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NONLINEAR TRANSPORT PHENOMENA IN MX_3

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It was confirmed from experiments of the Hall effect that the non-linear conductivity of NbSe_3 results from depinning of CDW's. The non-linear conductivity was also observed in the monoclinic phase of TaS_3 . The field dependence of the conductivity was very similar to that observed in the CDW state of NbSe_3 .

1) INTRODUCTION

Among the transition metal trichalcogenide MX_3 , the charge-density wave (CDW) states have been discovered in NbSe_3 ^{1,2)} and TaS_3 ^{3,4)}. The highly nonlinear conductivities have been found in these CDW states of MX_3 .^{5,6)}

It was first recognized by Fröhlich⁷⁾ that incommensurate CDW condensates are possible to slide as a whole and carry currents under the application of the electric field. In a real system, impurities or commensurability with a host lattice will pin down such a sliding motion of CDW's, and no dc conductivity from CDW is expected at low electric fields.⁸⁾ However, as the applied electric field is increased to the value necessary to overcome the pinning force, the depinning of CDW's will occur and derive the system highly conducting. The non-Ohmic behaviors observed in MX_3 have led to the subject of much interest concerning to sliding motion of depinned CDW's.

In this paper, we report on the Hall effect in the

non-Ohmic regimes of NbSe_3 and the nonlinear conductivity observed in the new phase of TaS_3 .

2) HALL EFFECT IN THE NON-OHMIC REGIMES OF NbSe_3

NbSe_3 undergoes CDW transitions at 142 K (T_1) and 58 K (T_2).²⁾ Just below T_1 and T_2 , the conductivity increases remarkably under an application of very small dc electric field of order 100 mV/cm.⁵⁾ Such a non-Ohmic behavior has been believed resulting from depinning of CDW's. Here, we show results of the Hall effect in the non-Ohmic regimes can be clearly interpreted in terms of parallel resistors, as shown in Fig. 1, corresponding to conduction due to normal carriers and CDW's after depinning. The Hall effect

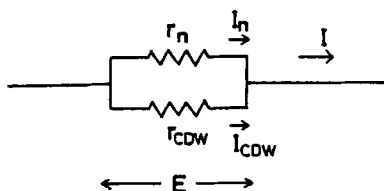


FIGURE 1 A parallel resistor model for the transport in the non-Ohmic regimes of NbSe_3 . E and I represent the electric-field applied to a crystal and total current, respectively.

experiments have been carried out in two non-Ohmic regimes just below T_1 and T_2 .^{9,10)} The Hall voltage measured along the c -axis with dc current parallel to the b -axis below T_1 was reproduced in Fig. 2 as a function of current and field.

The motion of CDW must be restricted in the direction along the b -axis in the T_1 -regime, at least, because the nesting vector has only the b -axis component in this regime.²⁾ Then currents due to depinned CDW's I_{CDW} can not generate the Hall voltage, and so V_H is proportional only to currents due to normal carriers I_n ,

$$V_H \propto I_n \quad (1)$$

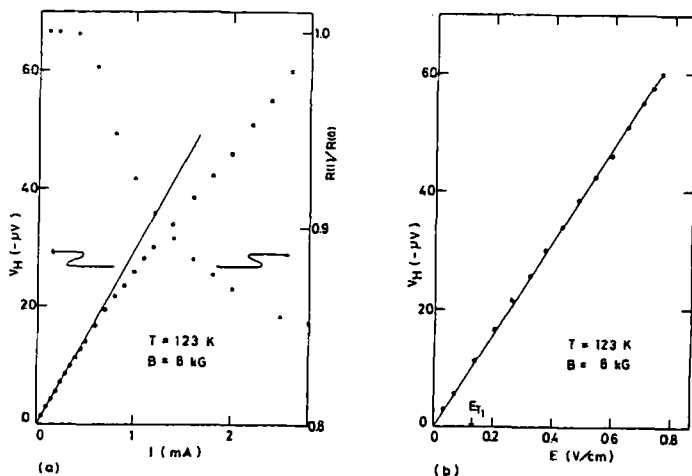


FIGURE 2 The Hall voltage of NbSe_3 , observed below T_1 , plotted as a function of a) current and b) electric-field.

In a parallel circuit, shown in Fig. 1, we obtain simple relations,

$$I = I_n + I_{\text{CDW}} \quad (2)$$

$$E = r_n I_n = r_{\text{CDW}} I_{\text{CDW}} \quad (3)$$

If depinning occurs above a threshold field E_T , the V_H - I curve is expected to deviate downward from a linear relation. On the other hand, the V_H - E relation can be rewritten, by using eq.(3), as V_H - $r_n I_n$ one. According to eq.(1), this relation will be linear. These predictions for V_H - I and V_H - E relations are consistent with the experimental results shown in Fig. 2. The same result is obtained in the T_2 regime.

In above discussion, r_n is assumed to be constant. But, the linear V_H - E relation observed, inversely, means that r_n remains constant below and above E_T , that is, no change occurs in the transport due to normal carriers above E_T . We can conclude that there exists extra currents generating no Hall voltage above E_T and attributed to depinning

of CDW's.

