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NON-LINEAR CONDUCTIVITY IN THE SPIN DENSITY WAVE PHASE OF $(TMTSF)_2ClO_4$

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Electrical conductivity was measured in the spin-density wave phase of $(TMTSF)_2ClO_4$, which is induced in zero magnetic field when ordering of ClO_4 ions is suppressed by rapid quenching. The conductivity along the one-dimensional axis increases above a threshold field, in the range of 10–40 mV cm^{-1}. The threshold field increases slightly with increasing temperature. The conductivity increase is presumably assigned to the sliding motion of the spin-density wave, as suggested by Tomic *et al.* for $(TMTSF)_2NO_3$. Significant sample dependence is observed in threshold field-temperature and conductance-field relations; important effects of the interaction between the deformable spin-density wave and pinning centers are inferred.

A RICH variety of phenomena associated with the charge-density wave (CDW) dynamics has been studied for the recent ten years. On the other hand, much less experimental evidences have been accumulated for the similar collective transport in the spin-density wave (SDW). Walsh *et al.* [1] found that the conductivity in the SDW phase of $(TMTSF)_2PF_6$ increases with the applied electric field and proposed that it is associated with the SDW sliding. Subsequent studies [2, 3] made clear that the conductivity increase is rather related to single-particle effects, e.g. hot electron which are coupled with an effect of break of conducting path in highly anisotropic media, however. Osada *et al.* [4] observed, in magnetic field-induced SDW phases of $(TMTSF)_2ClO_4$, that the conductivity perpendicular to the one-dimensional axis is non-ohmic and proposed that is associated with the SDW sliding. Tomic *et al.* [5] found, for the first time, that the conductivity parallel to the one-dimensional *a*-axis increases above a clear threshold in $(TMTSF)_2NO_3$, and suggested that the SDW sliding is responsible for this phenomenon.

It is well known [6] that the SDW appears in $(TMTSF)_2ClO_4$ in zero magnetic field by rapid quenching through the ClO_4 anion ordering temperature ~ 22 K. With increasing quenching rate, the SDW transition temperature (T_N) increases up to 5–6 K. When slowly cooled (in the so-called relaxed state) the SDW is suppressed and the superconductivity appears below 1.2 K. Under strong magnetic field, successive transitions into SDW phases are observed in the relaxed sample. This phenomenon has been subject to current study.

We measured the electrical conductance along the *a*-axis, of quenched $(TMTSF)_2ClO_4$ as functions of both electric field and temperature in zero magnetic field. Clear increase of the conductivity was observed above a threshold electric field (E_T). It is indicative of the sliding motion of the SDW, though direct evidence has not been obtained yet. The threshold field increases with increasing temperature but only weakly, in contrast with the CDW systems. In the latter strong temperature dependence of E_T has been observed. Both the conductance-field relation, in relatively weak E-field, and the threshold field-temperature relation are sample dependent. It is inferred that the SDW shows multiple metastable configuration in the presence of various pinning centres, as well as the CDW.

The standard four probe configuration was used for conductance measurement. Annealed gold wires of 20 μm diameter were attached with silver paint. Distances between the inner contacts were 2–5 mm. Both d.c. and pulse methods were used and these results were compared to exclude any effect of Joule heating. For most samples, d.c. data were found as reliable only below 2 K and in weak electric field. Since crystals of $(TMTSF)_2X$ family are brittle under thermal stress and frequently show sudden resistance increase (resistance jump) during pre-cooling, samples were cooled in helium atmosphere down to 80 K in 3–4 days. On further cooling, samples were quickly immersed into liquid helium after prolonged holding at 40 K. The quenching rate at ~ 20 K was typically larger than $100\,\mathrm{K\,sec^{-1}}$.

