Determination of deformation-potential constants by Brillouin scattering of light

E. Käräjämäki, R. Laiho, and T. Levola

Wihuri Physical Laboratory, University of Turku, 20500 Turku 50, Finland

B. H. Bairamov, A. V. Gol'tsev, and T. Toporov

A. F. Ioffe Physico-Technical Institute, Academy of Sciences of the U.S.S.R., 194021 Leningrad, U.S.S.R.

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The theory of resonance Brillouin scattering has been developed for diamond and zinc-blende-type crystals by the consideration of the intermediate electronic states involved in the scattering process as discrete exciton and electron-hole continuum states. The intraband and interband transitions involving the conduction band, the valence band, and the split-off band have been taken into account in the calculation of the resonance Brillouin scattering tensor. From the scattering tensor the expressions of the photoelastic coefficients and those of the deformation-potential constants have been derived. The results are applied to Brillouin scattering data of ZnSe measured at frequencies below the fundamental absorption-band edge. For the deformation-potential constants the values $\tilde{a} = -14.6$ eV, $b = -0.7$ eV, and $d = -4.9$ eV are obtained.

I. INTRODUCTION

During the past few years resonant Raman scattering (RRS) investigations of semiconductors for the determination of deformation-potential constants have received considerable attention. It has been found that the observed first- and second-order Raman scattering intensities are in good agreement with results of pseudopotential calculations. Accordingly, as discussed in the review article by Cardona,[1] both the electron—one-phonon and electron—two-phonon deformation-potential constants have been determined for the E_0, $E_0 + \Delta_0$, and the other next-higher-lying absorption edges in a number of diamond and zinc-blende structure semiconductors with either a direct or indirect energy gap.

It has been shown that resonance Brillouin scattering (RBS) from coupled phonon-exciton energy $(\hbar\omega_i > E_0)$ is a powerful technique to obtain exciton-polariton spatial dispersion curves. In this case Brillouin shifts exhibit strong dependence on excitation frequency. At excitation energies $\hbar\omega_i < E_0$ the dispersion curve of light inside the crystal is linear, and the frequencies of acoustic phonons do not depend on the excitation frequency. When the energy of the exciting light approaches the band edge, provided that the absorption of light does not increase too steeply, a resonance enhancement of the cross section for Brillouin scattering can be observed. In cases where small differences in the population of thermal phonons with $\vec{k} = 0$ can be allowed, resonance Brillouin scattering can be conveniently investigated by tuning the energy-band edge of the crystal through the energy of the laser line. This method was actually used in the first observation of RBS, which was made by Pine[2] in CdS for LA phonons propagating along the C axis of the crystal.

The first theoretical description of resonance Raman and Brillouin scattering was given by Loudon[3] under the assumption of an interaction between a phonon with a virtual electron-hole pair created by light in the crystal.

Ganguly and Birman[4] and Zeyher *et al.*[5] further developed the theory of RRS by taking into account the exciton states. To interpret the results of resonance Brillouin scattering from acousto-electrically amplified phonon domains in certain crystals Adachi *et al.*[6] have used a quasistatic approximation.

In this paper we briefly consider the theory of resonant Brillouin scattering assuming the intermediate states for the scattering process as exciton states belonging to discrete exciton zones and continuum spectra. We give an expression for the resonant term $R_{\mu\nu}$ of Brillouin scattering and compare it with our recent experimental data for ZnSe at $T = 295$ K.[7] The main purpose of our investigation is a more adequate theoretical and experimental study of Brillouin scattering in the nonresonant region which allows the Coulomb interactions between electrons and holes to be neglected. We derive explicit expressions for the scattering tensor and photoelastic coefficients, relating them to the deformation-potential constants. The results are applicable to cubic crystals with either diamond or zinc-blende structure. Accordingly, the fourfold-degenerate valence band Γ_8, which leads to the existence of heavy and light holes, and the twofold spin-orbit-split band Γ_7 are taken into account. Both the interband and the intraband transitions for the conduction and the valence bands are involved in our expressions.

II. EXPERIMENTAL PROCEDURE

The single crystals of ZnSe were cut to shapes of parallelepipeds with typical dimensions of $2 \times 3 \times 4$ mm³. The faces of the samples were oriented perpendicular to the crystallographic directions [100], [011], and [01$\bar{1}$]. The (011) and (0$\bar{1}$1) surfaces were as-grown and the (100) surface was carefully optically polished. The measurements of Raman scattering from TO and LO phonons in the temperature range from 6 to 600 K show that there are no additional contributions to the scattering by LO-

phonon—plasmon modes. From this we can conclude that the concentration of free carriers in our samples was less than 10^{15} cm^{-3} and therefore the influence of the free carriers through the acousto-electric interaction can be neglected in the investigated spectra. Observations of luminescence spectra in the region of fundamental absorption edge at $T=6$ K show that the intensity caused by exciton-impurity complexes and donor-acceptor pairs was weak, which also indicates the high quality of the samples.

Brillouin scattering was excited with the aid of single-mode Kr$^+$ and Ar$^+$ lasers. The spectrum of the scattered light was analyzed by a piezoscanned Fabry-Perot interferometer, which could be used either in a triple-pass or in a five-pass configuration. The detection system consisted of a cooled photomultiplier followed by a multichannel analyzer. The free spectral range of the interferometer was calibrated to an accuracy better than 0.1% by observing Brillouin scattering from fused quartz or directly by measuring the separation between the mirrors of the interferometer. Fused quartz was also used to determine the absolute values of the photoelastic coefficients of ZnSe by comparing the intensities of Brillouin scattering from both materials.

