

COMPARATIVE ANALYSIS OF FAR INFRARED TRANSMISSION AND  
REFLECTIVITY SPECTRA IN LAYER CRYSTALS

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**ABSTRACT:** Far infrared transmission and reflectivity spectra are compared for phonon frequency measurements in multimode layer crystals. For a typical layered semiconductor (single crystal GeS) these measurements have been done using polarized light. Four oscillators were observed for E//a and E//b, using either reflectivity or transmission data. The necessary conditions for obtaining transmission data free of spurious interference effects were determined.

#### 1. INTRODUCTION

Germanium sulphide is a typical layered semiconductor compound. It has an orthorhombic structure with lattice parameters:  $a=4.30\text{\AA}$ ;  $b=3.65\text{\AA}$  and  $c=10.44\text{\AA}$  and the space group  $D_{2h}^{16}$ . The technique of preparation of samples has been given elsewhere /1/.

A number of workers have investigated the optical properties of GeS in the far infrared range using only reflectivity measurements. Preliminary results on the reflectivity of GeS in the range between 40 and  $400\text{cm}^{-1}$  were obtained using unpolarized light /2/, /3/, but it was not possible to differentiate between the contributions due to the different crystallographic directions. The recent work of Wiley et al. /4/ consisted of an investigation of the reflectivity of GeS at room temperature in the far infrared using light polarized parallel to each principal axis in turn. The authors identified all but one of the predicted infrared active phonons. The missing  $B_{1u}$  phonon has been observed more recently by Mihajlović et al. /5/, when the reflectivity was measured at 77K.

In this work attention is drawn to an experimental problem associated with far infrared lattice vibrational spectroscopy of layer materials such as GeS. It is also shown under which conditions transmission spectra of germanium sulphide can provide reliable experimental information which can be compared with the reflectivity data.

## 2. EXPERIMENTAL AND DISCUSSION

Transmission measurements in the infrared and far infrared ranges were performed on several samples of various thicknesses between 4.3 and 50  $\mu\text{m}$ . These samples were cleaved perpendicular to the c axis. When the transmission technique is used in systems exhibiting complex phonon spectra it is particularly important that the sample thickness is sufficiently small for the first interference fringe to be beyond the far infrared range (e.g.  $\geq 400\text{cm}^{-1}$ ) so that false conclusions about mode positions are not drawn from convoluted interference/phonon spectra. In Figure 1a and 1b are given polarized (E//a) near infrared (2000-400  $\text{cm}^{-1}$ ) transmission spectra of two single crystal specimens 35.7  $\mu\text{m}$  and 4.3  $\mu\text{m}$  thick respectively.

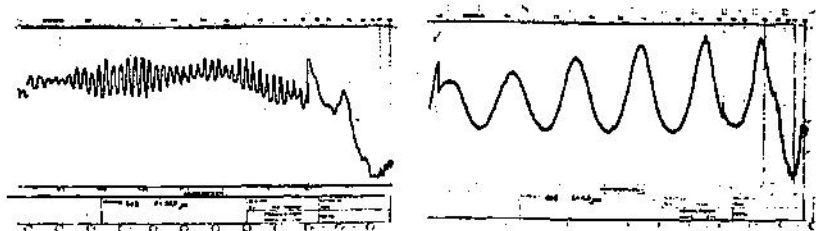


Figure 1a and 1b. Near infrared transmission spectra of 4.3  $\mu\text{m}$  (a) and 35.7  $\mu\text{m}$  (b) thick single crystal specimens of GeS in the range 2000-300  $\text{cm}^{-1}$ .

The thicker specimen displays pronounced interference peaks while the first fringe for the very thin sample occurs at about 420  $\text{cm}^{-1}$ .

Figure 2a displays the data obtained for the thicker sample and E//a in the far infrared range, where it is possible to

see that there is a well exposed interference/phonon interaction at about  $120 \text{ cm}^{-1}$ . This discrepancy does not appear for the same sample in the E//b plane because there is no oscillator in the range between 50 and  $200 \text{ cm}^{-1}$ . This is shown in Figure 2b.

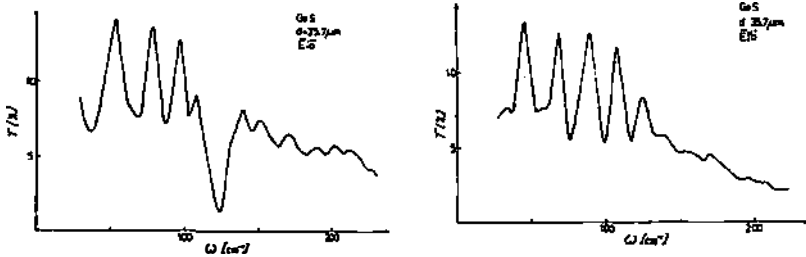


Figure 2a and 2b. Far infrared transmission spectra of  $4.3 \mu\text{m}$  (a) and  $35.7 \mu\text{m}$  (b) thick specimens of GeS in the range  $50\text{--}400 \text{ cm}^{-1}$ .

The reflectivity of GeS in the far infrared measured at room temperature and at 77K is shown in Figure 3. The room temperature curve corresponding to the a direction is consistent with the existence of two TO phonons having frequencies at about  $120$  and  $255 \text{ cm}^{-1}$ . On lowering the temperature, however, a third feature emerges which indicates the existence of a TO phonon with a frequency of about  $315 \text{ cm}^{-1}$ . The room temperature transmission measurements of the thin sample (in the same range) for both E//a and E//b, are given with the broken line in Figure 3.

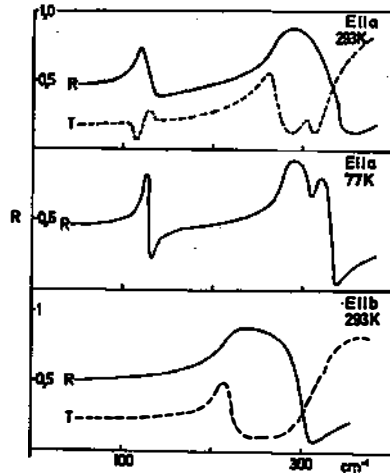


Figure 3. Reflectivity and transmission of GeS in the far infrared

Here one can see that room temperature transmission data have

given as much detail as reflectivity measurements at 77K.

### 3.CONCLUSION

In conclusion we wish to emphasise that the transmission method is a superior means of obtaining maximum information about lattice vibrations in semiconductor systems of the layer type using far infrared spectroscopy. It is necessary to use sufficiently thin optical samples so that interference fringe-phonon interaction do not occur in the observed range.

Although this discussion is confined to the layer semiconductor compound, GeS, similar results have been obtained with other materials with a multimode system (GeSe<sub>2</sub>, GeS<sub>2</sub>) and the same general comments apply /6/.

### 4.REFERENCES

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