Quantum Oscillation of Hall Resistance in the Extreme Quantum Limit of an Organic Conductor $(TMTSF)_2ClO_4$

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We report resistance and magnetic torque experiments under a high magnetic field up to 45 T in a three dimensional quantum Hall (QH) system $(TMTSF)_2ClO_4$, where TMTSF = tetramethyltetraselenafulvalene. The Hall resistance shows huge oscillations accompanied with sign reversal after the final QH state, where the Landau level filling factor is unity, is removed above 26 T. The magnetic torque also oscillates with the field. The results suggest that a novel quantum state, where the character of the carriers periodically changes with the field, is stabilized in the extreme quantum limit.

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Low dimensional electronic systems have been extensively studied because of their fascinating phenomena, especially under a high magnetic field. It is well known that a pure two dimensional electron gas goes into an integer quantum Hall (QH) phase and then to a fractional QH phase when the magnetic field is applied perpendicular to the plane [1]. At higher fields where the filling factor ν is much less than unity, it is believed that the state becomes insulating [2]. However, in a three dimensional (3D) QH state, the electronic state in the extreme quantum limit has not been clarified yet.

The $(TMTSF)_2X$ series of the organic conductors, where $X = ClO_4$, PF_6, etc., is known as the first 3D QH system [3]. The crystal structure of $(TMTSF)_2X$ has a triclinic unit cell and the stacked platelike TMTSF molecules yield a pair of open Fermi surface (FS) sheets, which are characterized by the highly anisotropic transfer integrals, $t_a/t_b/t_c \approx 0.2/0.02/0.001$ eV, respectively. The magnetic field along the c^* axis (perpendicular to the conducting ab plane) in $(TMTSF)_2PF_6$ leads to cascadelike field-induced spin-density-wave (FISDW) transitions, which are associated with the QH effect [4,5]. The overall features of the FISDW transitions are well understood by the so-called standard theory [6] which requires imperfect nesting of the two open sheets of the FS. Because of the imperfect nesting, a small closed orbit is formed and the energy levels become discrete by the Landau quantization under the field B. The electronic state is more stabilized by the field dependent nesting vector so that the Fermi level is always pinned between the adjacent Landau levels. This mechanism enables us to observe the QH effect in a wide field region. For $(TMTSF)_2ClO_4$ salts, however, the phase diagram has not been fully understood yet although intensive theoretical works have been done [7–14].

In $(TMTSF)_2ClO_4$, the ClO_4 anions order below 24 K, which causes a superlattice potential with a wave vector $(0, \pi/b, 0)$. The potential separates the original Fermi surface into two zones, forming four open sheets of the FS [Fig. 1(a)]. The final FISDW transition to $\nu = 1$ state occurs at about 8 T and survives up to about 26 T, where a subphase boundary is present [Fig. 1(b)] [7]. For convenience, we designate the lower (higher) FISDW phase as the SDW I (II) phase. The main purpose of this study is to investigate what happens in the extreme quantum limit where $\nu < 1$.

The platelike single crystals of $(TMTSF)_2ClO_4$ were synthesized electrochemically. The resistance and magnetic torque experiments were performed in dc magnetic fields up to 45 T at National High Magnetic Field Laboratory (NHMFL). The resistance was measured by a low frequency ac technique. The Hall resistance in the conducting ab plane was obtained by reversing the magnetic field and averaging the signals asymmetrically. The magnetic torque was measured by a microcantilever technique [15]. The sample temperature was precisely controlled in ^{3}He and ^{4}He cryostats by the vapor pressure and a calibrated Cernox sensor. We slowly cooled the samples from 30 to 18 K at about 10 mK/min to obtain a well ordered state. Both the resistances R_{xx} and R_{xy} were simultaneously measured for the same sample, but R_{zz} and the torque were measured for different ones.

