Broad Band Noise of monoclinic TaS_3 and $NbSe_3$

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Abstract

The first observation of non-linear conductivity and broad band noise associated with the non-linear conduction in monoclinic TaS_3 are reported. The results of broad band noise measurements in $NbSe_3$ are also reported for comparison.

The number of degrees of freedom for the CDW motion was estimated from the analysis of the results of broad band noise measurements. The result surprisingly indicates that about $10^{11-12}/cm^3$ of degrees of freedom exist in both of monoclinic TaS_3, which is semiconducting at low temperatures, and of $NbSe_3$, which is metallic.

1.Introduction

The transition metal tri-chalcogenide TaS_3 is one of the linear chain conductors. These quasi-one dimensional conductors show very interesting properties due to the collective motion of the depinned charge-density-waves (CDW's) proposed by Frohlich(1) and Lee, Rice, Anderson(2).

These properties were originally found in the other member of tri-chalcogenides, $NbSe_3$. That is, the observation of non-linear conduction at extremely weak field of tens of mV/cm only below the CDW transition temperature(3), and the observation of very large noise only above an onset of the non-linear conduction(4),(5), and so on. The recent interest of many researchers seems to be focused on the phenomena suggesting the existence of metastable state such as curious responses to short pulses(6)(7)(8),hysterisis and switching(9).

Especially, the low frequency broad band roise seems to be interesting because the low frequency broad band noise(10) may reflect the dynamical mechanism of the CDW motion.

Though the studies of the interesting propegrties of $NbSe_3$ have brought a lot of fruitful results, we think that the pace of the study for TaS_3 was much slower than that of $NbSe_3$. This may be partly because the sample of TaS_3 is finer. However, the main reason is thought to be that it is difficult to separate two different types of crystal structure in the crystal growth process, that is, one is orthorhombic[11] and another is monoclinic[12].

In particular, as for monoclinic TaS_3, few electrical measurements were made. Two years ago, we observed very strong non-linear conductivity and large broad band noise associated with this non-linear conduction[13].

Though the crystal structure of monoclinic TaS_3 is the same as that of $NbSe_3$, monoclinic TaS_3 is semiconducting below the CDW transition temperature while $NbSe_3$ remains metallic at low temperatures. So we consider that the monoclinic TaS_3 may be the best candidate to investigate the dynamics of depinned CDW systems. Orthorhombic TaS_3 shows a metall- insulator transition due to the formation of the CDW, too[14],[15], and also shows non-linear conductivity[16],[17] and noise phenomena[18][19] and so on. But as is shown later, we have found that the non-linear phenomena in monoclinic TaS_3 are more pronounced as compared with orthorhombic one.

In this paper, we report the experimental results of the measurements of non-linear conductivity and broad band noise of monoclinic TaS_3, and the results of broad band noise measurements of $NbSe_3$ were also reported for comparison. Brief discussion of the results will also be made.

2. Experiment

Unfortunately, we have not succeeded in obtaining the definite condition for the growth of monoclinic TaS_3 yet. But thanks to the valuable suggestion by Prof.Rouxel, we could obtain relatively good results under the conditions reported elsewhere[13].

Electrical conductivity measurements has been made using an ordinary four-probe method, flowing dc continuous current or dc pulse current. Pulse width was varied from 2μsec to 400μsec, and the frequency of the pulse was typically 100Hz.

Broad band noise measurements have been made by detecting the signal generated at the voltage leads of the sample when only dc current flows through the specimen. Detection was made by a lock-in amplifier in ac voltmeter mode after passing through a pre-amplifier. The frequency range is between 1Hz and 100kHz, and the Q value was varied between 20 and 100. The choice of the Q value did not affect the results at all.

3. Experimental results

Figure 1 shows the temperature dependence of the low field conductivity of monoclinic TaS_3. Two independent peaks at T_1=240K and T_2=160K in the temperature derivative indicated clearly that this sample is monoclinic, comparing with the results already reported by Roucau et al[20].

