

Neutron Diffraction Determination of Mean-square Atomic Displacements in InAs and GaSb

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Abstract

Integrated intensities for Bragg reflection of neutrons from single crystals of the III-V compounds InAs and GaSb have been measured at room temperature. The data were collected at two wavelengths, 0.947 and 1.241 Å, in order to establish the adequacy of a correction for moderate to severe anisotropic extinction. Data were also obtained for InAs at four temperatures from 408 to 933 K. Corrections for thermal diffuse scattering were applied. The results were analysed in the one-particle potential perturbation approximation with terms to fourth order in the atomic displacements $u \equiv (u_1, u_2, u_3)$. At 296 K, the mean-square components $\langle u_i^2 \rangle$ determined were: In, 0.0116(2) Å²; As, 0.0102(1) Å²; and Ga, 0.0120(3) Å²; Sb, 0.0107(3) Å². The third-order coefficients for InAs are comparable with those for Si and Ge, while those for GaSb are comparable with those for zinc chalcogenides. Below 400 K, the mean-square displacements in InAs decrease faster than predicted by the present perturbation approach.

1. Introduction

Indium arsenide and gallium antimonide are an isoelectronic pair of III-V semiconductors. They crystallize into the cubic zinc blende or sphalerite structure (space group $F\bar{4}3m$) in which neither the anion nor the cation sublattice contains a centre of inversion symmetry. Experimental estimates of the mean-square (m.s.) amplitudes of vibrations for the individual ions have varied greatly, particularly for GaSb, as will be seen from Table 3 in Section 5 below.

With the understanding that the analysis of diffraction data from simple, yet acentric, structures can provide evidence of anisotropic anharmonic vibration (see the review by Willis and Pryor 1975), several accurate studies of sphalerite structures have been undertaken. The largest anharmonic effects occur in the copper halides (CuBr, Harada *et al.* 1976; CuCl, Sakata *et al.* 1974 (with neutrons) and Valvoda and Ječný 1978 (with X-rays)) and the zinc chalcogenides (ZnS, Cooper *et al.* 1973, Moss *et al.* 1980; ZnSe, McIntyre *et al.* 1980; ZnTe, Cooper *et al.* 1973). Weak but similar effects have been observed in the related diamond structure elements (Si, Roberto *et al.* 1974; Ge, Mair and Barnea 1975).

The III-V semiconductors as a family fall naturally between the group IV elements and the zinc chalcogenides. The two compounds chosen here are of similar ionicity (0.37 for InAs and 0.29 for GaSb on Phillips's (1970) scale) but differ in their ratio of cation mass to anion mass (1.53 and 0.57 respectively). Comparisons of the m.s. amplitudes of vibration and any observable anharmonic effects therefore shed light on the influence of mass (and the related atomic size).

In this account we present the first determination of the thermal vibrational amplitudes in III-V compounds in which allowance for anharmonic effects has been attempted. For InAs, data for five temperatures from 296 to 933 K have been analysed to provide a complete and consistent picture of third and fourth order anharmonicity. For GaSb, room temperature data collected at two wavelengths have been analysed jointly to ensure that an adequate description of the degree of crystal perfection has been achieved. The experimental results are compared with theoretical values of the atomic Debye-Waller factors computed from current models used to analyse phonon dispersion data.

2. Theory

For cubic structures, in the particular formalism developed by Dawson (1967) the structure factor for a scattering vector $\mathbf{Q} = 2\pi a_0^{-1}(h, k, l)$ may be written

$$F(\mathbf{Q}) = \sum_j b_j(\mathbf{Q}) T_j(\mathbf{Q}) \exp(i\mathbf{Q} \cdot \mathbf{r}_j),$$

where for neutron scattering b_j is the scattering length and $T_j(\mathbf{Q})$ the temperature factor for the atom at position $\mathbf{r}_j$ in the cubic cell having side a_0 . In the zinc blende structure, where each atom occupies a site with $\bar{4}3m$ symmetry, the reflections may be divided into three groups with different structure-factor expressions. If, in the basis of the f.c.c. lattice, the cation (In or Ga) is allocated to the origin site and the anion (As or Sb) to $(\frac{1}{4}, \frac{1}{4}, \frac{1}{4})a_0$, then these structure factors are

