

Theory of Brillouin scattering in anisotropic, piezoelectric semiconductors

Ole Keller

Physics Laboratory, Royal Veterinary and Agricultural University, Copenhagen, Denmark

(Received 6 May 1975)

A theory of Brillouin scattering in piezoelectric semiconductors is presented. The formula derived for the differential-scattering cross section is valid for crystals of any symmetry and of any optical or acoustic anisotropy in any direction. The scattered light intensity is calculated on the basis of a new dyadic Green's function for radiation in anisotropic conducting media. The expression for the phonon-induced fluctuation in the dielectric-constant tensor is extended to incorporate the contributions from the free-carrier-screened indirect photoelastic effect and from the free-carrier plasma. By using the Boltzmann equation, the phonon-induced self-consistent electric field arising from the piezoelectric coupling and the deformation potential coupling is calculated. The influence of a spatial exponential growth or decay in the phonon beam intensity on the scattering cross section is considered.

I. INTRODUCTION

In the last decade Brillouin scattering has been extensively used to investigate acoustoelectric domains produced in piezoelectric semiconductors by acoustoelectric amplification of a band of lattice waves from the thermal-equilibrium spectrum.¹ The main emphasis of the analyses has been devoted to (a) the evolution in time and space of the various frequency components and angular components of the phonon gas caused by the linear electron-phonon interaction,²⁻⁵ (b) the lattice-vibration attenuation of the amplified modes originating in the nonelectronic interaction with the thermal background phonons,^{4,6,7} (c) the nonlinear phonon-phonon interaction due to the coupling of bunched electrons of one frequency with the piezoelectric fields associated with other frequencies,^{2,3,8,9} (d) the phonon focusing or defocusing arising from the elastic and the acoustoelectric angular dispersion effects,¹⁰⁻¹² and (e) the properties of acoustoelectrically inactive domains generated by mode conversion.^{6,13}

Parallel with the experimental studies of the domains progress has been made to generalize the theory of Brillouin scattering. The central points in a theoretical analysis are the scattering kinematics and the scattering cross section.

In optically isotropic solids the scattering kinematics is determined by the normal Bragg law if one neglects the small change in the frequency of the scattered light arising from the inelastic nature of the diffraction process.¹⁴ In optically anisotropic solids deviations from the normal Bragg law occur if (i) there is a polarization change of light in the scattering process,¹⁵ (ii) the refractive index depends on the propagation direction of light,^{16,17} or (iii) the scattering event involves a combination of (ii) and (iii).¹⁴ In conducting crystals an additional deviation from the isotropic kinematics can occur if the conductivity is aniso-

tropic.¹⁸

A theory of light scattering from an equilibrium thermal distribution of acoustic phonons in cubic crystals was given by Benedek and Fritsch.¹⁹ Using the autocorrelation function for the scattered field the theory predicts the spectral distribution of light scattered from a single acoustic mode of finite lifetime. The scattering theory for cubic crystals was used to investigate the acoustoelectric coupling in GaAs.^{3,6} Using an integral-equation method the theories of Brillouin scattering were extended to incorporate optical birefringence and depletion of the incident light beam by Hope.²⁰ Since this theory is rather complicated to apply to elastic waves with arbitrary polarization and propagation directions Hamaguchi²¹ derived, based on the method of Benedek and Fritsch, and a model of anisotropic scattering in hexagonal crystals, and applied it to acoustic waves propagating parallel or perpendicular to the *c* axis. Recently, the theory of Hamaguchi has been extended by the present author²² to account for the rotational contribution to the direct photoelastic effect and the angular deviation of the Poynting vector from the wave vector of the diffracted light. Partly to comply with the increasing interest in off-axis acoustic waves in hexagonal crystals, the author calculated Brillouin scattering cross sections of off-axis phonons in CdS for a number of important scattering geometries. For the complicated piezoelectrically inactive, pure transverse phonon type especially detailed results were given. Furthermore, based on the zeros for the quasilongitudinal phonon scattering cross section a simple method of determining the relative signs of the photoelastic tensor elements was proposed. By means of a Green's-function technique a theory of Brillouin scattering valid for nonconducting crystals of any symmetry has been published by Nelson *et al.*²³ In contrast to the above-mentioned works this theory takes into account the indirect photoelastic

effect, that is, the succession of the piezoelectric and the electro-optic effects.^{24,25} Recently, San'ya and Hamaguchi²⁶ have evaluated the contribution from the indirect photoelastic effect to the scattering cross section for acoustic waves propagating in the basal plane of CdS with the atomic displacement parallel to the optic axis. An experimental separation of the direct and the indirect photoelastic effect is not possible in this configuration.

Although all the previous Brillouin scattering theories have been concerned with nonconducting media they have been applied to semiconducting crystals. This can be justified for a number of important experimental investigations, but is in general not allowed. The present theory, which takes into account the piezoelectricity and the conductivity of the solid, thus has a much wider range of validity. Furthermore, this theory is valid for crystals of any symmetry and of any optical or acoustic anisotropy in any direction. In Sec. II the inhomogeneous wave equation for the diffracted electric field is established. The time-independent wave equation is solved by a Green's-function technique and an explicit expression for the dyadic Green's function for Brillouin scattering in anisotropic, semiconducting crystals is given. Finally, the time-averaged Poynting vector for the scattered field is evaluated. In Sec. III the phonon-induced perturbations of the optical dielectric-constant tensor is calculated taking into account a spatial decay or amplification of the acoustic mode considered. For the direct photoelastic effect both the Pöckel contribution²⁷ and the rotational contribution²⁸ to the dielectric constant are considered. For the indirect photoelastic effect we have included the screening caused by the free carriers by calculating the phonon-induced self-consistent field arising from the piezoelectric coupling²⁹ and the deformation-potential coupling.³⁰ The frequency- and wave-vector-dependent conductivity is calculated by means of the Boltzmann equation, so that the treatment becomes valid for acoustic wavelengths comparable with the electron mean free path. The effect of an external dc field is also included. The screened indirect photoelastic effect is especially important when the Brillouin scattering cross section due to the direct photoelastic effect vanishes. By eliminating the direct effect Brillouin scattering via the indirect effect provides a possibility of studying several aspects of the free-carrier screening in piezoelectric semiconductors. Also the fluctuation in the dielectric constant caused by the free-carrier plasma,²² which is most important for long optical wavelengths, has been calculated. In Sec. IV the phase-matched Brillouin scattering kinematics is discussed, and the influence of a depletion

(or an amplification) of the incident optical and acoustic beams on the scattering intensity for non-phase-matched scattering configurations is considered. In Sec. V the equation of motion for the lattice vibrations is established. Including the piezoelectric coupling and the deformation-potential coupling, approximate expressions for the acoustic phase velocity and the linear electron-phonon damping (or amplification) coefficient in tensorial notation is obtained. In Sec. VI the squared amplitude of the atomic displacement for a damped sound wave is expressed in terms of a phonon occupation number. In Sec. VII the final expression for the effective differential Brillouin scattering cross section in an anisotropic, piezoelectric semiconductor is given.

The theoretical formulas for the scattering kinematics and the scattering cross section derived in the present work apply inside the crystal, whereas experimental measurements are always made outside the medium. A detailed, accurate analysis of the surface effects for anisotropic crystals has been made by Lax and Nelson,³¹ who in addition to the transmission loss, took into account solid-angle expansion of the scattered beam and source-volume demagnification. In the theory of Hope²⁰ also the effect of internal reflections of both the scattered and the unscattered light beams was calculated. Simplified expressions for the reflectivity corrections in cubic and hexagonal crystals have been given by several authors.^{3,4,21}

II. GREEN'S-FUNCTION CALCULATION OF THE SCATTERED LIGHT INTENSITY

The present theory is based on a classical calculation of the scattered light intensity. The classical approach is valid if the solid can be regarded as a continuum, i. e., if the electromagnetic wavelength is large compared to a characteristic interatomic spacing. This condition restricts the treatment to electromagnetic waves having frequencies in or below the ultraviolet region. Thus, we exclude x-ray Brillouin scattering involving a reciprocal-lattice vector.^{32,33} The appearance of an upper limit on the optical wave frequency implies that the acoustic wave vectors, detectable by the considered spectrum of the electromagnetic radiation, are small compared to the Brillouin zone-boundary wave vectors.^{14,15,34} This, in turn, justifies that elastic velocity dispersion effects,³⁴ in contrast to elastic and acoustoelectric velocity dispersion effects,^{35,36} can be neglected. Since the resonant enhancement of the Brillouin scattering in the vicinity of the intrinsic absorption edge³⁷⁻⁴⁰ is not considered, the analysis becomes invalid for electromagnetic wavelengths below a certain limit.

Combining Maxwell's equations and the piezoelectric equations of state, we obtain the following

equation for the average electric field $\vec{E}(\vec{r}, t)$ in a piezoelectric semiconductor:

$$\vec{\nabla} \times [\vec{\nabla} \times \vec{E}(\vec{r}, t)] + \frac{1}{c^2} \frac{\partial^2}{\partial t^2} (\vec{\epsilon}_r^L(\vec{r}, t) \cdot \vec{E}(\vec{r}, t)) + \frac{1}{\epsilon_0} \vec{\sigma} \cdot \vec{S}(\vec{r}, t) + \frac{1}{c^2} \frac{\partial}{\partial t} \left(\frac{1}{\epsilon_0} \vec{\sigma}(\vec{r}, t) \cdot \vec{E}(\vec{r}, t) \right) = 0, \quad (1)$$

where \vec{r} is the position vector, t is the time, $\vec{\nabla}$ is the gradient operator, c is the velocity of light in vacuum, $\vec{\epsilon}_r^L$ is the contribution from the lattice and the bound electrons to the relative dielectric-constant tensor, ϵ_0 is the dielectric constant of vacuum, $\vec{\sigma}$ is the piezoelectric coefficient tensor, $\vec{\sigma}$ is the electric conductivity tensor, and \vec{S} is the strain associated with the electromagnetic wave due to the piezoelectric coupling. In the derivation of Eq. (1) it has been assumed that the elastic wave propagates adiabatically,⁴¹ so that the terms involving entropy changes do not occur.

As discussed by Kyame,^{41,42} the mechanical energy in the mixed electromagnetic-mechanical wave, which travels with a phase velocity close to the electromagnetic phase velocity, will be small compared with the electromagnetic energy. In the following we shall neglect the mechanical deformation \vec{S} in Eq. (1).

The presence of an elastic disturbance, propagating with a velocity close to the acoustic velocity, produces a time-space fluctuation in the lattice dielectric constant^{24,25,27,28} and, for piezoelectrically active waves, also in the conductivity.^{1,29,43,44} Thus, we may write

$$\vec{\epsilon}_r^L(\vec{r}, t) = \vec{\epsilon}_{r,0}^L + \delta \vec{\epsilon}_r^L(\vec{r}, t) \quad (2)$$

and

$$\vec{\sigma}(\vec{r}, t) = \vec{\sigma}_0 + \delta \vec{\sigma}(\vec{r}, t), \quad (3)$$

where $\delta \vec{\epsilon}_r^L$ and $\delta \vec{\sigma}$ are the perturbations of the lattice dielectric constant ($\vec{\epsilon}_{r,0}^L$) and the dc conductivity ($\vec{\sigma}_0$) caused by the acoustic disturbance. Furthermore, let us decompose the average electric field as follows:

$$\vec{E}(\vec{r}, t) = \vec{E}_i(\vec{r}, t) + \vec{E}_d(\vec{r}, t), \quad (4)$$

where \vec{E}_i and \vec{E}_d are the incident and the diffracted electric field.

Inserting Eqs. (2)–(4) into Eq. (1), neglecting the higher order products $\delta \vec{\epsilon}_r^L \cdot \vec{E}_d$ and $\delta \vec{\sigma} \cdot \vec{E}_d$, which correspond to a limitation of the theory to first-order scattering processes, and utilizing the fact that \vec{E}_i satisfies Eq. (1) in the absence of the acoustic disturbance, one obtains the inhomogeneous wave equation

$$c^2 \vec{\nabla} \times [\vec{\nabla} \times \vec{E}_d(\vec{r}, t)] + \left(\vec{\epsilon}_{r,0}^L \frac{\partial^2}{\partial t^2} + \frac{1}{\epsilon_0} \vec{\sigma}_0 \frac{\partial}{\partial t} \right) \cdot \vec{E}_d(\vec{r}, t)$$

$$= - \frac{\partial^2}{\partial t^2} [\delta \vec{\epsilon}_r^L(\vec{r}, t) \cdot \vec{E}_i(\vec{r}, t)] - \frac{\partial}{\partial t} \left(\frac{1}{\epsilon_0} \delta \vec{\sigma}(\vec{r}, t) \cdot \vec{E}_i(\vec{r}, t) \right) \quad (5)$$

for the diffracted field.

