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[10] The current seems also to saturate with CdS as the emitter. Saturation is unusual and interesting in internal photoemission experiments, and it is under study.

[11] The errors of B and ω_1 are linearly correlated.

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Ionic Brillouin Effect*

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A theory is presented of the inelastic scattering of monochromatic infrared radiation by the long-wavelength acoustic vibration modes of an arbitrary crystal possessing infrared-active optical modes. Central to the theory is a lattice-dynamical calculation of the ionic contribution to the photoelastic constants of a crystal, i.e., the fourth-rank-tensor coefficients which relate the change in the dielectric tensor to the parameters describing a homogeneous deformation of the crystal. The results of this calculation show that the photoelastic tensor is not symmetric in the second pair of indices, in general, in agreement with recent predictions of Nelson and Lax. The general expression for the ionic contribution to the photoelastic tensor has a resonant character for frequencies of the incident light close to the frequencies of the infrared-active optical modes of the crystal. This general expression is specialized to the case of diatomic cubic crystals, and the magnitude of the ionic contribution to the photoelastic constants is estimated for GaAs and KCl on the basis of simple lattice-dynamical models. From these results the scattering efficiencies for these two crystals are determined. It is found for each of these crystals that the integrated scattering efficiency for Stokes (or anti-Stokes) scattering exceeds 10^{-10} cm^{-1} sr^{-1} for frequencies of the incident light within 5% of the frequency of the transverse-optical mode.

I. INTRODUCTION

The ionic contribution to the Raman, or inelastic, scattering of light by the optical phonons in a crystal has recently been shown to dominate the electronic Raman effect when the incident light is in the infrared region.[1] This raises the question of the significance of the ionic contribution to the inelastic scattering of light by the long-wavelength acoustic phonons (Brillouin scattering) when the incident light is in the infrared. In this paper we present a theory of the ionic Brillouin effect.

The phenomenological theory of the Brillouin effect has been given by Born and Huang[2] who derive an expression for the intensity of scattered light. If their result is expressed in terms of a scattering efficiency [the ratio of the number of photons scattered into unit solid angle in the frequency interval $(\omega_s, \omega_s + d\omega_s)$ to the number of incident photons], it can be written

$$\mathcal{S} = L\left(\frac{\omega_0}{c}\right)^4 \frac{k_B T}{32\pi^2 \rho} \sum_{j=1}^{3} \frac{[f(\vec{q}j)]^2}{\omega_j^2(\vec{q})} \times [\delta(\Omega - \omega_j(\vec{q})) + \delta(\Omega + \omega_j(\vec{q}))], \quad (1.1)$$

where

$$f(\vec{q}j) = \sum_{\alpha\gamma\mu\nu} n_\alpha n_{0\gamma} k_{\alpha\gamma\mu\nu} e_\mu(\vec{q}j) q_\nu . \quad (1.2)$$

In these expressions, L is the path length of the light in the crystal, ω_0 is the frequency of the incident light, c is the speed of light, k_B and T are the Boltzmann constant and absolute temperature, respectively, and ρ is the mass density of the crystal. $\omega_j(\vec{q})$ is the frequency of the normal mode of vibration defined by the wave vector $\vec{q}$ and branch index j, $\vec{q} = \vec{k}_s - \vec{k}_0$, where $\vec{k}_s$ and $\vec{k}_0$ are the wave vectors of the scattered and incident light, and the summation on j runs over the three acoustic modes of the crystal. The first term on the right-hand side of Eq. (1.1) describes anti-Stokes scattering, while the second term describes Stokes scattering. In Eq. (1.2), $\vec{n}_0$ and $\vec{n}$ are unit vectors which specify the polarization of the incident and scattered light, respectively, the coefficients $\{k_{\alpha\gamma\mu\nu}\}$ are the photoelastic constants, and $e_\mu(\vec{q}j)$ is the unit polarization vector for the acoustic mode $(\vec{q}j)$. The harmonic approximation has been employed in obtaining the results expressed by Eq. (1.1). The small difference between the frequencies of the incident and scattered light has also been neglected in obtaining Eq. (1.1).

