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Zone-Boundary Acoustic Phonons in Adamantine Compounds from Far-Infrared Absorption Measurements

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Optical-absorption studies in the far infrared at 4.2 °K have been made on single crystals of cubic ZnS, ZnSe, ZnTe, CdTe, and GaAs. Absorption peaks are seen which correspond to the creation of 2TA(*L*) and 2TA(*X*) phonons, allowed two-phonon processes in the zinc-blende structure. These same processes are forbidden in the diamond, rock-salt, and cesium-chloride structures. The TA(*L*), TA(*X*), LA(*L*), and LA(*X*) phonon energies for 12 different adamantine compounds can readily be correlated with the measured elastic constants by assuming that the phonon-dispersion curves for all the crystals are similar. From this correlation, some predictions are made for acoustic-phonon energies in GaSb, InP, and InAs. This correlation does not work well for either SiC or diamond crystals.

I. INTRODUCTION

Infrared studies of II-VI, III-V, IV-IV compounds and IVth column elements yield information about the phonon energies at critical points in the Brillouin zone. Studies of surface reflectivity or Raman scattering on bulk samples or absorption on thin films give values for the zone-center phonon energies TO(Γ) and LO(Γ). Since we will be concerned with only the cubic crystals, there are at most these two energies at the zone-center point Γ. Many optical-absorption studies at photon energies $\bar{\nu}$ above TO(Γ), i.e.,

$$\bar{\nu} > \mathrm{TO}(\Gamma), \tag{1}$$

show absorption peaks corresponding to the simultaneous generation of two or three or more phonons at various critical points. However, if we consider the photon region

$$\bar{\nu} < \mathrm{TO}(\Gamma), \tag{2}$$

some single- and multiple-phonon-generation peaks can also be present for the lower-energy phonons. Several authors have seen such absorption peaks, and have attributed them to one- and two-phonon-generation processes. Such data exist for ZnSe, CdTe, GaP, and InSb. These results as well as the results of Raman, neutron, and x-ray scattering are collected[1-42] in Table I. The labels TA and LA designate the transverse and longitudinal acoustic phonons at the various critical points[43] *L*, *X*, *W*, Σ. The identification of the zone-boundary critical points involved is made by a comparison of the optical results with those of neutron scattering for the crystals where both types of data are available. For the other crystals, the identification is less certain, and is based on analogies in the shape of the optical-absorption curves and in the values of the elastic constants of these adamantine compounds.

II. PRESENT RESULTS—FAR INFRARED

The optical-absorption curves of five different crystals in the far infrared are given in Fig. 1. All of the samples were single crystals. The CdTe was grown in this laboratory[44] from the melt.[45] The ZnTe and ZnSe were melt-grown crystals purchased from Eagle-Picher,[46] the ZnS was a natural cubic crystal,[47] and the GaAs was a melt-grown crystal from Monsanto.[48] The GaAs crystal was doped with about 10^{17} cm^{-3} of oxygen in order to give it a high electrical resistivity, and hence a very small free-

TABLE I. Far-infrared absorption bands due to acoustic phonons in pure cubic adamantine compounds. The corresponding Raman- and neutron-scattering results are also listed. The phonon energies are in wave numbers (cm^{-1}).

Crystal	Far infrared Present work	 Literature	Raman literature	Neutron literature	Assignment
GaP			151[a]	171[b]	2TA(*L*)
		212[c]	209[a]	214[b]	2TA(*X*)
			285[a]		?
		366[d]	366[a]	366[b]	TO(Γ)
GaAs	161.5			161[e]	2TA(*X*)
		268[f]	270[g]	267[e]	TO(Γ)
InSb		85.5[h]		86[i]	2TA(*X*)
		187[j]	180[k]	186[i]	TO(Γ)
ZnS	148		143[l]	146[m]	2TA(*L*)
	182		182[l]	186[m]	2TA(*X*)
		276[n]	278[l]	271[m]	TO(Γ)
ZnSe		105[o]	114[p]		2TA(*L*)
	141.5	141[o]	146[p]		2TA(*X*)
	176				LA(*L*)
		208[q]	205[p]		TO(Γ)
ZnTe	110		107[r]		2TA(*X*)
	142		142[r]		?
	155.5				LA(*L*)
		181[s]	177[r]		TO(Γ)
CdTe		50[t]	61[u]		2TA(*L*)
	71.3	72[t]	72[u]		2TA(*X*)
	92	94[t]			$2A_2(\Sigma)$
	104.5	104[t]	105[u]		LA(*L*)
		145[t]	140[u]		TO(Γ)

