Density-of-State Oscillation of Quasiparticle Excitation in the Spin Density Wave Phase of $(TMTSF)_2ClO_4$

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Systematic measurements of the magnetocaloric effect, heat capacity, and magnetic torque under a high magnetic field up to 35 T are performed in the spin density wave (SDW) phase of a quasi-one-dimensional organic conductor $(TMTSF)_2ClO_4$. In the SDW phase above 26 T, where the quantum Hall effect is broken, rapid oscillations (ROs) in these thermodynamic quantities are observed, which provides clear evidence of the density-of-state (DOS) oscillation near the Fermi level. The resistance is semiconducting and the heat capacity divided by temperature is extrapolated to zero at 0 K in the SDW phase, showing that all the energy bands are gapped, and there is no DOS at the Fermi level. The results show that the ROs are ascribed to the DOS oscillation of the quasiparticle excitation.

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Electronic states with closed Fermi surfaces (FSs) undergo Landau quantization under magnetic fields. As the magnetic field increases, the equally spaced Landau levels periodically cross the Fermi level, and consequently, the free energy of the electronic state oscillates with the magnetic field. Quasi-one-dimensional (Q1D) organic conductors, $(TMTSF)_2X$ series, where TMTSF = tetramethyltetraselenafulvalene and $X = ClO_4$, PF_6, etc., [1] are known to show so-called rapid oscillations (ROs) in the spin-density-wave (SDW) phases. The ROs are quite similar to the conventional quantum oscillations due to the Landau quantization. However, the original FSs have no closed orbits, and the characteristic temperature and field dependence shows that the ROs are unconventional quantum oscillations.

For $(TMTSF)_2ClO_4$, the ClO_4 anion order below 24 K makes a superlattice potential, which separates the original FS into two zones, and then two pairs of the Q1D FS are formed [Fig. 1(a)]. In a magnetic field along the c^* axis (perpendicular to the conducting ab plane), cascadelike field-induced spin density wave (FISDW) transitions appear [1], which are associated with the integer quantum Hall (QH) effect [2]. The integer QH state with the quantum number $N = 1$ is stabilized in a wide field region above 8 T (SDW I), and then the first order phase transition to the final SDW phase (SDW II) takes place at 26 T (subphase boundary) [3]. The overall features of the FISDW transitions up to the SDW I phase have been reasonably understood by the so-called standard theory [4] improved by Osada [5], where the nesting instability of the two pairs of the Q1D FS plays a crucial role.

In the SDW II phase, the QH effect is completely broken, and the Hall resistance (R_{xy}) shows huge oscillations accompanied by the sign reversal [6,7]. The R_{xy} oscillations above 26 T (SDW II) are reproduced in this work [Fig. 1(c)]. Such oscillations are quite anomalous, and can never be understood in terms of conventional Landau quantization. The R_{xy} oscillation increases with increasing field up to 45 T or with decreasing temperature down to 0.8 K [7]. The in-plane resistance R_{xx} ($I//a$ axis) and the interplane resistance R_{zz} ($I//c^*$ axis) also show the large ROs in the SDW II phase [7,8]. The detailed phase

