Field-induced spin-density wave transition of the quasi-one-dimensional organic conductors

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Abstract. Magnetoresistance and Hall resistance measurements have been carried out along the highly conducting \(a\) axis in the field-induced spin-density wave (FISDW) phase of deuterated (TMTSF)\(_2\)ClO\(_4\) for various cooling rates through the anion ordering temperature. With increasing the cooling rate, (i) the high-field phase boundary \(\beta_{HI}\), observed at 27 T in hydrogenated samples for slowly cooled, is shifted towards a lower field, (ii) the last semimetallic SDW phase below \(\beta_{HI}\) is suppressed, and (iii) the FISDW insulating phase above \(\beta_{HI}\) is enhanced. The Hall resistance between 9 and 15 T, observed a very stable quantum Hall state above 9.0 T for slowly cooled, shows a drastically decrease with increasing the cooling rate. This result suggests that there is a new phase transition around 17 T in deuterated (TMTSF)\(_2\)ClO\(_4\). A possible ground state of the FISDW phase of (TMTSF)\(_2\)ClO\(_4\) for various cooling rates is discussed from the viewpoint of the peculiar SDW nesting vector stabilized by the dimerized gap due to anion ordering.

In the quasi one-dimensional (Q1D) organic compound deuterated (TMTSF)\(_2\)ClO\(_4\) (abbreviated as \(d\)-ClO\(_4\) hereafter), the ordering of the noncentrosymmetric anion ClO\(_4^−\) occurs at an anion ordering temperature \(T_{AO} \sim 27\) K [1]. For the ClO\(_4\) salt slowly cooled (relaxed) through \(T_{AO}\), the anion ordering creates a superlattice potential with a wave vector \(Q=(0,1/2,0)\) [2] and the superconducting phase appears below 1.2 K. When the sample is rapidly cooled (quenched) through \(T_{AO}\), the orientations of anions are frozen and the spin-density-wave (SDW) phase is induced below 6 K [2]. When the magnetic field is applied along the \(c^∗\) axis for relaxed ClO\(_4\), a cascade of field-induced SDW (FISDW) states is observed. The phase diagram of the FISDW phase in the PF\(_6\) salt, with quantized Hall resistance \(\rho_{xy} \sim h/(n2e^2)\) in the sequence \(n=\ldots,4,3,2,1,0\) as the magnetic field is increased, is successfully explained by the mean-field theory named the “standard model” based on the nesting of a pair of slightly warped parallel sheets of the Q1D Fermi surface. The states labeled with integer \(n\) have been identified as semimetallic FISDW states while that with \(n=0\) is a FISDW insulating state. However, the phase diagram of the FISDW phase in hydrogenated (TMTSF)\(_2\)ClO\(_4\) (abbreviated as \(h\)-ClO\(_4\) hereafter) for the case of slow cooling is known to show disagreements with the standard model i.e., (i) in the low-field cascade of FISDW transitions, the sequence of Hall plateaus is not in the expected order. (ii) a very stable quantum Hall state is observed from 7.5 to 27 T [2]. The deuteration of the TMTSF salt is thought to work as a positive chemical pressure in the crystal [1]. When the positive chemical pressure by deuteration is applied to the ClO\(_4\) salt, the nesting of the Q1D Fermi surface becomes more imperfect and the stabilization of the SDW phase in the intermediate cooled states is suppressed. As a result, the \(d\)-ClO\(_4\) salt is expected to show the FISDW phase in a broad range of cooling rates in contrast with the case of hydrogenated ones.

In this extended abstract, we describe the cooling rate dependence of the magnetoresistance [1] and the Hall resistance [3] in \(d\)-ClO\(_4\); we discuss a possible ground state of the FISDW phase of (TMTSF)\(_2\)ClO\(_4\) for various cooling rates from the viewpoint of the peculiar SDW nesting vector stabilized by the dimerized gap due to anion ordering.