Assuming the incident light and the acoustic beams to be monochromatic with angular frequencies ω_i and Ω we introduce the complex time-independent amplitude $\delta \vec{\epsilon}_r^L(\vec{r}, \Omega)$, $\delta \vec{\sigma}(\vec{r}, \Omega)$, $\vec{E}_i(\vec{r}, \omega_i)$, and $\vec{E}_d(\vec{r}, \omega_d)$ through

$$\delta \vec{\epsilon}_r^L(\vec{r}, t) = \text{Re}[\delta \vec{\epsilon}_r^L(\vec{r}, \Omega) e^{i\Omega t}], \quad (6)$$

$$\delta \vec{\sigma}(\vec{r}, t) = \text{Re}[\delta \vec{\sigma}(\vec{r}, \Omega) e^{i\Omega t}], \quad (7)$$

$$\vec{E}_i(\vec{r}, t) = \text{Re}[\vec{E}_i(\vec{r}, \omega_i) e^{-i\omega_i t}], \quad (8)$$

and

$$\vec{E}_d(\vec{r}, t) = \text{Re}[\vec{E}_d(\vec{r}, \omega_d) e^{-i\omega_d t}], \quad (9)$$

where the angular frequency of the scattered light is given by

$$\omega_d = \omega_i \pm \Omega. \quad (10)$$

It is well known that this relation in a particle picture expresses the conservation of energy in the phonon-photon scattering process. If a phonon is created in the collision the frequency of the scattered light will be less than that of the incident light, producing the Stokes component, whereas if a phonon is annihilated in the process, the scattered light has a higher frequency, the anti-Stokes component.

In general Ω can be complex in order to describe an exponential decay or amplification of the elastic disturbance as a function of time. The imaginary part of Ω gives rise to a linewidth in frequency of the Brillouin-scattered component.¹⁹ For a thermal or a nonthermal distribution of piezoelectrically inactive phonons the linewidth determines a phonon lifetime arising from elastic anharmonicity^{13,45} and boundary scattering.^{4,45} For a nonthermal distribution of piezoelectrically active phonons (eventually an acoustoelectric domain), the linewidth can give substantial information about the acoustic growth rate in the weak- and strong-flux regions,⁴⁶ and especially about the rate of parametric subharmonic or higher-harmonic phonon generation via the nonlinear phonon-electron interaction.^{47,48} The starting point for a calculation of the spectral distribution of the scattered radiation is the autocorrelation function for the scattered electric field.^{19,49} In the present work we shall confine the analysis to time-independent phonon distributions. Thus, the scattered power calculated in this section refers to the scattered power per unit frequency range integrated over the experimental line shape of a particular Brillouin-frequency component.

Inserting Eqs. (6)–(10) in Eq. (5) one obtains for the diffracted field the driven or inhomogeneous time-independent wave equation

$$\begin{aligned} \vec{\alpha}(\vec{\nabla}, \omega_d) \cdot \vec{E}_d(\vec{r}, \omega_d) \\ = \{ (c/\omega_d)^2 [\vec{\nabla}\vec{\nabla} - \vec{1}(\vec{\nabla} \cdot \vec{\nabla})] - \vec{\epsilon}_{r,0}(\omega_d) \} \cdot \vec{E}_d(\vec{r}, \omega_d) \\ = (1/\epsilon_0) \delta \vec{P}(\vec{r}, \omega_d), \end{aligned} \quad (11)$$

where $\vec{1}$ is the unit tensor.

In the above equation we have introduced the complex frequency-dependent relative dielectric-constant tensor

$$\vec{\epsilon}_{r,0}(\omega_d) = \vec{\epsilon}_{r,0}^L(\omega_d) + i \vec{\sigma}_0(\omega_d)/\epsilon_0 \omega_d, \quad (12)$$

containing the contributions from the lattice and the free carriers. In general both contributions can be complex and frequency dependent,⁵⁰ i. e.,

$$\vec{\epsilon}_{r,0}^L(\omega_d) = \text{Re}[\vec{\epsilon}_{r,0}^L(\omega_d)] + i \text{Im}[\vec{\epsilon}_{r,0}^L(\omega_d)],$$

and

$$\vec{\sigma}_0(\omega_d) = \text{Re}[\vec{\sigma}_0(\omega_d)] + i \text{Im}[\vec{\sigma}_0(\omega_d)].$$

The complex amplitude $\delta \vec{P}(\vec{r}, \omega_d)$ of the nonlinear driving polarization has been introduced through

$$\delta \vec{P}(\vec{r}, \omega_d) = \epsilon_0 \delta \vec{\epsilon}_r(\vec{r}, \Omega) \cdot \vec{E}_i(\vec{r}, \omega_i), \quad (13)$$

where

$$\delta \vec{\epsilon}_r(\vec{r}, \Omega) = \delta \vec{\epsilon}_r^L(\vec{r}, \Omega) + i \delta \vec{\sigma}(\vec{r}, \Omega)/(\epsilon_0 \omega_d) \quad (14)$$

is the phonon-induced fluctuation in the total relative dielectric-constant tensor. For nonconducting crystals Eq. (11) is in agreement with Eq. (2.1) in the paper of Nelson *et al.*²³

A calculation of the Brillouin-scattered light intensity requires a solution of the driven wave equation [Eq. (11)]. In a typical geometry used to observe Brillouin scattering the nonlinear polarization is nonzero over a small but finite region in the medium under study. Since outgoing-plane-wave solutions are inappropriate for such a geometry, Eq. (11) is best solved by a Green's-function technique.

Decomposing the operator $\vec{\alpha}(\vec{\nabla}, \omega_d)$ into a real part and an imaginary part, i. e.,

$$\vec{\alpha}(\vec{\nabla}, \omega_d) = \vec{\alpha}_R(\vec{\nabla}, \omega_d) - i \vec{\alpha}_I(\omega_d), \quad (15)$$

where

$$\vec{\alpha}_R(\vec{\nabla}, \omega_d) = (c/\omega_d)^2 [\vec{\nabla}\vec{\nabla} - \vec{1}\nabla^2] - \text{Re}[\vec{\epsilon}_{r,0}(\omega_d)] \quad (16)$$

and

$$\vec{\alpha}_I(\omega_d) = \text{Im}[\vec{\epsilon}_{r,0}(\omega_d)], \quad (17)$$

the solution of the driven time-independent wave equation is given by^{51,52}

$$\vec{E}_d(\vec{r}, \omega_d) = \frac{1}{\epsilon_0} \int_{\vec{r}'} \vec{G}(\vec{r}, \vec{r}') \cdot \delta \vec{P}(\vec{r}', \omega_d) d\vec{r}', \quad (18)$$

where the dyadic Green's function for Brillouin

scattering in anisotropic, conducting crystals is¹⁸

$$\begin{aligned} \vec{G}(\vec{r}, \vec{r}') = \int_{-\infty}^{\infty} [\vec{\alpha}_R(\vec{k}_R, \omega_d) - i \vec{\alpha}_I(\omega_d)]^{-1} \\ \times e^{i\vec{k}_R \cdot (\vec{r} - \vec{r}')} \frac{d\vec{k}_R}{(2\pi)^3}. \end{aligned} \quad (19)$$

The matrix $\vec{\alpha}_R(\vec{k}_R, \omega_d)$ in Eq. (19) is given by

$$\vec{\alpha}_R(\vec{k}_R, \omega_d) = n_R^2 (\vec{1} - \hat{s}_R \hat{s}_R) - \text{Re}[\vec{\epsilon}_{r,0}(\omega_d)], \quad (20)$$

with the definitions $n_R = ck_R/\omega_d$ and $\hat{s}_R = \vec{k}_R/k_R$ (\vec{k}_R real).

If $\vec{\alpha}_I$ vanishes the Green's function reduces to that obtained by Lax and Nelson⁵³ for nonabsorbing crystals.

Since the sound field associated with an optical wave in a piezoelectric crystal is of negligible importance,⁴¹ the effective optical equation of motion for a phonon-unperturbed propagating plane wave in a lossless medium $\{\text{Im}[\vec{\epsilon}_{r,0}(\omega_d)] = 0\}$ takes the form

$$\vec{\alpha}_R(\vec{k}_R, \omega_d) \cdot \vec{E}_i = 0. \quad (21)$$

The eigenvalue problem in Eq. (21) leads to an introduction of the real electric field eigenvectors $\vec{e}_R^\varphi = \vec{e}_R^\varphi(\hat{s}, \omega_d)$ with eigenvalues $[1/n_R^\varphi(\hat{s}, \omega_d)]^2$ defined by

$$(\vec{1} - \hat{s}\hat{s}) \cdot \vec{e}_R^\varphi = (1/n_R^\varphi)^2 (\text{Re} \vec{\epsilon}_{r,0}) \cdot \vec{e}_R^\varphi \quad (\varphi = 1, 2, \infty), \quad (22)$$

where \hat{s} is a unit wave vector. Besides the two electric field solutions ($\varphi = 1, 2$) which correspond to freely propagating, linearly polarized, undamped waves, we have included the third solution (infinite refractive index), which can exist only as a forced wave and be important in coupling various optical processes in high-order nonlinear optical interactions.^{53,54} It should be noticed that the eigenvectors can be chosen to obey the biorthogonality requirement⁵³

$$\vec{e}_R^\varphi(\hat{s}, \omega_d) \cdot [\text{Re} \vec{\epsilon}_{r,0}(\omega_d)] \cdot \vec{e}_R^\theta(\hat{s}, \omega_d) = \delta_{\varphi\theta}. \quad (23)$$

It has been shown by Lax and Nelson⁵³ that the inverse matrix $\vec{\alpha}_R^{-1}(\vec{k}_R, \omega_d)$ can be expressed in terms of the real eigenvectors and eigenvalues for $\vec{\alpha}_R(\vec{k}_R, \omega_d)$. Explicitly, the inverse operator can be written in the form

$$\vec{\alpha}_R^{-1}(\vec{k}_R, \omega_d) = \sum_{\varphi=1,2} \frac{\vec{e}_R^\varphi \vec{e}_R^\varphi}{(n_R/n_R^\varphi)^2 - 1} - \frac{\hat{s}\hat{s}}{\hat{s} \cdot (\text{Re} \vec{\epsilon}_{r,0}) \cdot \hat{s}}, \quad (24)$$

where the last term represents the contribution from the longitudinal forced plane wave. Inserting Eq. (24) into Eq. (19) the Green's function $\vec{G}(\vec{r}, \vec{r}')$ is expressed in terms of the real eigenvectors \vec{e}_R^φ ($\varphi = 1, 2, \infty$). Note that the integrand of Eq. (19) becomes infinite only when \vec{k}_R is "on the energy shell," i. e., appropriate to a free undamped plane-wave solution.

Above the Green's function was evaluated in terms of eigenvalues and eigenfunctions for the optical equation of motion in a lossless medium. In an absorbing crystal the effective optical equation of motion for a phonon-unperturbed plane wave takes the form

$$\vec{\alpha}(\vec{k}, \omega_d) \cdot \vec{E}_{e1} = [n^2(\hat{1} - \hat{s}\hat{s}) - \vec{\epsilon}_{r,0}] \cdot \vec{E}_{e1} = 0, \quad (25)$$

where $n = ck/\omega_d$ and $\hat{s} = \vec{k}/k$. Since the wave vectors turn out to be complex in this case, i. e., $\vec{k} = (\text{Re}k + i\text{Im}k)\hat{s}$, the eigenvalue problem in Eq. (25) leads to solutions in the form of damped plane waves ($\vec{k} = \vec{k}^\varphi$ and $n = n^\varphi = ck^\varphi/\omega_d$, $\varphi = 1, 2, \infty$). The propagation direction of the phase is given by the real unit wave vector \hat{s} . In a semiconductor the complex part of the dielectric constant arises mainly from the real part of the free-carrier conductivity for photon energies less than the bandgap energy. Thus, one usually has $[\text{Im}\vec{\epsilon}_{r,0}(\omega_d)]_{ij} \ll [\text{Re}\vec{\epsilon}_{r,0}(\omega_d)]_{ij}$ for every pair i, j . Using this relation it can be shown⁴⁹ that to first order in $[\text{Im}\vec{\epsilon}_{r,0}(\omega_d)]_{ij}/[\text{Re}\vec{\epsilon}_{r,0}(\omega_d)]_{ij}$, $\text{Re}k^\varphi = k_R^\varphi$ and $\text{Re}n^\varphi = n_R^\varphi$. This implies that the damping of the optical wave is determined by $\text{Im}\vec{\epsilon}_{r,0}(\omega_d)$ and the wavelength by $\text{Re}\vec{\epsilon}_{r,0}(\omega_d)$. Furthermore, it is well known⁴⁹ that the principal vibrations in an absorbing crystal in general are elliptically polarized, and that the electric displacement vectors no longer are perpendicular to the wave normal \hat{s} . Finally, it can be shown that the expansion of $\vec{\alpha}^{-1}(\vec{k}, \omega_d)$ in terms of the eigenfunctions and eigenvalues for the absorbing crystal becomes completely analogous to that in Eq. (24).