The Brillouin shift Δv of the frequency of light due to the inelastic scattering between photons and phonons can be given by the equation

$$\Delta v = \frac{v}{\lambda}(n_i^2 + n_s^2 - 2n_i n_s \cos\phi)^{1/2} , \tag{1}$$

where v is the velocity of phonons, λ is the wavelength of the incident light, and n_i and n_s are the refractive indices of the incident and scattered lights, which propagate with the mutual angle ϕ. The refractive indices were calculated from Marple's equation.[8] The velocity of phonons was determined from measurements of Δv made at 568.2 nm and using the value $n = 2.631$ in Eq. (1).

III. THEORY OF BRILLOUIN SCATTERING IN SEMICONDUCTORS

In the following we briefly outline the development of theoretical models for the cross section of Brillouin scattering. The final goal of this section is to derive formulas needed to obtain the values of photoelastic coefficients and deformation-potential constants in semiconductors with the diamond or zinc-blende structure.

Classical models for deriving the cross section of Brillouin scattering have been described in the book of Fabelinskii.[9] A detailed quantum-mechanical treatment, based on the use of third-order time-dependent perturbation theory, has been given by Loudon.[3] He obtained the cross section δ_B for light scattered into a solid angle $d\Omega$ as

$$\delta_B = V_0 \left[\frac{e}{m_0 c}\right]^4 \left[\frac{n_s}{n_i}\right]\left[\frac{\omega_s}{\omega_i}\right]\left[\frac{kT}{2\rho v^2}\right]$$

$$\times |e_{s\mu}R_{\mu\nu}e_{i\nu}|^2 . \tag{2}$$

Here V_0 and ρ are the volume and the density of the crystal, respectively, $n_{s,i}$, $\omega_{s,i}$, and $\bar{e}_{s,i}$ are the refractive index, the frequency, and the unit vector of polarization of the scattered (s) or incident (i) light, and v is the velocity of acoustic phonons in the crystal.

The components of the scattering tensor may be given as

$$R_{\mu\nu} = \frac{1}{V_0}\sum_{\alpha,\beta,\vec{k}}\left[\frac{\langle 0|p_\mu|\beta\vec{k}\rangle\langle\beta\vec{k}|H_{\alpha\beta}(\epsilon'_{\vec{q}j})|\alpha\vec{k}\rangle\langle\alpha\vec{k}|p_\nu|0\rangle}{[\hbar\omega_s - E_\beta(\vec{k})][\hbar\omega_i - E_\alpha(\vec{k})]} + \frac{\langle 0|p_\nu|\beta\vec{k}\rangle\langle\beta\vec{k}|H_{\alpha\beta}(\epsilon'_{\vec{q}j})|\alpha\vec{k}\rangle\langle\alpha\vec{k}|p_\mu|0\rangle}{[\hbar\omega_i + E_\beta(\vec{k})][\hbar\omega_s + E_\alpha(\vec{k})]}\right] , \tag{3}$$

where $|0\rangle$ is the wave function of an electron in the ground state, $|\alpha\vec{k}\rangle$ and $|\beta\vec{k}\rangle$ are the wave functions of an electron-hole pair in zones α and β, where the wave vector of an electron is denoted by $\vec{k}$ and the wave vector of a hole by $-\vec{k}$. $E_\alpha(\vec{k})$ and $E_\beta(\vec{k})$ are the energy of an electron in the zones α and β. Further, $\langle|p|\rangle$ is a momentum matrix element and $H(\epsilon_{\vec{q}j})$ is the Hamiltonian of the interaction between electrons and long-wavelength acoustic phonons of the j branch with the wave vector $\vec{q}$. The deformation caused by phonons is given as

$$\epsilon_{\mu\nu} = \frac{1}{2}\left[\frac{\partial u_\mu}{\partial x_\nu} + \frac{\partial u_\nu}{\partial x_\mu}\right] = \sum_{qj}(iqa^-_{\vec{q}j}\epsilon'_{\vec{q}j,\mu\nu}e^{i\vec{q}\cdot\vec{x}} - iqa^+_{\vec{q}j}\epsilon'_{\vec{q}j,\mu\nu}e^{-i\vec{q}\cdot\vec{x}}) , \tag{4}$$

where

$$\epsilon'_{\vec{q}j,\mu\nu} = \frac{1}{2q}[e^j_\mu(\vec{q})q_\nu + e^j_\nu(\vec{q})q_\mu]$$

and $a^-_{\vec{q}j}$ and $a^+_{\vec{q}j}$ are the annihilation and creation operators of the acoustic phonons with the unit polarization vector $e^j(\vec{q})$. In Eq. (2), as throughout in this paper, it is

assumed that the wave vector $\vec{q}$ of phonons is negligible in comparison with the wave vector $\vec{k}$ of electrons.

The frequency dependence of tensor $R_{\mu\nu}$ is a particularly interesting feature, especially from the experimental point of view. It is usually supposed that the frequency dependence of $R_{\mu\nu}$ arises mainly from transitions between states located near the top of the valence band and the bottom of the conduction band. We shall call this contribution $\Delta R_{\mu\nu}$. In addition, the higher valence- and conduction-band states contribute a slowly-frequency-dependent term called $R_{\mu\nu}^{0}$.