$$F(\mathbf{Q}) = 4(b_{\text{In}} T_{\text{In}} + b_{\text{As}} T_{\text{As}}), \quad h+k+l = 4n; \quad (1a)$$

$$= 4(b_{\text{In}} T_{\text{In}} - b_{\text{As}} T_{\text{As}}), \quad = 4n+2; \quad (1b)$$

$$= 4(b_{\text{In}} T_{\text{In}} \pm i b_{\text{As}} T_{\text{As}}), \quad = 4n \pm 1; \quad (1c)$$

where the T_j have real and imaginary parts related to the symmetric and antisymmetric parts of their Fourier transforms, the thermal smearing functions.

For the present case a perturbation expansion about a harmonic one-particle potential (OPP) is suitable. For an atom j at a site with $\bar{4}3m$ symmetry, the expansion to fourth order in the displacement $\mathbf{u}_j(u_1, u_2, u_3)$ is

$$V_j = V_{0j} + \alpha_j \left\{ \frac{1}{2}(u_1^2 + u_2^2 + u_3^2) + \beta'_j u_1 u_2 u_3 + \gamma'_j (u_1^2 + u_2^2 + u_3^2)^2 \right. \\ \left. + \delta'_j (u_1^4 + u_2^4 + u_3^4 - \frac{3}{5}(u_1^2 + u_2^2 + u_3^2)^2) \right\}. \quad (2)$$

The primes on the anharmonic parameters indicate that they differ from the more commonly used $\beta_j \equiv \alpha_j \beta'_j$, $\gamma_j \equiv \alpha_j \gamma'_j$ and $\delta_j \equiv \alpha_j \delta'_j$. The form provides a simpler reciprocal space description and the parameters are constant in the quasi-harmonic approximation.

The real and imaginary parts T_{cj} and T_{aj} from this potential are (Mair 1980)

$$T_{cj}(\mathbf{Q}) = \cos(\beta'_j \langle u_{sj}^2 \rangle^2 (2\pi/a_0)^3 hkl) \exp \left\{ \left(\frac{1}{2} \langle u_{sj}^2 \rangle - \langle u_{sj}^2 \rangle^2 (10\gamma'_j - \frac{1}{2}\beta_j'^2) \right) |\mathbf{Q}|^2 \right. \\ \left. + \langle u_{sj}^2 \rangle^3 (\gamma'_j |\mathbf{Q}|^4 + \frac{2}{5}\delta'_j f_4(\mathbf{Q}) - \frac{1}{2}\beta_j'^2 g_4(\mathbf{Q})) \right\}, \quad (3a)$$

$$T_{aj}(\mathbf{Q}) = \tan(\beta'_j \langle u_{sj}^2 \rangle^2 (2\pi/a_0)^3 hkl) T_{cj}(\mathbf{Q}); \quad (3b)$$

where the m.s. displacements $\langle u_{sj}^2 \rangle$ are equal to kT/α_j in the classical regime. The terms

$$g_4(Q) = (2\pi/a_0)^4(h^2k^2 + k^2l^2 + l^2h^2), \quad f_4(Q) = (2\pi/a_0)^4(h^4 + k^4 + l^4) - 3g_4(Q)$$

describe the anisotropic part of the quartic anharmonicity. For these temperature factors, the leading anharmonic contribution occurs in the structure-factor equation (1c) via the temperature equation (3b). The product hkl varies widely for the odd-ordered reflections leading to detectable deviations from harmonic behaviour if β' is sufficiently large. In the present experiments we have

$$\beta'_j \langle u_{sj}^2 \rangle^2 \ll \{(2\pi/a_0)^3 hkl_{\max}\}^{-1} \approx (\frac{1}{3} |Q_{\max}|)^{-3},$$

so only the difference $\beta'_1 \langle u_{s1}^2 \rangle^2 - \beta'_2 \langle u_{s2}^2 \rangle^2$ may be refined (Cooper *et al.* 1973).

Matsubara (1975) has discussed the problem of bounding the integration of the potential obtained by equation (2) when the quartic terms are ignored and suggests that the parameters $\beta'_j \langle u_{sj}^2 \rangle^2$ increase and α_j decrease rapidly at high temperatures. For the present approach no such increase is predicted. For III-V compounds, m.s. displacements are moderate and the relative merits of the two theories are not tested. Recent measurements on ZnS (Moss *et al.* 1980) favour the present approach.