[a] References 1, 5–8.
[b] Reference 9.
[c] Reference 1.
[d] References 1–4.
[e] References 14–16.
[f] References 4, 7, 10, 11.
[g] References 3, 12, 13.
[h] References 17, 18.
[i] No neutron data available. See Refs. 20–22.
[j] References 10, 17.
[k] References 12, 19.
[l] References 24–28.
[m] Reference 29.
[n] References 23, 24.
[o] References 18, 30–32
[p] References 8, 19, 24, 27, 36.
[q] References 24, 30, 31, 33, 35.
[r] References 8, 19, 27, 36.
[s] References 31, 33–35.
[t] References 11, 37–42.
[u] Reference 40.

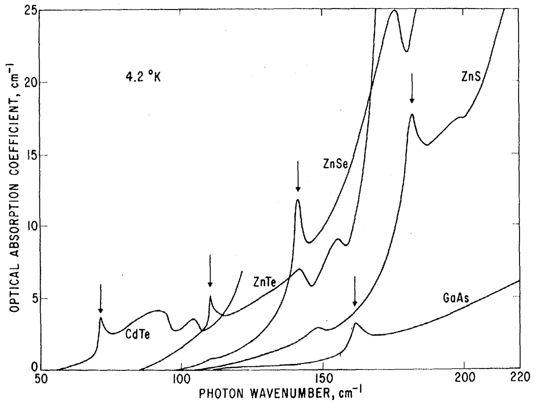

FIG. 1. Optical-absorption coefficient at 4.2 °K vs photon wave number in the far infrared for five different crystals. The vertical arrows designate the peaks associated with 2TA(*X*) phonons.

carrier optical absorption. The absorption curves were measured with an apparatus previously described.[49]

In Fig. 1 all of the crystals have a characteristic peak marked with a vertical arrow. For ZnS[29] and GaAs[14,15] the neutron-scattering results clearly identify this peak as the 2TA(*X*) peak. Such a peak occurs in the two-phonon density of states as calculated for CdTe[41] and GaAs.[50] The 2*TA*(*X*) overtone absorption band is an allowed[16,51] optical transition for all crystals with the zinc-blende structure. It is not allowed[51,52] for diamond-structure crystals such as Si and Ge, even though a peak occurs[50] in the two-phonon density of states. This is in agreement with the far-infrared optical-absorption studies on Ge.[53]

The other two peaks seen in the far-infrared absorption in Table I are associated with the allowed

TABLE II. Impurity-induced one-acoustic-phonon absorption peaks. The phonon energies are given in wave numbers (cm^{-1}).

Crystal	Observed far-infrared peak	Assignment[a]
Si[b]	114	TA(L)
	155	TA(X)
	212	$A_2(\Sigma)$
	330	LA(L)
ZnS[c]	70	TA(L)
ZnSe[c]	49	TA(L)
	70.5	TA(X)
	96	$A_2(\Sigma)$
	176	LA(L)
CdTe[d]	36	TA(X)
	46	$A_2(\Sigma)$
	104.5	LA(L)

[a] A_2 (Σ) designates the next-to-lowest acoustic branch at the Σ point in the Brillouin Zone. It has neither T nor L polarization.

[b] References 52, 55–58.

[c] Reference 32.

[d] References 32, 42.

TABLE III. Acoustic-phonon energies in cubic adamantine compounds derived from infrared absorption, spectral emittance, x-ray scattering, and neutron scattering. The energies are given in wave numbers (cm^{-1}).