The a axis resistance R_{xx} for fields perpendicular to the conducting ab plane ($B \| c^*$) is shown in Fig. 2. At 5 K, R_{xx} shows an increase due to the FISDW transition at about 13 T and then oscillations, commonly called rapid oscillations (RO) appear at higher fields. As temperature decreases, the FISDW transition takes place at lower fields, and the RO become more evident. Above 26 T, the electronic state enters the SDW II phase from the SDW I phase, where a kink or minimum is observable. For comparison, the c axis (interplane) R_{zz} is also shown in Fig. 2. The value of R_{zz} is much bigger than R_{xx}, showing the strong anisotropy of the transfer integrals. We also see a similar subphase transition at 26 T and RO in R_{zz}. These results are

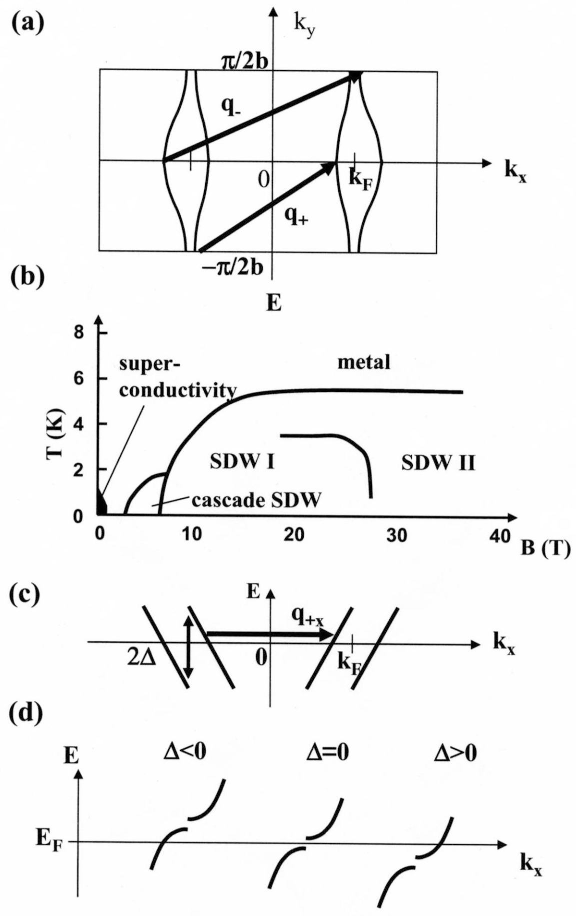

FIG. 1. (a) Schematic picture of the Fermi surface in $(TMTSF)_2ClO_4$. Two pairs of 1D Fermi surfaces appear after the anion order with the wave number $(0, \pi/b, 0)$. Two possible intrasubband nesting vectors, denoted as $\mathbf{q}_+$ and $\mathbf{q}_-$ are shown. (b) Temperature-magnetic field phase diagram for $B \| c^*$. In the final SDW phase, a subphase transition is present around 26 T in the low temperature region. (c) Two pairs of the magnetic subbands around the Fermi level. (d) Provided that only a pair of the subbands is fully nested with $\mathbf{q}_+$ ($\nu = 0$) in the SDW II phase, the other subbands are partially gapped. The partially gapped subbands are shown, whose energy levels periodically change with field.

consistent with the pulsed field experiments up to 50 T [16]. The RO, which were first observed by Ulmet *et al.* [17], are periodic in the inverse field with the frequency $F = 260$ T for $B \| c^*$, reminiscent of conventional quantum oscillations, i.e., the Shubnikov–de Haas (SdH) effect. However, unlike the SdH effect, the temperature dependence of the RO in the SDW I phase is anomalous. In the SDW I phase, the RO amplitude has a maximum around 2 to 3 K and then steeply decreases with decreasing temperature [18]. However, in the SDW II phase, the amplitude monotonically increases with decreasing temperature, accompanied with a steep increase in the background resistance.

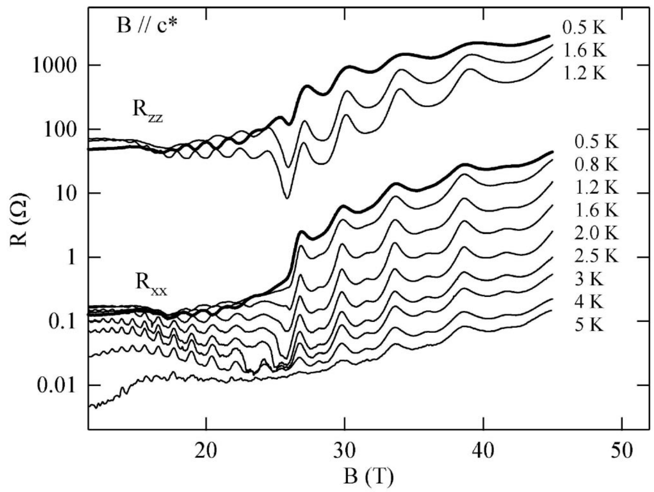

FIG. 2. Resistance along the a axis (R_{xx}) and the c axis (R_{zz}) for $B \| c^*$. The base temperature data are shown by thick lines.