Combining Eqs. (19), (21), and (25) yields the dyadic Green's function for a semiconducting crystal,

$$\vec{G}(\vec{r}, \vec{r}') = \int_{-\infty}^{\infty} \left(\lim_{\vec{k}_I \rightarrow 0} \vec{\alpha}^{-1}(\vec{k}, \omega_d) \right) e^{i\vec{k}_R \cdot (\vec{r} - \vec{r}')} \frac{d\vec{k}_R}{(2\pi)^3}, \quad (26)$$

where $\vec{k} = \vec{k}_R + i\vec{k}_I$.

In the present theory the Green's function has been expressed in terms of an integral over the real part (\vec{k}_R) of the complex \vec{k} space. Alternatively, \vec{G} could be evaluated as integrals along lines parallel to the real axes ($\vec{k}_I = \vec{\eta} \neq 0$) in analytical half-planes of the complex \vec{k} space, if one utilizes a three-dimensional Laplace transformation. Using a procedure analogous to the preceding one obtains

$$\vec{G}(\vec{r}, \vec{r}') = \int_{-i\vec{\eta}_\infty}^{-i\vec{\eta}_+\infty} \vec{\alpha}^{-1}(\vec{k}, \omega_d) e^{i\vec{k} \cdot (\vec{r} - \vec{r}')} \frac{d\vec{k}}{(2\pi)^3}. \quad (27)$$

In the following we shall use the expression for $\vec{G}(\vec{r}, \vec{r}')$ involving integrals over the real part of \vec{k} space [i. e., Eq. (19) or (26)].

In the next step, the integral in Eq. (26) is evaluated assuming the observation point \vec{r} to be in the far-field ("Fraunhofer") region. Using an

asymptotic method by Eckart⁵⁵ one obtains

$$\vec{G}(\vec{r}, \vec{r}') = \frac{1}{(2\pi)^2} \int_{-\infty}^{\infty} \vec{\alpha}^{-1}(k_R \hat{1}_R, \omega_d) \frac{e^{ik_R R}}{ik_R R} k_R^2 dk_R, \quad (28)$$

where $\vec{R} = \vec{r} - \vec{r}'$ and $\hat{1}_R = \vec{R}/R$. The approximation involved in obtaining the far-field Green's function, namely, $k_R R \gg 1$, is an excellent one in the optical region. Expanding $\vec{\alpha}^{-1}(k_R \hat{1}_R, \omega_d)$ to first order in $\vec{\alpha}_I(\omega_d)$ one finds

$$\vec{\alpha}^{-1}(k_R \hat{1}_R, \omega_d) = \vec{\alpha}_R^{-1}(k_R \hat{1}_R, \omega_d) + i\vec{\alpha}_I^{-1}(k_R \hat{1}_R, \omega_d) \cdot \vec{\alpha}_I(\omega_d) \cdot \vec{\alpha}_R^{-1}(k_R \hat{1}_R, \omega_d). \quad (29)$$

By inserting Eq. (29) into (28) and by using Eq. (24) one obtains, neglecting the nonpropagating mode, after straightforward contour integrations (see Appendix A) the following first-order approximation to the far-field dyadic Green's function⁷⁸:

$$\begin{aligned} \vec{G}(\vec{r}, \vec{r}') &= \sum_{\varphi=1,2} \vec{e}_R^\varphi \vec{e}_R^\varphi \frac{e^{ik_R^\varphi R}}{4\pi R} \frac{1}{1/(k_R^\varphi)^2} \\ &- i \sum_{\varphi, \theta=1,2} \vec{e}_R^\varphi \vec{e}_R^\theta \cdot [\text{Im}\vec{\epsilon}_{r,0}(\omega_d)] \cdot \vec{e}_R^\theta \vec{e}_R^\varphi \\ &\times \frac{e^{ik_R^\varphi R} - e^{ik_R^\theta R}}{4\pi R} \frac{1}{1/(k_R^\varphi)^2 - 1/(k_R^\theta)^2}, \end{aligned} \quad (30)$$

where $k_R^\varphi = (\omega_d/c)n_R^\varphi(\hat{1}_R, \omega_d)$, $\varphi = 1, 2$, and $\vec{e}_R^\varphi = \vec{e}_R^\varphi(\hat{1}_R, \omega_d)$. Suppressing the tensor notation a rough estimate of the numerical ratio between the second term and the first term in Eq. (30) gives, assuming $k_R^1 \approx k_R^2 (=k_R^0)$, $n_R^1 \approx n_R^2 (=n_R^0)$, $|\vec{e}_R^\varphi| \approx 1/n_R^\varphi$, and $\text{Im}\epsilon_{r,0} \approx \sigma_0/\epsilon_0 \omega_d$, the result

$$\begin{aligned} \frac{\sigma_0}{(n_R^0)^2 \epsilon_0 \omega_d} \left| \frac{1 - e^{i(k_R^0 - k_R^0)R}}{1 - (k_R^0/k_R^0)^2} \right| \\ \leq \frac{\sigma_0}{(n_R^0)^2 \epsilon_0} \frac{k_R^0 R}{\omega_d} \approx \frac{\tau_r}{\tau_c}, \end{aligned} \quad (31)$$

where τ_r is the time retardation of the diffracted electromagnetic wave between the interaction region and the crystal surface, and $\tau_c(\omega_d) = 1/\Omega_c(\omega_d)$ is the dielectric relaxation time at ω_d . Thus, if the retardation time is small compared to the appropriate dielectric relaxation time, i. e., for

$$\Omega_c(\omega_d)\tau_r \ll 1, \quad (32)$$

the electronic contribution to $\vec{G}(\vec{r}, \vec{r}')$ can be neglected.

When the source dimensions are small compared to the observation distance ($r' \ll r$) we can in Eq. (30) replace R by r and $\hat{1}_R$ by $\hat{1}_r = \vec{r}/r$ except in the exponential, where we make the sagittal approximation $R \approx r - \hat{1}_r \cdot \vec{r}'$. Doing this, and inserting Eq. (30) in (18), one obtains the diffracted electric field⁷⁸

$$\vec{E}_d(\vec{r}, \omega_d) = \sum_{\varphi=1,2} \frac{e^{ik_R^\varphi r}}{4\pi\epsilon_0 r} [k_R^\varphi(\hat{1}_r, \omega_d)]^2 \vec{e}_R^\varphi(\hat{1}_r, \omega_d) C^\varphi, \quad (33)$$

where C^ν with the abbreviation

$$\vec{\Gamma}^m = \int_{\vec{r}'} \exp[-ik_R^m(\hat{\mathbf{1}}_r, \omega_d) \hat{\mathbf{1}}_r \cdot \vec{r}'] \delta \vec{P}(\vec{r}', \omega_d) d\vec{r}' \quad (m = \varphi, \theta) \quad (34)$$

is given by

$$C^\nu = \vec{e}_R^\nu \cdot \left[\vec{\Gamma} - i[\text{Im} \vec{\epsilon}_{r,0}(\omega_d)] \cdot \sum_{\theta=1,2} \left(\frac{\vec{e}_R^\theta \vec{e}_R^\theta}{1 - (k_R^\theta/k_R^\nu)^2} \right) \cdot [\vec{\Gamma} - e^{i(k_R^\theta - k_R^\nu)r} \vec{\Gamma}^\theta (\vec{\Gamma}^\nu)^{-1}] \right] \cdot \vec{\Gamma}^\nu. \quad (35)$$

Using Maxwell's equation $\vec{\nabla} \times \vec{E}_d(\vec{r}, \omega_d) = i\omega_d \vec{H}_d(\vec{r}, \omega_d)$ and neglecting terms of order $(k_R^\nu r)^{-1}$, the diffracted complex magnetic field amplitude $\vec{H}_d(\vec{r}, \omega_d)$ becomes

$$\vec{H}_d(\vec{r}, \omega_d) = \sum_{\nu, \nu'=1,2} \frac{e^{ik_R^{\nu'} r}}{4\pi\epsilon_0 r} \frac{[k_R^{\nu'}(\hat{\mathbf{1}}_r, \omega_d)]^3}{\omega_d \mu_0} \times \hat{\mathbf{1}}_r \times \vec{e}_R^{\nu'}(\hat{\mathbf{1}}_r, \omega_d) C^{\nu'}. \quad (36)$$

Combining Eqs. (33) and (36) the time-averaged Poynting vector $\langle \vec{S} \rangle = \frac{1}{2} \text{Re}[\vec{E}_d(\vec{r}, \omega_d) \times \vec{H}_d^*(\vec{r}, \omega_d)]$ is found to be⁷⁸

$$\langle \vec{S} \rangle = \sum_{\nu, \nu'=1,2} \frac{c^2 (k_R^{\nu'})^3 (k_R^\nu)^2}{32\pi^2 \epsilon_0 r^2 \omega_d} \text{Re}[C^\nu (C^{\nu'})^* e^{i(k_R^\nu - k_R^{\nu'})r}] \times [\hat{\mathbf{1}}_r (\vec{e}_R^\nu \cdot \vec{e}_R^{\nu'}) - \vec{e}_R^{\nu'} (\vec{e}_R^\nu \cdot \hat{\mathbf{1}}_r)]. \quad (37)$$

Making use of Eq. (22) and the biorthogonality requirement [Eq. (23)], the power into a detector at \vec{r} with solid angle $d\Omega$ can be written in the form

$$\langle \vec{S} \rangle \cdot \hat{\mathbf{1}}_r r^2 d\Omega = \frac{\omega_d^4}{32\pi^2 \epsilon_0 c^3} \times \sum_{\nu=1,2} [n_R^\nu(\hat{\mathbf{1}}_r, \omega_d)]^3 |C^\nu|^2 d\Omega, \quad (38)$$

which contains no cross terms between the two polarizations. Equation (38) is the starting point for a calculation of the Brillouin scattering kinematics and cross section.

III. PHONON-INDUCED PERTURBATIONS OF THE DIELECTRIC CONSTANT

As discussed in Sec. II an evaluation of the scattered light intensity requires a determination of the thermal or nonthermal time-space fluctuations of the total dielectric constant of the crystal. Assuming the fluctuations caused by the acoustic disturbance to be describable by a plane, monochromatic wave over the interaction volume, the elastic displacement vector $\vec{u}^\mu(\vec{r}, t)$ may be written as

$$\vec{u}^\mu(\vec{r}, t) = \text{Re}[\vec{u}^\mu(\vec{r}, \Omega) e^{i\Omega t}], \quad (39)$$

where the complex time-independent displacement

is given by

$$\vec{u}^\mu(\vec{r}, \Omega) = u_0^\mu(\Omega, \vec{K}^\mu) \hat{\eta}^\mu \times \exp\{[\pm iK^\mu(\Omega) - \Gamma^\mu(\Omega, \vec{K}^\mu)] \hat{\mathbf{k}} \cdot \vec{r}\}. \quad (40)$$

The index μ labels the different branches in the phonon dispersion relation connecting the real phonon wave vector $\vec{K}^\mu(\Omega) = K^\mu(\Omega) \hat{\mathbf{k}}$ and the angular frequency Ω . The spatial decay (or amplification) of the phonon beam is governed by the frequency- and direction-dependent damping (or amplification) coefficient $\Gamma^\mu(\Omega, \vec{K}^\mu)$. The displacement amplitude is $\vec{u}_0^\mu(\Omega, \vec{K}^\mu)$, $\hat{\eta}^\mu$ denotes a unit vector in the direction of polarization of the sound wave, and $\hat{\mathbf{k}}$ is a unit vector in the direction of the acoustic wave vector. For shortness, we shall in the following usually omit the index μ on the wave vector and the damping coefficient.