By increasing the electric field, non-linear conductivity shown in

Fig.2(a) can be obtained. The rise of the field dependent conductivity is more strong than that of orthorhombic TaS_3 which is shown in Fig.2(b). Those non-linear conduction can be observed only below the CDW transition temperature T_1.

A definite threshold field E_T exists in the non-ohmic conductivity at each temperature. This can also be confirmed more clealy by measureing the differential resistivity (Fig.3). Below T_2, at first E_T decreases with decreasing temperature and shows a minimum of about 80mV/cm at about 140K, then E_T increases with further decreasing temperature. This behavior of the temperature dependence of E_T in monoclinic TaS_3 resembles that in $NbSe_3$[(5)] very much. At each temperature, high field conductivity seems to approach the ohmic value at the room temperature. These features of non-ohmic conduction are reproducible.

Accompanied by the non-linear conduction, large noise is observed. As is observed in $NbSe_3$, this noise also consists of both narrow band noise and broad band noise, and we have made both narrow and broad band noise measurements.

As for the narrow band noise, the linear relation between the characteristic frequency and the current carried by the CDW was obtained as in the case of $NbSe_3$[(21)], and using the elementary relation[(21)(22)], the carrier density condensed into the CDW is estimated as $n_c=1.7\times10^{21}/cm^3$.

But we believe that the low frequency broad band noise may be a more important clue to the depinning mechanisms of the CDW's. Figure 4 shows the example of the current dependence of broad band noise in monoclinic TaS_3. With increasing the current, noise suddenly appeared at the threshold of non-linear conduction, and after having a maximum just above the threshold field, the magnitude of the noise became almost independent of the current. The current which gives the maximum is almost independent of the frequency. This fact indicates that this peak does not originate from narrow band noise. Qualitatively similar behavior was obtained at other temperatures, but the shape of the curve becomes broader at 95K. At 80K, the magnitude of the noise is so large. Comparing with the thermal noise at the same temperature, this noise is 10^8 times larger at 500Hz.

When the value of current was fixed at the peak position in order to measure the frequency dependence of broad band noise, as is shown in Fig.5, a 1/f spectrum can be obtained at each temperature. Even if we fix the current at other position, we also obtained the 1/f spectrum. The power of noise strongly increases with decreasing temperature.

Up to now, the origin of low-frequency broad band noise is not clarified. Before we further discuss this problem, it seems better that the experimental results of the broad band noise measurements in $NbSe_3$ are shown.

As for the broad band noise in $NbSe_3$, Richard et al[23] have already reported the following results in the last year.

1) They found that the noise power is proportional to the length of the sample. So they concluded that this broad band noise should not be a contact effect, and that noise generators are statistically independently distributed.

2) The frequency dependence of noise obeys a 1/f law, more precisely, $f^{-0.8}$.

We also performed the same measurements for $NbSe_3$ as was made in monoclinic TaS_3. Since $NbSe_3$ is metallic even at low temperatures, it is possible to estimate the purity of samples by the residual resistivity ratio RRR as the parameter. So we can get the information on the purity dependence of broad band noise.

Table 1 is the list of the samples used by us. We summarize our results of the broad band noise measurements for $NbSe_3$.

1) In the temperature range above 47K, the current dependence of noise is almost similar to that observed in monoclinic TaS_3. That is, after having a maximum just above the threshold, the magnitude of noise decreases and becomes almost independent of the current(Fig.6(a)).

2) The height of the maximum value of the noise becomes temperature independent above 47K. At low temperatures, different behavior is observed. The magnitude of the noise has a tail to the lower field side below the threshold, and becomes a monotonically increasing function of the field in the measured field range(Fig.6(b)).

3) The frequency spectrum is f^{-a}. The value of a is ranged between 0.8 to 1.2. The value of a does not depend on the temperature and the current either. This frequency dependence is in good agreement with the result of Richard et al.