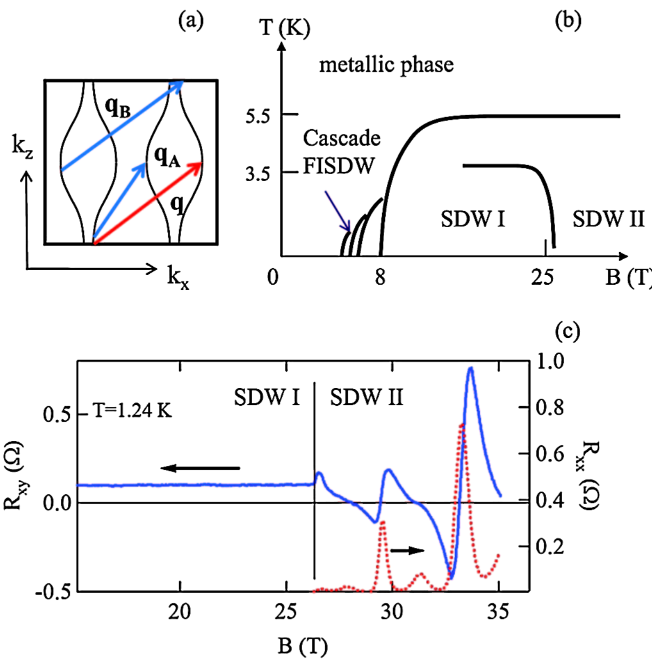

FIG. 1 (color online). (a) Schematic picture of two pairs of the Q1D FS in $(TMTSF)_2ClO_4$. Interband nesting vector $\mathbf{q}$, and intraband nesting vectors $\mathbf{q_A}$ and $\mathbf{q_B}$ are shown. (b) Temperature-magnetic field phase diagram for $B//c^*$. (c) Hall resistance R_{xy} and in-plane resistance R_{xx} as a function of field for $B//c^*$ at 1.24 K.

diagram, especially the subphase boundary between SDW I and II, and the mechanism of the ROs observed in both phases are still long-standing open questions although intensive studies have been done so far [9–15]. To obtain further insight of the RO mechanism, we have performed systematic measurements of thermodynamic properties, magnetocaloric effect, heat capacity, and magnetic torque measurements in fields up to 35 T.

The platelike single crystals of $(TMTSF)_2ClO_4$ were synthesized electrochemically. The heat capacity is measured by an ac technique employing a miniature vacuum cell [16], in which a single crystal of 0.65 mg with a thermometer and heater is weakly thermally isolated from the cell (heat bath) by thin Pt(W) wires. The magnetocaloric effect is measured by the same vacuum cell. In this measurement, the temperature of the cryostat is controlled to fixed temperatures and the field is swept up and back down at a constant rate of 50 mT/s. The sample is slightly cooled down (heated up) when the entropy of the electronic state increases (decreases). This sample temperature variation ΔT is measured as a function of field. The magnetic torque was measured by a microcantilever technique [17]. For comparison, R_{xx}, R_{zz} and R_{xy} were also measured. The sample temperature was precisely controlled in ^{3}He and ^{4}He cryostats by the vapor pressure. All the samples used for these measurements were mounted in the same probe and were slowly cooled from 30 K to 18 K at about 10 mK/ min simultaneously, to obtain the well ordered state.

Figure 2(a) shows magnetic field dependence of the magnetocaloric effect ΔT at various bath temperatures T_0. The magnetocaloric effect ΔT is directly related to the field dependence of the magnetic entropy S,

$$\Delta T = -\tau \frac{d(\Delta T)}{dt} - \frac{dB}{dt}\frac{T}{K}\left(\frac{\partial S}{\partial B}\right)_{T_0}, \quad (1)$$

where $\tau = C/K$ is the thermal relaxation time between the sample and bath, and C is the heat capacity of the sample and addenda, and K is the thermal conductivity. The ΔT curves should be symmetric around $T = T_0$ between up- and down-sweeps; the sign of ΔT is opposite because of the dB/dt term. Some hysteresis may be observed at a first order phase transition. The magnetocaloric effect of $(TMTSF)_2ClO_4$ was first measured down to 1.65 K and up to 30 T [18]. Our data well agree with theirs in this region. At low temperatures, large positive peaks of ΔT are observable below 8 T, which arise from the cascade FISDW transitions. In the whole field region up to 35 T, we observe ROs, which are periodic with the inverse field [Fig. 2(b)].

It is newly found that the ΔT curves are almost symmetric between the up- and down-sweeps above 1.6 K, but asymmetric below it. At 0.65 K, only positive ΔT peaks are observable above 7 T. Similar asymmetric behavior is reported in a heavy Fermion compound [19]. A possible reason is friction heating between the sample and holder, or