The observed integrated intensity I_0 for a reflection at Q is related to the structure factor by

$$I_0(Q) = (J/\omega)(V_0 \lambda^3/a_0^6) A \operatorname{cosec} 2\theta f(y, \alpha) |F(Q)|^2, \quad (4)$$

where the beam flux J , the angular scan rate ω , the sample volume V_0 , the neutron wavelength λ and the lattice constant are normally incorporated in a scale factor K . The Lorentz factor $\operatorname{cosec} 2\theta$ and the absorption A are calculated for each reflection together with the mean path length for neutrons in the crystal used to calculate the extinction factor y .

The manner in which y and the contribution of thermal diffuse scattering (TDS) to the integrated intensity (represented by α , the fraction of TDS included in integration of an unextinguished reflection) should enter equation (4) is not immediately clear. Mair *et al.* (1974) assumed that the TDS (which is weak and incoherent and hence described by kinematic theory) originates at the incident surface of the crystal, i.e.

$$f(y, \alpha) = y + \alpha. \quad (5)$$

On the other hand, Moss *et al.* (1980) used

$$f(y, \alpha) = y(1 + \alpha), \quad (6)$$

which assumes that all TDS is generated at the exit surface of the crystal. The actual situation lies between these simplifications. The total TDS is the incoherent sum of phonon scattering events throughout the crystal, the probability of each event being proportional to the local intensity of the beam in direction Q . Thus we expect the actual situation to lie between the extremes

$$f(y, \alpha) = y + \xi\alpha, \quad \text{where } 1 \geq \xi \geq y. \quad (7)$$

In the extreme situation of high temperature and high extinction, uncertainty is introduced by a lack of understanding of the interaction between diffuse scattering and dynamical effects. We have used equation (6), this being simpler computationally. For the levels of extinction in our InAs data the difference is not significant. For the GaSb data we take up this point in the discussion in Section 5 below.

Table 1. Crystallographic data

Compound (sample)	V (cm ³)	T (K)	a_0 (Å)	λ (Å)	μ (cm ⁻¹)	$\sin \theta_{\max}/\lambda$ (Å ⁻¹)	Reflections		
							Total	Independent	
InAs (I-5)	0.127	296(2)	6.05834 ^A	0.855	1.74(4)	1.04	393	71	
InAs (I-2)	0.036	296(2)	6.05838 ^A	1.241	2.53(6) ^B	0.660	50	16	
				0.947	1.93(5) ^B	0.973	74	30	
			408(5)	n.d.	0.947	1.93(5) ^B	0.973	137	32
			543(10)	n.d.	0.947	1.93(5) ^B	0.973	88	32
			813(15)	6.071(5)	0.947	1.93(5) ^B	0.960	60	28
			933(15)	6.080(5)	0.947	1.93(5) ^B	0.822	38	17
GaSb (G-1)	0.048	296(2)	6.09593 ^A	1.241	0.044 ^C	0.656	197	25	
				0.947	0.033	0.928	176	55	

^A From Casey and Trumbone (1970).

^B Calculated from μ measured at $\lambda = 0.855$ Å (sample I-5). ^C From Bacon (1975).

3. Experiment

InAs

In an initial experiment, 71 independent reflections plus equivalents out to $\sin \theta/\lambda = 1.04$ were collected from a $5 \times 5 \times 5$ mm cube (sample I-5) cut from a single crystal of InAs mounted on the four-circle automatic diffractometer at the 6HA beam from the Australian Atomic Energy Commission's HIFAR reactor. The same crystal was used to determine the absorption coefficient μ from transmission measurements with the beam finely collimated: a value of $\mu = 1.74(4)$ cm⁻¹ at $\lambda = 0.855$ Å was obtained (cf. 1.69 cm⁻¹ calculated from cross section data; Hughes 1965).

A flux determination by gold foil irradiation revealed that low-angle reflections were severely and anisotropically extinguished. Subsequent analysis indicated that both beam inhomogeneity and perhaps multiple diffraction were also contributing to the observed variance in low-angle reflections. Large beam divergences precluded experimental investigation of the presence of multiple diffraction.