In a piezoelectric semiconductor the phonon perturbation of the complex time-independent optical dielectric constant can be decomposed into²²

$$\delta \vec{\epsilon}_r(\vec{r}, \Omega) = \delta \vec{\epsilon}_r^D + \delta \vec{\epsilon}_r^I + \delta \vec{\epsilon}_r^C. \quad (41)$$

The first term, $\delta \vec{\epsilon}_r^D$, arising from the fluctuations in the strain tensor²⁷ and the mean rotation tensor,²⁸ gives the direct photoelastic effect. The second term, $\delta \vec{\epsilon}_r^I$, represents the screened indirect photoelastic effect, that is, the succession of the free-carrier screened piezoelectric effect and the electro-optic effect.^{24,25} The third term, $\delta \vec{\epsilon}_r^C$, gives the free-carrier contribution to the dielectric fluctuations.^{18,22}

The constitutive equations relating the dielectric and elastic properties of the medium combine the change in the inverse dielectric tensor and the elastic displacement. To link the constitutive relations to Eq. (41) one utilizes the fact that the derivative of the product of a matrix and its inverse is zero, deducing

$$\delta \vec{\epsilon}_r(\vec{r}, \Omega) = -\vec{\epsilon}_{r,0}(\omega_i) \cdot \delta \vec{\epsilon}_r^{-1}(\vec{r}, \Omega) \cdot \vec{\epsilon}_{r,0}(\omega_i). \quad (42)$$

Note, that $\delta \vec{\epsilon}_r(\vec{r}, \Omega)$ represents the change, caused by the elastic wave, in the dielectric tensor at the input optical frequency ω_i or the output frequency ω_d , since $\omega_i \gg \Omega$.¹⁴

A. Direct photoelastic effect

For small strains, the contribution to the inverse dielectric tensor arising from the direct photoelastic effect can be expressed as a linear function of the symmetric and antisymmetric combination of displacement gradients, i. e.,

$$[\delta \vec{\epsilon}_r^D(\vec{r}, \Omega)]^{-1} = \vec{p}^s(\omega_i) \cdot \vec{S}(\vec{r}, \Omega) + \vec{p}^{as}(\omega_i) \cdot \vec{R}(\vec{r}, \Omega), \quad (43)$$

where the first term gives Pöckels contribution²⁷ to the direct photoelastic effect and the second term gives the rotational contribution.²⁸ The infinitesimal strain \vec{S} and the mean rotation \vec{R} are defined

by $\vec{S} = \frac{1}{2}[\vec{\nabla}\vec{u}^\mu + (\vec{\nabla}\vec{u}^\mu)^T]$ and $\vec{R} = \frac{1}{2}[\vec{\nabla}\vec{u}^\mu - (\vec{\nabla}\vec{u}^\mu)^T]$, where $(\vec{\nabla}\vec{u}^\mu)_{ij} = \partial u_i^\mu(\vec{r}, \Omega)/\partial r_j$, and $(\vec{\nabla}\vec{u}^\mu)^T$ is the transpose of the matrix $\vec{\nabla}\vec{u}^\mu$. The photoelastic tensor \vec{p}^s is symmetric upon interchange of the acoustic (last two) indices, whereas \vec{p}^{as} is antisymmetric in these indices. The antisymmetric tensor \vec{p}^{as} can be calculated simply from the optical dielectric tensor. Hence in component form²⁸

$$\begin{aligned} p_{ijkl}^{as} = & \frac{1}{2}[(\epsilon_{rr,0}^{-1})_{il} \delta_{kj} + (\epsilon_{rr,0}^{-1})_{ij} \delta_{lk} \\ & - (\epsilon_{rr,0}^{-1})_{ik} \delta_{lj} - (\epsilon_{rr,0}^{-1})_{kj} \delta_{li}]. \end{aligned} \quad (44)$$

Equation (43) may be written

$$[\delta\epsilon_r^D(\vec{r}, \Omega)]^{-1} = \vec{p}(\omega_i) \cdot \vec{\nabla}\vec{u}^\mu(\vec{r}, \Omega), \quad (45)$$

where the single photoelastic tensor $\vec{p}(\omega_i)$ is defined by

$$\vec{p}(\omega_i) = \vec{p}^s(\omega_i) + \vec{p}^{as}(\omega_i). \quad (46)$$

It is seen from Eq. (45) that the natural measure of elastic deformation relevant to the photoelastic effect is the displacement gradient. The tensor $\vec{p}(\omega_i)$ is neither symmetric nor antisymmetric in the acoustic indices.

Combining Eqs. (40), (42), and (45) one obtains from the direct photoelastic effect the contribution

$$\begin{aligned} \delta\epsilon_r^D(\vec{r}, \Omega) = & -i\vec{\epsilon}_{r,0}(\omega_i) \cdot \vec{p}(\omega_i) \cdot \hat{\pi}^\mu \hat{\kappa} \cdot \vec{\epsilon}_{r,0}(\omega_i) \\ & \times [\pm K(\Omega) + i\Gamma(\Omega, \vec{K})] u^\mu(\vec{r}, \Omega). \end{aligned} \quad (47)$$

B. Screened indirect photoelastic effect

An acoustic wave propagating through a piezoelectric semiconductor will be accompanied by an electrostatic field arising from the piezoelectric polarization and an electromagnetic wave produced by the displacement current. According to Kyame^{41,42} and Hutson and White⁴³ it is quite justifiable to neglect the latter in comparison with the former. In the case of plane waves the only electrostatic fields of importance will be longitudinal.²⁹

The self-consistent time-independent part $\vec{F}_{sc}(\vec{r}, \Omega)$, of the electric field calculated on the basis of the longitudinal electrostatic field, causes, via the linear electro-optic effect (small-strain limit), a fluctuation in the inverse dielectric constant given by

$$[\delta\epsilon_r^I(\vec{r}, \Omega)]^{-1} = \vec{r}(\omega_i) \cdot \vec{F}_{sc}(\vec{r}, \Omega), \quad (48)$$

where $\vec{r}(\omega_i)$ is the electro-optic-constant tensor. Below, the self-consistent electric field is calculated (in full tensor notation) from Maxwell's equations, the continuity equation, the constitutive equation for the ac current caused by the carriers, plus the piezoelectric equations of state.^{56,57}

Calculating the constitutive equation for the ac current \vec{j}^{e1-ph} on basis of the Boltzmann equation one obtains (see Appendix B)⁴⁴

$$\vec{j}^{e1-ph} = \vec{\sigma}_{eff}^{e1-ph}(\Omega, \vec{K}) \cdot \vec{F}_{eff}^{e1-ph}, \quad (49)$$

where the effective ac conductivity tensor $\vec{\sigma}_{eff}^{e1-ph}$ is given by

$$\vec{\sigma}_{eff}^{e1-ph}(\Omega, \vec{K}) = (\vec{1} - \vec{R})^{-1} \cdot \vec{\sigma}^{e1-ph}(\Omega, \vec{K}). \quad (50)$$

The critical quantity in determining the free-carrier screening of the interaction is the frequency- and wave-vector-dependent conductivity tensor $\vec{\sigma}^{e1-ph}(\Omega, \vec{K})$. Explicit expressions for the tensor \vec{R} arising from the diffusion of the non-uniformly distributed carriers and for $\vec{\sigma}^{e1-ph}$ are given in Appendix B.

The effective field \vec{F}_{eff}^{e1-ph} acting on the free carriers in a piezoelectric semiconductor originates from the piezoelectric coupling^{29,43} and the deformation-potential coupling.^{30,44} At low acoustic frequencies the first coupling mechanism tends to dominate, whereas at high frequencies the second is the more important, even in strong piezoelectric materials. The net field \vec{F}_z exerted on the free electrons because of the deformation potential is given by

$$\vec{F}_z(\vec{r}, \Omega) = (1/q)\vec{\nabla}\vec{\Xi} \cdot \vec{\Xi} \cdot \vec{u}^\mu(\vec{r}, \Omega), \quad (51)$$

where $\vec{\Xi}$ is the deformation-potential tensor and q is the numerical magnitude of the electron charge. Using Eq. (40) the effective field may be written

$$\vec{F}_{eff}^{e1-ph} = \vec{F}_{sc} - (\hat{\kappa}\hat{\kappa} \cdot \vec{\Xi} \cdot \hat{\pi}^\mu/q)(\pm K + i\Gamma)^2 u^\mu(\vec{r}, \Omega). \quad (52)$$

The adiabatic piezoelectric equation of state takes the form

$$\vec{D} = \epsilon_0 \vec{\epsilon}_r^{L,s}(\Omega) \cdot \vec{F}_{sc} + \vec{e}(\Omega) \cdot \vec{\nabla}\vec{u}^\mu, \quad (53)$$

where \vec{D} is the electric displacement, $\vec{\epsilon}_r^{L,s}(\Omega)$ is the low-frequency relative dielectric tensor, and \vec{e} is the piezoelectric tensor.

Combining the Maxwell equation $\vec{\nabla} \cdot \vec{D} = \rho$, the continuity equation $\vec{\nabla} \cdot \vec{j}^{e1-ph} = \pm i\Omega\rho$, and Eqs. (40), (49), and (53), one finds since the self-consistent electric field is almost longitudinal⁵⁸

$$\begin{aligned} \vec{F}_{sc} \approx & F_{sc} \hat{\kappa} \\ = & \left(\frac{\hat{\kappa} \cdot \vec{\sigma}_{eff}^{e1-ph} \cdot \hat{\kappa}}{\pm i\Omega\epsilon_0} \frac{\hat{\kappa} \cdot \vec{\Xi} \cdot \hat{\pi}^\mu}{q} (\pm K + i\Gamma)^2 \right. \\ & \left. - \frac{\hat{\kappa} \cdot \vec{e} \cdot \hat{\pi}^\mu \hat{\kappa}}{i\epsilon_0} (\pm K + i\Gamma) \right) \\ & \times \left[\hat{\kappa} \cdot \left(\frac{\vec{\sigma}_{eff}^{e1-ph}}{\pm i\Omega\epsilon_0} - \vec{\epsilon}_r^{L,s} \right) \cdot \hat{\kappa} \right]^{-1} u^\mu(\vec{r}, \Omega) \hat{\kappa}. \end{aligned} \quad (54)$$

Finally, inserting Eqs. (48) and (54) into Eq. (42), one obtains from the free-carrier screened indirect photoelastic effect the contribution

$$\begin{aligned} \delta\epsilon_r^I(\vec{r}, \Omega) = & -\vec{\epsilon}_{r,0}(\omega_i) \cdot \vec{r}(\omega_i) \cdot \hat{\kappa} \cdot \vec{\epsilon}_{r,0}(\omega_i) \\ & \times \left(\frac{\hat{\kappa} \cdot \vec{\sigma}_{eff}^{e1-ph}(\Omega, \vec{K}) \cdot \hat{\kappa}}{\pm i\Omega\epsilon_0} \frac{\hat{\kappa} \cdot \vec{\Xi} \cdot \hat{\pi}^\mu}{q} [\pm K(\Omega) + i\Gamma(\Omega, \vec{K})]^2 \right) \end{aligned}$$

$$-\frac{\hat{\kappa} \cdot \vec{e}(\Omega) \cdot \hat{\pi}^\mu \hat{\kappa}}{i\epsilon_0} [\pm K(\Omega) + i\Gamma(\Omega, \vec{K})] \\ \times \left[\hat{\kappa} \cdot \left(\frac{\vec{\sigma}_{\text{eff}}^{\text{el-ph}}(\Omega, \vec{K})}{\pm i\Omega\epsilon_0} - \vec{\epsilon}_r^{\text{L},s}(\Omega) \right) \cdot \hat{\kappa} \right]^{-1} u^\mu(\vec{r}, \Omega), \quad (55)$$

to the phonon-induced fluctuation in the permittivity.