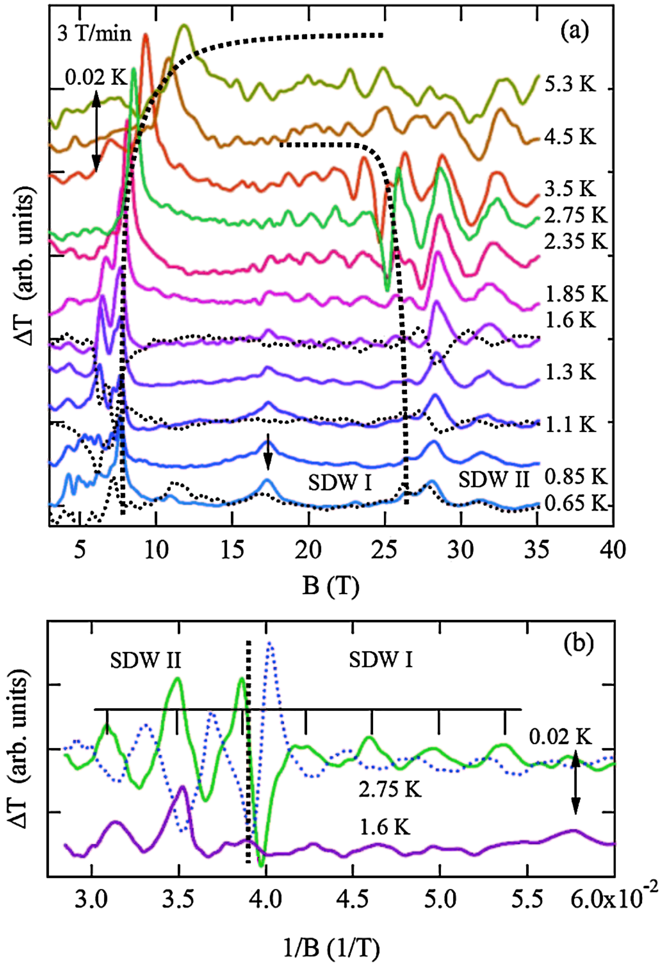

FIG. 2 (color online). (a) Magnetocaloric effect vs B plot for $B//c^*$. (b) Magnetocaloric effect vs $1/B$ plot for $B//c^*$. All the data are taken at a rate of 3 T/ min. The solid and dotted curves show the up- and down-sweep data, respectively. The thick dotted curves show the SDW phase boundaries. The anomaly at 17 T is indicated by the arrow.

between the main and second phases at transitions associated with volume changes. It is reasonable that such effect becomes dominant at low temperatures because of the low heat capacity of the sample and addenda. If the ROs are accompanied by the oscillation of the volume, the ROs will be obscured at low temperatures by the friction heating.

A characteristic feature is that the entropy uptake and loss with increasing field near 26 T is strongly enhanced for $T \sim 2.75$ K [Fig. 2(b)] although there is no such feature at 8 T. It is theoretically discussed that the entropy accumulates at quantum critical point (B_c), $(\partial S/\partial B)_{B_c} = 0$ which causes a sign change of the magnetocaloric effect with field [20]. Our data also suggest that the entropy has a maximum at about 26 T for $T \sim 2.75$ K although no evidence of the quantum criticality is obtained. At lower temperatures, we see no strong evidence of the subphase transition at 26 T. This fact may suggest that the subphase transition takes place in synchronization with the RO or the entropy change at the subphase transition is very small. Another new feature is a small peak (entropy loss) at 17 T [arrow in Fig. 2(a)], suggesting a phase transition. Anomalies at 17 T are reported in some other quantities [21,22]. However, SDW I is recognized as a uniform phase because the well quantized QH effect is observed [Fig. 1(c)]. At present, the origin of the anomalies at 17 T is an open question.

Figure 3 shows the heat capacity C/T at constant temperature as a function of field. Smoothly varying addenda, which are independently measured up to 35 T, are subtracted from the data. Since the lattice heat capacity is field independent, the observed field dependence is ascribed to the electronic origin, the density-of-state (DOS) change. The high field heat capacity measurements were first reported by Fortune *et al.*, down to 0.78 K and up to 30 T [23]. The overall features are consistent with theirs: (1) large peaks at 8 T and 28 T, (2) smaller heat capacity in the SDW I phase than that in the metallic phase, and (3) the presence of RO at high fields. The C/T curves show no strong anomaly at 26 T. The RO is periodic with inverse field in the whole field region up to 35 T without phase shift at 26 T (subphase boundary) [Fig. 3(b)]. A small peak is newly found at 17 T below 1.2 K. These features are consistent with the magnetocaloric data. The RO up to 35 T provides a clear evidence of the DOS oscillation near the Fermi level in the SDW II phase.