The above uncertainties led to a new measurement being undertaken with greatly reduced divergences and a spherical sample. The extinction appeared to be as severe as previously until a broken seal led to surface oxidation at 995 K. After grinding the oxidized layer from the specimen (now designated I-2, radius 2.05 mm) integrated intensities were measured at room temperature for a range of reflections out to $\sin \theta/\lambda = 0.97$ for $\lambda = 0.947$ Å. Similar ranges were covered in data collection at 408, 543, 813 and 933 K (Table 1). The collection was halted at the last temperature when the intensity of the standard reflection began to diminish. The variances in these intensities were consistent with Poisson statistics, indicating that many of the instabilities had been reduced, as had the level of extinction.

In order to test the adequacy of our extinction correction procedure, a further room temperature data set was collected for $\lambda = 1.241$ Å on a diffractometer operating at the 2TanB beam at HIFAR.

GaSb

Integrated intensities were collected for 55 independent reflections from a ground sphere of GaSb (sample G-1, radius 2.25 mm) at 0.947 Å. The extinction was again severe and anisotropic so a second data set was collected at 1.241 Å. The details of these measurements are summarized in Table 1. Between three and five equivalents were collected for each independent reflection in order to establish the nature and extent of the anisotropy in the crystal texture that was responsible for the extinction.

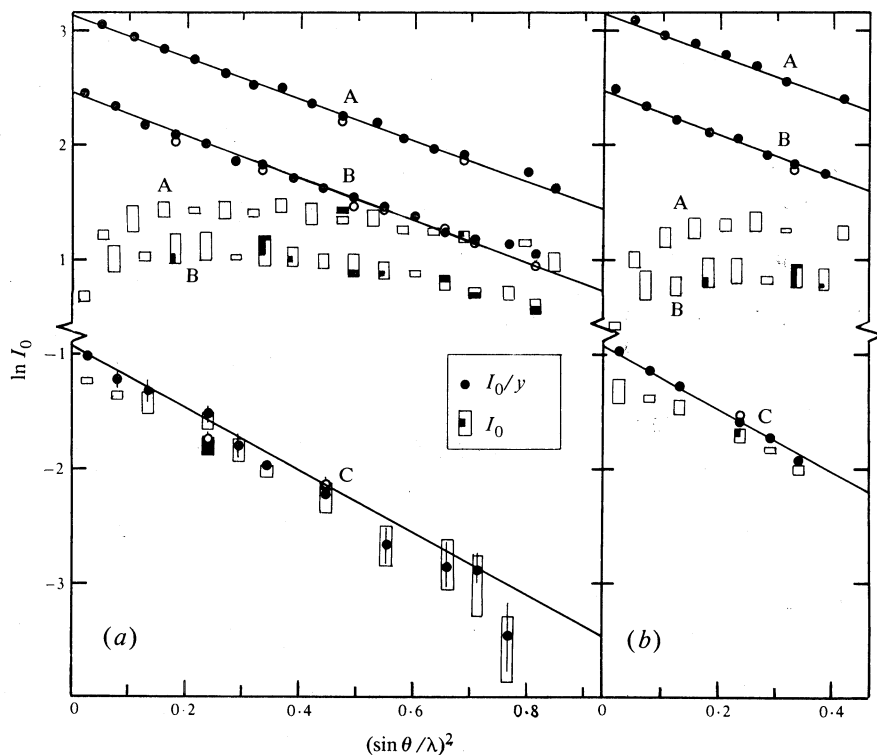


Fig. 1. Observed intensities I_0 versus $(\sin \theta / \lambda)^2$ for GaSb at wavelengths λ of (a) 0.947 Å and (b) 1.241 Å, together with the values of I_0/y after correction for extinction. The data are for reflections with (A) $h+k+l = 4n$, (B) $4n \pm 1$ and (C) $4n + 2$. The vertical dimension represents the spread of harmonically equivalent reflections. The filled areas represent the intensities for reflections with a second, higher value of the product hkl occurring at the same value of $\sin \theta / \lambda$. The oblique lines represent the best fit to the data. Note the logarithmic intensity scale.