In general, the photoelastic constants contain both an electronic and an ionic contribution. In the past, Brillouin scattering experiments have been performed with the incident light in the visible region. In this region, the electronic contribution is the dominant one and the ionic contribution is usually neglected. However, there are now available lasers which emit at frequencies in the infrared region where the ionic contribution becomes important. Therefore, in this paper, we consider the theory of the ionic, in addition to the electronic, Brillouin effect and estimate the relative importance of the two scattering processes.

In a recent paper, Nelson and Lax[3] have presented a very general theory of the photoelastic effect. Although this theory appears to be general enough to contain the dispersion of the photoelastic constants in the infrared as a special case, this topic is not discussed by Nelson and Lax.

II. MICROSCOPIC THEORY OF THE PHOTOELASTIC CONSTANTS

In this section, we derive an expression for the electronic and ionic susceptibilities of a homogeneously deformed crystal, and from them the corresponding contributions to the photoelastic constants. We then specialize our theory to the case of cubic crystals containing two inequivalent ions in a primitive unit cell, such as crystals of the rocksalt and zinc-blende structures.

For generality, we consider initially an arbitrary crystal consisting of N primitive unit cells, each of which contains r ions. We begin our analysis by expanding the potential energy of the crystal in powers of the displacements of the ions from their equilibrium positions, and in powers of the components of the macroscopic field:

$$\begin{aligned}\Phi = \Phi_0 &+ \tfrac{1}{2}\sum_{l\kappa\alpha}\sum_{l'\kappa'\beta} \Phi_{\alpha\beta}(l\kappa;l'\kappa')\,\xi_\alpha(l\kappa)\,\xi_\beta(l'\kappa') + \tfrac{1}{6}\sum_{l\kappa\alpha}\sum_{l'\kappa'\beta}\sum_{l''\kappa''\gamma} \Phi_{\alpha\beta\gamma}(l\kappa;l'\kappa';l''\kappa'')\,\xi_\alpha(l\kappa)\,\xi_\beta(l'\kappa')\,\xi_\gamma(l''\kappa'') + \cdots \\ &- \sum_\lambda E_\lambda\Big[\sum_{l\kappa\alpha} M_{\lambda\alpha}(l\kappa)\,\xi_\alpha(l\kappa) + \tfrac{1}{2}\sum_{l\kappa\alpha}\sum_{l'\kappa'\beta} M_{\lambda\alpha\beta}(l\kappa;l'\kappa')\,\xi_\alpha(l\kappa)\,\xi_\beta(l'\kappa') + \cdots\Big] \\ &- \tfrac{1}{2}\sum_{\lambda\mu} E_\lambda E_\mu \Big[P^{(0)}_{\lambda\mu} + \sum_{l\kappa\alpha} P_{\lambda\mu\alpha}(l\kappa)\,\xi_\alpha(l\kappa) + \cdots\Big] - \cdots . \qquad (2.1)\end{aligned}$$

In this expansion Φ_0 is the potential energy of the static lattice in the absence of a macroscopic field, $\xi_\alpha(l\kappa)$ is the α Cartesian component of the displacement of the κth atom in the lth primitive unit cell, and E_λ is the λ component of the macroscopic electric field. The $\{\Phi_{\alpha\beta}(l\kappa;l'\kappa')\}$ are the short-range atomic force constants, which include the contribution from the Lorentz field, but not from the macroscopic electric field. The latter contribution has been separated off explicitly in Eq. (2.1). The $\{\Phi_{\alpha\beta\gamma}(l\kappa;l'\kappa';l''\kappa'')\}$ are the cubic anharmonic force constants. The coefficients $\{M_{\lambda\alpha}(l\kappa)\}$, $\{M_{\lambda\alpha\beta}(l\kappa;l'\kappa')\}, \ldots,$ are the first-, second-, etc., order dipole-moment expansion coefficients, and the coefficients $\{P^{(0)}_{\lambda\mu}\}$, $\{P_{\lambda\mu\alpha}(l\kappa)\}, \ldots$ are the coefficients in the expansion of the electronic polarizability of the crystal in powers of the atomic displacements.

In our treatment we will ignore the frequency dispersion of the various coefficients appearing in Eq. (2.1). This means that all frequencies entering the present discussion will be well below the frequency of the lowest electronic transition in the crystal.

