

PIEZOBIREFRINGENCE IN GaP AND InP

F. Canal,* M. Grimsditch and M. Cardona

Max-Planck-Institut für Festkörperforschung, 7 Stuttgart 80, West Germany

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We report measurements of piezobirefringence in GaP and InP in the region below the direct gap. For GaP this includes the region above the indirect gap where the sample is opaque. The experimental data are compared with theoretical calculations of the contributions to the piezobirefringence arising from the E_0 , E_1 and E_2 transitions.

1. INTRODUCTION

THE INVESTIGATION of stress induced birefringence in solids is an old topic of crystal optics [1]. Only relatively recently, however, has a systematic investigation of the dispersion of the piezooptical constants of a number of materials been undertaken [2]. Prominent among these materials are the zinc-blende and diamond-like semiconductors. Due to the rather detailed understanding of the microscopic mechanisms underlying their optical properties it is possible to obtain for these materials theoretical expressions for the piezooptical constants and their dispersion. Data in the region of transparency have been presented for Ge, Si [3], AlSb [4], GaAs [3], GaSb, InAs, InSb [5], ZnS, ZnSe, ZnTe, CdTe [6], CuCl, CuBr, and CuI [7]. More recently a technique has been developed to investigate the piezo-optical constants *above* the absorption edge and applied to silicon [8]. In this paper we complete existing data by reporting the dispersion of the birefringence induced by a [111] and a [100] stress for InP, a direct gap material, in the region of transparency and for GaP, an indirect gap material, all the way up to the lowest direct gap.

From these and previous measurements the following picture emerges: the piezooptical constants in indirect gap materials (Si, GaP, AlSb) are not strongly dispersive as this gap is approached. They show, however, a strong dispersion in all materials of the family as the lowest direct gap is approached. The overall dispersion observed can be synthesized as the sum of three dispersion mechanisms. The $\Gamma_{15} \rightarrow \Gamma_1$ gap (E_0), the $\Lambda_3 \rightarrow \Lambda_1$ gap (E_1) and a gap at higher energies (E_2) which corresponds to the maximum of ϵ_2 in the material. The E_0 contributions have the same sign for [100] and for [111] stress and opposite sign to the sum of the E_1 and E_2 contributions. The E_1 and E_0

contributions have the same sign for [100] stress and opposite sign for [111] stress. The E_0 and E_1 contributions can be expressed analytically in terms of deformation potentials which are in semi-quantitative agreement with those found with other methods.

2. EXPERIMENTAL

The experimental method used to determine the piezooptical constants in the region where the sample is transparent has been described in the literature [9]. We also used a new technique that extends measurements into the region where the sample is opaque. This technique has been sufficiently described [8] and the details will not be repeated here. The measurements were made at room temperature using the stress apparatus described in [10].

In the transparent region of the samples the combinations of piezo-optical constants ($\pi_{11} - \pi_{12}$) and π_{44} were determined by stressing single crystal parallelepipeds along the [100] and [111] directions, respectively.

In the region where the samples are opaque we used the scattering geometries labelled (a) and (b) in [8] to determine the combinations π_{44} and ($\pi_{11} - \pi_{12}$), respectively. The experimentally determined depolarization of the Raman line as given by the ratio of intensities of the non-allowed to allowed polarizations (β) was fitted to an expression of the form:

$$\beta = \frac{1}{1 + \frac{1}{2(A\bar{X})^2}} + C, \quad (1)$$

where X is the applied stress and

$$A = \frac{\pi n^3 \pi_{ij}}{\lambda(\alpha_i + \alpha_j)}. \quad (2)$$

* Present address: Escuela Técnica Superior de Ingenieros de Telecomunicación, Barcelona, Spain.

In equation (2), n and λ are the average refractive index and the wavelength for the incident and scattered light,

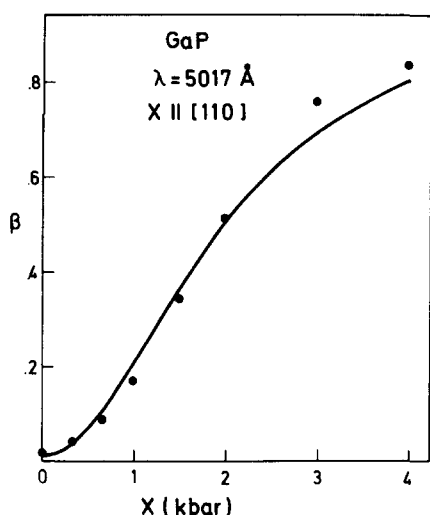


Fig. 1. Depolarization ratio (β) of the LO phonon of GaP observed from a (001) face as a function of applied stress along [110]. The wavelength of the incident radiation is 5017 Å.

