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II-2. Far Infrared Lattice Bands in Indium Antimonide

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Spectral emittance measurements were made on high purity single crystal *n*-type indium antimonide from 9.9 to 26.5 meV at 4.2, 32, and 77°K. In addition to the reststrahlen band, emission bands were observed at 10.3, 12.2, 13.75, 15.4, 16.8, 19.7, and 25.4 meV and a broad band from 15.5 to 18 meV. The temperature dependence indicates that the broad band and the 12.2 meV band are phonon difference bands and the rest are phonon summation bands. Using the selection rules for allowed dipole transitions in the zincblende lattice these bands have been assigned to two and three phonon combinations at the *X*, *L*, and *W* points of the Brillouin zone.

The spectral emittance of single crystal *n*-type indium antimonide was measured at several temperatures by the method described in earlier reports.[1-3] A Beckman IR-3 spectrophotometer was extensively modified for this experiment. The prisms were replaced by a small crystal quartz foreprism and an eight line per millimeter grating. The potassium bromide lenses were replaced by one polyethylene window. The detector was a boron doped germanium photoconductor operating at 4.2°K. The instrument covers the spectral range 9.9 to 26.5 meV (80 to 214 cm^{-1}).

The samples were cooled by thermal conduction from a cold finger. Two sample holders were used. For 4.2 and 77°K measurements a dewar with a liquid nitrogen cooled radiation shield was used. Either liquid helium or liquid nitrogen was used in the cold finger.

To study the temperature dependence of the lattice bands other sample temperatures were required so a Cryotip[4] Joule-Thompson expansion refrigerator was obtained. This device may be operated from 20 to 200°K using nitrogen and hydrogen.

The temperature dependence of multiple phonon bands has been discussed by Johnson.[5] For two phonons of energy E_1 and E_2 the temperature dependence of the summation band is given by $(1+F_1)(1+F_2)-F_1F_2$ or $1+F_1+F_2$ while that of the difference band is $(1+F_1)F_2-(1+F_2)F_1$ or F_2-F_1 where $F_n=[\exp(E_n/kT)-1]^{-1}$. Thus, the difference band has a much stronger temperature dependence than the summation band and disappears at very low temperature ($kT \ll E_n$).

The samples were all cut from the same boule of single crystal indium antimonide having a carrier concentration of about 7×10^{13} cc^{-1}. Sample thicknesses were 0.293, 1.23, and 5.66 millimeters. Figure 1 shows emittance spectra of the 1.23 millimeter sample at 4.2, 32, and 77°K. Temperature dependence of the multiple phonon bands can be observed, as well as the reststrahlen band at 23 meV.

Figure 2 shows the spectra of three samples of different thickness at 4.2°K. The weak bands

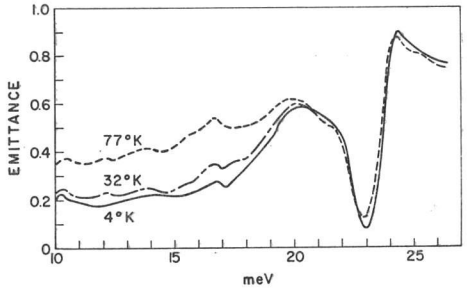

Fig. 1. Spectral emittance of indium antimonide at 4.2, 32, and 77°K. Sample was *n*-type 1.23 millimeters thick having a carrier concentration of 7×10^{13} cc^{-1}.

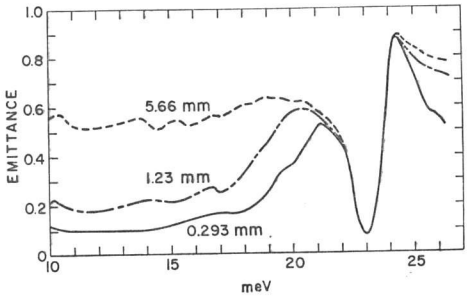

Fig. 2. Spectral emittance of *n*-type indium antimonide at 4.2°K for sample thicknesses of 0.293, 1.23, and 5.66 millimeters. Carrier concentration was 7×10^{13} cc^{-1}.

are more easily observed in the thick sample while the strong bands are more prominent in the thinner samples. Note that in the reststrahlen region, all of the samples are opaque and the emittance is the same for all three. Under this condition $E=1-R$ so that the emittance spectrum is just an inverted reflectance curve. The lower curve of Fig. 2 has been smoothed to eliminate the interference fringes which were observed from 10 to 18 meV.

