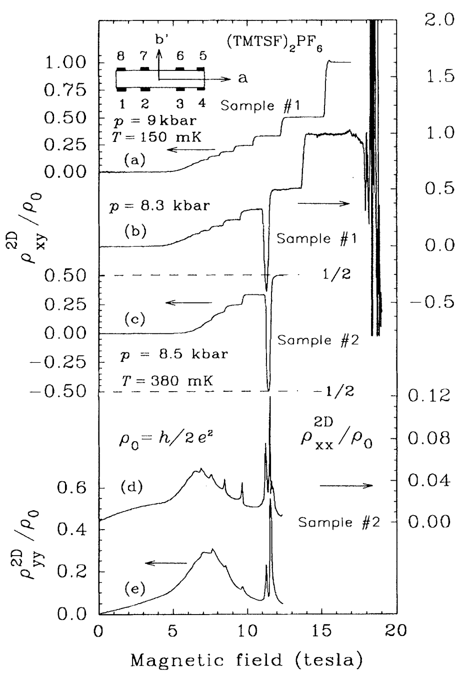

FIG. 1. Magnetic field dependence of the components of the resistance tensor in the conducting planes. The measured resistance is first multiplied by the number of conducting layers to obtain the one-layer resistance ρ_{ij}^{2D}, then this is normalized to the resistance quantum ρ_0. The inset shows the arrangement of the 8 electrode contacts similar on both samples.

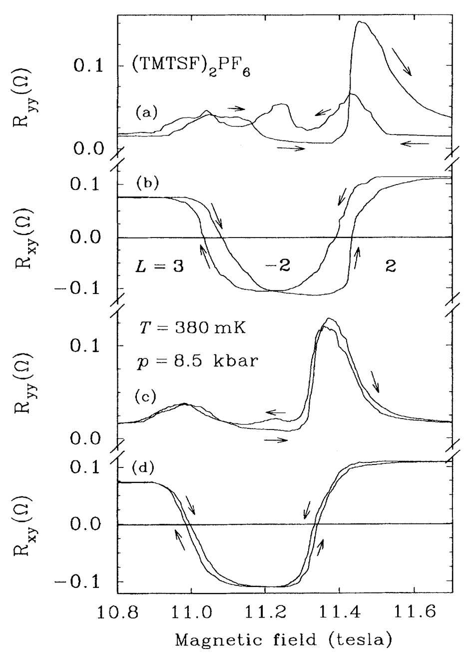

FIG. 2. Magnetic field dependence of the Hall resistance [(a) and (c)] and longitudinal resistance [(b) and (d)] in the neighborhood of the $L = -2$ phase. Arrows show the direction of field scans. Curves (a) and (b) have been obtained without depinning the FISDW. To obtain curves (c) and (d), the FISDW's have been depinned and repinned before every resistance measurement.

complex fine structure. We stress that the irregular line shapes are *not* due to instrumental noise; the noise can be judged from regions far from the transitions. It is also seen that the Hall resistance is not strictly constant in the negative phase and varies from one scan to another. To have further insight to the origin of the fine structure, we made use of some recent results on the FISDW transport.

If the current is low, the electrons condensed to the FISDW do not contribute to the electric transport because the FISDW is pinned to the defects of the crystal lattice. We have shown recently [12] that by applying a sufficiently high current the FISDW can be depinned from the lattice defects. The depinned FISDW carries charge along the conducting chains, and since this charge transport is dissipative the depinning is observed as the breakdown of the QHE: R_{ii} increases and $|R_{xy}|$ decreases.

As the FISDW wave vector changes with magnetic field [see Eq. (1)], the FISDW-defect interaction leads to metastable FISDW configurations because the rearrangement of the phase of the FISDW is impeded by the interaction with the defects. These metastable FISDW states may well be the origin of the observed hysteresis. Our method is based on the assumption that if the metastable FISDW is depinned from the lattice defects it will repin in the stable configuration. This method of "conditioning" has been successfully applied to eliminate the temperature hysteresis of the conductivity in charge-density-wave systems [13].

Figures 2(c) and 2(d) show the diagonal resistance and Hall resistance vs magnetic field in the conditioned state. These curves have been obtained the following way. After changing the magnetic field, first the FISDW's are depinned by applying a train of high-amplitude current pulses, then the resistances are measured at a low current. The amplitude of the conditioning pulses was 10 mA, i.e., 5 times higher than the threshold current for depinning obtained from an independent measurement. To avoid Joule heating, the duration of the pulses was as short as 1 μs. The pulses were repeated 20 times with a separation of 100 ms. The figure demonstrates that both the hysteresis and the fine structure are drastically reduced by the conditioning. Between the two transitions R_{xy} is constant, and its value agrees well with $L = -2$.