The Hall resistance R_{xy} for $B \| c^*$ is shown in Fig. 3. At 0.8 K, the quantized Hall resistance is observable in the SDW I phase [19]. The absolute value of R_{xy} per layer is about 6 kΩ, which is a factor of 2 less than the ideal value ($\nu = 1$), $h/2e^2$, = 12.9 kΩ, which may be due to nonideal current paths as reported previously [19]. The factor 1/2 in $h/2e^2$ comes from the spin degeneracy due to the SDW formation. The RO in the SDW I phase are most evident around 2.5 K, but not visible at 0.8 K as shown in the inset.

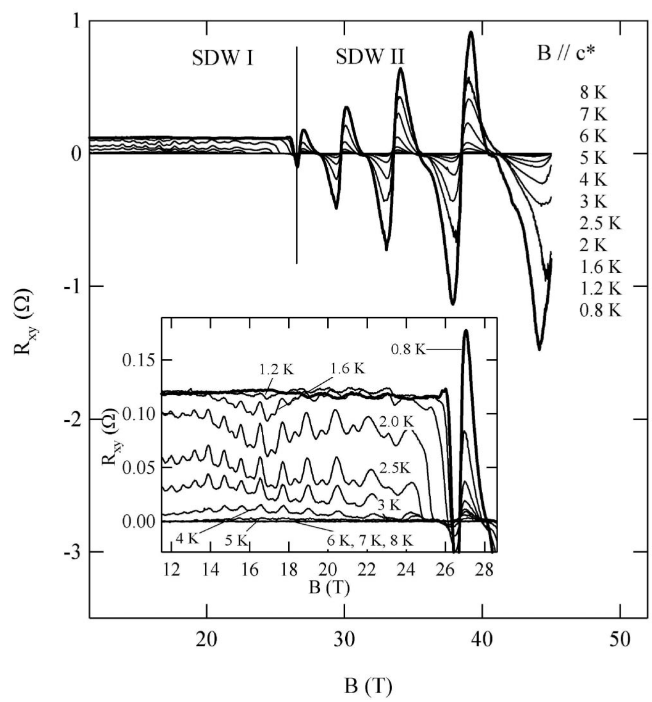

FIG. 3. Hall resistance R_{xy} for $B \| c^*$ at various temperatures. Inset: data below 28 T.

The striking feature is that the QH effect is removed around 26 T and then R_{xy} starts oscillating in the SDW II phase. This observation is the main experimental result of this Letter. The subphase transition at 26 T is slightly hysteretic, suggesting a first order phase transition [7]. The RO of R_{xy} in the SDW II phase are accompanied with a sign reversal and the amplitude increases with decreasing temperature or increasing field. Although high field experiments have been done [19,20], so far no clear evidence of such an oscillation in R_{xy} has been obtained. According to our knowledge, the Hall resistance oscillation accompanied with a sign reversal has not been observed in any other materials. The RO of R_{xy} in the SDW II phase have a sawtooth shape in contrast to R_{xx} and R_{zz}. Both the frequency and phase are completely the same in the SDW I and II phases.

The oscillatory part of the magnetic torque as a function of the field component along the c^* axis, $B_{\parallel c^*}$, is shown in Fig. 4. To increase the torque signal, the sample was tilted by $\theta = 26°$, as shown in the inset. Angular dependent measurements show that the transition field and the frequency of the RO completely scale with $B_{\parallel c^*}$. The RO amplitude is enhanced in the SDW II phase, as is the resistance. Since the magnetization is given by $-\partial F/\partial H$, the torque result shows that the free energy F of the electronic state oscillates with the field. The RO signal in the torque is not observed in the metallic state above 5.5 K [21]. A lot of theoretical works on the origin of the RO have been done, but the mechanism is still controversial [8,9,11,14,22].