Single crystals of (TMTSF)\(_2\)ClO\(_4\) were synthesized by the standard electrochemical method. The magnetoresistance and Hall resistance measurements were carried out using a standard six-probe dc method. The measurements in the fields to 28 T were done in a resistive magnet at the Grenoble High Magnetic Field Laboratory.
Figure 3 shows the Hall resistance in different FISDW states. These results mean that the last semimetallic FISDW phase and the FISDW insulating phase correspond towards a lower field, (ii) the last semimetallic FISDW phase between 9.7 T and the conclusions that, with increasing the cooling rate for from 5.5 to 4 K when the cooling rate \( T \) with increasing temperature above 2.5 K. In Figure 1 shows the magnetoresistance along the highly conducting direction for 0.0009, 0.018, and 0.67 K/s cooling rates, respectively.

Figure 2 shows the FISDW phase diagram in deuterated (TMTSF)$_2$ClO$_4$ constructed from many temperature and field sweeps for various cooling rates. The bold, dashed-dotted, and dashed lines are guides to the eye for 0.0009, 0.018, and 0.67 K/s cooling rates, respectively.

Figure 1 shows the magnetoresistance along the highly conducting \( a \) axis in hydrogenated (TMTSF)$_2$ClO$_4$ (the dashed line) for the slowly cooled state and deuterated (TMTSF)$_2$ClO$_4$ for two cooling rates with the magnetic field parallel to the lowest conductivity direction \( c^* \) at 1.5 K.

Figure 2 shows the FISDW phase diagram of deuterated (TMTSF)$_2$ClO$_4$ for two cooling rates at 1.5 K. Magnetic field up to 28 T was applied parallel to the lowest conductivity direction \( c^* \). For h-ClO$_4$, it is found from the sudden increase of resistance that the transitions to the first and last \((n=1)\) semimetallic SDW phase take place at about 6.5 and 8 T, respectively. Above 27 T the field which was proposed by McKernan et al. as a new phase boundary \( \beta_{HI} \) of a first-order transition, both the nonoscillatory background resistance \( R_0 \) and the amplitude of the rapid oscillation (RO) suddenly increases. On the other hand, by deuteration of the ClO$_4$ salt, the field of the broad peak shifts to the high-field side and \( \beta_{HI} \) is not observed below 28 T. This indicates that deuteration of the ClO$_4$ salt moves the FISDW phase boundary towards the high pressure side and \( \beta_{HI} \) is pushed out above 28 T. With increasing cooling rate \( R_C \), \( \beta_{HI} \) is shifted towards a lower field with a large hysteresis in the magnetoresistance. The nonoscillatory background resistance \( R_0 \) of h-ClO$_4$ shows hysteresis between 14 and 21 T. Moreover, the magnetoresistance of d-ClO$_4$ for the relaxed state shows a steplike change from the phase between 10 and 17 T to the phase above 20 T with hysteresis between 14 and 21 T. With increasing cooling rate, this boundary with hysteresis is shifted towards a lower magnetic field. These results suggest that there is a new phase transition around 17 T.

Figure 2 shows the FISDW phase diagram of d-ClO$_4$ determined from magnetoresistance measurements for various \( R_C \). For a slow cooling, the FISDW transition temperature \( T_{FISDW} \), almost independent of field above 15 T in h-ClO$_4$, slightly decreases with increasing field above 20 T in deuterated ones. In d-ClO$_4$ for 0.018 K/s, the magnetic field dependence of \( T_{FISDW} \) is almost the same as that for 0.0009 K/s. We observe, however, that \( \beta_{HI} \) (27 T) which is independent of temperature below 2.5 K decreases with increasing temperature above 2.5 K. In d-ClO$_4$ for 0.67 K/s, \( \beta_{HI} \) is observed at about 23.5 T and \( T_{FISDW} \) increases with increasing \( B \). The last semimetallic SDW phase between 9.7 T and \( \beta_{HI} \) is reduced from 5.5 to 4 K when the cooling rate \( R_C \) is increased. This result is consistent with the previous report [5] for h-ClO$_4$, the interpretation of which will be discussed later. Thus the experimental results lead to the conclusions that, with increasing the cooling rate \( R_C \), (i) the high-field phase boundary \( \beta_{HI} \) shifts towards a lower field, (ii) the last semimetallic FISDW phase between 9.7 T and \( \beta_{HI} \) is suppressed, (iii) the FISDW insulating phase above \( \beta_{HI} \) in which the Hall voltage becomes almost zero [4] is enhanced. These results mean that the last semimetallic FISDW phase and the FISDW insulating phase correspond to different FISDW states.