C. Free-carrier plasma

The traveling elastic wave causes, in general, a bunching of the free carriers due to piezoelectric coupling and the deformation-potential coupling. This bunching, in turn, gives rise to a modulation of the complex frequency-independent optical dielectric constant. For a nondegenerate solid-state plasma one finds in the low-wave-

vector limit^{50,59,60}

$$\delta\vec{\epsilon}_r^{\text{FC}}(\vec{r}, \Omega) = (iq/\epsilon_0\omega_d)n_1(\vec{r}, \Omega)\vec{\mu}(w, \omega_d), \quad (56)$$

where $n_1(\vec{r}, \Omega)$ is the complex amplitude of the carrier density modulation, and $\vec{\mu}(w, \omega_d)$ is the free-carrier mobility tensor at the diffracted-light frequency. As indicated, $\vec{\mu}$ depends on the energy w of the free carriers.

To derive the phonon-induced free-carrier-plasma contribution to the fluctuation in the permittivity we combine the Maxwell equation $\vec{\nabla} \cdot \vec{J}^{\text{el-ph}} = \mp i\Omega qn_1$, the constitutive equation $\vec{J}^{\text{el-ph}} = \vec{\sigma}_{\text{eff}}^{\text{el-ph}} \cdot \vec{F}_{\text{eff}}^{\text{el-ph}}$, and Eq. (40) to find the relation $qn_1 = -(K \mp i\Gamma)\hat{\kappa} \cdot \vec{\sigma}_{\text{eff}}^{\text{el-ph}} \cdot \vec{F}_{\text{eff}}^{\text{el-ph}}/\Omega$. Inserting this expression in Eq. (56) and utilizing Eqs. (52) and (54) one obtains

$$\delta\vec{\epsilon}_r^{\text{FC}}(\vec{r}, \Omega) = \frac{-i[K(\Omega) \pm i\Gamma(\Omega, \vec{K})]^2}{\epsilon_0\omega_d\Omega} \frac{\hat{\kappa} \cdot \vec{\sigma}_{\text{eff}}^{\text{el-ph}}(\Omega, \vec{K}) \cdot \hat{\kappa}}{\hat{\kappa} \cdot [\vec{\sigma}_{\text{eff}}^{\text{el-ph}}(\Omega, \vec{K})/(\pm i\Omega\epsilon_0) - \vec{\epsilon}_r^{\text{L},s}(\Omega)] \cdot \hat{\kappa}} \\ \times \left(\frac{1}{q} [K(\Omega) \pm i\Gamma(\Omega, \vec{K})] \hat{\kappa} \cdot \vec{\epsilon}_r^{\text{L},s}(\Omega) \cdot \hat{\kappa} \hat{\kappa} \cdot \vec{E} \cdot \hat{\pi}^\mu \pm \frac{i}{\epsilon_0} \hat{\kappa} \cdot \vec{e}(\Omega) \cdot \hat{\pi}^\mu \hat{\kappa} \right) \vec{\mu}(w, \omega_d) u^\mu(\vec{r}, \Omega). \quad (57)$$

IV. BRILLOUIN SCATTERING KINEMATICS

Assuming the incident light beam to be describable by a plane, monochromatic wave over the interaction volume V one has

$$\vec{E}_i^\theta(\vec{r}, \omega_i) = \vec{E}_i^\theta(0, \omega_i) e^{(i\kappa_i^\theta - \gamma_i^\theta)\hat{s}_i \cdot \vec{r}}, \quad (58)$$

where $\vec{k}_i^\theta = \kappa_i^\theta \hat{s}_i$ is the (real) wave vector for the incident light, and γ_i^θ is the attenuation coefficient. The polarization state of the beam is denoted by θ . The last quantity can be decomposed into

$$\gamma_i^\theta = \gamma_i^{\theta,0}(\omega_i, \vec{k}_i^\theta) + \gamma_i^{\theta,\text{Br}}, \quad (59)$$

where $\gamma_i^{\theta,0}(\omega, \vec{k}_i^\theta)$ gives the damping of a freely propagating optical wave in an absorbing medium. If one knows \hat{s}_i and $\vec{\epsilon}_{r,0}(\omega_i)$, \vec{k}_i^θ and $\gamma_i^{\theta,0}$ can be calculated from Eq. (25). The weak absorption implies that one can neglect the ellipticity of the vibrations in almost all crystallographic directions.⁴⁹ The regions of appreciable ellipticity are in general restricted to the neighborhood of four special directions, close to the optic axes, where the polarization is circular. In this approximation the directions of the electric displacement eigenvectors belonging to a given \hat{s}_i are the same as for a nonabsorbing crystal which has the same real dielectric constant. An additional depletion (or amplification) of the incident beam γ_i^{Br} arises from the Brillouin scattering process. In the present work we shall omit giving the explicit expression for γ_i^{Br} and only notice that γ_i^{Br} must be taken into account when dealing with intense scattering effects.^{3,20,61}

Next, let us write the phonon-induced fluctuation in the dielectric tensor on the form

$$\delta\vec{\epsilon}_r(\vec{r}, \Omega) = \delta\vec{\epsilon}_r(0, \Omega) e^{(i\kappa - \Gamma)\hat{\kappa} \cdot \vec{r}}. \quad (60)$$

Combining Eqs. (45), (47), (55), and (57) it is easy to write down the explicit expression for the space-independent factor $\delta\vec{\epsilon}_r(0, \Omega)$.

The most important contributions to the frequency- and direction-dependent sound attenuation coefficient $\Gamma(\Omega, \vec{K})$ arise from the electron-phonon interaction ($\Gamma^{\text{el-ph}}$), from the elastic anharmonicity ($\Gamma^{\text{ph-ph}}$), from the Brillouin scattering process (Γ^{Br}), and from boundary scattering (Γ^{B}). Thus,

$$\Gamma(\Omega, \vec{K}) = \Gamma^{\text{el-ph}} + \Gamma^{\text{ph-ph}} + \Gamma^{\text{Br}} + \Gamma^{\text{B}}. \quad (61)$$

In a piezoelectric semiconductor the main contribution to $\Gamma^{\text{el-ph}}$ is due to the piezoelectric coupling and the deformation-potential coupling. It is well known that a stimulated phonon emission occurs, i. e., $\Gamma^{\text{el-ph}} < 0$, for the acoustoelectrically active modes if the component of the free-carrier drift velocity in the direction $\hat{\kappa}$ exceeds the phase velocity of sound in this direction. In a certain region of phonon frequencies this may lead to a net gain of the acoustic wave, i. e., $\Gamma < 0$. The explicit expression for $\Gamma^{\text{el-ph}}$ is given in Sec. V.

Various theoretical expressions have been proposed for the nonelectronic lattice loss constant $\Gamma^{\text{ph-ph}}$. Here we shall only refer to the literature on that subject.⁶²⁻⁶⁹ Most of these theories predict a Ω^2 dependence of $\Gamma^{\text{ph-ph}}$. Unfortunately, this frequency dependence is inconsistent with many experiments performed with single-frequency

waves^{70,71} or with a spectrum of acoustoelectrically active or inactive waves amplified directly, or indirectly via mode conversion, from the thermal background.^{4,6,7,13,72} For many-frequency waves several experiments indicate that $\Gamma^{\text{ph-ph}}$ follows a $\Omega^{3/2}$ law over more than three decades at room temperature.^{4,7,13} A few experiments seem to confirm the Ω^2 dependence.^{73,74}

The depletion of the phonon beam caused by the coupling to the photon beam can normally be neglected when dealing with weak-scattering effects. A simple phenomenological treatment of the boundary scattering has been given by Ziman⁴⁵ and applied in a study of off-axis acoustoelectric domain propagation in semiconducting CdS.⁴

Combining Eqs. (13), (58), and (60) the integral in Eq. (34) can be written on the form

$$\vec{I}^\varphi = \epsilon_0 \delta \vec{\epsilon}_r(0, \Omega) \cdot \vec{E}_i^\varphi(0, \omega_i) V \Phi^\varphi \quad \varphi = 1, 2. \quad (62)$$

If the scattering volume is a rectangular parallelepiped ($V = L_1 L_2 L_3$) the numerical magnitude of Φ^φ can be found by straightforward calculations. The result is [integration ranges $(-\frac{1}{2}L_j, \frac{1}{2}L_j)$]

$$|\Phi^\varphi|^2 = \frac{1}{V^2} \left| \int_V \exp[i(\vec{k}_i^\varphi \pm \vec{K} - \vec{k}_R^\varphi) - (\gamma_i^\varphi \hat{s}_i + \Gamma \hat{k})] \cdot \vec{r}' d\vec{r}' \right|^2 = \prod_{j=1}^3 \left(\frac{\sin^2(\frac{1}{2}\Delta k_j^\varphi L_j) + \sinh^2(\frac{1}{2}\beta_j^\varphi L_j)}{(\frac{1}{2}\Delta k_j^\varphi L_j)^2 + (\frac{1}{2}\beta_j^\varphi L_j)^2} \right). \quad (63)$$

In Eq. (63) we have introduced the abbreviation $\Delta \vec{k}^\varphi = \vec{k}_i^\varphi \pm \vec{K} - \vec{k}_R^\varphi$, and $\vec{\beta}^\varphi = \gamma_i^\varphi \hat{s}_i + \Gamma \hat{k}$. For $\vec{\beta}^\varphi = 0$, Eq. (63) reduces to the corresponding equation for nondecaying incident photon and phonon beams obtained by Nelson *et al.*^{23,53} It should be noticed that L_j ($j = 1, 2, 3$) depends on the scattering geom-

etry, since L_j is a function of the angle between the Poynting vectors of the incident light and sound waves. It can be shown that $|\Phi^\varphi|^2$, independent of the vectorial attenuation coefficient $\vec{\beta}^\varphi$, attains its maximum value $|\Phi_{\text{max}}^\varphi|^2 = \sinh^2(\frac{1}{2}\beta_j^\varphi L_j)/(\frac{1}{2}\beta_j^\varphi L_j)^2$ for $\Delta \vec{k}^\varphi = 0$. This means that a significant scattered signal occurs only if the wave vector of the scattered light, \vec{k}_d^φ , satisfies the Bragg (or phase matching) condition

$$\vec{k}_d^\varphi = \vec{k}_i^\varphi \pm \vec{K}^\mu. \quad (64)$$

In a particle picture this relation represents the conservation of pseudomomentum in an inelastic first-order scattering event between the polarization states θ and φ . The plus sign corresponds to a phonon absorption, the minus sign to a phonon emission. Combined with the energy-conservation law

$$\omega_d = \omega_i \pm \Omega. \quad (65)$$

Eq. (64) forms the basis for a calculation of the Brillouin scattering kinematics.¹⁴

V. EQUATION OF MOTION FOR THE LATTICE VIBRATIONS

In a piezoelectric crystal, the equation of motion for the lattice vibrations is given by

$$\rho_0 \frac{\partial^2}{\partial t^2} \vec{u}^\mu(\vec{r}, t) = \vec{\nabla} \cdot \vec{c} \cdot \vec{\nabla} \vec{u}^\mu(\vec{r}, t) - \vec{\nabla} \cdot \vec{e} \cdot \vec{F}_{\text{sc}}(\vec{r}, t), \quad (66)$$

where \vec{c} is the elastic stiffness tensor for constant electric field, and ρ_0 is the mass density of the ions. Inserting Eqs. (39), (40) with Γ^μ replaced by $\Gamma^{\mu, \text{el-ph}}$, and Eq. (54) in Eq. (66) one obtains after a few straightforward calculations

$$\rho_0 \left(\frac{\Omega}{\pm K + i\Gamma^{\text{el-ph}}} \right)^2 \hat{\pi}^\mu = \vec{c} \cdot \hat{\pi}^\mu \hat{k} \cdot \hat{k} + \frac{1}{\epsilon_0} \vec{e} \cdot \hat{k} \cdot \hat{k} \left(\frac{\hat{k} \cdot \vec{e} \cdot \hat{\pi}^\mu \hat{k} - (1/q) \hat{k} \cdot \vec{\sigma}_{\text{eff}}^{\text{el-ph}}(\Omega, \vec{K}) \cdot \hat{k} \hat{k} \cdot \vec{\Xi} \cdot \hat{\pi}^\mu [(\pm K + i\Gamma^{\text{el-ph}})/(\pm \Omega)]}{\hat{k} \cdot [\vec{\epsilon}_r^{L,s}(\Omega) \pm i\vec{\sigma}_{\text{eff}}^{\text{el-ph}}(\Omega, \vec{K})]/(\epsilon_0 \Omega)} \cdot \hat{k} \right). \quad (67)$$

For any given direction of the acoustic wave vector, specified by \hat{k} , the polarization vectors $\hat{\pi}^\mu$ can be determined by solving the eigenvalue problem of Eq. (67). A detailed discussion of phonon propagation in anisotropic piezoelectric semiconductors will be given elsewhere.