We now assume that the displacement $\xi_\alpha(l\kappa)$ is compounded of two displacements: The first describes a homogeneous deformation of the crystal, and the second gives the displacements from the new equilibrium positions in the homogeneously deformed crystal:

$$\begin{aligned}\xi_\alpha(l\kappa) &= \textstyle\sum_\beta \epsilon_{\alpha\beta}\, x_\beta(l\kappa) + v_\alpha(l\kappa) \\ &\equiv s_\alpha(l\kappa) + v_\alpha(l\kappa) . \qquad (2.2)\end{aligned}$$

The displacement gradients $\{\epsilon_{\alpha\beta} = \partial\xi_\alpha/\partial x_\beta\}$ are the components of a second-rank tensor, which is not assumed to be symmetric. When we substitute Eq. (2.2) into Eq. (2.1) we obtain for the equations of motion of the crystal

$$-M_\kappa \ddot{v}_\alpha(l\kappa) = \frac{\partial\Phi}{\partial v_\alpha(l\kappa)} = \sum_{l'\kappa'\beta} \Phi_{\alpha\beta}(l\kappa;l'\kappa')\,[\,s_\beta(l'\kappa') + v_\beta(l'\kappa')]$$

$$+\tfrac{1}{2}\sum_{l'\kappa'\beta}\sum_{l''\kappa''\gamma}\Phi_{\alpha\beta\gamma}(l\kappa;l'\kappa';l''\kappa'')[s_\beta(l'\kappa')+v_\beta(l'\kappa')][s_\gamma(l''\kappa'')+v_\gamma(l''\kappa'')]+\cdots$$

$$-\sum_{\lambda}E_\lambda M_{\lambda\alpha}(l\kappa)-\sum_{\lambda}E_\lambda\sum_{l'\kappa'\beta}M_{\lambda\alpha\beta}(l\kappa;l'\kappa')[s_\beta(l'\kappa')+v_\beta(l'\kappa')]-\cdots-\tfrac{1}{2}\sum_{\lambda\mu}E_\lambda E_\mu P_{\lambda\mu\alpha}(l\kappa)-\cdots, \qquad (2.3)$$

where M_κ is the mass of an atom of type κ. At the same time the macroscopic polarization is given by

$$P_\lambda=-\frac{1}{JV_0}\frac{\partial\Phi}{\partial E_\lambda}=\frac{1}{JV_0}\left[\sum_{l\kappa\alpha}M_{\lambda\alpha}(l\kappa)[s_\alpha(l\kappa)+v_\alpha(l\kappa)]+\tfrac{1}{2}\sum_{l\kappa\alpha}\sum_{l'\kappa'\beta}M_{\lambda\alpha\beta}(l\kappa;l'\kappa')[s_\alpha(l\kappa)+v_\alpha(l\kappa)]\right.$$

$$\left.\times[s_\beta(l'\kappa')+v_\beta(l'\kappa')]+\cdots+\sum_{\mu}E_\mu\left(P^{(0)}_{\lambda\mu}+\sum_{l\kappa\alpha}P_{\lambda\mu\alpha}(l\kappa)[s_\alpha(l\kappa)+v_\alpha(l\kappa)]+\cdots\right)\right]. \qquad (2.4)$$

In this equation, JV_0 is the volume of the deformed crystal, where V_0 is the volume of the undeformed crystal and J is given by

$$J=\det(\vec{\mathrm{I}}+\vec{\epsilon})=1+\sum_\alpha\epsilon_{\alpha\alpha}+O(\epsilon^2). \qquad (2.5)$$

In the presence of a static external strain the displacement $v_\alpha(l\kappa)$ can be written as the sum of a static contribution and of a dynamic contribution, which is also varying in space:

$$v_\alpha(l\kappa)=d_\alpha(l\kappa)+u_\alpha(l\kappa;t). \qquad (2.6)$$

The static displacements $\{d_\alpha(l\kappa)\}$ are the inner displacements of the ions (more correctly, of the sublattices) induced by the static strain, and the $\{u_\alpha(l\kappa;t)\}$ are the vibrational displacements of the ions about their new equilibrium positions. Because $\operatorname{div}\vec{P}=0$ for that part of the macroscopic polarization arising from the static deformation of the crystal, the macroscopic electric field in the crystal is that set up by the long-wavelength vibrations of the ions, and therefore has the same time dependence as that of $u_\alpha(l\kappa;t)$.