4) We have made the same measurements in many samples with different purity, but systematic difference in the magnitude of the noise is not found among these samples. That is, the current fluctuation $\langle(\Delta I)^2\rangle$ at the peak position, which is obtained from the experimental data by the procedure shown later, does not depend on the temperature above 47K (Fig.7) nor the RRR value (Fig.8).

4. Discussion

Several models for the low-frequency broad band noise were proposed up to now. That is, the effect due to the internal deformation in the moving CDW with a constant velocity v_s [19], the beat among many CDW domains which generates the oscillating current whose frequency is slightly different from each other [23], and the effect which takes place when the transformation from condensed carrier to normal carrier takes place [24]. But there is no complete explanation. If we focus on the number of degrees of freedom of the CDW motion, there seem to be two view points which seem to oppose each other. One is to regard that the CDW moves as a single deformable domain, that is, the degree of freedom of the motion is only one. The ground for this idea is the observation of coherent current oscillation in real time [8][25]. Another is to consider the excess current to be carried by many degrees of freedom of the motion. When we consider the problem of broad band noise, we take the latter view point, because we think that the low frequency broad band noise cannot be generated by only a single degree of freedom. As is shown above, the magnitude of broad band noise is largest near the threshold field E_T. In this field region, the statistical fluctuation of the depinning motion of each CDW domain may be remarkable. If we assume each domain can be depined with the depinning probability P, which is an increasing function of the electric field, then the relative fluctuation of the current carried by the CDW is represented as follows.

$$\langle(\Delta I)^2\rangle / I_{CDW}{}^2 = (1/NV)(1-P/P) \ ,$$

where N is the density of the number of degrees of freedom of the motion, V is the volume of the sample. Using this equation, we can estimated the value of N. On estimating the value $\langle(\Delta I)^2\rangle$, we integrated the spectrum of our broad band noise, using Wiener-Kintchine's theorem with some assumptions [26]. The estimated value of N is almost $10^{11-12}/cm^3$, that is, 10^{3-4} in the sample for monoclinic TaS_3.

If we carry out the same procedure for the results in $NbSe_3$, the value of 10^{3-4} in the sample is obtained. This value does not seem to depend on the temperature nor the RRR value. That is, although the magnitude of the voltage fluctuation is very different between the two materials, the number of degrees of freedom is almost of the same order of magnitude between the two. This surprising result obtained for semiconducting monoclinic TaS_3 and metallic $NbSe_3$ may be consistent with other similarities in the CDW properties. More explicitly,

1) Both undergo two independent incommensurate CDW transitions.

2) Below the CDW transition temperature, both show non-ohmic conduction with a definite threshold field E_T. At low temperatures, the threshold field for the non-linear conductivity increases with decreasing temperature, after having a

minimum at about $T=0.9xT_2$.

3)Both show narrow band noise. The density of the condensed carrier estimated from the narrow band noise spectrum is of the same orders of magnitude, $10^{21}/cm^3$.

But the most difficult point for this model is how to reconcile with the explanation for the observation of current oscillation in real time. Moreover, no satiafactory explanation has been given for the 1/f spectrum. Ordinary 1/f noise in other systems has been explained as due to an effect in the equilibrium state(27). But the 1/f noise in $NbSe_3$ and TaS_3 is an effect in the non-equilibrium state. So this 1/f noise is essentially different from the ordinary 1/f noise observed so far in other systems.

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10) The term "broad band noise" seems to have been used in two meanings. One literaly means "the noise that is distributed over the wide frequency range", and another is "the low-frequency noise". In this paper, we use the term "broad band noise" in the latter sence.