Since the SDW I phase is in the quantum limit ($\nu = 1$), one may expect that the system enters an insulating phase at higher fields. Actually, both R_{xx} and R_{zz} steeply increase with the field in the SDW II phase. However, the temperature dependence below 2 K is much weaker than thermally activated behavior $R \sim \exp(\delta/T)$, where δ is the energy gap. Moreover, it is very likely that the RO originate from some quantization of the electronic state, suggesting that a fraction of the density of states remains at the Fermi level. In addition, the values of R_{xy} remain finite in the SDW II phase. Therefore, the SDW II phase is probably not insulating in the whole field region. In the classical picture, the sign of the Hall resistance is determined by the nature of the major carriers, i.e., positive for holes and negative for electrons. In this sense, the oscillation of R_{xy} suggests that the major carries periodically alternate between holes and electrons with increasing field.

Figure 5 shows the oscillatory parts of R_{zz}^{osc}, R_{xx}^{osc}, and R_{xy} as a function of $B_{\parallel c^*}$, which allows us to compare the phases of the oscillations with each other. We note that R_{zz}^{osc} and R_{xx}^{osc} show maxima at fields where R_{xy} turns from minimum (negative) to maximum (positive), crossing zero. The behavior suggests that the Fermi level lies in the energy gap (no carriers) when the major carriers turn from electrons to holes.

In $(TMTSF)_2ClO_4$, four open FS sheets are formed by the anion superlattice potential, which is written as $U = V\cos(\pi y/b)$, where b is the lattice constant. In this case, there are two possible intrasubband nesting vectors, denoted as $\mathbf{q}_+$ and $\mathbf{q}_-$ [Fig. 1(a)]. Assuming small anion potential as compared with t_b, Osada *et al.* first discussed the SDW stability due to such intrasubband nesting [8]. They argue that the SDW phases with even quantum numbers ν are periodically suppressed with $1/B$, causing the collapse of the QH effect, whereas the SDW phases with odd ν are not affected by the anion order gap; i.e., the Hall resistance is well quantized. This model is consistent with the observation of the QH effect only with odd ν but explains neither the phase boundary between the SDW I and II phases nor the oscillation of R_{xy}.

Reexamining various experimental data, Mckernan *et al.* first pointed out the possibility that the transitions of the intrasubband nesting take place separately [7]. They argue that only a pair of the intrasubbands is nested in the SDW II phase, whereas both pairs are nested in the SDW I phase. Some other models have been proposed, which treat intra-

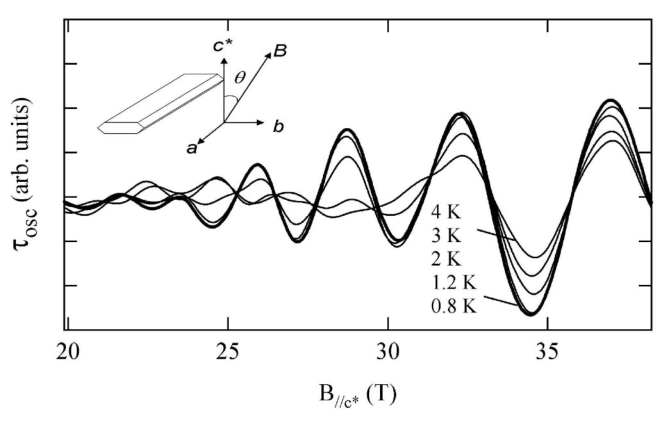

FIG. 4. Oscillatory part of the magnetic torque τ as a function of the field component along the c^* axis, $B_{\parallel c^*}$. The sample is tilted by $\theta = 26°$ as shown in the inset.

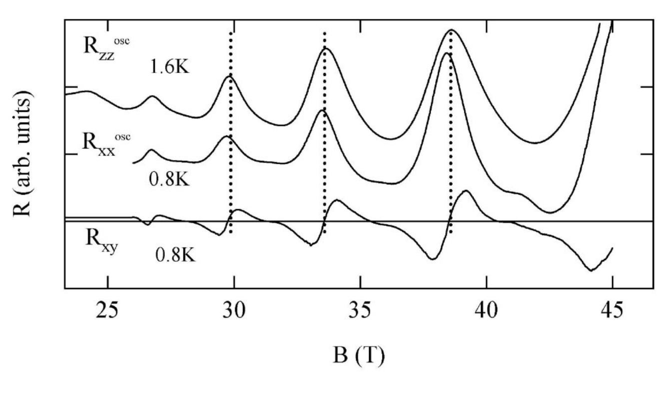

FIG. 5. Oscillatory parts of R_{zz}^{osc}, R_{xx}^{osc}, and R_{xy} as a function of $B_{\parallel c^*}$. The field for R_{zz} is slightly corrected because the sample is tilted by $\sim 2°$.