Emission bands were observed at 10.3, 12.2, 13.75, 15.4, 16.8, 19.7 and 25.4 meV (83, 98, 111, 124, 136, 159, and 205 cm^{-1}). The temperature dependence of these bands indicates that the one at 12.2 meV is a phonon difference band and the rest are phonon summation bands. A broad difference band was also observed from about 15.5 to 18 meV.

Birman[6] has calculated the allowed two and three phonon processes for the zincblende lattice at the Γ, X, L, and W critical points. Using his tables, spectra of indium antimonide from 25 to 50 meV were analyzed and reported.[7] The data presented here have been used to modify that analysis. Table I lists the observed bands and the phonon combinations assigned to them.

Table I

Assignment	Observed $h\nu$	Calc. $h\nu$
2TA(X)	10.3 meV	10.3 meV
TO(Γ)−2TA(X)	12.2	12.3
2TA(L)+TA(X)	13.75	13.55
3TA(X)	15.4	15.45
LO(L)−TA(L)		15.6
TO(X)−TA(X)		16.65
LA(L)+TA(L)	16.8	16.9
TO(L)−TA(L)		17.0
LA(W)+TA(W)	19.7	19.5
2LA(L)	25.4	25.4
TO(L)+TA(L)	25.4	25.4

All of the allowed two phonon difference bands that are within the range of our instruments fall within the 15.5 to 18 meV band mentioned above. Of all the allowed two phonon processes, the only one within the range of our measurements which was definitely not observed was the LO(X) −TA(X) transition which should have been observed at 10.85 meV.

The assignment of three transverse acoustic phonons for the 13.75 and 15.4 meV bands seems justified. Although it is difficult to measure the temperature dependence precisely, these two bands seem to have a stronger temperature dependence than one would expect for a two phonon band. The assignment for the 12.2 meV is less satisfactory. The energy is a reasonable fit with the observed data but the temperature dependence is much less than one would predict for this combination.

The assignments listed in Table I plus those at higher energy lead to the critical point energy values given in Table II.

Table II. Critical point energies for InSb.

	Γ		L		X		W	
	meV	cm^{-1}	meV	cm^{-1}	meV	cm^{-1}	meV	cm^{-1}
LO	24.2	195	19.8	160	16.0	129	16.4	132
TO	22.6	182	21.2	171	21.8	176	22.6	182
LA			12.7	102	15.0	121	13.5	109
TA			4.2	34	5.15	41.5	6.0	49
$\sum E_i^2$	1607		1487		1485		1545	

The phonon energies at the Γ point agree with those derived from analysis of reststrahlen band data.[8]

One of the criteria used in making the assignments was that the sum rule[9] should apply. This rule states that the squares of the phonon energies summed over the branches is a constant. Mitra and Marshall[10] have pointed out that this relation should hold for III-V compounds. This set of phonon energies fits the sum rule within a few percent. The last line of Table II gives the values of these sums.

The W point energies are based on six rather weak bands but the values are about what one would predict, being quite close to the X point values. Although one cannot say that these are the only possible assignments that will fit the data, it is unlikely that a different set would shift the energy values significantly.

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DISCUSSION

Matumura, O.: Usually to measure a thermal radiation spectrum, the temperature of sample should be higher than the temperature of chopper, dispersion system and detector. I would like to know how the situation is in such a low temperature case.

Stierwalt, D. L.: The temperature of the sample may be above or below the temperature of the chopper, dispersion system, and detector as long as there is sufficient difference to give a usable signal. The only difference will be in the polarity of the signal.