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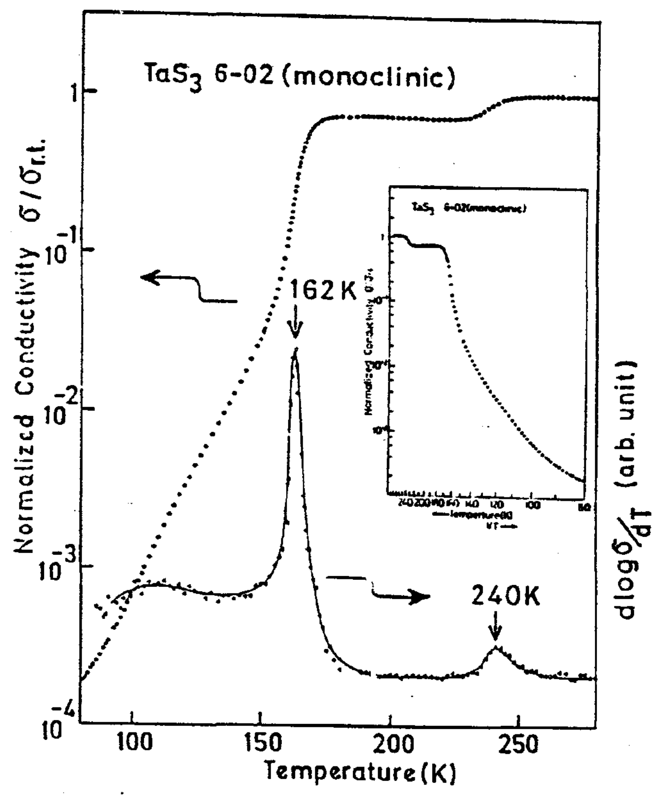
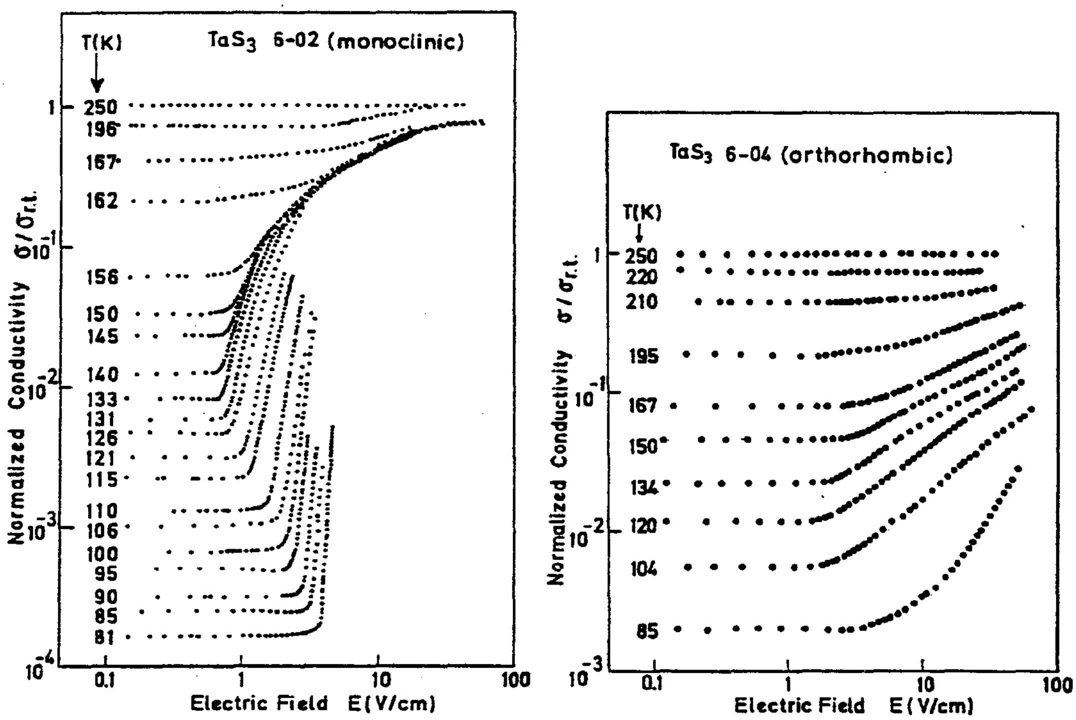


Figure 1 The temperature dependence of the normalized conductivity of monoclinic TaS_3, together with the temperature derivative. The inset shows the Arrhenius plot of the same sample.