For cubic crystals with zinc-blende structure the lower conduction band is nondegenerate and therefore

$$\langle c\vec{k} \mid H_{\alpha\beta}(\epsilon'_{\vec{q}j}) \mid c\vec{k}\rangle = C\epsilon'_{\vec{q}j} = C\sum_{\mu}\epsilon'_{\vec{q}j,\mu\mu} , \qquad (5)$$

where C is the corresponding deformation-potential constant.

A. Excitonic theory of resonant Brillouin scattering

Under the conditions $\hbar\omega_i \approx E_g$ the exciting light incident on the crystal may create virtual electron-hole pairs. The electrons in the conduction band and the holes in the valence band interact with acoustic phonons

through the deformation-potential mechanism. When the electron-hole pair is recombined, the emitted light has the frequency shift corresponding to the energy of the acoustic phonons. Neglecting the Coulomb intraction between electrons and holes the following expression has been derived[2,3] for the resonance part of $R_{\mu\nu}$:

$$R_{\mu\nu} = \frac{1}{2\pi^2}\sum_{\alpha,\beta}\left[\frac{2\mu}{\hbar}\right]^{3/2}\frac{p_{\beta 0}H_{\beta\alpha}(\epsilon'_{\vec{q}j})p_{\alpha 0}}{\omega_{g\beta} - \omega_{g\alpha} + \omega_q}$$

$$\times[(\omega_{g\beta} - \omega_s)^{1/2} - (\omega_{g\beta} - \omega_i)^{1/2}] , \qquad (6)$$

where μ is the reduced mass of the electron-hole pair, $\omega_{q\alpha}$ and $\omega_{q\beta}$ are frequencies corresponding to the optical gaps for the incident and scattered light, respectively.

Ganguly and Birman[4] modified Loudon's[3] treatment by taking into account discrete and continuum exciton transitions in their theory of resonant Raman scattering (RRS). Zeyher *et al.*[5] have further developed this theory and given an explicit expression for the RRS tensor in the case of one valence and one continuum exciton state. In the same manner we have developed the theory of RBS for a three-band process. By using hydrogenlike exciton states for the conduction, valence, and the split-off bands the following expression is obtained for the RBS tensor:

$$R_{is} = \sum_{\alpha,\beta}\frac{p_{\beta 0}H_{\beta\alpha}(\epsilon'_{gj})p_{\alpha 0}}{\omega_{g\beta}\omega_{g\alpha} + \omega_q}\left\{\frac{1}{2\pi a_0^3}\sum_{n=1}^{\infty}\frac{1}{n^3}\left[\frac{1}{-\omega_{g\alpha} - \omega_i - R/n^2} - \frac{1}{\omega_{g\beta} - \omega_s - R/n^2}\right]\right.$$

$$+ \frac{1}{4\pi}\left[\frac{2\mu}{\hbar}\right]^{3/2}R^{1/2}\ln\frac{\omega_{g\beta} - \omega_s}{\omega_{g\alpha} - \omega_q} + \frac{1}{4}\left[\frac{2\mu}{\hbar}\right]^{3/2}R^{1/2}\left[\cot\left[\frac{\pi^2 R}{\omega_{g\beta} - \omega_s}\right]^{1/2}\right.$$

$$\left.\left. - \cot\left[\frac{\pi^2 R}{\omega_{g\beta} - \omega_i}\right]^{1/2}\right]\right\} , \qquad (7)$$

where $a_0 = \epsilon\hbar^2/\mu^2$ is the exciton Bohr radius and $R = \mu e^4/2\hbar^2\epsilon^2$ is the exciton Rydberg constant. The first term inside the curly brackets represents the contribution of the discrete excitonic states to light scattering and the last two terms are due to the continuum states. It should be noted that this expression differs from that derived by Adachi and Hamaguchi.[10] In the derivation of Eq. (7) we have assumed that a_0 is the same for all the three excitons formed from states of the conduction and the three valence bands. Details of the evaluation of Eq. (7) and results of contour integration of scattering amplitude in the complex plane will be given elsewhere.[11]

B. Photoelastic and deformation-potential constants

In cubic crystals with diamond or zinc-blende structure the valence-band edge at $\vec{k} = 0$ is a sixfold-degenerate multiplet. The orbital symmetry is $\Gamma_{25'}$ for the diamond and Γ_{15} for the zinc-blende structure. The degeneracy is removed by the spin-orbit interaction resulting in the forma-

tion of a fourfold $p_{3/2}$ multiplet with $J = \frac{3}{2}$, $m_J = \pm\frac{3}{2}, \pm\frac{1}{2}$ and a twofold multiplet $p_{1/2}$ with $J = \frac{1}{2}$, $m_J = \pm\frac{1}{2}$. For the Γ_{15} representation the Hamiltonian of the electron-phonon interaction in Eq. (4) has been given by Bir and Picus[12(a)] in the form

$$H(\epsilon') = (-a + \tfrac{5}{4}b)\epsilon' - b\sum_{\mu}J_{\mu}^2\epsilon'_{\mu\mu}$$

$$- \frac{d}{\sqrt{3}}\sum_{\mu,\nu}[J_{\mu}J_{\nu}]\epsilon'_{\mu\nu} , \qquad (8)$$

where a, b, and d are the deformation-potential constants. The angular momentum matrices J_{μ} and $[J_{\mu}J_{\nu}]$ are formed by using the functions $\Psi_m^{3/2}$ $(m = \pm\frac{1}{2}, \pm\frac{3}{2})$ and $\Psi_m^{1/2}$ $(m = \pm\frac{1}{2})$ as basis functions. The explicit wave functions of the fourfold Γ_8 state are given by Bir and Picus.[12(b)]