When Eq. (2.6) is substituted into Eq. (2.3), and only terms of first order in the dynamic displacements and the dynamic macroscopic field on the right-hand side are retained, the equations of motion of the crystal take the form

$$-M_\kappa\ddot{u}_\alpha(l\kappa)=\sum_{l'\kappa'\beta}\hat{\Phi}_{\alpha\beta}(l\kappa;l'\kappa')u_\beta(l'\kappa')-\sum_\lambda E_\lambda\hat{M}_{\lambda\alpha}(l\kappa), \qquad (2.7)$$

where

$$\hat{\Phi}_{\alpha\beta}(l\kappa;l'\kappa')=\Phi_{\alpha\beta}(l\kappa;l'\kappa')+\sum_{l''\kappa''\gamma}\Phi_{\alpha\beta\gamma}(l\kappa;l'\kappa';l''\kappa'')s_\gamma(l''\kappa'')+\sum_{l''\kappa''\gamma}\Phi_{\alpha\beta\gamma}(l\kappa;l'\kappa';l''\kappa'')d_\gamma(l''\kappa''), \qquad (2.8\text{a})$$

$$\hat{M}_{\lambda\alpha}(l\kappa)=M_{\lambda\alpha}(l\kappa)+\sum_{l'\kappa'\beta}M_{\lambda\alpha\beta}(l\kappa;l'\kappa')s_\beta(l'\kappa')+\sum_{l'\kappa'\beta}M_{\lambda\alpha\beta}(l\kappa;l'\kappa')d_\beta(l'\kappa'). \qquad (2.8\text{b})$$

The equations of motion take the form given by Eq. (2.7) only if the subsidiary condition

$$0=\sum_{l'\kappa'\beta}\Phi_{\alpha\beta}(l\kappa;l'\kappa')s_\beta(l'\kappa')+\sum_{l'\kappa'\beta}\Phi_{\alpha\beta}(l\kappa;l'\kappa')d_\beta(l'\kappa') \qquad (2.9)$$

is satisfied. This equation expresses the condition that no force act on any ion when all the ions are at their equilibrium positions in the absence of a dynamic macroscopic field. It relates the inner displacements $\{d_\alpha(l\kappa)\}$ to the deformation parameters to the first order of accuracy.

When Eq. (2.6) is substituted into Eq. (2.4), the macroscopic polarization can be expressed as the sum of a static contribution and a dynamic contribution. It is only the latter which is of interest here, and it is given by

$$P_\pi=\frac{1}{JV_0}\sum_{l\kappa\alpha}\hat{M}_{\lambda\alpha}(l\kappa)u_\alpha(l\kappa)+\frac{1}{JV_0}\sum_\mu\hat{P}_{\lambda\mu}E_\mu, \qquad (2.10)$$

where $\hat{M}_{\lambda\alpha}(l\kappa)$ is defined by Eq. (2.8b), and $\hat{P}_{\lambda\mu}$ is given by

$$\hat{P}_{\lambda\mu}=P^{(0)}_{\lambda\mu}+\sum_{l\kappa\alpha}P_{\lambda\mu\alpha}(l\kappa)[s_\alpha(l\kappa)+d_\alpha(l\kappa)]. \qquad (2.11)$$

The coefficient $M_{\lambda\alpha}(l\kappa)$ is seen to be the coefficient of the term linear in the atomic displacements in the expression for the dipole moment of the deformed crystal in the absence of the macroscopic electric field. By definition it is therefore the transverse effective charge tensor for the deformed crystal.

Inasmuch as we have assumed that the macroscopic field is uniform, the preceding results are applicable only to the long-wavelength vibrations of a crystal, in which the constituent sublattices vibrate rigidly against each other, and the displacement amplitude $u_\alpha(l\kappa)$ becomes independent of the cell index l. If we take this last fact into account explicitly by replacing the amplitude $u_\alpha(l\kappa)$ by $u_\alpha(\kappa)$, we can rewrite Eqs. (2.7) and (2.10) in the forms

$$-\hat{M}_\kappa\ddot{u}_\alpha(\kappa)=\sum_{\kappa'\beta}\hat{f}_{\alpha\beta}(\kappa\kappa')u_\beta(\kappa')-\sum_\lambda E_\lambda\hat{M}_{\lambda\alpha}(\kappa), \qquad (2.12)$$