(a) (b)

Figure 2 The electric field dependence of the conductivity of monoclinic TaS_3((a)) and that of orthorhombic TaS_3((b)).

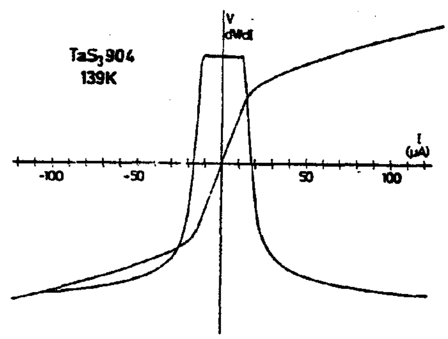


Figure 3 The electric field dependence of the differential resistance of monoclinic TaS_3, together with the I-V curve.

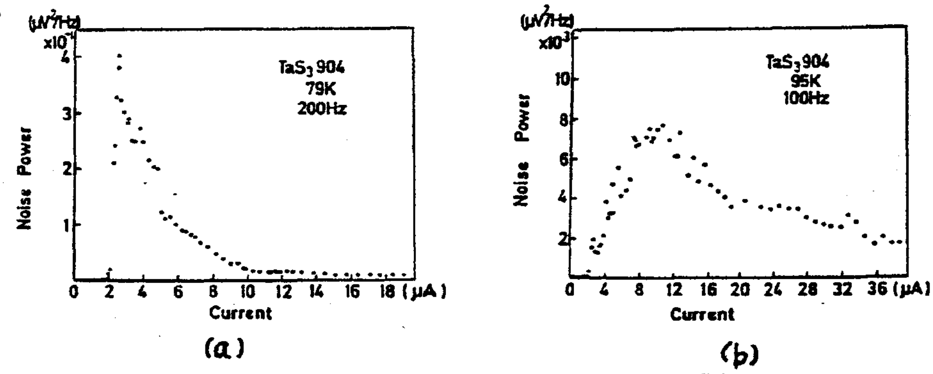


Figure 4 Broad band noise of monoclinic TaS_3 as a function of the current through the sample. A sharp increase in the noise is observed at the current corresponding to the threshold of the non-ohmic conduction.

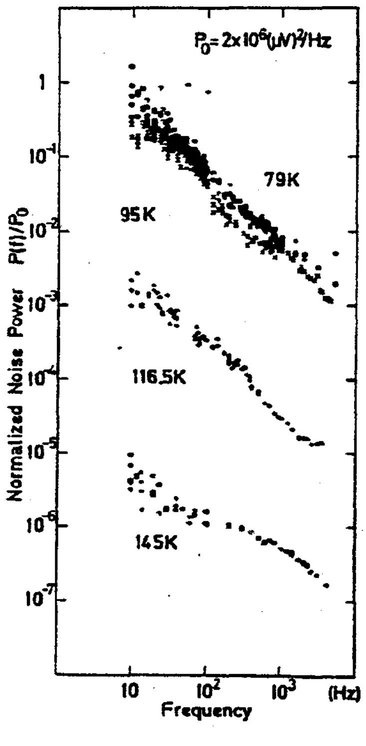


Figure 5 The frequency dependence of the noise of monoclinic TaS_3.