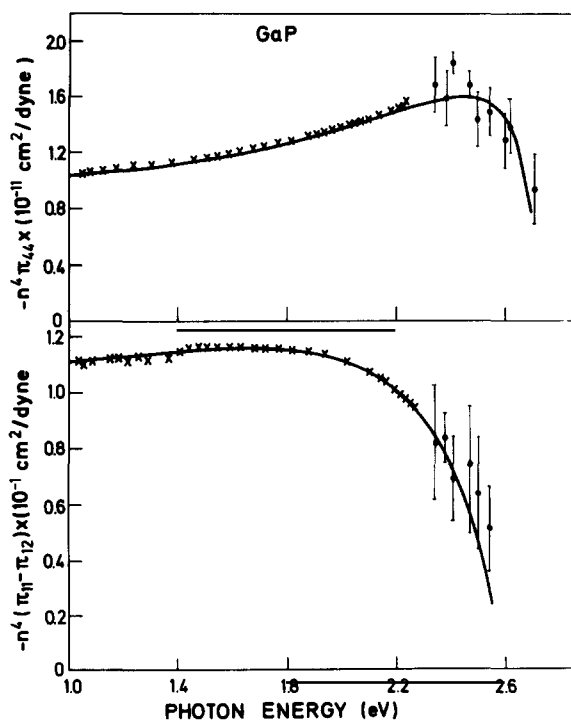


Fig. 2. Measured values of $n^4 \pi_{44}$ and $n^4 (\pi_{11} - \pi_{12})$ for GaP as a function of photon energy. The crosses indicate values determined by conventional techniques where the sample is transparent, the dots are values obtained from the depolarization of the Raman spectra in the region where the sample is opaque. The full line represents a fit to the experimental values due to the various contributions to the birefringence of the various critical points in the band structure.

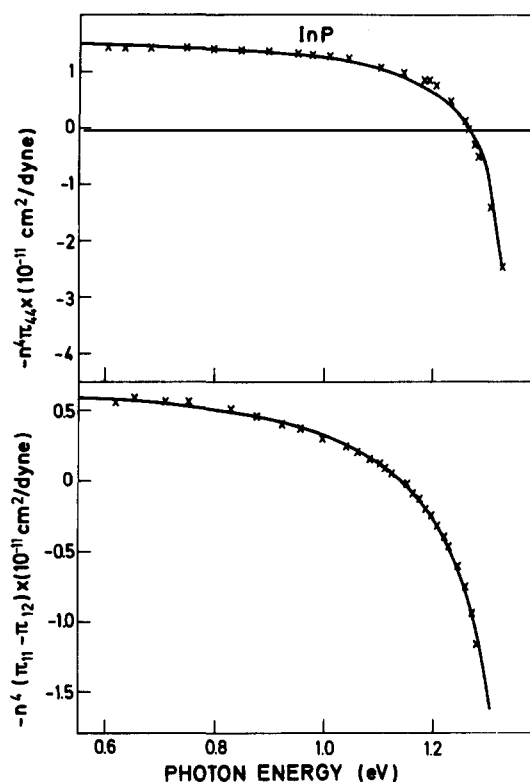


Fig. 3. Measured values of $n^4 \pi_{44}$ and $n^4 (\pi_{11} - \pi_{12})$ for InP as a function of photon energy. The meaning of the crosses and full line is the same as in Fig. 2.

π_{ij} is π_{44} for case (a) and $\pi_{11} - \pi_{12}$ for case (b), α_i and α_s are the absorption coefficients for the incident and scattered light. The constant C in equation (1) is included in the fit to account for any leakage (i.e. spurious depolarization) that may occur at zero stress.

3. RESULTS

The results obtained in the opaque region for β at a wavelength of 5017 Å, for stress along [110], by observing the Raman active LO phonon from a (001) face of GaP are shown in Fig. 1. By fitting equation (1) to these results we obtain $A = 0.35 \text{ kbar}^{-1}$, which in turn leads to $\pi_{44} = 11 \times 10^{-14} \text{ cm}^2/\text{dyne}$. In the analysis of our data for GaP we have used values of the absorption coefficient measured for our crystals. These absorption coefficients are in agreement with the values given in [11]. We have interpolated the refractive index between the values given in [12] and [13].