3) NONLINEAR CONDUCTIVITY OF THE NEW PHASE OF TaS_3

Orthorhombic crystal of TaS_3 is more one-dimensional than NbSe_3 and undergoes the CDW transition to a semiconducting state at 218 K.³⁾ The anisotropy of resistivity is about 120 at room temperature, as shown in Fig. 3. The non-Ohmic behaviors observed in this material are similar to those in TTF-TCNQ .¹¹⁾ The CDW has commensurate period of $4c$ and the order of the critical field where I-E curves at different temperatures tend to converge can be understood by the commensurability energy.⁶⁾ Recently, another phase of TaS_3 has been reported by French group.¹²⁾ Its crystal structure is monoclinic and essentially the same with that of NbSe_3 . Monoclinic crystal undergoes two incommensurate-CDW transitions at 240 K and 160 K, very similar to NbSe_3 except the semiconducting state at low temperatures.¹²⁾

Monoclinic crystals were obtained here by a direct reaction of Ta and S at 580 K for three months. Although a crystal obtained often contains both monoclinic and orthorhombic TaS_3 , crystals used in measurements were confirmed to be monoclinic by the single-crystal X-ray diffractometer. The lattice constant along the a -axis is larger by 3 % than that reported by French group.¹²⁾ The resistivities

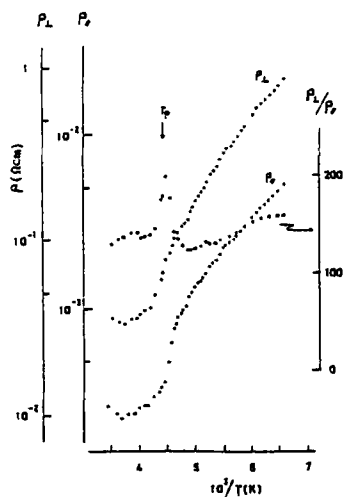


FIGURE 3 Resistivities of orthorhombic TaS_3 , along the b and c axes, measured by the Montgomery method.

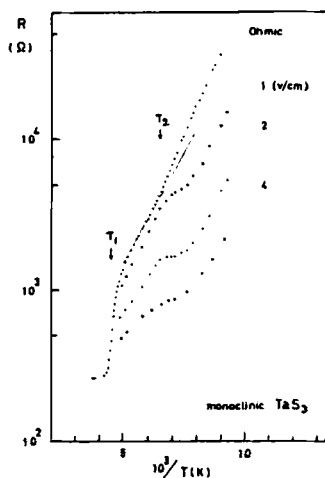


FIGURE 4 The temperature dependence of resistivity of monoclinic TaS_3 .

measured under different electric-fields are plotted in Fig. 4 by semi-log scale as a function of $1/T$. The metal-semiconducting transition occurs at 222 K (T_1). The change of the slope in the $\ln R - 1/T$ curve is observed around 154 K

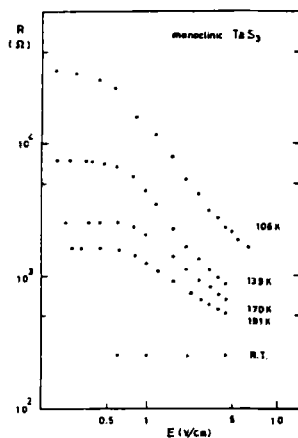


FIGURE 5 The field dependence of the resistivity of monoclinic TaS_3 .

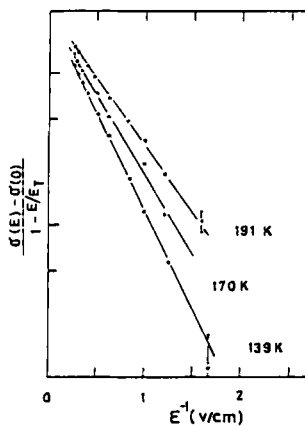


FIGURE 6 The field dependence of the conductivity of monoclinic TaS_3 .

(T_2). The anomalous behavior around T_2 is clear in the data obtained at high electric fields. In the orthorhombic crystal, no anomaly was observed around 154 K.

The resistivity of monoclinic crystals exhibits highly non-Ohmic behaviors below T_1 . The field dependence of the resistivity is shown in Fig. 5. The threshold field is smaller by one-order at least than that observed in orthorhombic crystal. The field dependence of the conductivity, except one at 106 K, can be fitted to the relation

$$\sigma = \sigma(0) + \sigma_0(1 - E_T/E)e^{-E_0/E}, \quad (4)$$

based on the coherent tunneling of CDW's.¹³⁾ The same field dependence has been observed in the non-Ohmic regimes of NbSe₃.

The temperature dependence and non-Ohmic behaviors of the resistivity, mentioned above, are very similar to those obtained by Thompson *et al.*,¹⁴⁾ although the crystal used was reported as orthorhombic.

The temperature dependence of the resistivity, in the monoclinic phase, around T_1 and T_2 is different from those obtained by French group. A possible origin of the difference may exist in the degree of stoichiometry.

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