



ON THE NONLINEAR CHARGE DENSITY WAVE CONDUCTIVITY OF TaS<sub>3</sub>

A. Zettl and G. Grüner

Department of Physics, University of California, Los Angeles, California 90024

and

A. H. Thompson

Corporate Research Laboratory, Exxon Research and Engineering Company,  
Linden, New Jersey 07036

(Received 10 June 1981 by A. Zawadowski)

The conductivity of the charge-density-wave semiconductor TaS<sub>3</sub> is shown to consist of temperature dependent ohmic, and field dependent but temperature independent, contributions at temperatures below the Peierls transition  $T_P = 215$  K. The field dependent conductivity can be described by  $P$  a tunneling formalism proposed by Bardeen.

The linear chain compound TaS<sub>3</sub> (orthorhombic phase) undergoes a phase transition to a Peierls-Frölich semiconducting state<sup>1,2</sup> at  $T_P = 215$  K. The superstructure<sup>2</sup> is at  $0.5 a^*$ ,  $0.125 b^*$ ,  $0.25 c^*$  where  $c$  is the chain direction, and thus the charge density wave (CDW) is commensurate with the underlying lattice. We have shown before that the conductivity is strongly nonlinear<sup>3</sup> and frequency dependent<sup>4</sup> below  $T_P$ , and is accompanied by a giant dielectric constant<sup>3</sup> in the CDW state. These observations, together with the appearance of narrow band noise<sup>5</sup> with fundamental frequency  $f_n \sim I_{CDW}$ , where  $I_{CDW}$  is the excess current due to the CDW, give conclusive evidence that the CDW's are pinned at low fields but can be depinned at high electric field strengths.

In this communication we report detailed field dependent conductivity studies in the CDW state. We demonstrate that the dc conductivity  $\sigma$  below  $T_P$  can be decomposed into two parts: an ohmic  $P$  conductivity which depends strongly on the temperature, and a field dependent conductivity which is independent of temperature above 150 K. We conclude that finite temperature effects are unimportant to describe the CDW dynamics in TaS<sub>3</sub>.

The low field dc conductivity is shown in Fig. 1. The Peierls transition, evidenced by the temperature derivative<sup>3</sup> of  $\sigma$ , is shown in the same figure by the arrow. Detailed field dependent conductivity measurements, using pulse widths as short as 100 nsec, were performed at selected temperatures both above and below  $T_P$ . Above  $T_P$  we have found ohmic behavior up to  $P$  fields  $P$  of approximately 1 V. Below  $T_P$ ,  $\sigma$  is strongly non-ohmic at small electric  $P$  fields. Down to about 130 K we observe a sharp threshold  $E_T$  for the onset of nonlinear conductivity. At  $T$  low temperatures the sharp threshold disappears, most probably due to inhomogeneous electric field distributions. In this communication we focus on experiments performed

in the temperature range where a sharp threshold is observed.

A typical  $\sigma(E)$  curve measured in this temperature region is shown in the insert of Fig. 2.  $\sigma$  measured at 700 mV at various temperatures is also shown in Fig. 1, together with  $\sigma_{ac}$  measured at 2.4 GHz. It is evident from  $\sigma_{ac}$  Fig. 1 that the conductivity is enhanced both at high electric fields and at high frequencies. This is indicative of a strong relation between

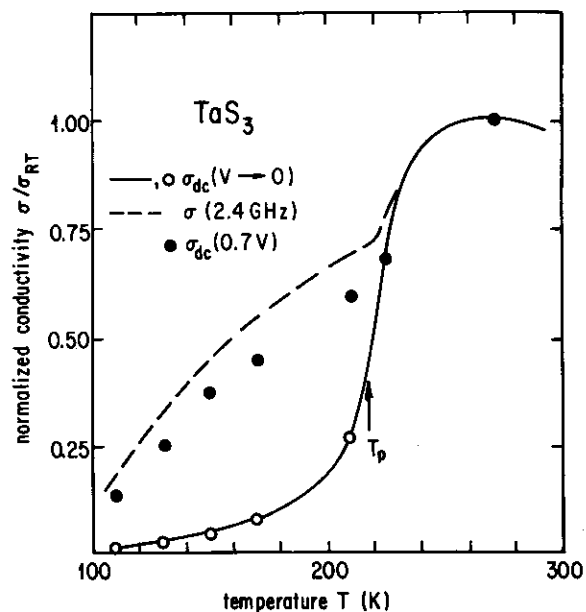


Fig. 1. Temperature dependence of the conductivity of TaS<sub>3</sub>.  $\sigma(2.4 \text{ GHz})$  is from Ref. 4.

field and frequency dependent response of the CDW condensate.