The values we obtain for π_{ij} for GaP can be compared with those obtained by Dixon [14] at $0.63 \mu\text{m}$ which are $\pi_{44} = -9.8$ and $\pi_{11} - \pi_{12} = -8.4$ in units of $10^{-14} \text{ cm}^2/\text{dyne}$. Our values for the same constants are $\pi_{44} = -11.6$ and $\pi_{11} - \pi_{12} = -9.7$ in the same units.

In Figs. 2 and 3 we present our values for $n^4 \pi_{44}$ and

$n^4(\pi_{11} - \pi_{12})$ for GaP and InP, respectively. The crosses were obtained by conventional techniques, the dots from the depolarization of the Raman spectra. For InP we were unable to use the new technique of determining the piezo-optical constants where the sample is opaque. This is due to a combination of reasons; the Raman scattering is extremely weak, which leads to noisy signals. Furthermore, the small penetration depth arising from the direct nature of the gap, leads to small values of A [see equation (2)]. The small depolarization expected, together with the noisy signals made measurements impracticable. In the analysis of the data of InP we have used the refractive indices of [15].

4. THEORETICAL INTERPRETATION

The band structures of most of the tetrahedrally bonded semiconductors are well known. It is also known that the main contributions to the dielectric constant arise from the spin-orbit split band at zone center E_0 and $E_0 + \Delta_0$ the spin-orbit split bands along the $\langle 111 \rangle$ directions E_1 and $E_1 + \Delta_1$, and last but not least a large contribution from higher bands which is customarily labelled [2] E_2 . In the analysis of piezobirefringence in Si, Chandrasekhar *et al.* [8] have considered only contributions to the piezobirefringence due to the $E_1, E_1 + \Delta_1$ and E_2 gaps. The E_0 and $E_0 + \Delta_0$ gaps in Si have been neglected because they are almost degenerate with the E_1 gap and have a much smaller oscillator strength. However, in GaP and InP, where the E_0 band lies below E_1 , this contribution can no longer be neglected. We give here the contributions to piezobirefringence arising from the $E_0, E_0 + \Delta_0, E_1, E_1 + \Delta_1$ and E_2 gaps [2, 16]. For each gap we consider the two cases $X \parallel [100]$ and $X \parallel [111]$.

E_0 and $E_0 + \Delta_0$ gap

$$\left(\frac{\epsilon_{\parallel} - \epsilon_{\perp}}{X} \right)^{100} = C_0 \left\{ -g(x_0) + \frac{4E_0}{\Delta_0} \times \left[f(x) - \left(\frac{E_0}{E_0 + \Delta_0} \right)^{3/2} f(x_{0s}) \right] \right\}, \quad (3)$$

where ϵ_{\parallel} and ϵ_{\perp} are the dielectric constants for radiation polarized parallel and perpendicular to the applied stress, $x_0 = \hbar\omega/E_0$, $x_{0s} = \hbar\omega/(E_0 + \Delta_0)$ and $g(x)$ and $f(x)$ are defined by

$$g(x) = x^{-2} [2 - (1+x)^{-1/2} - (1-x)^{-1/2}], \quad (4)$$

$$f(x) = x^{-2} [2 - (1+x)^{1/2} - (1-x)^{1/2}]. \quad (5)$$

The constant C_0 is given by

$$C_0 = \frac{3}{4} \frac{C_0''(S_{11} - S_{12})b}{E_0}, \quad (6)$$

where S_{ij} are the elastic compliance constants and b the deformation potential of the Γ_{15} valence bands for [100] stress. The constant C_0'' is related to the contribution to the real part of the dielectric constant of the E_0 and $E_0 + \Delta_0$ gaps by

$$\epsilon_r - 1 \approx C_0'' [f(x_0) + 0.435f(x_{0s})]. \quad (7)$$

The equivalent expression for [111] stress is obtained from equation (3) by replacing C_0 by C_0' where

$$C_0' = \frac{\sqrt{3}C_0''S_{44}d}{8E_0} \quad (8)$$

and d is the deformation potential of the Γ_{15} valence bands for a [111] stress.

