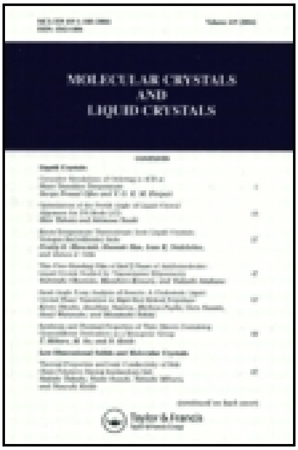

Molecular Crystals and Liquid Crystals
Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl16

Microwave and Hall Studies of TaS_3 and NbS_3

N. P. Ong [a], G. X. Tessema [a], G. Verma [a], J. C. Eckert [c a], J. Savage [a] & S. K. Khanna [b a]
[a] Department of Physics, University of Southern California
[b] Jet Propulsion Laboratory
[c] Physics Department, Harvey Mudd College, Pomona, CA
Version of record first published: 14 Oct 2011.

To cite this article: N. P. Ong, G. X. Tessema, G. Verma, J. C. Eckert, J. Savage & S. K. Khanna (1982): Microwave and Hall Studies of TaS_3 and NbS_3, Molecular Crystals and Liquid Crystals, 81:1, 41-47

To link to this article: http://dx.doi.org/10.1080/00268948208072549

Mol. Cryst. Liq. Cryst., 1982, Vol. 81, pp. 41–47
0026-8941/82/8101-0041$06.50/0

(Proceedings of the International Conference on Low-Dimensional Conductors, Boulder, Colorado, August 1981)

MICROWAVE AND HALL STUDIES OF TaS_3 and NbS_3

N.P. ONG, G.X. TESSEMA, G. VERMA
J.C. ECKERT*, J. SAVAGE
Department of Physics
University of Southern California

S.K. KHANNA
Jet Propulsion Laboratory

Received for publication August 27, 1981

The microwave conductivity of TaS_3 and NbS_3 have been measured. In both compounds the conductivity at 9.8 GHz shows very large enhancement over the dc value. A brief discussion based on contributions from charge-density-wave oscillation is given for TaS_3. The Hall effect at room temperature for TaS_3 is found to be $|R_H| = 6.3 \times 10^{-4}$ C/m^3. This corresponds to a carrier density of 1×10^{22} cm^{-3} and a mobility of 2 cm^2/vs. A comparison with the figure obtained from conduction noise measurements will be made.

MICROWAVE CONDUCTIVITY

High frequency measurements of the conductivity of the trichalcogenides have proved very powerful in revealing the contributions of the oscillating charge-density-wave (CDW) condensate.[1] We have performed measurements of the complex dielectric constant of TaS_3 and NbS_3 at 9.8 GHz from 4K to 300K using the well-known cavity perturbation equations[2] for high conductivity samples.

In Fig. 1 the conductivity at 9.8 GHz is compared with the dc for two samples of TaS_3. It is clear that below the transition at 210K there is a very large enhancement over the dc value. This enhancement grows with decreasing

*Present address: Physics Department, Harvey Mudd College, Pomona, CA.

