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FAR INFRARED REFLECTANCE AND CONDUCTIVITY OF TaS_3: COMPLETION OF THE AC RESPONSE SPECTRUM

W.N. CREAGER, P.L. RICHARDS, and A. ZETTL
Department of Physics, University of California at Berkeley, and Materials and Chemical Sciences Division of the Lawrence Berkeley Laboratory,
Berkeley, California 94720 U.S.A.

ABSTRACT

The far infrared reflectance of the charge density wave (CDW) conductor TaS_3 has been measured from 3 to 700cm^{-1} with incident radiation polarized parallel and perpendicular to the long axis of the crystals. For parallel polarization, a strong reflection edge correlated with the formation of the CDW appears near 70cm^{-1}. The associated conductivity shows no dramatic conduction mode in the far infrared. These results are in sharp contrast to the observed behavior in the related CDW materials $(TaSe_4)_2I$ and $K_{0.3}MoO_3$, and they rule out models of a "generic far infrared mode" in CDW excitations.

INTRODUCTION

One of the most outstanding features of relatively weakly pinned charge density wave (CDW) conductors is the unusual ac response spectrum. Three distinct excitation modes comprise the spectrum--dielectric relaxation, the pinned acoustic phason, and single particle excitations across the Peierls gap. Recent studies demonstrated that the CDW conductor $(TaSe_4)_2I$ and possibly the blue bronze $K_{0.3}MoO_3$ have in addition a giant conductivity mode in the far infrared (FIR) frequency range between the acoustic phason and the Peierls gap [1-2]. It has been suggested that the additional excitation mode in the FIR may be generic to all CDW conductors and, on this basis, several general CDW conductivity models have been constructed. On the other hand, both $(TaSe_4)_2I$ and $K_{0.3}MoO_3$ are relatively complicated CDW systems (as discussed below), and it is possible that the giant FIR mode arises from peculiarities of the materials. We report on FIR reflectivity measurements of a third CDW conductor TaS_3 which, together with previous microwave and optical measurements, complete the excitation spectrum for this material. Despite a rather complicated FIR reflectance in the CDW state, we show conclusively that no giant FIR mode exists in TaS_3. This rules out models which have predicted that the giant FIR excitation mode would be found in all CDW conductors. Subtle but important differences between the CDW structures of TaS_3 and $(TaSe_4)_2I$ (and $K_{0.3}MoO_3$) allow us to infer the origin of the previously unidentified giant FIR mode.

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EXPERIMENTS AND RESULTS

Fig. 1a shows the measured FIR reflectance of TaS_3 on a logarithmic frequency scale for light polarized parallel to the long crystal axis. The sample is made from several thousand high-purity single crystals of TaS_3, aligned by hand into an opaque mat 1cm in diameter. A trace amount of a plastic resin holds the crystals in place. The dashed line in Fig. 1 is the room temperature reflectance and the solid line is the reflectance in the CDW state well below T_p. The most prominent feature of the low temperature reflectance is the sharp drop from about 75% at $60cm^{-1}$ to about 6% at $76cm^{-1}$. This dramatic reflection edge begins to develop at about 200K, just below T_p, and it sharpens continuously as the temperature is lowered. Fig. 1b shows that the reflectance for transverse polarization is nearly temperature-independent. Neither polarization contains any outstanding feature near $500cm^{-1}$ that might be associated with the $500cm^{-1}$ anomaly [3] previously observed in transverse-polarized bolometric absorption spectra of TaS_3. The remainder of this discussion considers only the parallel polarization.

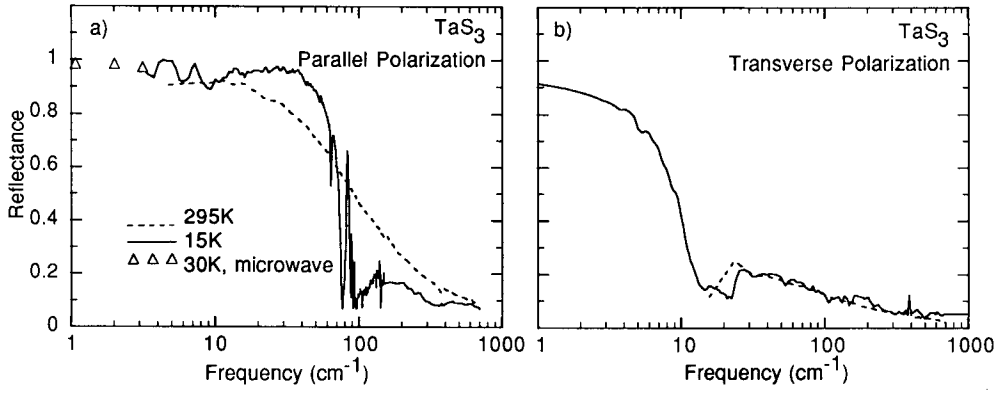

Fig. 1. Reflectance versus frequency for TaS_3 for light polarized parallel to the crystal axis (a) and transverse to the axis (b). The triangles in (a) are reflectances calculated from microwave frequency conductivities measured at 30K in Reference 4.