$$P_\lambda = \frac{1}{Jv_a} \sum_{\kappa\alpha} \hat{M}_{\lambda\alpha}(\kappa)\, u_\alpha(\kappa) + \frac{1}{JV_0} \sum_\mu \hat{P}_{\lambda\mu} E_\mu , \quad (2.13)$$

where

$$\hat{f}_{\alpha\beta}(\kappa\kappa') = \sum_{l'} \hat{\Phi}_{\alpha\beta}(l\kappa; l'\kappa') \quad (2.14)$$

and v_a is the volume of an undeformed primitive unit cell. In writing these equations we have also used the fact that since a homogeneously deformed crystal is still perfectly periodic, the coefficient $\hat{M}_{\lambda\alpha}(l\kappa)$ is independent of the cell index l, and we have consequently denoted it by $\hat{M}_{\lambda\alpha}(\kappa)$.

If we assume a harmonic time dependence for the atomic displacements, we can write the time-independent equations of motion of the crystal as

$$\sum_{\kappa'\beta} C_{\alpha\beta}(\kappa\kappa' \mid \omega^2) w_\beta(\kappa') = - \sum_\lambda E_\lambda \frac{\hat{M}_{\lambda\alpha}(\kappa)}{M_\kappa^{1/2}} , \quad (2.15)$$

where $w_\alpha(\kappa) = M_\kappa^{1/2} u_\alpha(\kappa)$ and

$$C_{\alpha\beta}(\kappa\kappa' \mid \omega^2) = \delta_{\kappa\kappa'} \delta_{\alpha\beta} \omega^2 - \frac{\hat{f}_{\alpha\beta}(\kappa\kappa')}{(M_\kappa M_{\kappa'})^{1/2}} . \quad (2.16)$$

Solving Eq. (2.15) for $\omega_\alpha(\kappa)$, we find that

$$\omega_\alpha(\kappa) = - \sum_{\kappa'\beta\lambda} E_\lambda C^{-1}_{\alpha\beta}(\kappa\kappa' \mid \omega^2) \frac{\hat{M}_{\lambda\beta}(\kappa')}{M_{\kappa'}^{1/2}} . \quad (2.17)$$

If we substitute Eq. (2.17) into Eq. (2.10) and use the relation $P_\lambda = \sum_\mu \chi_{\lambda\mu} E_\mu$, we find that the susceptibility $\chi_{\lambda\mu}$ is given by

$$\chi_{\lambda\mu}(\omega) = \frac{1}{JV_0} \hat{P}_{\lambda\mu} - \frac{1}{Jv_a} \times \sum_{\kappa\alpha} \sum_{\kappa'\beta} \frac{\hat{M}_{\lambda\alpha}(\kappa)\, C^{-1}_{\alpha\beta}(\kappa\kappa' \mid \omega^2)\, \hat{M}_{\mu\beta}(\kappa')}{(M_\kappa M_{\kappa'})^{1/2}} . \quad (2.18)$$

The first term on the right-hand side of this equation is the electronic contribution to the susceptibility, while the second term is the ionic contribution.

To obtain the photoelastic constants we must now expand each of these contributions to first order in the deformation parameters.

The solution of Eq. (2.9) can be written formally as

$$d_\alpha(l\kappa) = \sum_{l'\kappa'\beta} \sum_{\gamma\delta} \Gamma_{\alpha\beta}(l\kappa; l'\kappa')\, G_{\beta\gamma\delta}(l'\kappa')\, \epsilon_{\gamma\delta} , \quad (2.19)$$

where

$$G_{\alpha\beta\gamma}(l\kappa) = - \sum_{l'\kappa'} \Phi_{\alpha\beta}(l\kappa; l'\kappa')\, x_\gamma(l'\kappa') , \quad (2.20)$$

and $\underline{\Gamma}$ is an effective inverse to the matrix $\underline{\Phi}$.[4] It follows from infinitesimal translational invariance as expressed by the conditions

$$\sum_{l'\kappa'} \Phi_{\alpha\beta}(l\kappa; l'\kappa') = \sum_{l\kappa} \Phi_{\alpha\beta}(l\kappa; l'\kappa') = 0 , \quad (2.21)$$

that

$$\sum_{l\kappa} G_{\alpha\beta\gamma}(l\kappa) = 0 . \quad (2.22)$$

From Eq. (2.21) it also follows that we can write $G_{\alpha\beta\gamma}(l\kappa)$ in an equivalent form, which is better suited for its calculation:

$$G_{\alpha\beta\gamma}(l\kappa) = \sum_{l'\kappa'} \Phi_{\alpha\beta}(l\kappa; l'\kappa')\, x_\gamma(l\kappa; l'\kappa') , \quad (2.23)$$

where $x_\alpha(l\kappa; l'\kappa') = x_\alpha(l'\kappa') - x_\alpha(l'\kappa')$.