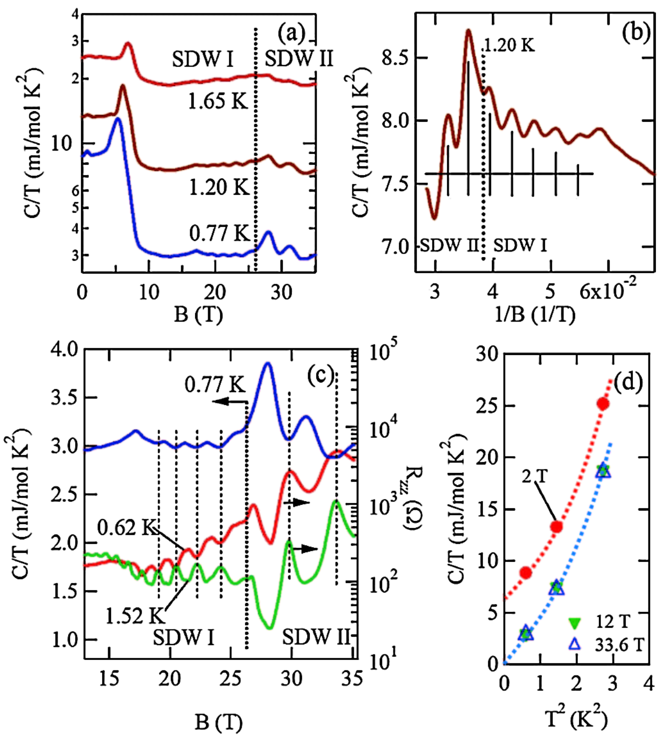

FIG. 3 (color online). (a) Magnetic field dependence of heat capacity divided by temperature C/T for $B//c^*$. (b) C/T vs $1/B$ plot at 1.20 K. (c) C/T and the interlayer resistance R_{zz} vs B plot. The thick dotted lines in (a), (b), and (c) denote the subphase boundary at 26 T. (d) C/T vs T^2 plot at 2 T (normal metal), 12 T (SDW I), and 33.6 T (SDW II). The dotted lines are guides for the eye.

In the SDW II phase, we clearly find that the minima of C/T correspond to the maxima of R_{zz} [Fig. 3(c)]. Since the tunneling rate between the layers ($\propto 1/R_{zz}$) is proportional to the square of the DOS near the Fermi level, the out-of-phase relation between the ROs of C/T and R_{zz} is reasonable. However, the RO of R_{zz} in SDW I changes the phase by π at about 1 K, the in-phase (out-of-phase) relation between C/T and R_{zz} for $T < 1$ K ($T > 1$ K). The RO of R_{xx} also shows the same phase reversal as R_{zz}. These results suggest that the dominant mechanism of the carrier scattering changes at about 1 K in SDW I. This is not a phase transition but a crossover because of no anomaly in the thermodynamic quantities.

Figure 3(d) shows the plot of C/T vs temperature. At 2 T (metallic state), the C/T curve is extrapolated to $\sim 7\ \mathrm{mJ/mol \cdot K^2}$ at 0 K, comparable to the previously reported values [24]. On the other hand, it is newly found that the C/T curves at 12 T (SDW I) and 33.6 T (SDW II) are extrapolated to zero at 0 K, showing that all the energy bands are gapped, and there is no DOS at the Fermi level. The results are consistent with the semiconducting behavior of the resistance in both SDW phases. In the SDW I phase (QH regime), the edge state has a finite DOS at the Fermi level [9], but the C/T value of the edge state is smaller than the bulk one by 3 orders of magnitude in this temperature range, which is within the noise level.

We also performed the magnetic torque measurements for $B//c^*$ (Fig. 4). The RO is observed in the SDW phases, but not in the metallic state above 5.5 K [25]. No anomaly is seen at 17 T. Here, we check the thermodynamic relation of the ROs. The Maxwell relation between the magnetic entropy S and magnetization M is given by $(\partial M/\partial T)_B = (\partial S/\partial B)_T \propto -\Delta T$. We obtain that $M = \zeta/B$ from $\zeta = M \times B$, where ζ is the magnetic torque. The relation requires that $\partial M/\partial T = 0$ when $\Delta T = 0$, which is seen in Fig. 4(a). In addition, the ROs in $\partial M/\partial T \sim \delta M(T) = M(6K) - M(T)$ and $-\Delta T \propto \partial S/\partial B$ show the in-phase relation and larger amplitude in SDW II than in SDW I as shown in Fig. 4(b). The results consistently show that the ROs in these quantities have the same origin.