We generated the complex dielectric function $\varepsilon(w)=\varepsilon 1 + i\varepsilon 2$ shown in Fig. 2 by using a Kramers-Kronig analysis of the measured reflectance along with low-frequency effective reflectance data converted from the previously measured microwave conductivity of TaS_3 [4]. Fig. 2 shows that ε_1 crosses from its large positive value at zero frequency to negative values at about $1cm^{-1}$, indicating the pinned acoustic phason mode near this frequency. At higher frequencies, ε_1 then recovers and slowly climbs to another zero crossing near $70cm^{-1}$ that corresponds to the dramatic infrared reflectance edge. Aside from the dominant phason mode near $1cm^{-1}$, several other identifiable modes modify the gradual rise in ε_1 and affect the location of the zero crossing. The largest of these smaller FIR modes is marked by an arrow. It has an oscillator strength $\Omega_p^2 \leq 6.2 \times 10^{26}$ at $10.6cm^{-1}$ which is a factor of 40 smaller than the oscillator strength of the giant FIR mode in $(TaSe_4)_2I$ at $38cm^{-1}$. We conclude that no giant FIR mode exists in TaS_3. The reflectance edge in TaS_3 is a result of a zero crossing of the real part of the dielectric function, but that zero crossing is a manifestation of the microwave pinned acoustic phason mode rather than any single mode at infrared frequencies.

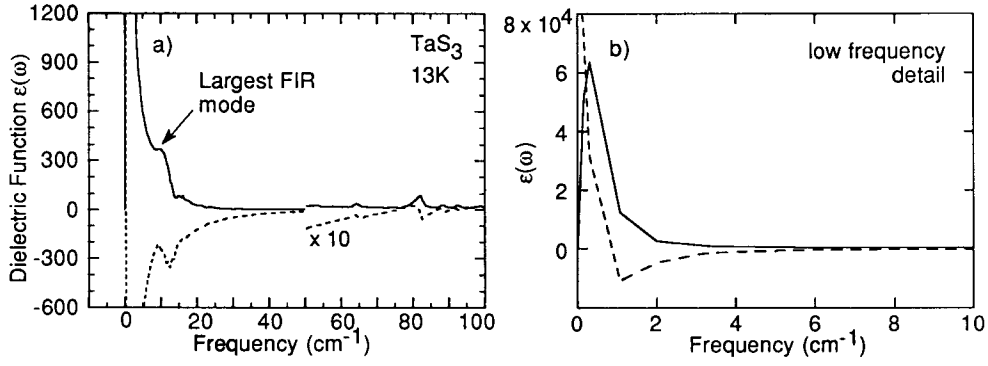

Fig. 2. The real part (dashed line) and imaginary part (solid line) of the dielectric function deduced by a Kramers-Kronig analysis of the 15K reflectivity data for TaS_3. Above $50cm^{-1}$, the scales are expanded by a factor of 10.

In Fig. 3, a comparison is made between the real part of the ac conductivity σ(w) of TaS_3 and that of $(TaSe_4)_2I$ over a wide frequency range. The solid line in Fig. 3a is the FIR conductivity determined from this study; curves at lower and higher frequencies for TaS_3 and all the data for $(TaSe_4)_2I$ have been adapted from data published previously by other groups [1, 3-5]. The intrinsic conductivity modes in each material are identified in the Figure, along with the giant FIR mode found at approximately 10^{12}Hz in $(TaSe_4)_2I$ but absent in TaS_3. The contrast between Figs. 3a and 3b dramatically shows the difference between the excitation spectra of these two similar CDW materials: the giant FIR mode is not a generic feature of CDW conductors. This rules out at least two very different explanations of the giant FIR mode in $(TaSe_4)_2I$. Sherwin et al. [1] considered an "optical phason" argument and Lyons and Tucker [6] have attributed the giant FIR mode to coherent oscillations of the CDW phase in tiny regions surrounding impurities. Each model implies that an infrared conductivity resonance should be found universally in CDW materials. These mechanisms clearly do not generate a large FIR oscillator strength in TaS_3 and are thus unlikely explanations for the giant mode seen in other CDW materials.