4. Analysis

Each set of intensities was corrected for the Lorentz effect and absorption and was scaled approximately from estimates of the beam flux. The TDS correction was based on the routine of Merisalo and Kurittu (1977) which sums phonon modes directly and thus allows those phonon modes not excited by the neutrons to be excluded from the sum. Elastic constants for InAs were taken from Burenkov *et al.*, Nikanorov (1976) and for GaSb from Lin and Wong (1972).

Least-squares analysis of the data sets described in Table 1 was undertaken with a version of the LINEX program (Becker and Coppens 1974) that had been modified

Table 2. Refined parameters and refinement statistics for InAs and GaSb

$wR(F^2)$ is the weighted R factor minimized in the refinements, while the goodness of fit (GOF) is the r.m.s. weighted residual per degree of freedom (a value near unity indicates that little further improvement in the R factor can be expected); the other parameters are defined in Section 2. Estimated standard deviations are quoted in parentheses, the absolute error in temperature being given in square brackets. The parameters for GaSb were obtained by joint refinement of data for $\lambda = 0.947$ and $1.241 \text{ \AA}$ in which the scale factor $K(0.947)$ was set identically to unity

Temp. T (K)	K	$\langle u_s^2(\text{III}) \rangle$, $\langle u_s^2(\text{V}) \rangle$ ($\text{\AA}^2$)	β'_{eff} ($\text{\AA}^{-1}$)	$\delta'(\text{III})$, $\delta'(\text{V})$ ($\text{\AA}^{-2}$)	$b(\text{III})^A$ (fm)	g^B	$wR(F^2)$ (%)	GOF
(a) InAs								
296[2]	0.904(7)	0.0116(2), 0.0102(1)	0.3(4)	$-0.4(2.6)$, $1.5(1.8)$	4.00(4)	0.24(2)	2.45	1.10
408[5]	0.918(5)	0.0175(1), 0.0145(1)	$-0.6(2)$	$-0.4(6)$, $0.4(4)$	4.03(3)	0.26(2)	2.75	1.12
543[10]	0.897(6)	0.0238(2), 0.0194(1)	$-0.5(3)$	$-0.1(4)$, $0.4(3)$	4.00(3)	0.23(2)	2.18	1.18
813[15]	0.88(1)	0.0361(5), 0.0296(2)	0.3(3)	$-0.4(5)$, $-0.1(3)$	3.98(5)	0.31(4)	4.36	1.50
933[15]	0.83(1)	0.0404(5), 0.0335(3)	$-0.7(3)$	$-0.4(3)$, $-0.6(3)$	3.94(4)	0.26(3)	1.89	1.05
(b) GaSb								
296[2]	1.52(1) ^C	0.0120(1) ^D , 0.0107(1) ^D	$-1.7(9)$	$3.5(1.2)$, $4.2(2.1)$	7.21(5)	0.017(1), 0.024(1), 0.025(6), 0.001(1), 0.002(1), 0.011(1)	3.53	2.02

^A With $b(\text{V})$ taken as $6.60, 5.65 \text{ fm}$ for As, Sb (Koester *et al.* 1980).
^B g is the extinction parameter; values of the anisotropy g_{ij} are given for GaSb.
^C $K(1.241)$ for $K(0.947) = 1$; standard deviation includes measured deviation in $K(0.947)$ of 0.03 .
^D Standard deviations are increased to $3 \times 10^{-4} \text{ \AA}^2$ by the uncertainty in TDS (see Section 5).

to allow the anharmonic coefficients β'_j , γ'_j and δ'_j (equation 3) to be refined and, if necessary, data obtained at two wavelengths to be refined simultaneously. Correlation between $\langle u_{sj}^2 \rangle$ and the γ'_j precluded direct determination of the latter in any data set, so they were set to zero in the refinement procedure.

Refinement of the data set from sample I-5 yielded values of $\langle u_s^2(\text{In}) \rangle = 0.0113(5)$ and $\langle u_s^2(\text{As}) \rangle = 0.0099(4)$ but the large variances in many reflections prevented reliable determination of the other parameters.

For sample I-2 an isotropic correction for extinction proved sufficient at all temperatures. The scattering length b of In was refined because it was less precisely known than the other values that had been determined by the Christiansen filter technique.* Furthermore, for temperatures above 600 K the possibility of oxidation and sublimation of either element could be monitored.