Figure 1 shows the temperature dependence of the ohmic conductance. The SDW transition temperature (T_N) is 5 K. Assuming that the conductance below 3 K is of Arrhenius-type, the SDW gap 2Δ is given as approximately 13 K. The smaller ratio $2\Delta/T_N \cong 2.6$, compared with the BCS value of 3.5, is presumably due to distribution of T_N and 2Δ, as the result of inevitable inhomogeneous quenching rate within the sample and internal strain. This estimate is partly supported by the broad plateau just above T_N shown in the inset of Fig. 1; if its broadness were to be attributed solely to the one-dimensional fluctuation, $2\Delta/T_N$ larger than the BCS value should be expected as observed in most CDW systems. The higher temperature data shown in the inset of Fig. 1 were obtained on heating, therefore the effect of gradual growth of anion ordering during measurement is found near T_N; the ordered phase shows smaller resistance than that extrapolated from above the transition temperature. The ratio $R_{min}/R(300\,\mathrm{K})$, where R_{min} is the minimum value of the ohmic resistance near 10 K is approximately equal to 100. This is much smaller than 750 found [5] in $(TMTSF)_2NO_3$ probably because of anion disorder.

Below T_N it was observed that the conductivity σ increases above a threshold field. Figure 2 shows the excess conductivity normalized by the Ohmic value, $(\sigma(E) - \sigma(E \cong 0))/\sigma(E \cong 0)$, at various temperature, of the same sample as that of Fig. 1. For this sample, gold lead wires attached onto evaporated gold pads. The distance between two inner voltage contacts, of width 0.1 mm, was 4.3 mm. Resistance jump of 1% had been observed near 150 K. Similar conductivity increase with a threshold was observed in all samples examined, irrespective of the magnitude of resistance

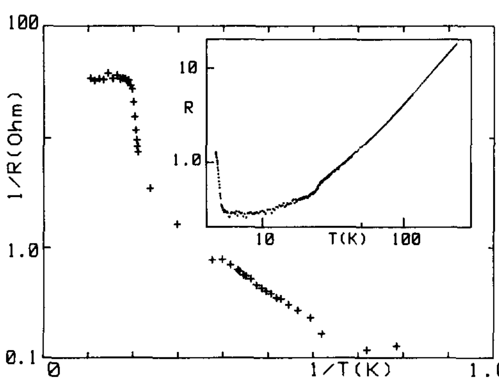

Fig. 1. Ohmic conductance of a $(TMTSF)_2ClO_4$ sample. Inset: Resistance measured on heating above 4 K.

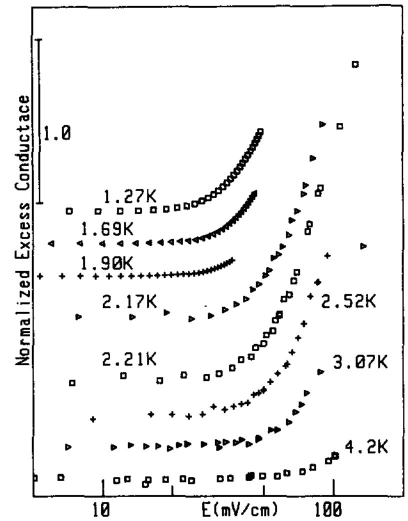

Fig. 2. Excess conductivity of the same sample as that of Fig. 1.

jump and contact distance. Because curves in Fig. 2 are rounded, especially at high temperature, E_T is determined but with relatively large limits of uncertainty. Figure 3 shows E_T as the function of temperature. It is $(25 \pm 5)\,\mathrm{mV\,cm^{-1}}$ below $T/T_N = 0.5$ and increases slightly up to 4 K ($0.8T_N$); $E_T(4\,\mathrm{K})/E_T(T < 2\,\mathrm{K}) \sim 1.4$. The gradual increase of E_T with temperature, and its value in the range of 20–40 mV $\mathrm{cm^{-1}}$ at low temperature as well, have been observed repeatedly in other samples.