Approximate expressions for the acoustic phase velocity $V_p^\mu(\Omega, \vec{K}) = \Omega/K$ and the attenuation coefficient $\Gamma^{\text{el-ph}}(\Omega, \vec{K})$ can be obtained by solving the equation

$$\rho_0 \left(\frac{\Omega}{\pm K + i\Gamma^{\text{el-ph}}} \right)^2 = \hat{k} \cdot \hat{\pi}^\mu \cdot \vec{c} \cdot \hat{k} \cdot \hat{\pi}^\mu + \frac{1}{\epsilon_0} \hat{\pi}^\mu \cdot \hat{k} \cdot \vec{e} \cdot \hat{k} \frac{\hat{\pi}^\mu \cdot \hat{k} \cdot \vec{e} \cdot \hat{k} - (1/q) \hat{k} \cdot \vec{\sigma}_{\text{eff}}^{\text{el-ph}} \cdot \hat{k} \hat{k} \cdot \vec{\Xi} \cdot \hat{\pi}^\mu [(\pm K + i\Gamma^{\text{el-ph}})/(\pm \Omega)]}{\hat{k} \cdot (\vec{\epsilon}_r^{L,s} \pm i\vec{\sigma}_{\text{eff}}^{\text{el-ph}}/\epsilon_0 \Omega) \cdot \hat{k}}, \quad (68)$$

with respect to $\Omega/(\pm K + i\Gamma)$, assuming that the polarization vectors $\hat{\pi}^\mu$ have been evaluated from the pure elastic eigenvalue problem

$$\rho_0 (\Omega/K)^2 \hat{\pi}^\mu = \vec{c} \cdot \hat{\pi}^\mu \hat{k} \cdot \hat{k}. \quad (69)$$

If the deformation-potential coupling can be neg-

lected Eq. (68) gives directly the anisotropic formulas

$$V_p^\mu(\Omega, \vec{K}, \vec{\Xi}=0) = \left(\frac{\hat{k} \cdot \hat{\pi}^\mu \cdot \vec{c} \cdot \hat{k} \cdot \hat{\pi}^\mu}{\rho_0} \right)^{1/2} \times \left\{ 1 + \frac{1}{2\epsilon_0} \frac{(\hat{k} \cdot \vec{e} \cdot \hat{\pi}^\mu \hat{k})^2}{\hat{k} \cdot \hat{\pi}^\mu \cdot \vec{c} \cdot \hat{k} \cdot \hat{\pi}^\mu} \times \text{Re} \left[\hat{k} \cdot \left(\vec{\epsilon}_r^{L,s} \pm i \frac{\vec{\sigma}_{\text{eff}}^{\text{ol-ph}}}{\epsilon_0 \Omega} \right) \cdot \hat{k} \right]^{-1} \right\}, \quad (70)$$

for the phase velocity, and

$$\Gamma^{\mu, \text{ol-ph}}(\Omega, \vec{K}, \vec{\Xi}=0) = \mp \frac{\Omega}{2V_p^\mu \epsilon_0} \frac{(\hat{\pi}^\mu \cdot \hat{k} \cdot \vec{e} \cdot \hat{k})^2}{\hat{k} \cdot \hat{\pi}^\mu \cdot \vec{c} \cdot \hat{k} \cdot \hat{\pi}^\mu}$$

$$\times \text{Im} \left[\hat{k} \cdot \left(\vec{\epsilon}_r^{L,s} \pm i \frac{\vec{\sigma}_{\text{eff}}^{\text{ol-ph}}}{\epsilon_0 \Omega} \right) \cdot \hat{k} \right]^{-1}, \quad (71)$$

for the amplitude attenuation coefficient in the weak-coupling approximation $\Omega/V_p^\mu(\Omega, \vec{K}) \gg \Gamma^{\text{ol-ph}}(\Omega, \vec{K})$. For $K \ll 1$ (1 being the mean free path of the free carriers) Eqs. (70) and (71) are reduced to the equations obtained by Klein⁷⁵ in his macroscopic theory of anisotropic acoustic amplification.

Including the deformation-potential coupling the general expressions for the amplitude attenuation coefficient and the phase velocity take the form

$$V_p^\mu(\Omega, \vec{K}) = \left(\frac{\hat{k} \cdot \hat{\pi}^\mu \cdot \vec{c} \cdot \hat{k} \cdot \hat{\pi}^\mu}{\rho_0} \right)^{1/2} \left\{ 1 + \frac{1}{2\epsilon_0} \frac{(\hat{k} \cdot \vec{e} \cdot \hat{\pi}^\mu \hat{k})^2}{\hat{k} \cdot \hat{\pi}^\mu \cdot \vec{c} \cdot \hat{k} \cdot \hat{\pi}^\mu} \text{Re} \left[\hat{k} \cdot \left(\vec{\epsilon}_r^{L,s} \pm i \frac{\vec{\sigma}_{\text{eff}}^{\text{ol-ph}}}{\epsilon_0 \Omega} \right) \cdot \hat{k} \right]^{-1} - \frac{\Omega \rho_0^{1/2}}{2q} \frac{(\hat{k} \cdot \vec{e} \cdot \hat{\pi}^\mu \hat{k})(\hat{k} \cdot \vec{\Xi} \cdot \hat{\pi}^\mu)}{(\hat{k} \cdot \hat{\pi}^\mu \cdot \vec{c} \cdot \hat{k} \cdot \hat{\pi}^\mu)^{3/2}} \text{Re} \left(\frac{\hat{k} \cdot (\vec{\sigma}_{\text{eff}}^{\text{ol-ph}}/\epsilon_0 \Omega) \cdot \hat{k}}{\hat{k} \cdot (\vec{\epsilon}_r^{L,s} \pm i \vec{\sigma}_{\text{eff}}^{\text{ol-ph}}/\epsilon_0 \Omega) \cdot \hat{k}} \right) \right\} \quad (72)$$

and

$$\Gamma^{\mu, \text{ol-ph}}(\Omega, \vec{K}) = \mp \frac{\Omega \rho_0^{1/2}}{2\epsilon_0} \frac{(\hat{k} \cdot \vec{e} \cdot \hat{\pi}^\mu \hat{k})^2}{(\hat{k} \cdot \hat{\pi}^\mu \cdot \vec{c} \cdot \hat{k} \cdot \hat{\pi}^\mu)^{3/2}} \text{Im} \left[\hat{k} \cdot \left(\vec{\epsilon}_r^{L,s} \pm i \frac{\vec{\sigma}_{\text{eff}}^{\text{ol-ph}}}{\epsilon_0 \Omega} \right) \cdot \hat{k} \right]^{-1} \pm \frac{\Omega^2 \rho_0}{2q} \frac{(\hat{k} \cdot \vec{e} \cdot \hat{\pi}^\mu \hat{k})(\hat{k} \cdot \vec{\Xi} \cdot \hat{\pi}^\mu)}{(\hat{k} \cdot \hat{\pi}^\mu \cdot \vec{c} \cdot \hat{k} \cdot \hat{\pi}^\mu)^2} \times \text{Im} \left(\frac{\hat{k} \cdot (\vec{\sigma}_{\text{eff}}^{\text{ol-ph}}/\epsilon_0 \Omega) \cdot \hat{k}}{\hat{k} \cdot (\vec{\epsilon}_r^{L,s} \pm i \vec{\sigma}_{\text{eff}}^{\text{ol-ph}}/\epsilon_0 \Omega) \cdot \hat{k}} \right), \quad (73)$$

in the weak-coupling approximation.

VI. QUANTUM-MECHANICAL DESCRIPTION OF A SPATIALLY DECAYING LATTICE WAVE

Describing the complex time-independent displacement [Eq. (40)] by its spatial Fourier components (denoted by \vec{q}), the Fourier amplitudes of the displacement,

$$\vec{u}_{\vec{q}}^\mu = \hat{\pi}^\mu \int_{\vec{r}} u_0^\mu \exp[i(\pm \vec{K}^\mu - \vec{q}) \cdot \vec{r} - \Gamma^\mu \hat{k} \cdot \vec{r}] d\vec{r}, \quad (74)$$

are given by [integration ranges $(-\frac{1}{2}a_i | \frac{1}{2}a_i)$]

$$|\vec{u}_{\vec{q}}^\mu|^2 = |u_0^\mu|^2 V_s^2 \prod_{i=1}^3 \frac{\sin^2(\frac{1}{2} \Delta K_i^\mu a_i) + \sinh^2(\frac{1}{2} \Gamma_i^\mu a_i)}{(\frac{1}{2} \Delta K_i^\mu a_i)^2 + (\frac{1}{2} \Gamma_i^\mu a_i)^2}, \quad (75)$$

where $\Delta \vec{K}^\mu = \vec{K}^\mu \mp \vec{q}$. In the derivation of Eq. (75) it has been assumed that the volume of the solid occupied by the acoustic wave is a rectangular parallelepiped, i. e., $V_s = a_1 a_2 a_3$. To obtain a quantum-mechanical description of the lattice vibrations one must make the following replacement for the squared Fourier amplitude of the displacement¹⁹

$$|\vec{u}_{\vec{q}}^\mu|^2 \rightarrow |\langle n \pm 1 | \vec{u}_{\vec{q}}^\mu | n \rangle|^2 = V_s \hbar \Omega N_{\vec{q}}^\mu \pm 2\rho_0 \Omega^2, \quad (76)$$

where the plus sign corresponds to the Stokes component (phonon creation) and the minus sign to the anti-Stokes component (phonon annihilation). The

occupation number for phonons of wave vector \vec{q} is $N_{\vec{q}}^\mu$, and $N_{\vec{q}}^{\mu*}$ is defined by $N_{\vec{q}}^{\mu*} = N_{\vec{q}}^\mu + 1$ and $N_{\vec{q}}^{\mu-} = N_{\vec{q}}^\mu$.

The squared amplitude of the displacement $|u_0^\mu|^2$ for the damped sound wave can be expressed in terms of the occupation number $N_{\vec{K}}^\mu$ by combining Eqs. (75) and (76). Thus,

$$|u_0^\mu|^2 = \frac{1}{V_s} \frac{\hbar \Omega N_{\vec{K}}^{\mu*}}{2\rho_0 \Omega^2} \prod_{i=1}^3 \frac{(\frac{1}{2} \Gamma_i^\mu a_i)^2}{\sinh^2(\frac{1}{2} \Gamma_i^\mu a_i)}. \quad (77)$$

VII. EFFECTIVE DIFFERENTIAL BRILLOUIN SCATTERING CROSS SECTION

Combining the time-averaged Poynting vector

$$\langle \vec{S}_i^\theta \rangle = \frac{1}{2} \text{Re} \{ [\vec{E}_i^\theta(\vec{r}, \omega_i)]^* \times \vec{H}_i^\theta(\vec{r}, \omega_i) \}$$

and the Maxwell equation

$$\vec{\nabla} \times \vec{E}_i^\theta(\vec{r}, \omega_i) = i\omega_i \mu_0 \vec{H}_i^\theta(\vec{r}, \omega_i),$$

the spatial-dependent intensity of the incident light beam, which polarization state is θ , can be written

$$|\langle \vec{S}_i^\theta(\vec{r}) \rangle| = \frac{1}{2} (\epsilon_0/\mu_0)^{1/2} |\text{Re}(n_i^\theta(\omega_i, \hat{s}_i) \{ \hat{s}_i \cdot \vec{E}_i^\theta(\vec{r}, \omega_i) \}^2 - \vec{E}_i^\theta(\vec{r}, \omega_i) \cdot [\hat{s}_i \cdot \vec{E}_i^\theta(\vec{r}, \omega_i)] \rangle| \approx \frac{1}{2} \epsilon_0 c n_{\vec{K}}^\theta(\omega_i, \hat{s}_i) |\vec{E}_i^\theta(\vec{r}, \omega_i)|^2 \cos \delta^\theta(\omega_i, \hat{s}_i), \quad (78)$$

where $n_i^\theta(\omega_i, \hat{s}_i)$ is the complex refractive index of the incident light, and $\delta^\theta(\omega_i, \hat{s}_i)$ is the angle between the Poynting vector and the wave vector. In the derivation of Eq. (78) we have neglected the depletion of the beam arising from the Brillouin scattering process.