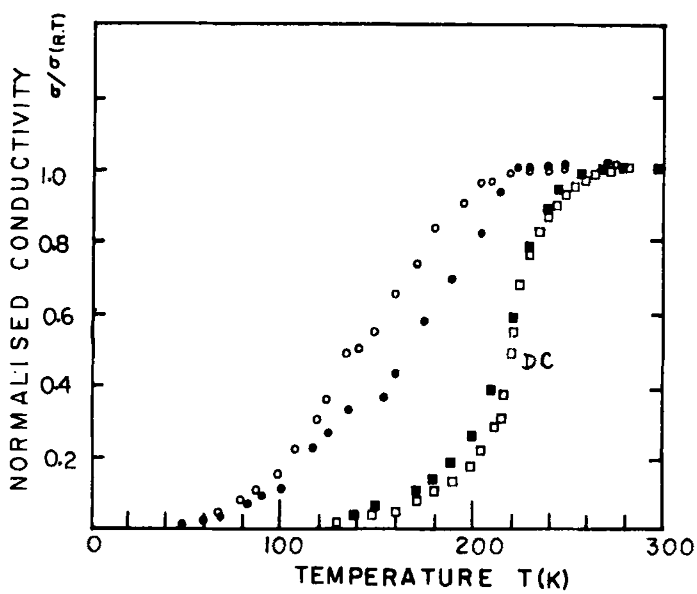

FIGURE 1 The conductivity of TaS_3 at 9.8 GHz (circles) and dc (squares). Solid (open) circles refer to Sample 1 (2).

temperature till it attains a factor of 250 at 30K. The dielectric constant (not shown) increases approximately linearly with temperature and is of the order 250,000 at 200K.

Tantalum trisulphide has been shown[3] to occur in two polytypes (orthorhombic and monoclinic). In the orthorhombic structure a metal insulator transition occurs[4] at ~ 210K. The charge-density-waves in neighbouring chains become locked in phase at this temperature and the dc conductivity decreases exponentially below 210K. In the monoclinic structure two phase transitions occur with the formation of incommensurate CDWs.[5] The situation is similar to that of $NbSe_3$ which also has a very similar crystal structure. On the basis of our dc measurements we have observed only one phase transition in our samples and therefore identify our samples as orthorhombic.

Aside from the large enhancement of the 9.8 GHz conductivity over the dc value the most interesting feature of the microwave conductivity is the flat temperature dependence above 200K. In the dc data (measured on the same samples) fluctuations into the CDW state are pronounced and extend to temperatures 60K above the Peierls transition temperature T_p. (This is due to the more quasi-one dimensional structure as compared with $NbSe_3$.) These fluctuations lead to a large enhancement of the dc resistivity well above T_p. (Above 260K the resistivity in our samples is almost temperature independent.) In sharp contrast, the microwave conductivity remains temperature independent until a few Kelvin below T_p, where it begins to decrease gradually. Thus, the high frequency conductivity is unaffected by the uncorrelated one-dimensional fluctuations into the CDW state. At present it is not known if the excess 9.8 GHz conductivity is due to the driven oscillation of the CDW condensate about pinning centers or due to the actual sliding of the uncorrelated one-dimensional fluctuations. The study of CDW transport and interaction of the condensate with rf fields will be very fruitful near T_p in TaS_3.

Figure 2 shows the microwave conductivity and dielectric constant of NbS_3 as a function of temperature. As in TaS_3 a very large enhancement over the dc value is observed in the 9.8 GHz conductivity. We have plotted both the 9.8 GHz and dc conductivity data normalized to the room temperature value. However, the observed absolute value at 9.8 GHz at 300K is 1000 $(\Omega\ cm)^{-1}$ compared with 200 $(\Omega\ cm)^{-1}$ for the dc. Thus the enhancement factor is larger than

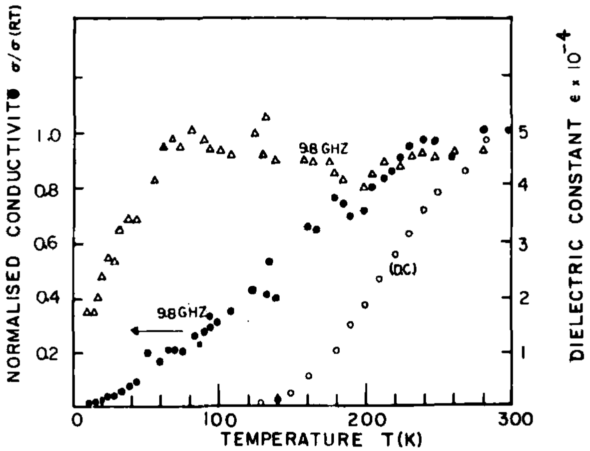

FIGURE 2 The conductivity (solid circles) and dielectric constant (triangles) of NbS_3 at 9.8 GHz. The dc conductivity is also shown (open circles).