By using Eqs. (7) and (8) the expression for the frequency-independent part, $\Delta R_{\mu\nu}$, of the matrix $R_{\mu\nu}$ can be written as

$$\left[\frac{3\pi^2}{p^2}\right]e_{s\mu}R_{\mu\nu}e_{i\nu}=\left[-\tilde{a}\sum_{n=1}^{3}f_{nn}-b\left(\tfrac{1}{10}f_{11}+\tfrac{1}{10}f_{22}+\tfrac{4}{5}f_{12}+f_{13}+f_{23}\right)\right]\epsilon'(\vec{e}_s,\vec{e}_i)$$

$$+\tfrac{3}{5}b\left[\tfrac{1}{2}f_{11}+\tfrac{1}{2}f_{22}+4f_{12}+5(f_{13}+f_{23})\right]\sum_{\mu}\epsilon'_{\mu\mu}e_{s\mu}e_{i\mu}$$

$$+\sqrt{3}d\left[\frac{1}{30}\left[\frac{D}{B}\right]^2(f_{11}+f_{22}-2f_{12})+f_{12}+f_{13}+f_{23}\right]$$

$$\times[\epsilon'_{xy}(e_{sx}e_{iy}+e_{sy}e_{ix})+\epsilon'_{xz}(e_{sx}e_{iz}+e_{sz}e_{ix})+\epsilon'_{yz}(e_{sy}e_{iz}+e_{sz}e_{iy})]\ . \tag{9}$$

In the derivation of this formula all possible interband and intraband transitions of the electrons for conduction, valence, and split-off bands with emission of acoustical phonons were taken into account.

In Eq. (9) D and B are constants of the valence band[12(c)] and $\tilde{a}=a+C$. The functions $f_{nl}\equiv f_{nl}(\omega_l)$ are defined as

$$f_{nl}\equiv f_{nl}(\omega_l)=\frac{2\pi\mu_n\mu_l}{\hbar^4}\left\{\left[\left[\frac{2\mu_n}{\hbar^2}(E_{gn}-\hbar\omega_i)\right]^{1/2}+\left[\frac{2\mu_l}{\hbar^2}(E_{gl}-\hbar\omega_i)\right]^{1/2}\right]^{-1}\right.$$

$$\left.+\left[\left[\frac{2\mu_n}{\hbar^2}(E_{gn}+\hbar\omega_i)\right]^{1/2}+\left[\frac{2\mu_l}{\hbar^2}(E_{gl}+\hbar\omega_i)\right]^{1/2}\right]^{-1}\right\}\ , \tag{10}$$

where E_{gn} is the energy separation between the conduction band and the three valence bands labeled with n ($n=1$ for the light-hole band, $n=2$ for the heavy-hole band, and $n=3$ for the spin-orbit split-off band). The reduced mass $\mu_n=m_e m_{hn}/(m_e+m_{hn})$ is given by the effective masses of electrons m_e and holes m_{hn}.

Equation (9) and (10) do not take into account the nonparabolicity of zones and may be not quite correct when the frequencies of incident and the scattered light approach the value of the energy gap. In this case the main contribution to $\Delta R_{\mu\nu}$ will arise from states near the Γ point where the terms linear in $\vec{k}$ become important.

The momentum matrix elements are given as

$$p^2=\frac{1}{2}m_0\left[\frac{m_0}{m_e}-1\right]\frac{E_g(E_g+\Delta)}{E_g+2\Delta/3}\ , \tag{11}$$

where E_g is the energy gap and Δ is the spin-orbit splitting of the valence band in the Γ point.

The photoelastic coefficients $p_{\mu\nu ij}$ and $R_{\mu\nu}$ are connected by the formula

$$R_{\mu\nu}=-\frac{m_0^2\omega_i^2}{e^2}\frac{n_\mu^2 n_\nu^2}{4\pi}p_{\mu\nu ij}e_i(\vec{q})\frac{q_j}{q}\ , \tag{12}$$

where n_μ is the refractive index corresponding to the μ axis. By using Eqs. (7) and (9) we obtain the expressions of the photoelastic coefficients Δp_{11}, Δp_{12}, and Δp_{44} as

$$\Delta p_{11}\equiv\Delta p_{1111}=\frac{4}{3\pi}\frac{e^2p^2}{m_0^2\omega_i^2 n_i^4}\left[\tilde{a}\sum_{n=1}^{3}f_{nn}-2b\left[\tfrac{1}{10}(f_{11}+f_{22}-2f_{12})+f_{12}+f_{13}+f_{23}\right]\right]\ ,$$

$$\Delta p_{12}\equiv\Delta p_{1122}=\frac{4}{3\pi}\frac{e^2p^2}{m_0^2\omega_i^2 n_i^4}\left[\tilde{a}\sum_{n=1}^{3}f_{nn}+b\left[\tfrac{1}{10}(f_{11}+f_{22}-2f_{12})+f_{12}+f_{13}+f_{23}\right]\right]\ , \tag{13}$$

$$\Delta p_{44}\equiv p_{1212}=-\frac{2\sqrt{3}}{3\pi}\frac{e^2p^2d}{m_0^2\omega_i^2 n_i^4}\left[f_{12}+f_{13}+f_{23}+\frac{1}{30}\left[\frac{D}{B}\right]^2(f_{11}+f_{22}-2f_{12})\right]\ .$$