* We thank Dr L. Koester for communicating, prior to publication, values of b for As, Sb and In. The average value we obtain for $b(\text{In}) = 4.01(4) \text{ fm}$ should be compared with his result $4.065(20) \text{ fm}$ (Koester and Knopf 1980).

The refinement of the GaSb measurements proved more difficult because the severe extinction ($y_{\min} \approx 0.15$) led to high correlation between the scale factor, the m.s. displacements and the extinction coefficients. In order to establish the extinction parameters, and hence the m.s. displacements with greater confidence, the data sets for the two wavelengths were refined jointly. The anisotropy in the extinction was particularly marked but consistently so in both sets of measurements. The model incorporating Type I (angular spread) mosaic with a Lorentzian distribution and the Thornley and Nemes (1974) description of anisotropy was clearly the most successful. The success of the model can be ascertained from Fig. 1 where the spread of the uncorrected intensities I_0 and the corrected values I_0/y are compared for both wavelengths.

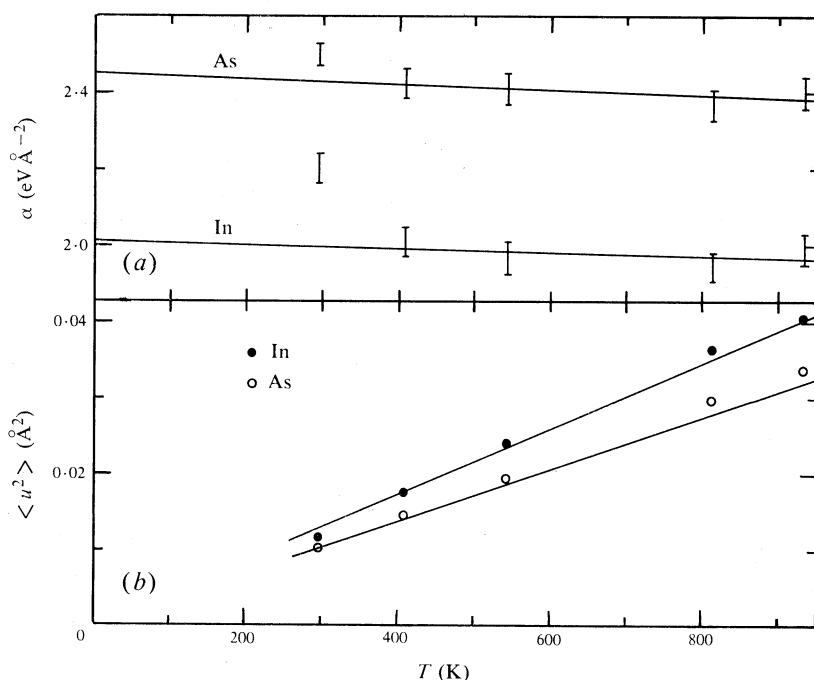


Fig. 2. Variation with temperature of (a) the harmonic one-particle potential parameters $\alpha(\text{In})$ and $\alpha(\text{As})$ and (b) the m.s. displacements $\langle u^2 \rangle$ for In and As. In (a) the slope of the line of best fit drawn through the high temperature points is used to estimate the fourth order anharmonic parameters, while the intercepts at $T = 0$ determine the harmonic model. The lines in (b) represent the harmonic model deduced from the intercept in (a).

A high correlation between the scale factors and the scattering length of Ga was overcome by refinements with $K(0.947 \text{ \AA})$ held at its experimental value of 1.00 and at 0.97 which represented a shift of one standard deviation. Only $K(1.241 \text{ \AA})$ and $b(\text{Ga})$ were affected and the standard deviations quoted for each of these in Table 2 include the added uncertainty.

5. Discussion

The m.s. displacements are well determined for all data sets with refined standard deviations of order 1%. The data from the original InAs measurement differed by

less than two standard deviations from the later value despite the very different texture and thermal history of the samples. Uncertainties in the experimental temperatures and partial correlation with the scale factors lead us to assign uncertainties of 2% to the values of the harmonic potential coefficients α_j . These are given graphically for InAs in Fig. 2a, together with a line fitted to the points for 408 K and above to assist in the estimation of the fourth order anharmonic parameter. The room temperature values are not included because they seem to represent the 20% decrease in the Debye temperature observed by Shumskii *et al.* (1972) in powder data. A perturbation OPP description cannot account for such behaviour. The existence of an anomaly is the only similarity with the powder result, since we found that $\langle u_s^2(\text{In}) \rangle$ was 15% greater than $\langle u_s^2(\text{As}) \rangle$ at all temperatures. A larger vibrational amplitude for the cation is a common feature of zinc blende structures, reflecting the smaller size of the ionized core of the cation and manifesting itself in the extreme case of the copper ion in the superionic copper halides (Sakata *et al.* 1974).