No.	RRR	length:L	cross section:A	volume:V
304	158	0.21 cm	4.0×10^{-7} cm^2	8.4×10^{-8} cm^3
305	218	0.26	1.1	2.9
406	127	0.25	1.7	4.4
407	221	0.23	3.3	8.4
509	54	0.11	1.9	3.8
510	137	0.16	0.72	1.1
611	83.9	0.18	1.0	1.8
612	89.4	0.17	1.8	3.1
613	86.8	υ.12	1.1	1.3
314	133	0.17	1.2	2.0

Table 1 The list of the $NbSe_3$ samples used for broad band noise measurement.

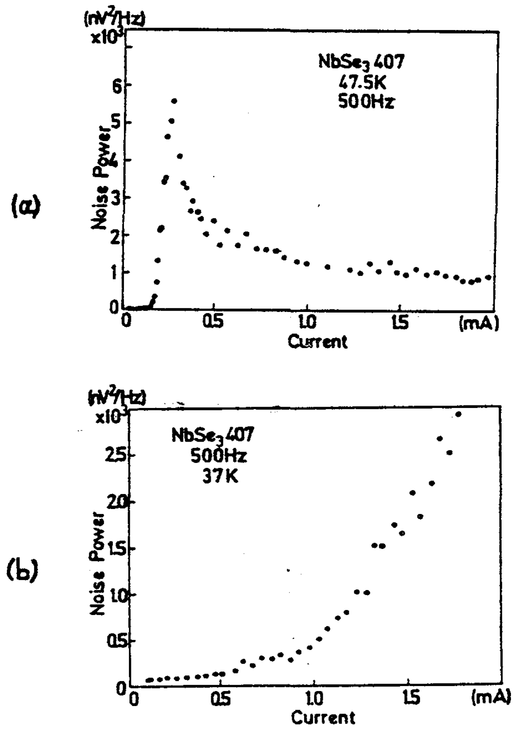


Figure 6 The current dependence of broad band noise of $NbSe_3$. Figure(b) is the data at 37K. The threshold current at this temperature obtained from the results of the ordinary conductivity measurements is 0.95mA.

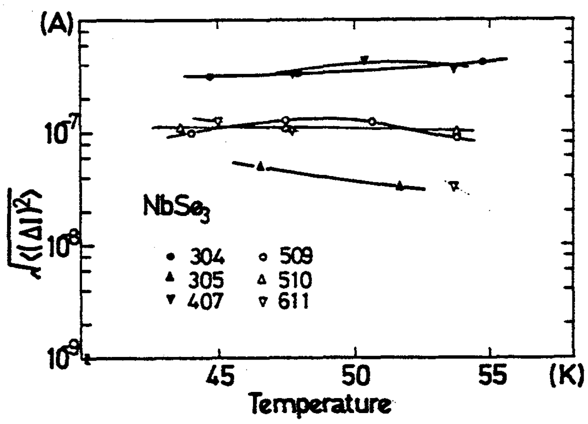


Figure 7 The temperature dependence of the fluctuation of the current $\langle(\Delta I)^2\rangle$ in $NbSe_3$. $\langle(\Delta I)^2\rangle$ is obtained by the integration of the experimental results, using Wiener-Kintchine's theorem.

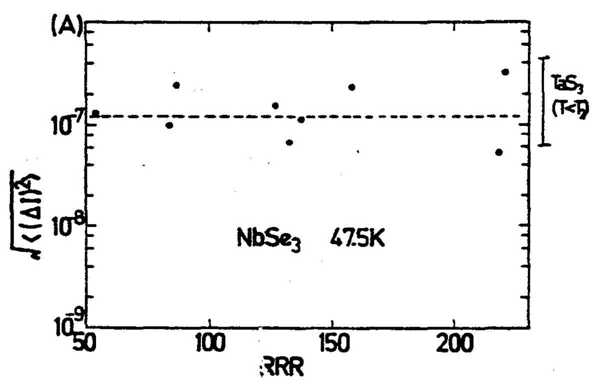


Figure 8 The comparison of the fluctuation of the current $\langle(\Delta I)^2\rangle$ among samples with different purity. The results for TaS_3 are also shown.