In the SDW I phase, the QH state ($N = 1$) is interpreted by the interband nesting of the two pairs of the FSs by the same nesting vector $\mathbf{q}$ [5]. In the SDW II phase, on the other hand, many theoretical models based on the intraband nesting (the inner or outer pair nesting) have been proposed [9–12,15,26]. When the inner pair is nested by $\mathbf{q}_A$ [Fig. 1(a)], the outer pair could be also nested by $\mathbf{q}_A - \mathbf{G}$, where $\mathbf{G} = (2\pi/a, 0, 0)$, causing an umklapp gap [26]. This vector is almost identical to $\mathbf{q}_B$. The small difference between $\mathbf{q}_A - \mathbf{G}$ and $\mathbf{q}_B$ arises from the higher order corrugation of the FSs.

The semiconducting behavior of the resistance and the temperature dependence of the heat capacity in the SDW

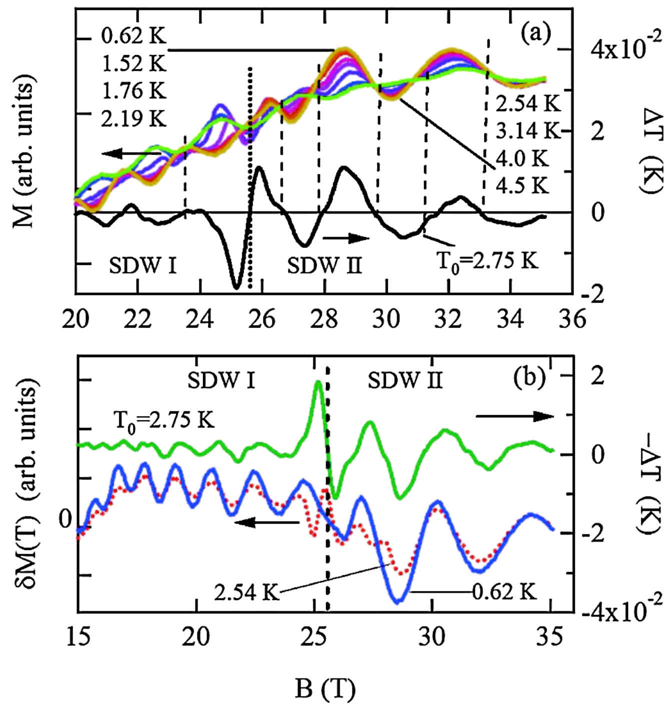

FIG. 4 (color online). (a) Comparison between the magnetization M given by ζ/B and the magnetocaloric data δT for $T_0 = 2.75$ K. (b) Comparison between the temperature variation of the magnetization $\delta M = M(6.0\text{ K}) - M(T)$ and the magnetocaloric data ΔT.

phases show that all the energy bands are gapped. On the other hand, the ROs in the resistance and thermodynamic quantities consistently show the DOS oscillation near the Fermi level. Therefore, we can conclude that the ROs are ascribed to the DOS oscillation of the quasiparticle excitation. The DOS oscillation may be associated with the energy gap oscillation, as has been discussed in much of the literature [5,10,12–14,26]. The SDW transition takes place at 5.5 K in high fields [Fig. 1(b)], so the energy gap caused by the main FS nesting is an order of several kelvin. The umklapp gap, if it exists, will be much smaller than that. Actually, we obtain a small energy gap of about 0.5 K from the Arrhenius plots of both R_{xx} and R_{zz} below 1 K, suggesting that the RO amplitude disappears at temperatures lower than the gap.

In the SDW II phase, the inner pair of the FSs may be fully nested in the SDW II phase ($N = 0$) by $\mathbf{q}_A$ and then the Fermi level is pinned in the energy gap [3,7,26]. Because of the periodic magnetic potential, the outer pair also has the periodic subband gaps with the period of $beBv_F$, where b is the lattice constant and v_F is the Fermi velocity. The outer pair of the energy bands shifts up and the subband gaps periodically cross the Fermi level with increasing field, whose frequency corresponds to the RO [7,26]. It should be noted that the outer pair could be always slightly gapped by the umklapp process at the Fermi level. This picture may explain the semiconducting behavior and the RO oscillation of R_{xy} accompanied by the sign change, but it is not clear whether this nesting state is the ground state. At present, no theory satisfactorily explains the phase diagram and the features of the ROs. Further theoretical studies are required.

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