It can be easily shown that the following relations exist between the photoelastic coefficients:

$$\Delta p_{11}-\Delta p_{12}=-\frac{4}{\pi}\frac{e^2p^2}{m_0^2\omega_i^2 n_i^4}b\left[\tfrac{1}{10}(f_{11}+f_{22}-2f_{12})\right.$$

$$\left.+f_{12}+f_{13}+f_{23}\right]\ , \tag{14}$$

and

$$\Delta p_{11}+2\Delta p_{12}=\frac{4}{\pi}\frac{e^2p^2}{m_0^2\omega_i^2 n_i^4}\tilde{a}\sum_{n=1}^{3}f_{nn}\ . \tag{15}$$

Another useful relation can be found in crystals for which the masses of light and heavy holes greatly exeed the effective mass of the electrons ($m_{hl},m_{hh}\gg m_e$). In this case $\mu_1\approx\mu_2$ and

$$\frac{\Delta p_{44}}{\Delta p_{11}-\Delta p_{12}}\approx\frac{1}{2\sqrt{3}}\frac{d}{b}\ . \tag{16}$$

The experimentally measured values of the photoelastic coefficients p_{ij} are given by

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$$p_{11} = \Delta p_{11} + \frac{1}{\tilde{\omega}_i^2} p_{11}^{(0)} ,$$

$$p_{12} = \Delta p_{12} + \frac{1}{\tilde{\omega}_i^2} p_{12}^{(0)} , \qquad (17)$$

$$p_{44} = \Delta p_{44} + \frac{1}{\tilde{\omega}_i^2} p_{44}^{(0)} ,$$

where $p_{ij}^{(0)}$ are the frequency-independent contributions to the photoelastic coefficients arising from higher valence and conduction bands. They can be determined from measurements made at different wavelengths of the exciting light. Thus we have obtained explicit expressions for the photoelastic coefficients taking into account the con-

tributions from the interband and the intraband transitions between the conduction and the valence bands. From the experimentally determined three independent photoelastic coefficients it is possible to obtain the values of the deformation-potential constants $\tilde{a}$, b, and d.

Tada *et al.*[13] have used the theory of Higginbotham *et al.*[14] to determine the values of the deformation potential constants from their experimental data for ZnSe. However, this theory is based on the assumption $m_h \gg m_e$, which is not valid in ZnSe, where we have $m_e/m_{hl} \sim 2$ (see Table II). Consequently, the values of the deformation-potential constants should be determined using Eqs. (10)—(13) which allow the proper values of the effective masses to be taken into account.
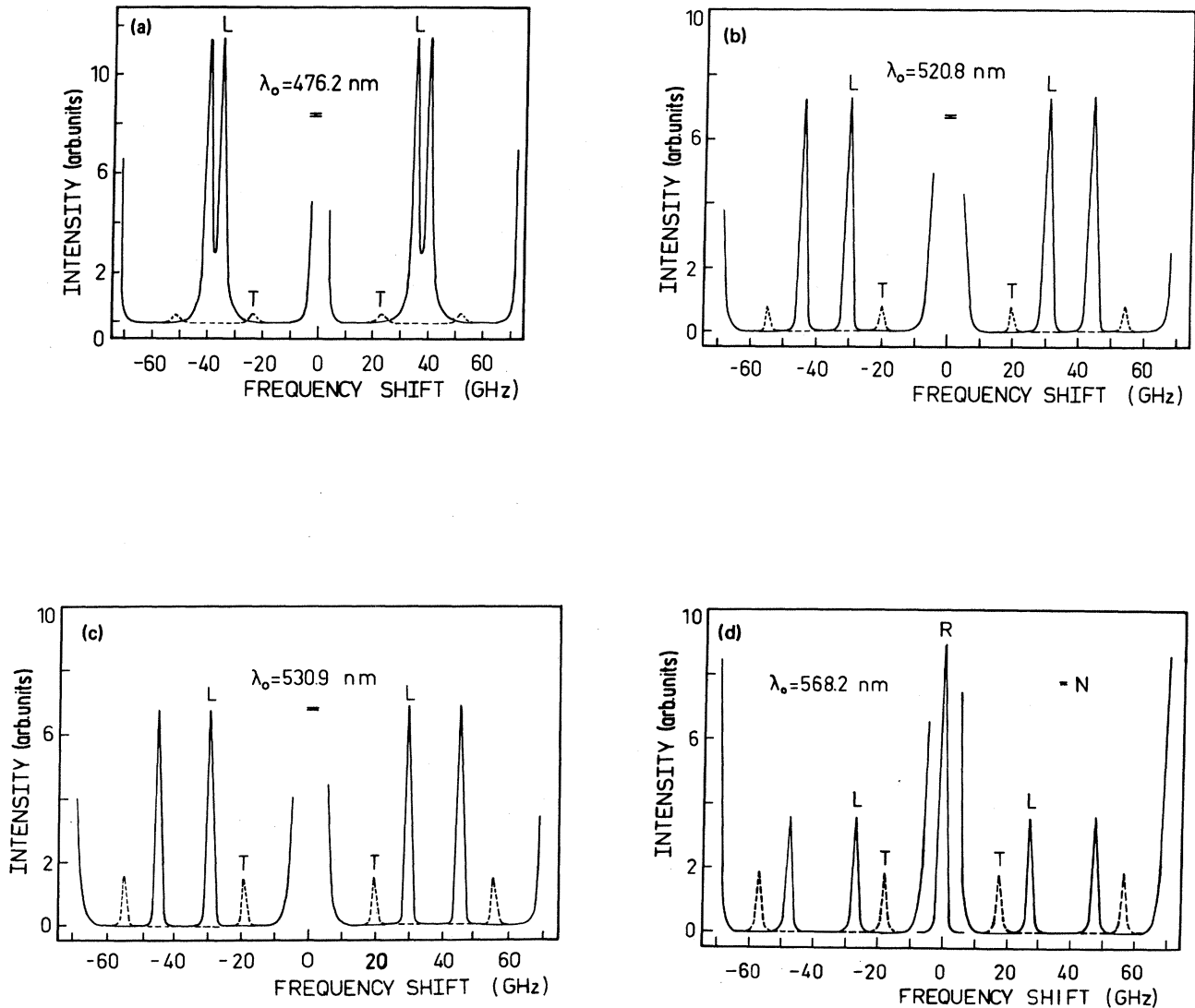