suggested by Fig. 2. The rather large value for the real part of the dielectric constant ε_1 (50,000 at 100K) is quite surprising and suggests a strong parallel with the tantalum compound. (The detailed temperature variation of ε_1 is sensitive to the choice of the filling factor α in the cavity perturbation equations. However, reasonable choices of α all give a value of ε_1 exceeding 10,000. We have adopted a uniform criterion for determining α based on the depolarization peak in the loss data. A detailed discussion of this point will be provided in a forthcoming full report of these experiments.)

The phase diagram of the Nb-S system is quite complicated. Crystals of stoichiometry NbS_2, Nb_3S_4, $Nb_{1+x}S$ and NbS_3 have been obtained by various groups[6]. The trisulphide NbS_3 has been reported by Rijnsdorp and Jellinek[7] to have a structure very close to the monoclinic form of $ZrSe_3$. The main difference from the $ZrSe_3$ structure is in the existence of a slight modulation which doubles the unit cell in the chain (b) direction. Again on the basis of our dc conductivity we tentatively identify our samples with the dimerized form.

Because of the enormous value of the dielectric constant and the enhanced microwave conductivity we argue that the dimerization is a periodic lattice distortion stabilized by a charge-density-wave. Such a large polarizability is consistent with a weakly pinned electron-phonon condensate. (The commensurability of the presumed CDW in NbS_3 would imply that pinning to the host is orders of magnitude stronger than for the incommensurate CDWs in $NbSe_3$. Nonetheless it is still sufficiently polarizable to give the large observed values of ε_1.) The presence of a periodic lattice distortion and the large polarizability at 9.8 GHz make the case for a CDW compelling in NbS_3. A test for this hypothesis will be provided by alloying with a group IV element or by intercalation (changing the Fermi wave vector.)

Non-Ohmicity in TaS_3 was first reported[8] by Sambongi's group. Recently Thompson et al[9] reported the observation in TaS_3 of a threshold field E_T beyond which non-Ohmicity takes place. Conduction noise was observed in the non-Ohmic regime by Grüner et al[10] and strong frequency dependence of the conductivity was also observed[11]. We have also performed non-Ohmic measurements on our samples of TaS_3 using pulses of widths under 1 μs. Our I-V curves are similar to those reported by Sambongi's group. No evidence of threshold field was observed in our samples. At present the question of whether the observations of a threshold field and conduction noise are associated with the monoclinic phase (where the CDW's are incommensurate) or with the orthorhombic phase remains open, and more investigations are necessary. No non-Ohmicity at room temperature was observed in NbS_3 up to fields exceeding 300 V/cm, aside from a smooth quadratic increase in the conductivity which was ascribed to self-heating.

HALL EFFECT

Turning to the Hall effect we felt that it is important to make comparisons in the case of TaS_3 because of the slight controversy surrounding the Hall measurements in $NbSe_3$ and the noise measurements of Monceau et al[12] (from which a condensate density can be extracted). Since the transition to the insulating state is complete in TaS_3 and the anisotropy (~ 150) is not excessively large only one kind of carrier exists above T_p (210K). The measured Hall constant R_H gives directly the condensate density. (In $NbSe_3$ the interpretation is complicated by two transitions and the presence of surviving pockets of electrons and holes.)[13]

The small transverse dimension of our samples (3mm x 45μm x 3μm) presented serious difficulties with two Hall leads. Consequently we dropped one Hall lead and adopted an ac bridge method (see inset Fig. 3). This three probe technique[14] more fully exploits lock-in methods because of the lack of an IR drop due to lead misalignment. The