The anharmonic parameters of InAs are barely significant. They are the smallest values yet observed for a zinc blende structure and are comparable with Si and Ge for which $\beta' \approx 0.3$ (Mair and Barnea 1975). Negative values of δ_j' represent a tendency for an atom to vibrate along the 100 directions, superimposed in this case on the preference (indicated by the negative value of β'_{eff}) for both atoms to move slightly apart, towards the octahedral holes in the structure. The magnitude of the isotropic fourth order parameter may be estimated from the temperature coefficient of α_j :

$$\alpha_j^{-1} \partial \alpha_j / \partial T = -2\chi\gamma_G + 10k\alpha_j^{-1} \gamma_j'.$$

From Fig. 2a the coefficient is approximately $-0.3 \times 10^{-4} \text{ K}^{-1}$ which is to be compared with the contribution $-2\chi\gamma_G$ due to the volume expansion of the lattice, where γ_G is the Grüneisen coefficient and χ the volume coefficient of expansion. From specific heat and compressibility data we obtain $-0.20 \times 10^{-4} \text{ K}^{-1}$ while from thermal conductivity and thermal expansion (Steigmeyer and Kudman 1963) we get $-0.13 \times 10^{-4} \text{ K}^{-1}$. In spite of the uncertain differences, values of γ_j' about $-0.2 \text{ \AA}^{-2}$ are indicated. This is similar to the anisotropic quartic component and indicates a slight softening of the OPP at larger amplitudes.

The m.s. displacements for GaSb quoted in Table 2 do not include all sources of uncertainty. The TDS correction α at high angle was 3% while y for the corresponding $4n$ reflection was 0.54. From equations (5) and (6) we obtain 0.57 and 0.556 respectively for $f(y, \alpha)$. While some of the 3% difference between these values may be absorbed into the refined value of y , the added uncertainty is included in the error in the m.s. displacement and becomes in fact the dominating uncertainty.

The GaSb results, although giving similar values for the m.s. displacements of the anion and cation, differ markedly from the foregoing in having larger values of the anharmonic terms. We note that the values are only slightly more significant, the larger errors being presumably due to our failure to take account of multiple diffraction which is more troublesome in crystals with severe extinction (Prager 1971). The magnitude of $\beta'_{\text{eff}} = 2$, which is comparable with the values for the zinc chalcogenides (McIntyre *et al.* 1980), reflects the small size of the cation with respect to the anion. On this evidence, the short-range steric considerations are more important than the long-range Coulombic effects in determining the anisotropic perturbation to harmonic behaviour in the zinc blende and diamond structures. In InAs, where the ion cores

are of approximately equal size, the anharmonic effects are similar to the diamond structure compounds while in GaSb, despite its lower ionicity (Phillips 1970), the smaller cation leads to behaviour similar to the II–VI compounds.

Theoretical calculations of Debye–Waller factors for zinc blende compounds have not had conspicuous success in predicting experimental values (Vetelino *et al.* 1972; Talwar and Agrawal 1974). This may be traced to the absence from the dynamical matrices of elements reflecting differences in the interactions between next-nearest neighbour ions, i.e. the In–In and As–As coupling parameters are usually set equal.

Table 3. Comparison of experimental and theoretical Debye–Waller factors at room temperature for InAs and GaSb

The Debye–Waller factor $B = 8\pi^2\langle u_s^2 \rangle$

$B(\text{In})$ ($\text{\AA}^2$)	$B(\text{As})$ ($\text{\AA}^2$)	$B(\text{Ga})$ ($\text{\AA}^2$)	$B(\text{Sb})$ ($\text{\AA}^2$)	Source
0.92(1)	0.803(8)	0.95(2)	0.85(2)	Present experiment
—	—	1.27	0.57	RIM
0.92	0.63	0.91	0.78	OVSM
—	—	1.12	0.75	SM
1.00	1.11	1.02	0.76	Shumskii <i>et al.</i> (1972)
1.0	1.0	1.48	1.24	Sirota (1969)
—	—	0.74	0.43	Orlova (1977)
0.99	0.99	0.67	0.67	Vetelino <i>et al.</i> (1972)
—	—	0.91	0.91	Talwar and Agrawal (1974)