FIG. 1. Examples of Brillouin scattering spectra observed at different excitation wavelengths λ in ZnSe at $T = 295$ K. The scattering spectra from longitudinal-acoustic phonons (L, solid curve) and from transverse-acoustic phonons (T, dashed curves) are observed in the geometries $y(xx)z$ and $y(xy)z$, respectively. The effect of resonance is seen as changes of the relative intensities of the (L) and (T) components when the energy of the laser line is varied near the band gap. The level of noise is indicated by horizontal bars.

TABLE I. Brillouin scattering data for ZnSe at 295 K. In the notation $a(bc)d$ the first and the last symbols stand for the directions of the incident and the scattered light and those in parentheses their polarizations, respectively. Definitions used: $x||[100]$, $y||[011]$, and $z||[01\bar{1}]$. The data are obtained at $\lambda=568.2$ nm. The combinations of the elastic constants equal to ρv^2 and the photoelastic coefficients $\propto I_{si}$ used in this experiment are also given.

Case	Δv (GHz)	v (m/s)	ρv^2	$\propto I_{si}$
$y(xx)\bar{y}$	42.61	4599	$\dfrac{C_{11}+C_{12}+2C_{44}}{2}$	$\dfrac{2p_{12}^2}{C_{11}+C_{12}+2C_{44}}$
$x(yy)\bar{x}$	38.09	4112	C_{11}	$\dfrac{p_{12}^2}{C_{11}}$
$y(xx)z$	27.22	4155	C_{11}	$\dfrac{p_{12}^2}{C_{11}}$
$y(xy)z$	17.97	2744	C_{44}	$\dfrac{p_{44}^2}{2C_{44}}$
$y(zy)z$	27.16	4146	C_{11}	$\dfrac{(p_{11}-p_{12})^2}{C_{11}}$

IV. RESULTS AND DISCUSSION

A. Experimental results

Some typical Brillouin scattering spectra containing both Stokes and anti-Stokes components for scattering from longitudinal (L) and transverse (T) acoustic phonons in ZnSe are shown in Fig. 1. In these spectra the effect of resonance scattering is qualitatively seen as changes of relative intensities between the (L) and (T) components when the energy of the laser line is varied in the vicinity of the fundamental band gap, $E_g=2.68$ eV (462.5 nm). The Brillouin shifts observed in different scattering geometries, the corresponding values of the velocities of phonons, and the combinations of the elastic coefficients appropriate to each scattering case are given in Table I.

As can be seen from Fig. 2 resonance effects are observed in the scattering intensity for all modes when the energy of the exciting light approaches the region of the band gap. The intensity of Brillouin scattering and the components of the photoelastic tensor, $p_{\nu\mu kl}$, are related as

$$I_{is}=\frac{8\pi^2 kT}{\lambda_i^4}\frac{(n_{i\mu}^2 n_{sv}^2 e_{sv} p_{\nu\mu kl} u_k q_l e_{i\mu})^2}{(n_i+1)^2(n_s+1)^2\rho v^2}I_0 L_e , \qquad (18)$$

where e_{sv} and $e_{i\mu}$ are the νth and μth components of the polarization vectors $\vec{e}_i$ and $\vec{e}_s$ of light and u_k and q_l are the components of the displacement and the wave vectors of phonons. I_0 is the intensity of the incident light and L_e is the effective scattering length in the crystal. The methods used to determine L_e for different scattering geometrics are discussed elsewhere.[7] It is evident from literature that various optical parameters of ZnSe determined by different authors differ considerably. Therefore, the values of L_e at different wavelengths were calculated from the absorption data measured specifically for the samples used in our scattering experiments.

In Fig. 2 the following relations between the scattering intensities and the photoelastic coefficients are valid: (a) $I_{LA}\propto p_{12}^2$, (b) $I_{TA}\propto p_{44}^2$, and (c) $I_{LA}\propto(p_{11}-p_{12})^2$. When the experimental data are compared with the Loudon's theory [Eq. (6), dashed line] and the present excitonic theory [Eq. (7), solid line] a somewhat better agreement is achieved with the excitonic theory at values of $\hbar\omega_i$ below resonances with discrete exciton states. The growth of the calculated intensities when $\hbar\omega_i$ approaches the absorption edge is due to the first term in Eq. (7). This term increases resonantly when approaching the lowest exciton state which is at 2.68 eV in ZnSe at 300 K. At high values of $\hbar\omega_i$ a better agreement with the experimental data can be obtained if the finite lifetime of the excitonic states is included in Eq. (7) by replacing ω by $\omega+i(\gamma/2h)$. The best result was achieved by assuming $\hbar\gamma=0.01$ eV but there still remains some discrepancy. To further develop the theory of RBS it would possibly be necessary to take into account the deviation of the excitonic states from hydrogenlike states as well as the polariton effects and the details of the band structure.