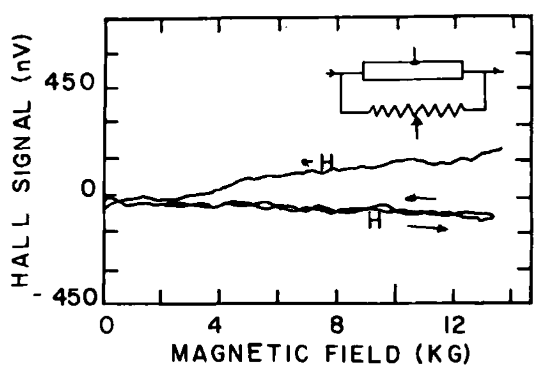

FIGURE 3 The Hall signal of TaS_3. The excitation current is 1 mA. Different sweep directions (arrows) indicate no hysteresis. Inset is the three-probe bridge used.

in-phase voltage is proportional to the Hall constant. Our measurements show that $|R_H| = 6.3 \times 10^{-4}$ C/cm^3 corresponding to 1.0×10^{22} carriers/cm^3. The sign of R_H was not determined. Also the tenuous nature of the silver paint contact precluded low temperature readings. The measured room temperature conductivity is 3400 $(\Omega\ \mathrm{cm})^{-1}$.

Grüner et al[10] have observed sliding CDW noise in TaS_3 and by plotting noise frequency versus the CDW contribution to the current I_{CDW} (following Monceau et al[12]) they report a value for n_s equal to 2×10^{21} cm^{-3}. Thus the agreement between the Hall result and the noise is satisfactory (bearing in mind that the crystals may belong to different morphologies).

This high carrier density implies a mobility of only ~ 2 cm^2/vs a fact which explains the flat variation of resistivity with temperature above 210K. The electron gas is so strongly scattered that further scattering by phonons causes no degradation in the conductivity. It is somewhat surprising that such a smeared out Fermi surface can support a Peierls instability at the high temperature of 210K.

We thank P. Monceau and G. Grüner for mailing manuscripts prior to publication. Helpful discussions with G. Grüner, M. Ido, Pat Lee, P. Monceau, Alan Portis and Meir Weger are acknowledged. This work was supported by NSF grant DMR 70-05418 and Caltech's President's Fund PF-167. Preliminary results of the microwave work were reported at the March Meeting of the American Physical Society at Denver, Colorado, 1981.

REFERENCES

1. N.P. Ong and P. Monceau, Phys. Rev. B16, 3443 (1977); G. Grüner, L.C. Tippie, J. Sanny, W.G. Clark and N.P. Ong, Phys. Rev. Lett. 45, 935 (1980).
2. L.J. Buranov and I.F. Shchegolev, Prib. Tekh. Eksp. 14, No. 2, Pt. 2, 171 (1971) [Instrum. Exp. Tech. (USSR) 14, 528 (1972)].
3. A. Meerschaut, J. Rouxel, P. Haen, P. Monceau, and M. Núñez-Regueiro. J. Phys. Lett. 40, L-157(1979).
4. T. Tsutsumi, T. Sambongi, S. Kagoshima, and T. Ishiguro, J. Phys. Soc. Japan 44, 1735 (1978).
5. J. Rouxel, preceeding paper in this conference.
6. F. Kadijk and F. Jellinek, J. of Less-Common Metals, 19 421 (1969).
7. J. Rijnsdorp and F. Jellinek, J. of Sol. St. Chem. 25,

325 (1978).
8. T. Takoshima, M. Ido, K. Tsutsumi, and T. Sambongi, Solid State Commun. 35, 911 (1980).
9. A.H. Thompson, A. Zettl and G. Grüner, Phys. Rev. Lett. 47, 64 (1981).
10. G. Grüner, A. Zettl, W.G. Clark, and A.H. Thompson, Phys. Rev. B23, 6813 (1981).
11. C. Jackson, A. Zettl, G. Grüner and A.H. Thompson, Solid State Commun. (submitted).
12. P. Monceau, J. Richard and M. Renard, Phys. Rev. Lett. 45, 43 (1980).
13. N.P. Ong, Phys. Rev. B18, 5272 (1978).
14. N.P. Ong, G.X. Tessema, unpublished.