We can combine the preceding results to write

$$\hat{\Phi}_{\alpha\beta}(l\kappa; l'\kappa') = \Phi_{\alpha\beta}(l\kappa; l'\kappa') + \sum_{\gamma\delta} \Phi_{\alpha\beta\gamma\delta}(l\kappa; l'\kappa')\, \epsilon_{\gamma\delta} , \quad (2.24)$$

$$\hat{M}_{\lambda\alpha}(\kappa) = M_{\lambda\alpha}(\kappa) + \sum_{\gamma\delta} M_{\lambda\alpha\gamma\delta}(\kappa)\, \epsilon_{\gamma\delta} , \quad (2.25)$$

where

$$\Phi_{\alpha\beta\gamma\delta}(l\kappa; l'\kappa') = \sum_{l''\kappa''} \Phi_{\alpha\beta\gamma}(l\kappa; l'\kappa'; l''\kappa'')\, x_\delta(l''\kappa'') + \sum_{l''\kappa''} \sum_{l'''\kappa'''} \sum_{\mu\nu} \Phi_{\alpha\beta\mu}(l\kappa; l'\kappa'; l''\kappa'')\, \Gamma_{\mu\nu}(l''\kappa''; l'''\kappa''')\, G_{\nu\gamma\delta}(l''\kappa'') , \quad (2.24')$$

$$M_{\lambda\alpha\gamma\delta}(\kappa) = \sum_{l'\kappa'} M_{\lambda\alpha\gamma}(l\kappa; l'\kappa')\, x_\delta(l'\kappa') + \sum_{l'\kappa'} \sum_{l''\kappa''} \sum_{\beta\mu} M_{\lambda\alpha\beta}(l\kappa; l'\kappa')\, \Gamma_{\beta\mu}(l'\kappa'; l''\kappa'')\, G_{\mu\gamma\delta}(l''\kappa'') . \quad (2.25')$$

We see from Eqs. (2.24′) and (2.25′) that the strain-induced changes in the harmonic force constants and the transverse effective charge, described by the coefficients $\Phi_{\alpha\beta\gamma\delta}(l\kappa; l'\kappa')$ and $M_{\lambda\alpha\gamma\delta}(\kappa)$, respectively, consist of two contributions. The first describes the change due to the homogeneous deformation of each sublattice, while the second describes the change due to the inner displacements of the sublattices induced by the homogeneous deformation.

We now obtain an expression for the electronic contribution to the susceptibility to first order in the deformation parameters. Starting with the first term on the right-hand side of Eq. (2.18), we expand J^{-1},

$$J^{-1} = 1 - \sum_{\alpha\beta} \delta_{\alpha\beta} \epsilon_{\alpha\beta} + \cdots . \quad (2.26)$$

We use this expansion, combine Eqs. (2.11) and (2.19), and substitute the results into Eq. (2.18). If we write the electronic susceptibility as

$$\chi^{(e)}_{\lambda\mu} = \chi^{(0e)}_{\lambda\mu} + \sum_{\rho\sigma} \chi^{(1e)}_{\lambda\mu\rho\sigma}\, \epsilon_{\rho\sigma}, \tag{2.27}$$

we find that

$$\chi^{(0e)}_{\lambda\mu} = P^{(0)}_{\lambda\mu}/V_0, \tag{2.28a}$$

$$\chi^{(1e)}_{\lambda\mu\rho\sigma} = \frac{1}{V_0}\Big[- P^{(0)}_{\lambda\mu}\, \delta_{\rho\sigma} + \sum_{l\kappa} P_{\lambda\mu\rho}(l\kappa)\, x_\sigma(l\kappa) + \sum_{l\kappa} \sum_{l'\kappa'} \sum_{\gamma\delta} P_{\lambda\mu\gamma}(l\kappa)\, \Gamma_{\gamma\delta}(l\kappa; l'\kappa')\, G_{\delta\rho\sigma}(l'\kappa')\Big] . \tag{2.28b}$$