Crystal	TA(L)	TA(X)	LA(L)	LA(X)	References
Si	119	155	377	407	a
Ge	64	82	217	232	a
AlSb	56	82	215	...	a
GaP	86	107	210	250	b
GaAs	58	76	207	219	a
InP	87	162	237	231	c
InAs	73	111	148	145	c
InSb	34	42	102	121	d
ZnS	73	93	179	222	e
ZnSe	49	71	176	...	f
ZnTe	39	55	156	...	g
CdTe	25	36	105	...	f

[a] From infrared data, Ref. 20.

[b] From neutron data, Ref. 9.

[c] From spectral-emittance data, Ref. 59.

[d] From spectral-emittance and x-ray data, Refs. 21, 22.

[e] From neutron data, Ref. 29.

[f] From Tables I and II.

[g] From Table I and Ref. 32.

two-phonon band at 2TA(L) and the impurity- or isotope- (see Refs. 42 and 54) induced absorption at LA(L). The assignments of the 2TA(L) band are reasonably certain; the LA(L) bands are questionable. The general agreement of the present phonon energies with those found in the literature is good.

III. IMPURITY-INDUCED ONE-PHONON PEAKS

Optical transitions corresponding to the generation of a single acoustic phonon are allowed in impure crystals.[54] Some data from the literature are collected in Table II for Si,[52, 55–58] NnS,[32] ZnSe,[32] and CdTe.[32, 42] The phonon energies found by this technique are in good agreement with those in Table I and with the neutron-scattering results for Si[20] and ZnS.[29]

IV. ACOUSTIC-PHONON ENERGIES

In Table III we have collected the best experimental values for the acoustic-phonon energies at the X and L critical points of 12 different adamantine compounds. The three previously unmentioned crystals are AlSb,[20] InP,[59] and InAs.[59] These 48 values come from infrared-optical-absorption, neutron-scattering, x-ray-scattering, and spectral-emittance studies. In each column of Table III the energy values cover a range of a factor of about 4. It would be useful to have a means of correlating these values to look for systematic trends. This is done in Sec. V.

V. ELASTIC CONSTANTS

The phonon-dispersion curves of Ge have been carefully studied by neutron-scattering techniques by Brockhouse and Iyengar.[60] The results are shown in Fig. 2 for the [100] direction in the lattice. The LA and TA phonon branches have values at the X critical point of

$$\mathrm{TA}(X) = 81\ \mathrm{cm}^{-1}, \quad \mathrm{LA}(X) = 230\ \mathrm{cm}^{-1}, \tag{3}$$

which are slightly lower than the optical values given in Table III. The slope of the energy vs momentum curves near $q = 0$ in Fig. 2 can be calculated from the second-order elastic constants of Ge.[61] These slopes can be extrapolated to the zone boundary at X to give the "linear-limit" values shown in Fig. 2 of

$$L,\ \mathrm{TA}(X) = 209\ \mathrm{cm}^{-1}, \quad L,\ \mathrm{LA}(X) = 290\ \mathrm{cm}^{-1}. \tag{4}$$

A similar analysis can be made for the [111] direc-

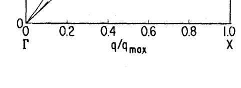

FIG. 2. Dispersion curves in the [100] direction for the two acoustic branches in Ge. The critical points Γ and X in the Brillouin zone are indicated. The points L, TA(X) and L, LA (X) denote the linear-limit extrapolation to the zone boundary at X of the initial slope at Γ.

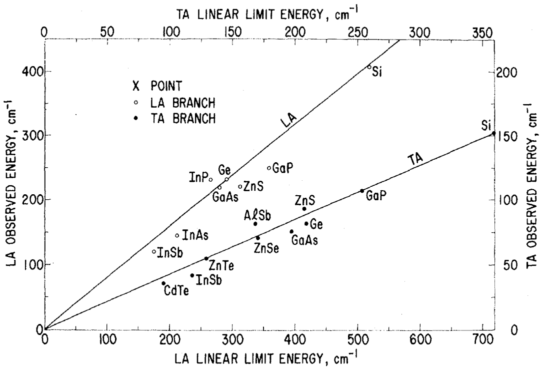

FIG. 3. Observed acoustic-phonon energies at the X point of the [100] direction vs the linear-limit value at the same point. The values for the various crystals are indicated. The solid lines are the "best-fit" lines through the plotted points.