Figure 3 shows the Hall resistance in d-ClO$_4$ for two cooling rates at 1.3 K. For relaxed d-ClO$_4$, in which the cooling rate is about 0.0008 K/s, a very stable quantum Hall state is observed above 9 T and the
Figure 3: Hall resistance in deuterated (TMTSF-d$_{12}$)$_2$ClO$_4$ for two cooling rates with the magnetic field parallel to the lowest conductivity direction $c^*$ at 1.3 K.

Figure 4: Schematic of the Fermi surfaces of (TMTSF)$_2$ClO$_4$, resulting from a dimerization of the system along the $b$ axis. $Q_{SDW} = (2k_F, \pi/b)$ = $(2k_F, 0)$ and $Q_{SDW}^{\pm} = (2k_F^{\pm}, \pi/2b)$ are the SDW nesting vectors.

FISDW insulating phase in which the Hall voltage becomes almost zero [4] is not observed below 28 T. With increasing the cooling rate, $\beta_{HI}$ determined from the sudden decrease of the Hall resistance is clearly visible and it is shifted towards a lower field. This result is consistent with the cooling rate dependence of the FISDW phase diagram as shown in Fig. 2. The Hall resistance between 17 and 25 T slightly decrease with increasing the cooling rate. On the other hand, the Hall resistance between 9 and 15 T drastically decrease with increasing the cooling rate. These results suggest that the phases below and above the new phase transition around 17 T correspond to different quantum states. Although magnetoresistance and Hall resistance measurements clearly show the new phase transition around 17 T, the origin of this phase transition is unsolved.

In order to explain this $R_C$ dependence of the FISDW phase, we will now discuss the role of the dimerized gap due to anion ordering (AO) and that of the SDW nesting vector. As discussed in a previous report, [5] the concentration of scattering centers associated with the boundaries between anion-ordered regions increases with increasing $R_C$. For the slowly cooled ClO$_4$ salt, AO creates a superlattice potential dividing the original Fermi surface into two pairs of open sheets. As a result, a dimerized gap due to AO is introduced in the electron band as shown in Fig. 4. Because the periodic anion potential is out of phase at the boundary between adjacent anion-ordered regions, these boundaries not only work as scattering centers but also suppress the dimerized gap due to AO. As a result, the effective dimerized gap due to AO decreases with increasing $R_C$. Using nonperturbative calculations Kishigi claimed that a new FISDW phase with a SDW nesting vector $Q_{SDW}^{\pm} = (2k_F^{\pm}, \pi/2b)$ as shown in Fig. 4 is stabilized for a large $V$. [6] From the cooling rate dependence of the FISDW phase diagram, it is reasonable to estimate that the respective ground states of the last semimetallic FISDW phase and of the insulating FISDW phase are a $n=1$ state with $Q_{SDW}^{\pm}$ or $Q_{SDW}$ and a $n=0$ insulating state with $Q_{SDW}$, respectively. Because the FISDW phase with $Q_{SDW}^{\pm}$ becomes more stable with increasing the value of $V$, the model with $Q_{SDW}^{\pm}$ can explain the decrease of $\beta_{HI}$ and the suppression of the last semimetallic SDW phase. We are therefore led to conclude that $\beta_{HI}$ of our experiment corresponds to the phase boundary between the $Q_{SDW}^{\pm}$ phase and the $Q_{SDW}$ phase.

References