Table 4. Comparison of Debye characteristic temperatures for InAs and GaSb

$\Theta(\text{In})$ (K)	$\Theta(\text{As})$ (K)	$\Theta(\text{Ga})$ (K)	$\Theta(\text{Sb})$ (K)	Source ^A
180	238	228	182	Present experiment
202		238		Elastic constants (a)
270		230		Specific heat (b)
240		235		Diffraction (c)

^A Values from: a, present work; b, Piesbergen (1966); c, Arnold and Nereson (1963).

For shell model (Dolling and Waugh 1965), deformable bond approximation (Kunc 1973; Kunc *et al.* 1975) and overlap valence shell model (Kunc and Bilz 1976) parameter sets this is not so. Indeed, Harada *et al.* (1976) used measurements of Debye–Waller factors to distinguish shell models which were otherwise indistinguishable. The recent availability of routines for evaluating eigenvectors and eigenvalues for these models (Kunc and Nielsen 1979*a*, 1979*b*) has allowed the evaluation of the Debye–Waller factors for published sets of parameters for III–V compounds (F. K. Larsen and O. H. Nielsen, personal communication). The results of these calculations for InAs and GaSb are included with the present results and earlier data in Table 3. The agreement with the OVSM values for GaSb adds confidence to our use of the two-wavelength procedure in treating extinction effects in our data.

For InAs the agreement is less satisfactory. The OVSM parameters (Borchards and Kunc 1978) were obtained by interpolation between model parameters for other III-V compounds. The present data, specific heat data (Piesbergen 1966) and, as already noted above, powder diffraction data (Shumskii *et al.* 1972) all exhibit similar behaviour, in that the characteristic temperature falls 20% between 200 and 400 K. In these circumstances simple interpolation between the phonon-spectral properties of InSb and InP is unlikely to provide an adequate description of the lattice dynamics of InAs.

In Table 4, room temperature values for the Debye characteristic temperature, obtained from the present experimental data and from the elastic constants used to calculate the TDS correction (Hewat 1972), are compared with values from specific heat measurements (Piesbergen 1966) and the original scattering length determination (Arnold and Nereson 1963).

6. Conclusions

We have measured neutron intensities from InAs and GaSb at room temperature and, for InAs, at four higher temperatures up to 933 K. The data were corrected for thermal diffuse scattering. Least-squares refinements determine the mean-square, third and fourth order anharmonic displacement parameters together with extinction corrections for each measurement. Data taken at two wavelengths and refined simultaneously improved the anisotropic extinction correction for GaSb. The major conclusions concerning the lattice dynamics of zinc blende structures are:

- (1) The vibration amplitude of the cation is greater than that of the anion in both compounds, as it is for all other zinc blende compounds for which single crystal data exist.
- (2) The close agreement between the present results for GaSb and recent calculations based on shell model parameters fitted to phonon dispersion data gives confidence that the extreme extinction evident in our data has been adequately described by the theory of Becker and Coppens (1974).
- (3) Between 200 and 400 K, the mean-square displacements for In and As increase rapidly, corresponding to a decrease in $\alpha(\text{In})$ and $\alpha(\text{As})$ which can only be described in the one-particle potential approach by introducing an explicit temperature dependence in these coefficients. The lack of agreement between calculated and measured mean-square displacements in this case suggests that the interpolation to obtain model parameters for InAs from models for other III-V compounds has failed to account for this behaviour.
- (4) Above 400 K, anisotropic effects in InAs are similar in magnitude to those in Si and Ge. For GaSb, the effects at 296 K are similar to those in the zinc chalcogenides. The ranking supports the importance of the steric considerations of ion size in controlling anharmonic effects in the zinc blende lattice.

A source of uncertainty arose from the application of simple thermal diffuse scattering corrections in the presence of severe extinction. The contribution of diffuse scattering to the measured intensity of Bragg peaks in the presence of dynamical diffraction requires closer study.

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