tion in Ge, and for the neutron-scattering results for Si,[20] GaP,[9] GaAs,[20] and ZnS.[29] In Figs. 3 and 4 we have plotted the observed phonon energies vs the linear-limit energies for the TA (L), LA (L), TA(X), and LA(X) phonons for these five crystals. There is a general correlation of the type

$$\mathrm{TA}(L) = [L,\ \mathrm{TA}(L)]\,[S(\mathrm{TA})]\ , \tag{5}$$

where S(TA) is some constant, $0 < S < 1$. Having found this correlation, which shows that the acoustic branches of the phonon-dispersion curves of these five crystals are all similar, we also tried it with the other seven crystals in Table III. The linear-limit energies given in Table IV have been computed from the measured elastic constants[61–69] according to the following formulas:

$$\begin{aligned} &L,\ \mathrm{TA}(X) = (c_{44}/\rho)^{1/2}(ca)^{-1}, \\ &L,\ \mathrm{LA}(X) = (c_{11}/\rho)^{1/2}(ca)^{-1}, \\ &L,\ \mathrm{TA}(L) = [(c_{11}+c_{44}-c_{12})/\rho]^{1/2}(2ca)^{-1}, \\ &L,\ \mathrm{LA}(L) = [(c_{11}+4c_{44}+2c_{12})/\rho]^{1/2}(2ca)^{-1}, \end{aligned} \tag{6}$$

where c_{11}, c_{12}, c_{44} are the elastic constants, ρ is the crystal density, a is the cubic x-ray lattice constant, and c is the velocity of light. The phonon energies are then given in wave numbers. The results of plotting the entries in Table III versus those in Table IV are shown in Figs. 3 and 4.

The solid lines in Figs. 3 and 4 show the best-fit values for the constant S in Eq. (5). These lines allow one to predict the acoustic-phonon energies at X and L from the known elastic constants to an accuracy of

$$\begin{aligned} &\pm 20\%\ \text{ for } L\text{-point phonons}, \\ &\pm 15\%\ \text{ for } X\text{-point phonons}, \end{aligned} \tag{7}$$

by using the empirical values

$$S(\mathrm{TA}) = 0.42, \quad S(\mathrm{LA}) = 0.78. \tag{8}$$

Note that the S(TA) value for the X and L points are essentially equal. This is also true for S(LA).

VI. PREDICTIONS

From these S values, we predict for InP and InAs that

$$\begin{aligned} \text{InP:}\quad &\mathrm{TA}(L) = 50 \pm 10\ \mathrm{cm}^{-1}, \\ &\mathrm{TA}(X) = 75 \pm 10\ \mathrm{cm}^{-1}, \\ \text{InAs:}\quad &\mathrm{TA}(L) = 40 \pm 10\ \mathrm{cm}^{-1}, \\ &\mathrm{TA}(X) = 60 \pm 10\ \mathrm{cm}^{-1}. \end{aligned} \tag{9}$$

The experimental values in Table III derived from spectral-emittance studies[59] are clearly much larger than these. We suggest that the values in Eq. (9) are more nearly correct, and that the spectral-emittance results need some reinterpretation. All the other results in Tables III and IV correlate quite well.

The elastic constants of GaSb have been measured,[70] and we have calculated the following values for GaSb using Eqs. (5), (6), and (8):

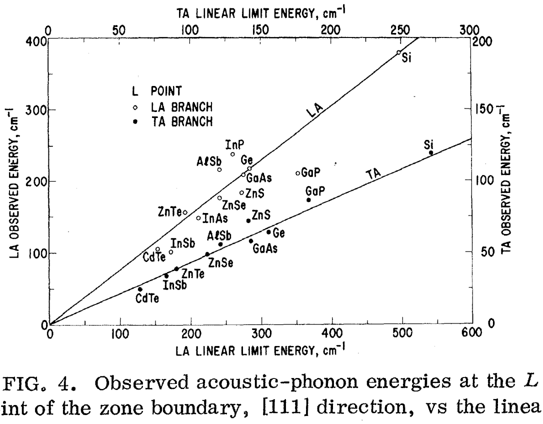

FIG. 4. Observed acoustic-phonon energies at the L point of the zone boundary, [111] direction, vs the linear-limit value at the same point. The values for the various crystals are indicated. The solid lines are the "best-fit" lines through the plotted points.