It has been shown by Nelson *et al.*³¹ that if $d\Omega_\epsilon$ is an element of solid angle of Poynting vector directions the corresponding element of solid angle of the wave vectors $d\Omega_p$ will be given by

$$d\Omega_p = \frac{\cos\delta^\theta(\omega_d, \hat{s}_d)}{\chi^\theta [k_R^\theta(\omega_d, \hat{s}_d)]^2} d\Omega_\epsilon, \quad (79)$$

where χ^θ is the Gaussian curvature in \vec{k}_R space of the $\omega(\vec{k}_R)$ surface, for the φ polarization at the point where the surface normal is parallel to the direction of observation. The normalization condition in Eq. (23) requires δ^θ to be given by $\cos\delta^\theta = [n_R^\theta |\vec{e}_R^\theta|]^{-1}$.

According to Eq. (79) the power per unit solid angle of Poynting vectors, $dP^\theta/d\Omega_\epsilon$, is related to the power per unit solid angle of wave vectors $dP^\theta/d\Omega_p$, via⁷⁸

$$dP^\theta/d\Omega_\epsilon = \{\cos\delta^\theta / [\chi^\theta (k_R^\theta)^2]\} dP^\theta/d\Omega_p.$$

The effective differential Brillouin scattering cross section between the polarization states θ and φ , $d\sigma_B^{\theta, \varphi}/d\Omega_\epsilon$, is defined through the equation

$$\frac{d\sigma_B^{\theta, \varphi}}{d\Omega_\epsilon} \equiv \frac{dP^\theta}{d\Omega_\epsilon} \frac{1}{|\langle \vec{S}_i^\theta(\vec{r}=0) \rangle| A}, \quad (80)$$

i. e., as the scattered power per unit solid angle of Poynting vectors divided by the incident power at the center of the interaction region ($\vec{r}=0$). The cross-sectional area of the incident beam has been denoted by A .

Introducing the unit field vectors $\hat{e}_i^\theta = \vec{E}_i^\theta(\vec{r}, \omega_i) / \vec{E}_i^\theta(\vec{r}, \omega_i)$ and $\hat{e}_d^\theta \equiv \hat{e}_R^\theta = \hat{e}_R^\theta(\omega_d, \hat{s}_d) = \vec{e}_R^\theta n_R^\theta \cos\delta^\theta$, the normalized fluctuation in the dielectric-constant tensor $\delta\vec{\epsilon}_{r,N}(\Omega) = \delta\vec{\epsilon}_r(\vec{r}, \Omega) / u^\mu(\vec{r}, \Omega)$, and the approximation $\omega = \omega_d \approx \omega_i$, Eqs. (35), (38), (62)–(64), and (77)–(80) can be combined to give the following expression for the effective differential Brillouin-scattering cross section between the polarization states θ and φ in an anisotropic, piezoelectric semiconductor:

$$\begin{aligned} \frac{d\sigma_B^{\theta, \varphi}}{d\Omega_\epsilon} &= \left(\frac{\omega}{4\pi c}\right)^2 \frac{|\hat{e}_d^\theta \cdot \{\vec{1} - [\text{Im}\vec{\epsilon}_{r,0}(\omega)] \cdot \sum_{\alpha=1,2} \hat{e}_d^\alpha \hat{e}_d^\alpha Z^{\theta, \eta}\} \cdot [\delta\vec{\epsilon}_{r,N}^D(\Omega) + \delta\vec{\epsilon}_{r,N}^I(\Omega) + \delta\vec{\epsilon}_{r,N}^{FC}(\Omega)] \cdot \hat{e}_i^\theta|^2}{\chi^\theta n_R^\theta(\omega, \hat{s}_i) n_R^\theta(\omega, \hat{s}_d) \cos\delta^\theta(\omega, \hat{s}_i) \cos\delta^\theta(\omega, \hat{s}_d)} \\ &\times \prod_{m=1}^3 \left(\frac{\Gamma_m^\mu a_m \sinh(\frac{1}{2} \beta_m^\theta L_m)}{\beta_m^\theta L_m \sinh(\frac{1}{2} \Gamma_m^\mu a_m)} \right)^2 \frac{V^2}{V_s A} \frac{\hbar \Omega N_{\vec{k}}^{\mu \pm}}{2\rho_0 \Omega^2}, \end{aligned} \quad (81)$$

where

$$Z^{\theta, \eta} = \frac{i}{\cos^2 \delta^\theta(\omega, \hat{s}_d)} \frac{1 - (\Phi^\theta / \Phi^\eta) \exp\{i(\omega/c)[n_R^\theta(\omega, \hat{s}_d) - n_R^\eta(\omega, \hat{s}_d)]r\}}{[n_R^\theta(\omega, \hat{s}_d)]^2 - [n_R^\eta(\omega, \hat{s}_d)]^2}. \quad (82)$$

Since usually $(\omega/c)|n_R^\theta - n_R^\eta| r \gg 1$ for $\eta \neq \varphi$ one cannot observe the spatial variation of the rapidly oscillating factor $\exp[i(\omega/c)(n_R^\theta - n_R^\eta)r]$. Thus, by averaging the cross section over the interval $(r|r + (2\pi c/\omega)(n_R^\theta - n_R^\eta))$ one obtains

$$\begin{aligned} \frac{d\sigma_B^{\theta, \varphi}}{d\Omega_\epsilon} &= \left(\frac{\omega}{4\pi c}\right)^2 \frac{1}{\chi^\theta n_R^\theta n_R^\varphi \cos\delta^\theta \cos\delta^\varphi} \prod_{m=1}^3 \frac{(\frac{1}{2} \Gamma_m^\mu a_m)^2}{\sinh^2(\frac{1}{2} \Gamma_m^\mu a_m)} \frac{V^2}{V_s A} \frac{\hbar \Omega N_{\vec{k}}^{\mu \pm}}{2\rho_0 \Omega^2} \\ &\times \left(|\Phi^\theta|^2 |\hat{e}_d^\theta \cdot \left\{ \vec{1} - \frac{1}{2} \omega \tau_r (\text{Im}\vec{\epsilon}_{r,0}) \cdot \vec{e}_d^\theta \vec{e}_d^\theta - i \left[1 - \left(\frac{n_R^\theta}{n_R^\varphi} \right)^2 \right]^{-1} (\text{Im}\vec{\epsilon}_{r,0}) \cdot \vec{e}_d^\theta \vec{e}_d^\theta \right\} \cdot \delta\vec{\epsilon}_{r,N} \cdot \hat{e}_i^\theta|^2 \right. \\ &\left. + |\Phi^\varphi|^2 \left[1 - \left(\frac{n_R^\varphi}{n_R^\theta} \right)^2 \right]^{-2} |\hat{e}_d^\varphi \cdot (\text{Im}\vec{\epsilon}_{r,0}) \cdot \vec{e}_d^{\theta'} \vec{e}_d^{\theta'} \cdot \delta\vec{\epsilon}_{r,N} \cdot \hat{e}_i^\theta|^2 \right), \quad \varphi' \neq \varphi, \end{aligned} \quad (83)$$

where $\delta\vec{\epsilon}_{r,N} = \delta\vec{\epsilon}_{r,N}^D + \delta\vec{\epsilon}_{r,N}^I + \delta\vec{\epsilon}_{r,N}^{FC}$ and $\tau_r = (r/c)n_R^\theta$. The phase-matching condition requires that one must insert $\Delta\vec{k}^\theta = 0$ and $\Delta\vec{k}^{\theta'} = (n_R^\theta - n_R^{\theta'}) (\omega/c) \hat{s}_d$ in the explicit expressions for $|\Phi^\theta|^2$ and $|\Phi^{\theta'}|^2$. On a rough estimate where the tensor notation is suppressed, the condition $(\omega/c)|n_R^\theta - n_R^{\theta'}| r \gg 1$ implies that terms containing the φ' polarization can be neglected. In this approximation the cross section takes the simple form⁷⁹

$$\frac{d\sigma_B^{\theta, \varphi}}{d\Omega_\epsilon} = \left(\frac{\omega}{4\pi c}\right)^2 \frac{|\hat{e}_d^\theta \cdot \delta\vec{\epsilon}_{r,N} \cdot \hat{e}_i^\theta|^2}{\chi^\theta n_R^\theta n_R^\varphi \cos\delta^\theta \cos\delta^\varphi} \left[1 - \frac{1}{2} \omega \tau_r |\vec{e}_d^\theta|^2 |\hat{e}_d^\theta \cdot \text{Im}\vec{\epsilon}_{r,0} \cdot \hat{e}_d^\theta|^2 \right] \prod_{m=1}^3 \left(\frac{\Gamma_m^\mu a_m \sinh(\frac{1}{2} \beta_m^\theta L_m)}{\beta_m^\theta L_m \sinh(\frac{1}{2} \Gamma_m^\mu a_m)} \right)^2 \frac{V L_\theta}{V_s} \frac{\hbar \Omega N_{\vec{k}}^{\mu \pm}}{2\rho_0 \Omega^2}, \quad (84)$$

where L_θ is the length of the scattering volume in the direction $\vec{S}_i^\theta / |\vec{S}_i^\theta|$. From Eq. (83) one obtains the interesting result that $d\sigma_B^{\theta, \varphi}/d\Omega_\epsilon \approx 0$ for $\tau_c/\tau_r \approx \pi$.

In the limit of zero conductivity the present anal-

ysis is in agreement with that of Nelson *et al.*²³ To compare the two results one must notice that the final formula of Nelson *et al.*²³ applies for a thermal-equilibrium distribution of phonons whereas

Eq. (84) of this work is valid for a decaying mode. By combining Eqs. (2.15) and (2.30) in Ref. 23 and by utilizing the fact that $\tilde{\mathbf{e}}^m = \hat{\mathbf{e}}^m / (n_R^m \cos \delta^m)$, $m = \theta, \varphi$, and $d\sigma_B^{\theta, \varphi} / d\Omega_{\mathbf{k}} = |\tilde{\mathbf{S}}_d^{\theta, \varphi}| r^2 / (|\tilde{\mathbf{S}}_i^{\theta, \varphi}| A)$, the agreement between the two papers is obvious.

A recent paper by the author²² treats Brillouin scattering from nondecaying modes in hexagonal, insulating crystals. In this paper the scattering cross sections were defined on basis of the components of the incident and scattered photon intensities along $\hat{\mathbf{k}}_i$ and $\hat{\mathbf{k}}_d$. A comparison of the anisotropic dipole approximation method used in Ref. 22 and the Green's-function formalism used in this work is given elsewhere.⁸⁰ For the calculation of relative scattering cross sections in CdS the difference between \hat{R} and \hat{k}_d is of no importance.

It should be recalled that the effective differential scattering cross section calculated in this section refers to the cross section per unit frequency range integrated over the line shape of the considered Stokes or anti-Stokes component.

ACKNOWLEDGMENTS

The author is very grateful to Professor K. Maack Bisgård for his interest in this work, and

to Professor P. O. Neerup for many stimulating discussions concerning the Green's-function formalism.

APPENDIX A: FAR-FIELD DYADIC GREEN'S FUNCTION IN A SEMICONDUCTOR

By means of the residue theorem the following relations (x real) can easily be verified:

$$\lim_{\eta \rightarrow 0} \int_{-\infty}^{\infty} \frac{x e^{i a x} dx}{x^2 - (\sigma + i \eta)^2} = i \pi e^{i \sigma a}, \quad (\text{A1})$$

$$\lim_{\eta \rightarrow 0} \int_{-\infty}^{\infty} \frac{x e^{i a x} dx}{[x^2 - (\sigma + i \eta)^2]^2} = \frac{\pi a}{2 \sigma} e^{i \sigma a}, \quad (\text{A2})$$

and

$$\begin{aligned} \lim_{\eta_1, \eta_2 \rightarrow 0} \int_{-\infty}^{\infty} \frac{x e^{i a x} dx}{\prod_{p=1}^2 [x^2 - (\sigma_p + i \eta_p)^2]} \\ = \frac{i \pi}{\sigma_1^2 - \sigma_2^2} (e^{i \sigma_1 a} - e^{i \sigma_2 a}). \end{aligned} \quad (\text{A3})$$

In order to obtain the usual outgoing Green's function a small imaginary part $\eta > 0$, limiting zero after the contour integration, has been added to the above denominators.