In order to examine the field dependent part of the dc conductivity in detail, we have

subtracted from  $\sigma(T, E)$  the conductivity measured below the threshold field  $E_T$ ,  $\sigma(T, E \rightarrow 0)$ . This is shown in Fig. 2, where  $\sigma(T, E) - \sigma(T, E \rightarrow 0)$  has been normalized to the pre-transition value  $\sigma(T = 221 \text{ K})$ . It is evident from Fig. 2 that  $\sigma$  can be written as

$$\sigma(T, E) = \sigma(T) + \sigma(E) \quad (1)$$

where  $\sigma(T)$  is the ohmic part of the conductivity. The field dependent part  $\sigma(E)$ , which represents the response of the CDW to a dc field, is independent of the temperature between 210 K and 150 K. The ohmic part changes by nearly one order of magnitude in this temperature region. I-V curves taken at temperatures below 150 K show deviations from those shown in the figure. At low temperatures independent evidence<sup>3,4</sup> suggests that a coherent response of the CDW does not apply, and that independent CDW segments, with a distribution of pinning energies, play an important role.

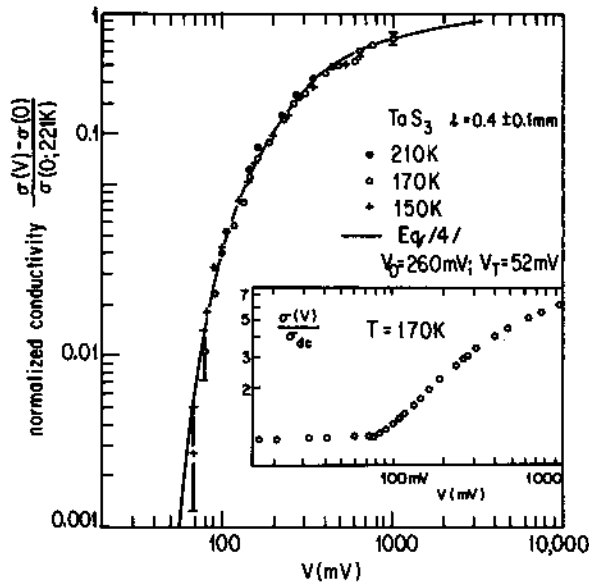


Fig. 2. Field dependence of the nonlinear part of the conductivity. The full line is Eq. (4), with parameters given on the figure. The insert shows  $\sigma(E)$  measured on the same sample.

The ohmic and strongly temperature dependent contribution to the conductivity is most probably due to single particle excitations across the Peierls-Frölich gap. Indeed,  $\sigma(T)$  can well be described by an activated conductivity  $\sigma(T) = \sigma_0 \exp[-\Delta(T)/kT]$ , where  $\Delta(T)$  is temperature dependent.  $\Delta(T \rightarrow 0) = 700 \text{ K}$  and goes to zero at the transition. From  $\sigma(T)$  the temperature dependence of  $\Delta$  can be evaluated.<sup>5</sup> Alternatively,<sup>7</sup>  $\Delta(T)$ , as given by the BCS form, accounts well for the observed  $\sigma(T)$ .

In describing nonlinear conductivity in NbSe<sub>3</sub>, the prototype material for CDW conduction, various formulas have been proposed. The majority of these are modifications of a tunneling formula, accounting also for the finite threshold field for the onset of CDW conductivity.

It was suggested recently by Bardeen<sup>9</sup> that for a state where the CDW can be represented by a finite correlation length  $L$ , two characteristic fields play an important role. The characteristic field appearing in the tunneling probability is given by

$$E_0 = \frac{\pi \epsilon_g^2}{4 \hbar e^* v_F} = \frac{\epsilon_g}{2 \xi_0 e^*} \quad (2)$$

where  $\epsilon_g$  is the pinning gap,  $e^*/e = m/M$  is the ratio of the band mass to the Frölich mass, and  $\xi_0 = 2 \hbar v_F / \hbar \epsilon_g$  is the coherence distance.