E_1 and $E_1 + \Delta_1$ gap

$$\left(\frac{\epsilon_{\parallel} - \epsilon_{\perp}}{X} \right)^{[100]} = C_1 \left[x_1^{-2} \ln(1 - x_1^2) - \left(\frac{E_1}{E_1 + \Delta_1} \right) x_{1s}^{-2} \ln(1 - x_{1s}^2) \right], \quad (9)$$

where $x_1 = \hbar\omega/E_1$ and $x_{1s} = \hbar\omega/(E_1 + \Delta_1)$. The constant C_1 is defined by

$$C_1 = \frac{16\sqrt{2}D_3^2(S_{11} - S_{12})}{3a_0\Delta_1E_1}, \quad (10)$$

where D_3^2 is a deformation potential constant and a_0 the lattice parameter. Also

$$\left(\frac{\epsilon_{\parallel} - \epsilon_{\perp}}{X} \right)^{[111]} = C_1' \left[(1 - x_1^2)^{-1} + \left(\frac{E_1}{E_1 + \Delta_1} \right)^2 (1 - x_{1s}^2)^{-1} \right] \quad (11)$$

and

$$C_1' = \frac{8}{9} \frac{S_{44}(D_1^5 + 2\sqrt{2}D_3^5)}{a_0E_1^2}. \quad (12)$$

E_2 gap

$$\left(\frac{\epsilon_{\parallel} - \epsilon_{\perp}}{X} \right)^{[100]} = C_2(1 + 2x_2^2), \quad (13)$$

where $x_2 = \hbar\omega/E_2$ and C_2 is an experimentally determined parameter. For stresses along [111] equation (13) remains valid with a different value of C_2 , i.e. C_2' .

5. DISCUSSION

It is easy to show that the terms $[(\epsilon_{\parallel} - \epsilon_{\perp})/X]^{[100]}$ and $[(\epsilon_{\parallel} - \epsilon_{\perp})/X]^{[111]}$ are equal to $-n^4(\pi_{11} - \pi_{12})$ and $-n^4\pi_{44}$, respectively. In Figs. 2 and 3 we give our experimental results for these constants as a function of

Table 1. Parameters entering the evaluation of the various contributions to birefringence

	E_0 (eV)	Δ_0 (eV)	E_1 (eV)	Δ_1 (eV)	E_2 (eV)	a_0 (Å)	$S_{11} - S_{12}$ (cm ² /dyne)	S_{44} (cm ² /dyne)	b (eV)	d (eV)	D_3^3 (eV)	$D_1^5 + 2\sqrt{2}D_3^5$ (eV)
GaP	2.78 ^a	0.12 ^a	3.69 ^a	0.08 ^a	5.3 ^a	5.45 ^b	1.27×10^{-12c}	1.42×10^{-12c}	-1.8 ^e	-4.6 ^e	-1.8 ^g	40.6 ^g
InP	1.34 ^a	0.21 ^a	3.12 ^a	0.15 ^a	5.0 ^a	5.87 ^b	2.24×10^{-12d}	2.17×10^{-12d}	-1.55 ^e	-4.4 ^f	-1.8 ^g	40.6 ^g

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^g Values for Si from [8].

Table 2. Contributions to piezobirefringence of the various gaps of GaP and InP (units 10^{-11} cm²/dyne)

	Experimental					Calculated	
	C_0	C'_0	C'_1	C_2	C'_2	C_1	C'_1
GaP	-0.38	-0.28	1.5	1.71	-1.36	-8	0.50
InP	-0.35	-0.38	-	0.94	0.19	-9	1.1

photon energy for GaP and InP. The crosses represent values determined by conventional techniques and the dots values determined from the depolarization of the Raman spectrum. The full lines are fits to the experimental values according to the expressions given in the previous section. Because of the relatively weak dispersion of the E_1 and E_2 contributions in the region studied, it has been possible to determine only C_0 and C'_0 from the experimental fits. Only in the case of GaP $X \parallel [111]$ was it possible to obtain an unambiguous result for C'_1 . For the other fits we have used calculated values of C_1 and C'_1 according to equations (10) and (12). The values of the constants entering into the calculations are summarized in Table 1. In the evaluation of C_1 and C'_1 we have used the values of D_3^3 , D_1^5 and D_3^5 measured for Si [8] because these constants have not been measured for either GaP or InP.

Our experimental values of C_0 , C'_0 , C'_1 , C_2 and C'_2 are given in Table 2 together with the calculated values of C_1 and C'_1 . Our values of C_0 and C'_0 allow C''_0 to be determined using equations (6) and (8). For GaP we obtain 6.2 and 5.5, for InP, 1.8 and 2.5. The difference between the values obtained for each substance are probably due to the simple model that has been used to fit the data.

However, it should be noted that the values obtained

fall within the range of those determined both experimentally and theoretically for other materials of this family [2].

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