and intersubband nesting [7,9,11,14] or magnetic breakdown effects [16,22]. When the anion potential V can be treated as a perturbation, the anion gap in the energy spectra oscillates with field [8] as $\Delta = VJ_0(4t_b/\hbar V_F G)$, where J_0 is the zeroth order Bessel function and $G = beB/\hbar$. The x components of the nesting vectors q_{+x} and q_{-x} are given by $q_{+x} = 2k_F + 2\nu G - 2\Delta/_{\hbar} v_F$ and $q_{-x} = 2k_F + 2\nu G + 2\Delta/_{\hbar} v_F$ for even ν and $q_{+x} = q_{-x} = 2k_F + 2\nu G$ for odd ν [8]. Since the SDW I phase is characterized by the quantum number $\nu = 1$, both pairs of the intrasubbands are nested by the same nesting vector $\mathbf{q}_+ (= \mathbf{q}_-)$, which is the reason why the QH effect is observed in the SDW I phase. Therefore, it may be likely that the quantum number ν is zero in the SDW II phase, i.e., at least one of the two pairs is fully nested.

If only a pair of the subbands is fully nested with $\mathbf{q}_+$ ($\nu = 0$) [7] as shown in Fig. 1(c), the other subbands are reconstructed by $\mathbf{q}_+$ and partially nested (gapped). The schematic picture of the subbands is illustrated in Fig. 1(d). The energy levels of the subbands are different from those of the fully nested subbands by the anion gap Δ. Since the gap Δ periodically oscillates with $1/B$, the energy levels of the subbands change with the field. For $\Delta < 0$ ($\Delta > 0$), the gap of the subbands lies above (below) E_F; thus the carriers become holes (electrons). For $\Delta = 0$, both the subbands are fully gapped, so the system becomes insulating (no carriers). Such a periodic change of the carrier number and character should cause large oscillations in R_{xy} accompanied with a sign reversal, and in R_{xx} and R_{zz} as well. However, this model doubles the frequency of R_{xx} and R_{zz}, which is not consistent with the experimental results.

On the other hand, it is pointed out that the anion potential V cannot be treated as a perturbation [11], because V is comparable to t_b [23]. Hasegawa *et al.* treated V nonperturbatively and discussed the SDW stability near the quantum limit [11]. According to their theory, the SDW phases with the same nesting vectors q_{+x} and q_{-x} as those obtained in the perturbation theory are stabilized, and the anion gap Δ also shows an oscillation with the same frequency as $J_0(4t_b/\hbar V_F G)$. It is also argued that the eigenstate of the $\nu = 0$ phase includes large amplitudes of the $\nu = \pm 1$ states. The amplitudes of the $\nu = \pm 1$ states are different and oscillate with field, whose frequency is equal to $J_0(4t_b/\hbar V_F G)$. Therefore, one may expect that large oscillations of R_{xy} accompanied with a sign reversal as well as those in R_{xx} and R_{zz}. As the field increases, the amplitudes of the $\nu = \pm 1$ states decrease, suggesting that the oscillation amplitude of R_{xy} increases with increasing fields. It is also consistent with the experimental results. However, the theory does not explain the phase boundary between the SDW I and II phases, either. The phase boundaries between different ν numbers are not well defined for such a large anion potential. Further theoretical investigation will be needed for a full understanding of the SDW II phase.

In summary, we observed large RO in R_{xy} in the SDW II phase, which are accompanied with the sign reversal. The RO amplitude in R_{xy} monotonically increases with increasing field or decreasing temperature as well as in R_{xx}, R_{zz}, and the magnetic torque. The RO in such quantities clearly show that the free energy of the electronic state as well as the character of the carriers periodically changes with field. The results suggest that a novel quantum state is stabilized in the extreme quantum limit for $(TMTSF)_2ClO_4$.

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