Using the relation $k_{\lambda\mu\rho\sigma} = 4\pi\chi^{(1)}_{\lambda\mu\rho\sigma}$, we obtain the final result for the electronic contribution to the photoelastic constants:

$$k^{(e)}_{\lambda\mu\rho\sigma} = \frac{4\pi}{V_0}\Big[- P^{(0)}_{\lambda\mu}\, \delta_{\rho\sigma} + \sum_{l\kappa} P_{\lambda\mu\rho}(l\kappa)\, x_\sigma(l\kappa) + \sum_{l\kappa} \sum_{l'\kappa'} \sum_{\gamma\delta} P_{\lambda\mu\sigma}(l\kappa)\, \Gamma_{\gamma\delta}(l\kappa; l'\kappa')\, G_{\delta\rho\sigma}(l'\kappa')\Big] . \tag{2.29}$$

We next consider the second term on the right-hand side of Eq. (2.18) which is the ionic contribution to the susceptibility. From Eqs. (2.14), (2.24), and (2.24′) we see that we can expand the matrix $\hat{f}_{\alpha\beta}(\kappa\kappa')$ to first order in the deformation parameters as

$$\hat{f}_{\alpha\beta}(\kappa\kappa') = \hat{f}^{(0)}_{\alpha\beta}(\kappa\kappa') + \sum_{\gamma\delta} \hat{f}^{(1)}_{\alpha\beta\gamma\delta}(\kappa\kappa')\, \epsilon_{\gamma\delta} + \cdots , \tag{2.30a}$$

where

$$\hat{f}^{(0)}_{\alpha\beta}(\kappa\kappa') = \sum_{l'} \Phi_{\alpha\beta}(l\kappa; l'\kappa'), \tag{2.30b}$$

$$\hat{f}^{(1)}_{\alpha\beta\gamma\delta}(\kappa\kappa') = \sum_{l'} \Phi_{\alpha\beta\gamma\delta}(l\kappa; l'\kappa) = \sum_{l'} \sum_{l''\kappa''} \Phi_{\alpha\beta\gamma}(l\kappa; l'\kappa'; l''\kappa'')\, x_\delta(l''\kappa'') + \sum_{l'} \sum_{l''\kappa''} \sum_{l'''\kappa'''} \sum_{\mu\nu} \Phi_{\alpha\beta\mu}(l\kappa; l'\kappa'; l''\kappa'')\, \Gamma_{\mu\nu}(l''\kappa''; l'''\kappa''')\, G_{\nu\gamma\delta}(l'''\kappa''') = \hat{f}^{(1a)}_{\alpha\beta\gamma\delta}(\kappa\kappa') + \hat{f}^{(1b)}_{\alpha\beta\gamma\delta}(\kappa\kappa') . \tag{2.30c}$$

Combining Eqs. (2.16) and (2.30) we see we can expand the matrix $C_{\alpha\beta}(\kappa\kappa' \mid \omega^2)$ to first order in the deformation parameters in the form

$$C_{\alpha\beta}(\kappa\kappa' \mid \omega^2) = C^{(0)}_{\alpha\beta}(\kappa\kappa' \mid \omega^2) + C^{(1)}_{\alpha\beta}(\kappa\kappa' \mid \omega^2) + \cdots , \tag{2.31a}$$

where

$$C^{(0)}_{\alpha\beta}(\kappa\kappa' \mid \omega^2) = \delta_{\kappa\kappa'}\, \delta_{\alpha\beta}\, \omega^2 - \frac{\hat{f}^{(0)}_{\alpha\beta}(\kappa\kappa')}{(M_\kappa M_{\kappa'})^{1/2}}, \tag{2.31b}$$

$$C^{(1)}_{\alpha\beta}(\kappa\kappa' \mid \omega^2) = - \sum_{\rho\sigma} \frac{\hat{f}^{(1)}_{\alpha\beta\rho\sigma}(\kappa\kappa')}{(M_\kappa M_{\kappa'})^{1/2}}\, \epsilon_{\rho\sigma} . \tag{2.31c}$$