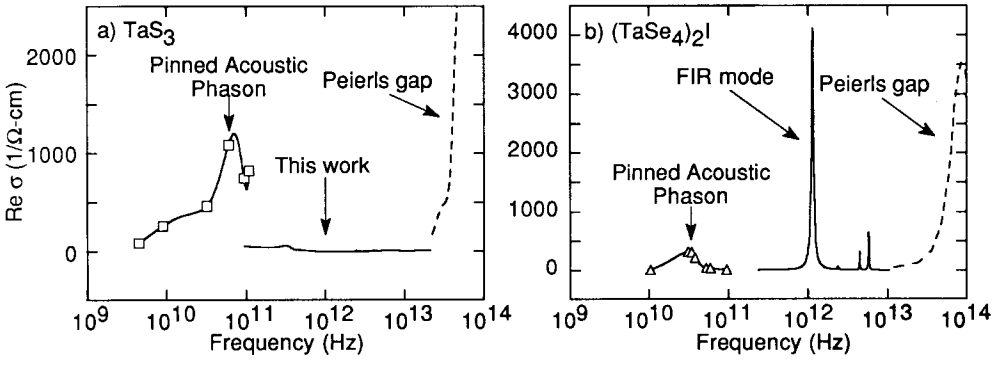

Fig. 3. Real part of the conductivity for (a) TaS_3 and (b) $(TaSe_4)_2I$. Solid line in (a) is this work; other data comes from References 1, 3, 4, and 5.

The absence of a giant resonance in TaS_3 suggests that there are only three intrinsic excitation modes in CDW conductors, and that the giant FIR mode in $(TaSe_4)_2I$ and $K_{0.3}MoO_3$ arises from peculiar properties of those materials. This finding give strong support to a model first considered by Sugai et. al. [7] in which the giant mode arises through zone-folding, which is sensitive to the relation of the Fermi surface to the Brillouin zone edge. Since $2k_F>\pi/a$ in $(TaSe_4)_2I$, the Kohn anomaly mode lies on a transverse optical branch above the transverse acoustic branch in the reduced zone scheme. As the temperature is decreased toward T_p, the TO Kohn anomaly begins to soften and triggers the CDW distortion in the usual way, but the interaction between the TO branch and the lower energy TA branch forces the TA branch to $\omega=0$ first. Thus, the pinned acoustic phason mode seen at microwave frequencies arises from the TA anomoly, and the giant FIR mode from the TO Kohn anomaly. An analogous situation would occur in any CDW material with $2k_F>\pi/a$ (such as $K_{0.3}MoO_3$), but not for TaS_3 which is in a simpler class of materials with $2k_F<\pi/a$. The Kohn anomaly for TaS_3 lies inside its Brillouin zone and generates a microwave mode directly without producing an infrared mode. Our interpretation suggests that a giant FIR excitation mode should be absent in other CDW materials with $2k_F<\pi/a$, such as $NbSe_3$, but may be present in KCP, to which a zone-folding mode generation argument applies.

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REFERENCES

1 M.S. Sherwin, A. Zettl, and P.L. Richards, Phys. Rev. B, 36 (1987) 6708.

2 G. Travaglini and P. Wachter, Phys. Rev. B, 30 (1984) 1971; H.K. Ng, G.A. Thomas, and L.F. Schneemeyer, Phys. Rev. B, 33 (1986) 8755.

3 G. Minton and J.W. Brill, Solid State Comm., 65 (1988) 1069.

4 S. Sridhar, D. Reagor, and G. Grüner, Phys. Rev. B, 34 (1986) 2223.

5 S. L. Herr, G. Minton, and J.W. Brill, Phys. Rev. B, 33 (1986) 8851; Tae Wan Kim, D. Reagor, G. Grüner, K. Maki, and A. Virosztek, Phys. Rev. B, 40 (1989) 5372; H.P. Geserich, G. Scheiber, M. Dürrler, F. Lévy, and P. Monceau, Physica, 143B (1986) 198.

6 W.G. Lyons and J.R. Tucker, Phys. Rev. B, 40 (1989) 1720.

7 S. Sugai, M. Sato, S. Kurihara, Phys. Rev. B, 32, (1985) 6809.