The combinations of the photoelastic coefficients appropriate to the used scattering geometries are compiled in Table I. The dispersion of the coefficients p_{11}, p_{12}, and p_{44} are shown in Fig. 3. It is obvious that their absolute values increase when the energy of the exciting light approaches the fundamental absorption edge. The dispersion curves of p_{11} and p_{22} are found to cross at $E\approx 2.5$ eV ($\lambda\approx 495.8$ nm) and the coefficient p_{44} changes its sign at $E\approx 2.53$ eV ($\lambda\approx 490.0$ nm). The positions of these "isotropic points" are in good agreement with the values obtained from piezobirefringence measurements[15,16] or from light scattering data from acousto-electrically amplified acoustic domains.[13]

B. Calculation of the deformation-potential constants

We have used Eqs. (12)–(16) to determine the values of the deformation-potential constants $\tilde{a}$, b, and d from the
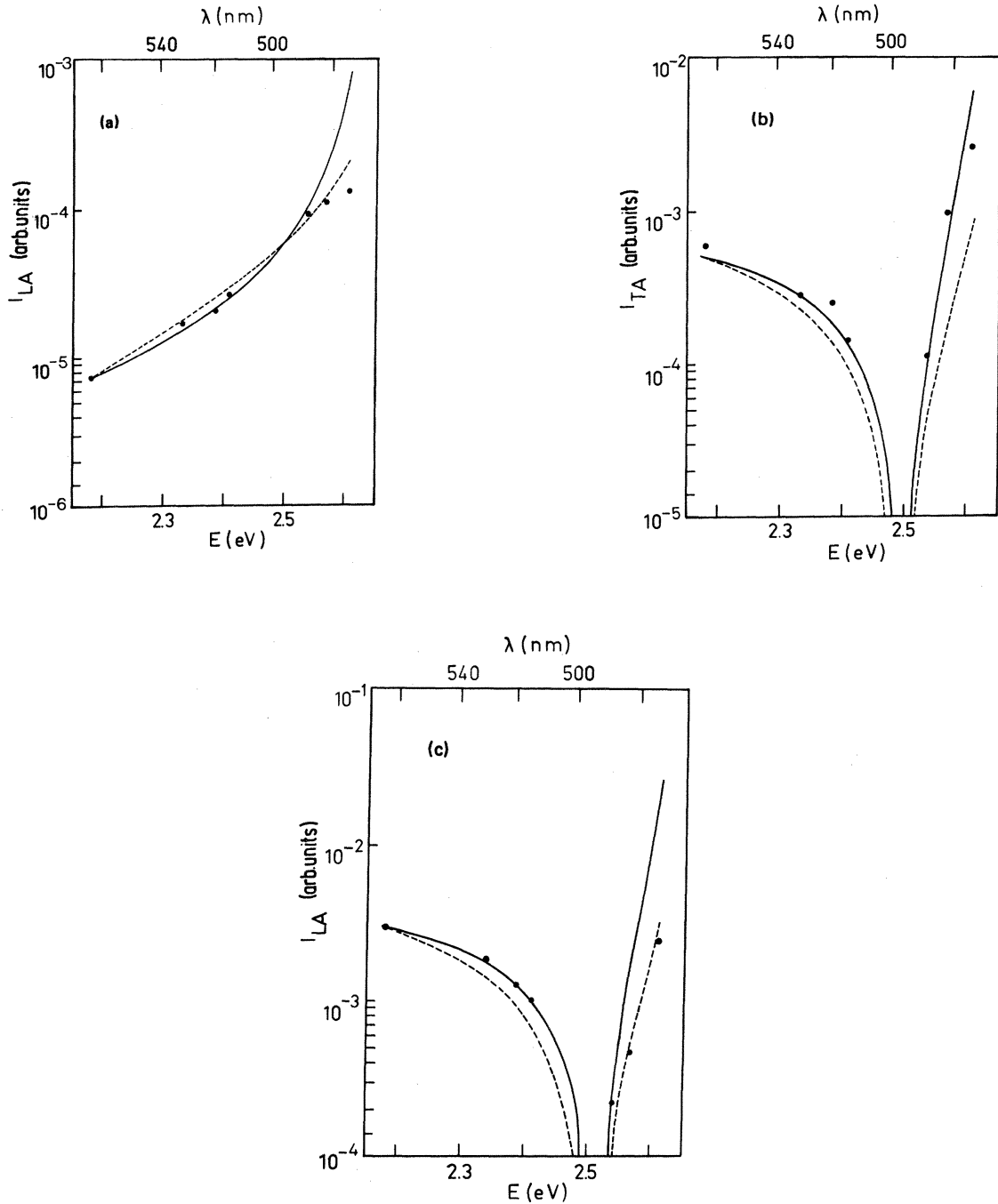
FIG. 2. Dependence of the intensity of Brillouin scattering on the excitation energy in the scattering geometries (a) $y(xx)z$, (b) $y(xy)z$, and (c) $y(zy)z$. The solid lines represent the dispersion of the intensity calculated with the aid of Eq. (7) and the dashed lines the intensity calculated from the theory of Loudon (Ref. 3).

Brillouin scattering data for ZnSe. The values of the photoelastic coefficients measured at excitation frequencies far from the resonance region ($\hbar\omega_i < E_g$) were used in this calculation. The other physical constants involved in the calculation are given in Table II.