The reproducible results obtained in the conditioned state allow an accurate determination of the temperature–magnetic field phase diagram (Fig. 3). The transitions between FISDW phases are defined as the maxima in dR_{xy}/dB at constant temperature, while the transition line between the metallic and FISDW phases $[T_c(B)]$ is determined from the breakpoint in constant-field temperature scans as illustrated in the figure.

The $L = -2$ phase has a shape in the phase diagram similar to other FISDW phases, except that its width in $1/B$ space is about 1 order of magnitude smaller than that of other phases. An important lesson of this phase diagram is the existence of a finite magnetic field range over which a transition occurs from the high-temperature metallic phase

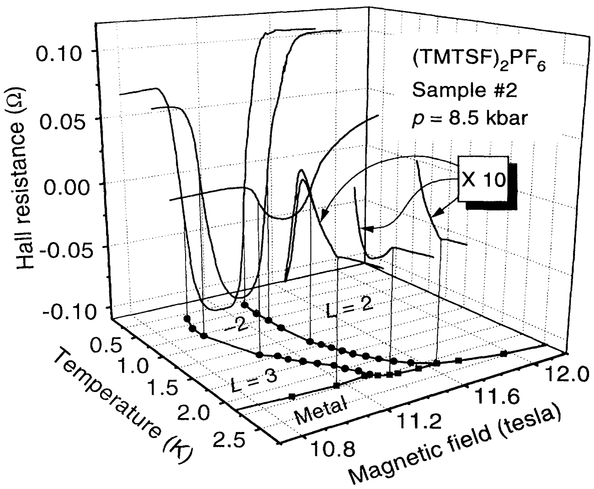

FIG. 3. Magnetic field dependence of the Hall resistance R_{xy} at constant temperatures and temperature dependence at constant fields. Resistance values of the temperature scans are multiplied by 10 for clarity. The temperature–magnetic field phase diagram, shown in the base plane of the figure, has been inferred from these and similar field scans (circles) and temperature scans (squares).

to the low-temperature $L = -2$ phase. This is clearly seen from the 11.4-T temperature scan where the Hall resistance decreases with decreasing temperature below the metal-FISDW transition. The 11.1-T scan demonstrates the temperature hysteresis of the $L = 3 \longrightarrow -2$ transition.

In the "standard model" of FISDW's [5,6], the metallic phase is unstable against the formation of an FISDW for any integer N in Eq. (1). To determine the actual sequence of phases, one needs to specify the electronic dispersion in the metallic phase. Because of the strong anisotropy, it is justified to linearize the dispersion along the chains, and to use a tight-binding formula perpendicular to the chains:

$$\varepsilon(\mathbf{k}) = \hbar v(|k_x| - k_F) - 2t_b \cos(k_y b) - 2t_b' \cos(2k_y b), \quad (2)$$

where v is the Fermi velocity and t_b and t_b' characterize the perpendicular coupling of the chains. The term $\propto t_b'$ is necessary to have an imperfect nesting. With this dispersion, the standard model yields a regular sequence of phases with decreasing magnetic field [6]: $|N| = 0, 1, 2, \ldots$ There is no sign reversal and $\mathrm{sgn}(N) = -\mathrm{sgn}(t_b')$. Our results at 9 kbar are well described by this model with $t_b' < 0$. For an even N, however, the energy difference between the $+N$ and $-N$ phases is small and decreasing with decreasing $|t_b'|$ [14]. If, in addition, the actual dispersion deviates from Eq. (2) to favor the $N = -2$ phase, this phase may become stable for sufficiently small $|t_b'|$, i.e., for sufficiently small pressure.

A further possibility is that the negative Hall number is the consequence of so-called multiple-order-parameter (MOP) states introduced independently by Lebed' [15] and Machida *et al.* [16]. The latter authors have shown by a numerical solution of a simplified model Hamiltonian that order parameters corresponding to all integers N are simultaneously present in the ground state, and several of these may be comparable in magnitude to the largest order parameter of index N_0. Yakovenko *et al.* [17] have shown that in a MOP state L is not certainly equal to N_0. Moreover, L may change while all the order parameters evolve continuously with magnetic field. In particular, L may become negative for a certain range of magnetic fields with N_0 constant and positive. Crucial in this respect is our observation of a transition line between the metallic and $L = -2$ FISDW phases. In the vicinity of the metal-FISDW transition, there exists a dominant order parameter of index N_0, and $L = N_0$. Therefore assuming MOP states does not bring us closer to the solution since we still have to explain why the sequence of the dominant order parameters is irregular.