The threshold field is given by the criterion that  $e^* E_T L > \epsilon_g$  for tunneling to occur. These assumptions lead to a tunneling probability

$$P(E) \sim [1 - (E_T/E)] \exp(-E_0/E) \quad (3)$$

and consequently to a conductivity given by

$$\sigma(T, E) - \sigma(T) = A [1 - (E_T/E)] \exp(-E_0/E). \quad (4)$$

The full line of Fig. 2 is Eq. (4) with parameters  $E_T = 1.3 \text{ V/cm}$ ,  $E_0 = 6.5 \text{ V/cm}$  calculated from  $V_0$  and  $V_T$  and  $l$  as given on the figure, and  $A = 0.8 \sigma_{RT}$ .

We have also measured the differential conductivity  $dI/dV$  in the presence of a dc bias voltage, using a lock-in technique. By comparing the conductivities measured by pulse and by dc technique, we have established that sample heating is not important up to fields approximately three to four times  $E_m$ . In Fig. 3 the full points are from a continuous  $dV/dI$  plot, and have been normalized to the same  $\sigma(E \rightarrow \infty)$  as given in Fig. 2. The tunneling probability [Eq. (3)] leads to

$$\frac{dI}{dV} \frac{1}{\sigma(E \rightarrow \infty)} = \left[ 1 + \frac{E_0(E - E_T)}{E^2} \right] \exp(-E_0/E). \quad (5)$$

Equation (5), plotted as a function of  $V/V_T$ , is shown as a full line in Fig. 3. The only adjustable parameter in the figure is  $V_T$ , which is determined by the length of the sample. We note that the differential conductance is a very sensitive function of the nonlinear characteristics. Although  $\sigma(E)$  derived on the basis of a classical description<sup>10</sup> leads to a conductivity which has the same overall behavior as the tunneling formula given by Eq. (4),  $dI/dV$  in the classical description diverges at  $E_m$ . This is in contrast to Eq. (5), which predicts an increasing differential conductance with increasing  $V$  in agreement with the experiment.

The measured characteristic fields  $E_0$  and  $E_1$  can be used to evaluate the parameters which characterize the CDW in the tunneling model.<sup>9</sup> From Eq. (2) and from the definition of the correlation length  $L$  it follows that  $E_0/E_T = L/2\xi_0$ . Then with  $E_0 = 6.5 \text{ V/cm}$  and  $E_T = 1.3 \text{ V/cm}$ , we obtain  $L = 10 \xi_0$ , a correlation distance which is ten times larger than the coherence distance. Assuming that  $e^*/e = m/M = 10^{-3}$ , and  $v_F = 10^7 \text{ cm/sec}$ , Eq. (2) leads to  $\epsilon_g = 1.1 \times 10^{-17} \text{ ergs}$ , orders of magnitude smaller than  $kT$ , and to  $\xi_0 = \epsilon_g / 2e^*E = 10^{-14} \text{ cm}$ .

We also note that a formula, slightly dif-

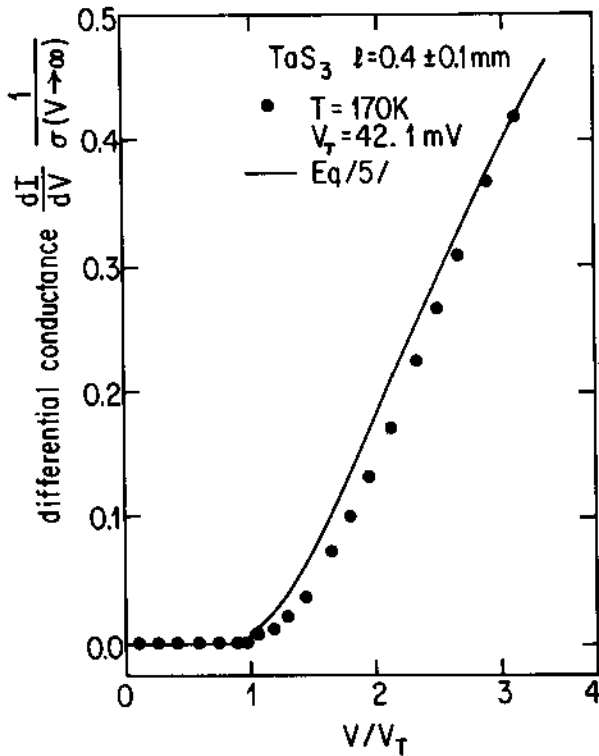


Fig. 3. Differential conductance  $dI/dV$  versus electric field. The full line is Eq. (5) with parameters given on the figure.

ferent from Eq. (4), was proposed to account for  $\sigma(E)$  observed in NbSe<sub>3</sub>. Fleming and Grimes<sup>8</sup> analyzed their experiments using an empirical expression

$$\sigma(E) = A[1 - (E_T/E)] \exp[-E_0/(E-E_T)]. \quad (6)$$

We also find good agreement between our experiments and Eq. (6).