Unfortunately the constants B and D in Eq. (12) are not known for ZnSe. It is usually assumed that the ratio D/B is approximately 2 to 4 in crystals with diamond or zinc-

blende structure. If this is also the case in ZnSe, then

$$\frac{1}{30}\left[\frac{D}{B}\right]^2 \sim 0.1-0.5$$

and as an approximation we will neglect this parameter in our calculations. The results for $\tilde{a}$, b, and d are given in Table III. It can be found that the deformation-potential
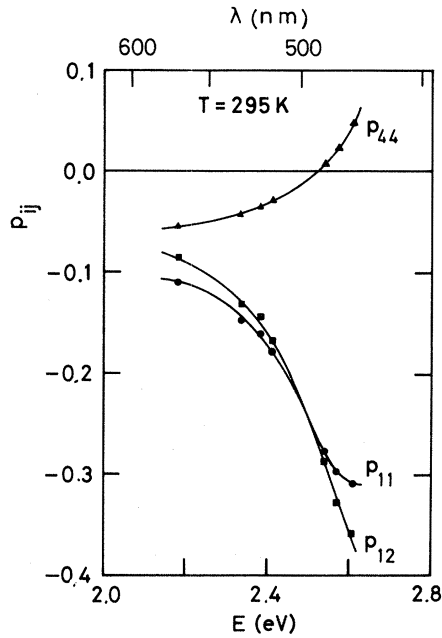
FIG. 3. Dispersion of the photoelastic coefficients.

constants do not show a large dispersion. This feature clearly supports the validity of our method.

In Table IV the final values of the deformation-potential constants are compared with the results obtained by other authors. It is likely that the differences between the present data and the other results quoted arise from different theoretical approaches used. This suggests that the contributions from the interband and intraband transitions between the conduction band and all the three valence bands must be considered in the calculation of the deformation-potential constants.

Finally, it should be noted that the values of the deformation-potential constants depend on several experimental factors including the impurity concentration and the quality of the bulk and the surfaces of the sample crystals. There is also much difference in the magnitude and the topology of the strain field applied in different experimental methods. In the present theory the differences between the effective masses of the light and heavy holes are taken into account. In principle further improvement could be achieved by properly adjusting the value of the parameter D/B and by allowing the values of the effective masses to depend on the direction of the wave vector in the crystal.

TABLE II. Numerical values of the physical constants of ZnSe used to calculate the values of the deformation-potential constants $(T=295$ K).

$E_g = 2.68$ eV	$m_e = (0.16 \pm 0.01)m_0{}^{a}$
$\Delta = 0.43$ eV	$m_{hl} = (0.33 \pm 0.06)m_0{}^{a}$
$E_{g1} = E_{g2} = E_g$	$m_{hh} = (2.2 \pm 0.4)m_0{}^{a}$
$E_{g3} = E_g + \Delta$	

[a]B. Sermage and G. Fishman, Phys. Rev. Lett. <u>43</u>, 1043 (1979).

TABLE III. Values of the deformation-potential constants for ZnSe when $\frac{1}{30}(D/B)^2 \ll 1$. All units are in eV.

$\hbar\omega_i$	$\tilde{a} = a + C$	b	d	d/b
2.18	−14.34	−0.70	−4.41	6.3
2.20	−14.43	−0.68	−4.62	6.8
2.25	−14.51	−0.70	−5.03	7.2
2.34	−15.11	−0.77	−5.42	7.0

IV. CONCLUSIONS

We have discussed the theory of resonance Brillouin scattering for diamond and zinc-blende structured crystals. In the three-band scattering process the intermediate electronic states have been described with discrete exciton states and with states belonging to the electron-hole continuum. By using hydrogenlike exciton states for the conduction, valence, and split-off bands we have obtained the expression for the resonance scattering tensor and compared it with experimental data for ZnSe at $T=295$ K when the energy of the exciting light approaches the fundamental absorption band edge E_g.

At energies $\hbar\omega_i < E_g$ a satisfactory agreement is obtained between the scattering intensities measured for the LA and TA phonons and values calculated from the expression for the scattering terms. Around 2.5 eV deep minima are observed in the scattering intensities from one of the LA modes and from the TA mode reflecting corresponding variations of the values of the photoelastic constants.

By using the formula of the scattering tensor, explicit expressions have been derived for the photoelastic coefficients p_{11}, p_{12}, and p_{44} relating them with the deformation-potential constants a, b, and d. In this calculation, which is valid at $\hbar\omega_i < E_g$, the interband and intraband transitions for the conduction and valence bands and the split-off band are taken into account. Different effective masses are allowed for the light and heavy holes. The discrepancies found between the existing values of the deformation-potential constants and those calculated from the present experimental data for ZnSe are attributed to differencies both in theoretical and experimental approaches used by different authors.

TABLE IV. Defromation-potential constants for ZnSe obtained by different methods (in units of eV).

	This work	a	b	c
$\tilde{a} = a + C$	−14.6	−6.8		
b	−0.7	−2.6	−2.3	−1.2
d	−4.9	−6.7	−5.2	−3.8

[a]Reference 13; acousto-optic technique using pulses of longitudinal ultrasonic phonons.
[b]Reference 14; piezobirefringence measurements.
[c]D. W. Langer, R. N. Euwema, K. Era, and T. Koda, Phys. Rev. B <u>2</u>, 4005 (1970); exciton-stress interaction.

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