TABLE IV. Linear-limit values for the acoustic-phonon energies in cubic adamantine crystals. The energies are given in wave numbers (cm^{-1}).

Crystal	L, TA(L)	L, TA(X)	L, LA(L)	L, LA(X)	Reference
Si	271	359	497	518	61
Ge	155	209	284	290	61
AlSb	121	168	241	246	62
GaP	184	253	352	358	63
GaAs	143	197	276	279	64
InP	124	176	260	263	65
InAs	102	145	211	211	66
InSb	83	118	173	175	67
ZnS	141	207	273	312	68
ZnSe	112	170	241	231	68
ZnTe	90	129	192	194	68
CdTe	64	95	153	155	69

$$\begin{aligned} &\mathrm{TA}(L)=46\ \mathrm{cm}^{-1},\ \ \mathrm{TA}(X)=64\ \mathrm{cm}^{-1},\\ &\mathrm{LA}(L)=166\ \mathrm{cm}^{-1},\ \mathrm{LA}(X)=169\ \mathrm{cm}^{-1}. \end{aligned} \tag{10}$$

The knowledge of these phonon energies from other sources is meager. Mitra[71] gives $\mathrm{TA}=49\ \mathrm{cm}^{-1}$, $\mathrm{LA}=134\ \mathrm{cm}^{-1}$, with the critical point unspecified.

The present model does not work well for either diamond[72–76] or SiC.[77] For these two crystals the approximate values of S are

$$S(\mathrm{TA})=0.65\pm0.05,\quad S(\mathrm{LA})=0.70\pm0.05\ . \tag{11}$$

These are not in good agreement with Eq. (8). Thus we have not included these two crystals in our analysis.

VII. ROCK-SALT-STRUCTURE COMPOUNDS

The phonon-dispersion curves have been measured by neutron spectroscopy for a number of compounds with the NaCl (rock salt) structure, and in particular for KBr.[78] The $2\mathrm{TA}(X)$ absorption peak has been reported[18] for KBr at $84\ \mathrm{cm}^{-1}$. This peak has been shown[79] to be a one-phonon peak for which the optical absorption is induced by the presence of trace impurities. The selection rules for two-phonon processes[80,81] in the rock-salt lattice forbid photon absorption for the two processes:

$$\bar{\nu}=2\mathrm{TA}(X),\quad \bar{\nu}=2\mathrm{TA}(L). \tag{12}$$

Thus, the interpretation of Hadni *et al.*[18] for KBr appears to be wrong. The alkali halides do show[80] some multiphonon optical-absorption peaks at photon energies greater than $\mathrm{TO}(\Gamma)$, but none at energies less than $\mathrm{TO}(\Gamma)$. Hence the present type of far-infrared studies at $\bar{\nu}<\mathrm{TO}(\Gamma)$ are not applicable to the rock-salt structure.

VII. CONCLUSIONS

The optical-absorption studies in the far infrared on adamantine compound semiconductors reveal absorption peaks associated with one- and two-phonon-generation processes of acoustic phonons. The phonon energies derived from the analyses of these results agree well with those derived by other techniques. The acoustic-phonon energies at the X and L critical points of the Brillouin zone can be correlated to within ± 20% with measured values of the elastic constants. The agreement is good for Si, Ge, GaP, GaAs, ZnS, AlSb, InSb, ZnSb, ZnSe, ZnTe, and CdTe. The published values for the TA phonons in InP and InAs are believed to be too large, and the new approximate values are given. Some phonon energies for GaSb are calculated.

Note added in proof. New data from Raman and neutron scattering have become available for ZnTe, ZnSe, and InSb.[82–84] The agreement of these new results with Table III is good except for ZnSe, where the new TA phonon energies appear to be too high.

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