By combining Eqs. (24), (28), (29), and (A1)–(A3) the far-field Green's function becomes in a first-order approximation

$$\begin{aligned} \bar{\mathbf{G}}(\vec{\mathbf{r}}, \vec{\mathbf{r}}') = \sum_{\nu=1,2} (k_R^\nu)^2 \frac{e^{i k_R^\nu R}}{4 \pi R} \tilde{\mathbf{e}}_R^\nu \tilde{\mathbf{e}}_R^\nu \cdot \left\{ \bar{\mathbf{1}} - \frac{1}{2} i k_R^\nu R [\text{Im} \bar{\boldsymbol{\epsilon}}_{r,0}(\omega_d)] \cdot \tilde{\mathbf{e}}_R^\nu \tilde{\mathbf{e}}_R^\nu \right\} \\ + \frac{(k_R^1 k_R^2)^2}{(k_R^1)^2 - (k_R^2)^2} \frac{e^{i k_R^1 R} - e^{i k_R^2 R}}{4 \pi R} \left\{ \tilde{\mathbf{e}}_R^1 \tilde{\mathbf{e}}_R^1 \cdot [\text{Im} \bar{\boldsymbol{\epsilon}}_{r,0}(\omega_d)] \cdot \tilde{\mathbf{e}}_R^2 \tilde{\mathbf{e}}_R^2 + \tilde{\mathbf{e}}_R^2 \tilde{\mathbf{e}}_R^2 \cdot [\text{Im} \bar{\boldsymbol{\epsilon}}_{r,0}(\omega_d)] \cdot \tilde{\mathbf{e}}_R^1 \tilde{\mathbf{e}}_R^1 \right\} \\ - \sum_{\nu=1,2} \left(\frac{\hat{\mathbf{1}}_R \hat{\mathbf{1}}_R \cdot [\text{Im} \bar{\boldsymbol{\epsilon}}_{r,0}(\omega_d)] \cdot \tilde{\mathbf{e}}_R^\nu \tilde{\mathbf{e}}_R^\nu + \tilde{\mathbf{e}}_R^\nu \tilde{\mathbf{e}}_R^\nu \cdot [\text{Im} \bar{\boldsymbol{\epsilon}}_{r,0}(\omega_d)] \cdot \hat{\mathbf{1}}_R \hat{\mathbf{1}}_R}{\hat{\mathbf{1}}_R \cdot [\text{Re} \boldsymbol{\epsilon}_{r,0}(\omega_d)] \cdot \hat{\mathbf{1}}_R} \right) (k_R^\nu)^2 \frac{e^{i k_R^\nu R}}{4 \pi R}. \end{aligned} \quad (\text{A4})$$

Neglecting the nonpropagating mode Eq. (A4) is reduced to

$$\bar{\mathbf{G}}(\vec{\mathbf{r}}, \vec{\mathbf{r}}') = \sum_{\nu=1,2} \tilde{\mathbf{e}}_R^\nu \tilde{\mathbf{e}}_R^\nu \frac{e^{i k_R^\nu R}}{4 \pi R} \frac{1}{1/(k_R^\nu)^2} - i \sum_{\nu, \theta=1,2} \tilde{\mathbf{e}}_R^\nu \tilde{\mathbf{e}}_R^\nu \cdot [\text{Im} \bar{\boldsymbol{\epsilon}}_{r,0}(\omega_d)] \cdot \tilde{\mathbf{e}}_R^\theta \tilde{\mathbf{e}}_R^\theta \frac{e^{i k_R^\nu R} - e^{i k_R^\theta R}}{4 \pi R} \frac{1}{1/(k_R^\nu)^2 - 1/(k_R^\theta)^2}, \quad (\text{A5})$$

where $k_R^\nu = (\omega_d/c) n_R^2(\hat{\mathbf{1}}_R, \omega_d)$ and $\tilde{\mathbf{e}}_R^\nu = \tilde{\mathbf{e}}_R^\nu(\hat{\mathbf{1}}_R, \omega_d)$. In Eq. (A5) the first term represents the Green's function obtained by Nelson *et al.*⁵³ for nonabsorbing crystals.

APPENDIX B: SEMICLASSICAL CALCULATION OF THE EFFECTIVE CONDUCTIVITY TENSOR $\bar{\mathcal{C}}_{\text{eff}}^{\text{el-ph}}(\Omega, \vec{\mathbf{K}})$ IN A SEMICONDUCTOR

The Boltzmann equation for electrons interacting with a nondecaying acoustic wave of frequency Ω and wave vector $\vec{\mathbf{K}}$ in the presence of a dc electric field $\vec{\mathbf{F}}_0$ is^{76,77}

$$\frac{\partial f}{\partial t} + \vec{\mathbf{v}} \cdot \frac{\partial f}{\partial \vec{\mathbf{r}}} - \frac{q}{m^*} (\vec{\mathbf{F}}_0 + \vec{\mathbf{F}}_{\text{eff}}^{\text{el-ph}}) \cdot \frac{\partial f}{\partial \vec{\mathbf{v}}} = - \frac{f - f_s}{\tau(E)}, \quad (\text{B1})$$

where the effective electric field acting on the free

electrons in the presence of the acoustic wave, $\vec{\mathbf{F}}_{\text{eff}}^{\text{el-ph}}$, may be written

$$\vec{\mathbf{F}}_{\text{eff}}^{\text{el-ph}} = \vec{\mathbf{F}}_{\text{sc}} - \frac{\hat{\mathbf{k}} \hat{\mathbf{k}} \cdot \vec{\mathbf{E}} \cdot \hat{\boldsymbol{\mu}}}{q} K^2(\Omega) u^\mu(\vec{\mathbf{r}}, t, \Omega). \quad (\text{B2})$$

The electronic charge has been denoted $-q$, the effective mass of the electron assumed isotropic m^* , the energy-dependent electron relaxation time $\tau(E)$,⁵⁶ and the distribution function to which the electrons relax in the presence of the wave f_s . In a semiconductor one obtains⁷⁶

$$f_s(\vec{v}) \approx f_0(\vec{v}) + n_1 \frac{\partial f_0(\vec{v})}{\partial n_0}, \quad (\text{B3})$$

where $f_0(\vec{v})$ is the equilibrium distribution of the electrons. Treating the electrons as obeying Boltzmann statistics one has

$$f_0(\vec{v}) = n_0 \left(\frac{m^*}{2\pi k_B T} \right)^{3/2} \exp\left(\frac{-m^* v^2}{2k_B T} \right), \quad (\text{B4})$$

where n_0 is the equilibrium electron density, k_B is Boltzmann's constant, and T is the temperature. The second term on the right-hand side of Eq. (B3) arises from the fact that the scattering is local. The fluctuating part of the electron concentration is n_1 .

To determine the electron distribution function, $f(\vec{r}, \vec{v}, t)$ is decomposed as follows:

$$f = f_{dc}(\vec{v}) + g(\vec{v}) e^{i(\vec{k} \cdot \vec{r} - \Omega t)}. \quad (\text{B5})$$

The first term in Eq. (B5) represents the electron distribution function in the presence of the dc electric field but in the absence of the acoustic wave. Solving the dc part of Eq. (B1) one finds, expanding $f_0(\vec{v} - \vec{v}_d)$ to first order in \vec{v}_d ,

$$f_{dc}(\vec{v}) = f_0(\vec{v} - \vec{v}_d), \quad (\text{B6})$$

where

$$\vec{v}_d = -[q\tau(E)/m^*] \vec{F}_0 \quad (\text{B7})$$

is the drift velocity of the electrons in the dc field.

Neglecting the nonlinear term $(q/m^*) \vec{F}_{eff}^{*1-ph} \cdot \partial g(\vec{v})/\partial \vec{v}$, and taking the direction of the dc electric field to be along the z axis of our coordinate system, the ac part of Eq. (B1) turns into a simple inhomogeneous first-order differential equation with the solution

$$g(\vec{v}) = \int_{-\infty}^{v_x} \frac{\tau(E)}{v_d} \left(\frac{q}{m^*} \vec{F}_{eff}^{*1-ph} \cdot \frac{\partial f_{dc}}{\partial \vec{v}} + \frac{n_1}{\tau(E)} \frac{\partial f_0}{\partial n_0} \right) \exp\left(- \int_{v'_x}^{v_x} [i(\vec{k} \cdot \vec{v} - \Omega) + \tau^{-1}(E)] \frac{\tau(E)}{v_d} dv'_x \right) dv'_x, \quad (\text{B8})$$

where now the amplitude of the electron density fluctuation has been denoted by n_1 .

The phonon-induced current is given by

$$\vec{J}^{*1-ph} = -q \int \vec{v} g(\vec{v}) d\vec{v} = \vec{\sigma}_{eff}^{*1-ph}(\Omega, \vec{k}) \cdot \vec{F}_{eff}^{*1-ph}. \quad (\text{B9})$$

Using the continuity equation $-qn_1 V_p^\mu(\Omega, \vec{k}) = \hat{k} \cdot \vec{J}^{*1-ph}$, the effective ac conductivity tensor can be written on the form

$$\vec{\sigma}_{eff}^{*1-ph}(\Omega, \vec{k}) = (\vec{I} - \vec{R})^{-1} \cdot \vec{\sigma}^{*1-ph}(\Omega, \vec{k}), \quad (\text{B10})$$

with

$$\vec{\sigma}^{*1-ph}(\Omega, \vec{k}) = - \int_{-\infty}^{\infty} \vec{v} \left[\int_{-\infty}^{v_x} \frac{\tau(E)}{v_d} \frac{q^2}{m^*} \frac{\partial f_{dc}}{\partial \vec{v}} \exp\left(- \int_{v'_x}^{v_x} [i(\vec{k} \cdot \vec{v} - \Omega) + \tau^{-1}(E)] \frac{\tau(E)}{v_d} dv'_x \right) dv'_x \right] d\vec{v} \quad (\text{B11})$$

and

$$\vec{R}(\Omega, \vec{k}) = \int_{-\infty}^{\infty} \vec{v} \hat{k} \left[\int_{-\infty}^{v_x} \frac{1}{v_d V_p^\mu(\Omega, \vec{k})} \frac{\partial f_0}{\partial n_0} \exp\left(- \int_{v'_x}^{v_x} [i(\vec{k} \cdot \vec{v} - \Omega) + \tau^{-1}(E)] \frac{\tau(E)}{v_d} dv'_x \right) dv'_x \right] d\vec{v}. \quad (\text{B12})$$

Explicit expression for $\vec{\sigma}^{*1-ph}$ and \vec{R} have been given by Spector for the case where the electron relaxation time is energy-independent.⁷⁶ Writing the oscillating part of the distribution function on the form $g(\vec{v}) e^{-i(\vec{k} \cdot \vec{r} - \Omega t)}$, the effective ac conductivity tensor in Eq. (B10) must be replaced by $[\vec{\sigma}_{eff}^{*1-ph}(\Omega, \vec{k})]^*$.

¹For an over-all description and extensive references see N. I. Meyer and M. H. Jorgensen, *Adv. Solid State Phys.* **10**, 21 (1970).

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- ⁷⁸The far-field Green's function (and thus also \vec{E}_d and $\langle \vec{S} \rangle$) obtained by the procedure followed in this work is only appropriate in a calculation of the fictitious scattered power into a solid angle $d\Omega_g$ associated with a bundle of wave vectors in \vec{k}_R space. It follows from Eqs. (38) and (79) that the correct Poynting vector $\langle \vec{S} \rangle_{\text{corr}}$ for scattering into a solid angle $d\Omega_g$ of the bundle of rays (Poynting vectors) in ordinary space is given by $\langle \vec{S} \rangle_{\text{corr}} = (f^\nu)^2 \langle \vec{S} \rangle$ with $f^\nu = \cos \delta^\nu / [k_R^\nu (\chi^\nu)^{1/2}]$. This result is in agreement with the direct calculation of the correction factor f^ν for the Green's function for nonconducting crystals recently presented by Lax *et al.* (Ref. 31). The expressions for the scattered power $dP^\nu/d\Omega_g$ obtained by the two procedures are in agreement. An extension of the method of Lax *et al.* (Ref. 31) to conducting media will be published elsewhere.
- ⁷⁹An equivalent calculation of the Brillouin scattering cross section can be obtained by resolving the time-independent part of the atomic displacement after its spatial Fourier components. This method, which will be used in a forthcoming paper, is preferable when the spatial fluctuation in the dielectric tensor cannot be described by a single damped wave.
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