Our observation of two close transitions $L = 3 \rightarrow -2$ and $-2 \rightarrow 2$ instead of the one transition $3 \rightarrow 2$ at high pressure is very likely the same phenomenon as the doubling of the transitions observed in magnetocaloric data in $(TMTSF)_2ClO_4$ [18,19]. In this latter case, the existence of a tetracritical point has been evidenced where the two split phases, the intruding phase, and the metallic phase meet. In our case the tetracritical point is missing. A further arborescence of the phase diagram with decreasing temperature has also been reported in the perchlorate compound [18]. The complicated structure of the magnetocaloric scans from which the new phase transitions have been inferred resembles the complex fine structure of our resistance scans without FISDW depinning [Figs. 2(a) and 2(b)]. The fact that most of this fine structure disappears upon conditioning suggests that the original structure is not a signature of new phases, but indicates the existence of metastable domains in the sample. Metastability arises from the existence of multiple and very close free energy minima of the FISDW phases, separated by energy barriers due to the SDW pinning by defects.

In conclusion, we have shown the existence of an FISDW phase with negative quantum Hall number intruding between two positive-quantum-Hall-number phases. Based on an analysis of the temperature–magnetic field phase diagram, we suggest that the negative phase has a dominant order parameter of negative index.

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[1] D.J. Thouless *et al.,* Phys. Rev. Lett. **49**, 405 (1982).
[2] J.R. Cooper *et al.,* Phys. Rev. Lett. **63**, 1984 (1989); S.T. Hannahs *et al., ibid.* **63**, 1988 (1989).
[3] M. Ribault, Mol. Cryst. Liq. Cryst. **119**, 91 (1985).
[4] J.F. Kwak *et al.,* Phys. Rev. Lett. **46**, 972 (1981); M. Ribault *et al.,* J. Phys. (Paris), Lett. **44**, L953 (1983); P.M. Chaikin *et al.,* Phys. Rev. Lett. **51**, 2333 (1983).
[5] L.P. Gor'kov and A.G. Lebed', J. Phys. (Paris), Lett. **45**, L433 (1984); M. Héritier, G. Montambaux, and P. Lederer, *ibid.* **45**, L943 (1984); K. Yamaji, J. Phys. Soc. Jpn. **54**, 1034 (1985); P.M. Chaikin, Phys. Rev. B **31**, 4770 (1985); K. Maki, *ibid.* **33**, 4826 (1986).
[6] A. Virosztek, L. Chen, and K. Maki, Phys. Rev. B **34**, 3371 (1986).
[7] D. Poilblanc *et al.,* Phys. Rev. Lett. **58**, 270 (1987).
[8] W. Kang, S.T. Hannahs, and P.M. Chaikin, Phys. Rev. Lett. **70**, 3091 (1993).
[9] L. Brossard *et al.,* Physica (Amsterdam) **143B**, 406 (1986); W. Kang, Ph.D. thesis, Princeton University, 1992.
[10] W. Kang, J.R. Cooper, and D. Jérome, Phys. Rev. B **43**, 11 467 (1991).
[11] A.M. Dykhne and I.M. Ruzin, Phys. Rev. B **50**, 2369 (1994); Igor Ruzin and Shechao Feng, Phys. Rev. Lett. **74**, 154 (1995).
[12] L. Balicas, N. Biškup, and G. Kriza, J. Phys. IV (France) **3**, C2-319 (1993); L. Balicas and G. Kriza (unpublished).
[13] Gy. Hutiray, G. Mihály, and L. Mihály, Solid State Commun. **47**, 121 (1983).
[14] G. Montambaux, Ph.D. thesis, Université Paris-Sud, Orsay, 1985.
[15] A.G. Lebed', Pis'ma Zh. Eksp. Teor. Fiz. **51**, 583 (1990) [JETP Lett. **51**, 663 (1990)].
[16] K. Machida and M. Nakano, J. Phys. Soc. Jpn. **59**, 4223 (1990); K. Machida, Y. Hori, and M. Nakano, Phys. Rev. Lett. **70**, 61 (1993).
[17] V.M. Yakovenko, Phys. Rev. B **43**, 11 353 (1991); K. Machida *et al., ibid.* **50**, 921 (1994).
[18] G. Faini, F. Pesty, and P. Garoche, J. Phys. (Paris), Colloq. **48**, C8-807 (1988); F. Tsobang *et al.,* J. Appl. Phys. **73**, 5651 (1993).
[19] U. Scheven, W. Kang, and P.M. Chaikin, J. Phys. IV (France) **3**, C2-287 (1993).