Tomic *et al.* [5] have found that E_T, in a good quality sample of $(TMTSF)_2NO_3$, is temperature independent at low T/T_N (< 0.5). Their results are in agreement with the theoretical prediction by Maki and Virosztek [7]. They pointed out that the phase fluctuation has little effect on the depinning process in the SDW systems of low T_N and large coherence

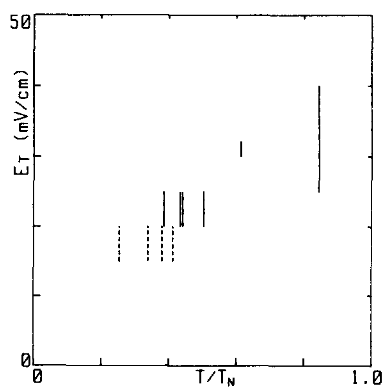

Fig. 3. Threshold electric field for the conductivity increase. Large limits of uncertainty are put because of the rounded conductance curves. Solid and dotted bars denote the pulse and d.c. data, respectively.

length, while the CDW depinning in, e.g. $NbSe_3$, is assisted by the phase fluctuation. As the results, strong increase of the E_T with lowering temperature is expected in the CDW systems as has been observed, while E_T in SDW systems of $(TMTSF)_2$ family is practically independent of temperature below $0.5T_N$. Our result shown in Fig. 3 is qualitatively consistent with the theory by Maki and Virosztek, in the sense that E_T is rather insensitive to temperature and that it does not show the strong increase at low temperature. On the other hand, we have observed, in another samples, that E_T shows gradual decrease with lowering temperature even for $T/T_N < 0.5$. Presumably, the detailed temperature dependence of E_T is subject to a subsidiary effect, such as of the multiplicity of the metastable spatial configuration of the phase which plays an important role in the CDW systems.

The detailed field dependence of the excess conductivity above E_T is sample dependent. In the paper by Tomic *et al.* [5], the normalized excess conductivity $(\sigma(E) - \sigma(E \simeq 0))/\sigma(E \cong 0)$, in $(TMTSF)_2NO_3$, is a universal linear function of $\log(E - E_T)$ in the range below $0.5T_N$ and up to $2E_T$. On the other hand, such a relation is not found in Fig. 2, in which the excess conductivity $\sigma(E) - \sigma(E \ll E_T)$ rather than the normalized excess conductivity, is a universal function of $(E - E_T)$ in a limited temperature range. Such a variety near E_T has been observed in the CDW systems.

Small increase of the conductivity with field was observed at $E < E_T$, especially in samples which suffered larger resistance jump during pre-cooling. Similar non-linearity in $(TMTSF)_2PF_6$ has been attributed to the hot-electron effect of thermally excited normal carriers, coupled with extrinsic effects such as break of current path; there is no threshold and the non-linearity becomes stronger with lowering temperature because of the increasing mobility of normal carriers. On the other hand, for $(TMTSF)_2ClO_4$, the increment of the conductivity below E_T is much smaller; for example, the conductivity shown in Fig. 2 is constant within 1% up to $0.5E_T$ at 1.2 K. The mobility of thermally excited normal carriers is presumably lower in $(TMTSF)_2ClO_4$ because of the anion disorder. The rounding near E_T becomes notably only at higher temperature, as shown in Fig. 2. Therefore, the latter should be attributed to, e.g. a distribution of pinning parameters.

In one sample, the discontinuous conductivity increase (switching) was observed at low temperature, as shown in Fig. 4. To this sample gold lead wires were

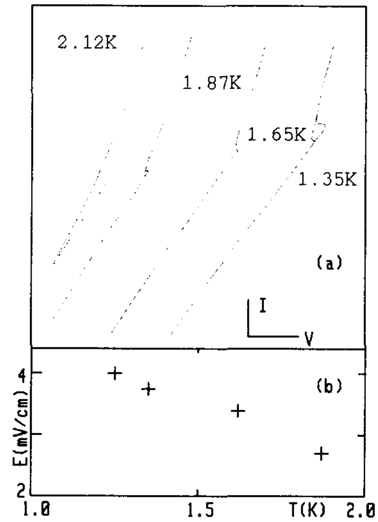

Fig. 4. Switching into the larger conductivity observed in another sample. (a) Current–voltage characteristics. (b) Temperature dependence of the switching field.

attached directly with silver paint and the distance between the inner voltage contacts was approximately 1 mm. Above 2 K it is merged into the smooth change. The switching field increases with lowering temperature, suggesting a role of thermal fluctuation.

Effects of controlled degree of anion order, conductivity at higher E-field and the narrow band noise, which is commonly observed in the CDW systems, are under study.

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