In conclusion, we have shown that the dc conductivity  $\sigma(T, E)$  of TaS<sub>3</sub> can be decomposed into a temperature dependent but ohmic part, and a field dependent part which does not appear to depend on the temperature. This observation suggests that the high field limit of the conductivity depends only slightly on the temperature somewhat below the Peierls transition. The analysis also suggests that  $\sigma$ , due to the sliding CDW, is close to the conductivity due to normal electrons observed above the transition temperature  $T_p$ .

The field dependent part of the conductivity can be described by the tunneling formula proposed by Bardeen, with parameters in agreement with those suggested by commensurability pinning.<sup>9</sup> The characteristic energy  $\epsilon$  is orders of magnitude smaller than  $kT$ , and the characteristic length  $L \gg c_0$ , where  $c_0$  is the lattice constant, providing direct evidence for transport by the collective mode. The observation that  $E_0$  and  $E_T$  are independent of the temperature is surprising. The coherence length is expected to decrease near  $T_p$ , leading to a divergent  $E_0$  (and also  $E_T$ ) at the transition temperature. While this has been observed in NbSe<sub>3</sub>,<sup>11</sup> both  $E_T$  and  $E_0$  are temperature independent up to  $(T_p - T)T^{-1} = 4 \times 10^{-2}$ .

We also note that the same analysis was previously applied by Bardeen<sup>9</sup> to account for the nonlinear conductivity of TaS<sub>3</sub> observed at low temperatures.<sup>12</sup> We have shown before<sup>3,4</sup> that at low temperatures randomness plays an important role and has a drastic influence on the I-V characteristics. We believe, therefore, that our evaluation of the CDW parameters is more relevant to the intrinsic coherent response of the charge density waves in TaS<sub>3</sub>.

We acknowledge useful conversations with J. Bardeen, W. G. Clark, and T. Holstein, and thank P. M. Chaikin for the use of the ac bridge and lock-in amplifier. This report is based upon work supported by NSF Grant DMR 81-03085 and a grant from the UCLA Academic Senate Research Committee.

#### REFERENCES

1. SAMBONGI, T., TSUTSUMI, K., SHIOZAKA, Y., YAMAMOTO, M., YAMAYA, K., and ABE, Y., *Solid State Commun.* **22**, 729 (1977).
2. TSUTSUMI, T., SAMBONGI, T., KAGOSHIMA, S., and ISHIGURO, T., *J. Phys. Soc. Japan* **44**, 1735 (1978).
3. THOMPSON, A. H., ZETTL, A., and GRÜNER, G., *Phys. Rev. Lett.* (submitted for publication).
4. JACKSON, C., ZETTL, A., GRÜNER, G., and THOMPSON, A. H., *Solid State Commun.* (submitted for publication).
5. GRÜNER, G., ZETTL, A., CLARK, W. G., and THOMPSON, A. H., *Phys. Rev. B*, June 15 (1981) (to be published).
6. LEE, P. A., RICE, T. M., and ANDERSON, P. W., *Solid State Commun.* **14**, 703 (1974).
7. TSANG, J. C., HERMANN, C., and SHAFER, M. W., *Phys. Rev. Lett.* **40**, 1528 (1978).
8. FLEMING, R. M., and GRIMES, C. C., *Phys. Rev. Lett.* **42**, 1423 (1979).
9. BARDEEN, J., *Phys. Rev. Lett.* **45**, 1978 (1980).
10. GRÜNER, G., ZAWADOWSKI, A., and CHAIKIN, P. M., *Phys. Rev. Lett.* **46**, 511 (1981).
11. FLEMING, R. M., *Phys. Rev. B* **22**, 25606 (1980).
12. TAKOSHIMA, T., IDO, M., TSUTSUMI, K., and SAMBONGI, T., *Solid State Commun.* **35**, 911 (1980).