The expansion can now be used to obtain the following expression for the inverse matrix $C^{-1}_{\alpha\beta}(\kappa\kappa' \mid \omega^2)$:

$$C^{-1}_{\alpha\beta}(\kappa\kappa' \mid \omega^2) = C^{(0)\,-1}_{\alpha\beta}(\kappa\kappa' \mid \omega^2) - \sum_{\kappa''\gamma} \sum_{\kappa'''\delta} C^{(0)-1}_{\alpha\gamma}(\kappa\kappa'' \mid \omega^2)\, C^{(1)}_{\gamma\delta}(\kappa''\kappa''' \mid \omega^2)\, C^{(0)\,-1}_{\delta\beta}(\kappa'''\kappa' \mid \omega^2) + \cdots . \tag{2.32}$$

In these expressions, the superscript denotes the order in the deformation parameters. We also consider the ionic susceptibility to be given by

$$\chi^{(i)}_{\lambda\mu}(\omega) = \chi^{(0i)}_{\lambda\mu}(\omega) + \sum_{\rho\sigma} \chi^{(1i)}_{\lambda\mu\rho\sigma}(\omega)\, \epsilon_{\rho\sigma} \tag{2.33}$$

and combine Eqs. (2.24)–(2.26) with Eqs. (2.31) and (2.32) to obtain

$$\chi^{(0i)}_{\lambda\mu}(\omega) = - \frac{1}{v_a} \sum_{\kappa\alpha} \sum_{\kappa'\beta} \frac{M_{\lambda\alpha}(\kappa)\, C^{(0)\,-1}_{\alpha\beta}(\kappa\kappa' \mid \omega^2)\, M_{\mu\beta}(\kappa')}{(M_\kappa M_{\kappa'})^{1/2}}, \tag{2.34a}$$

$$\begin{aligned}\chi^{(1i)}_{\lambda\mu\rho\sigma}(\omega) = {} & \frac{1}{v_a} \sum_{\kappa\alpha} \sum_{\kappa'\beta} \frac{M_{\lambda\alpha}(\kappa)\, C^{(0)\,-1}_{\alpha\beta}(\kappa\kappa' \mid \omega^2)\, M_{\mu\beta}(\kappa')}{(M_\kappa M_{\kappa'})^{1/2}}\, \delta_{\rho\sigma} - \frac{1}{v_a} \sum_{\kappa\alpha} \sum_{\kappa'\beta} \frac{M_{\lambda\alpha\rho\sigma}(\kappa)\, C^{(0)}_{\alpha\beta}(\kappa\kappa' \mid \omega^2) M_{\mu\beta}(\kappa')}{(M_\kappa M_{\kappa'})^{1/2}} \\ & - \frac{1}{v_a} \sum_{\kappa\alpha} \sum_{\kappa'\beta} \frac{M_{\lambda\alpha}(\kappa)\, C^{(0)\,-1}_{\alpha\beta}(\kappa\kappa' \mid \omega^2)\, M_{\mu\beta\rho\sigma}(\kappa')}{(M_\kappa M_{\kappa'})^{1/2}} - \frac{1}{v_a} \sum_{\kappa\alpha} \sum_{\kappa'\beta} \sum_{\kappa''\gamma} \sum_{\kappa'''\delta} \frac{M_{\lambda\alpha}(\kappa)\, C^{(0)\,-1}_{\alpha\gamma}(\kappa\kappa'' \mid \omega^2)\, \hat{f}^{(1)}_{\gamma\delta\rho\sigma}(\kappa''\kappa''')}{(M_\kappa M_{\kappa''})^{1/2}} \\ & \times \frac{C^{(0)\,-1}_{\delta\beta}(\kappa'''\kappa' \mid \omega^2)\, M_{\mu\beta}(\kappa')}{(M_{\kappa'''} M_{\kappa'})^{1/2}} .\end{aligned} \tag{2.34b}$$

If we introduce the eigenvalues and eigenvectors of the matrix $\hat{f}^{(0)}_{\alpha\beta}(\kappa\kappa')/(M_\kappa M_{\kappa'})^{1/2}$ by

$$\sum_{\kappa'\beta} \frac{\hat{f}^{(0)}_{\alpha\beta}(\kappa\kappa')}{(M_\kappa M_{\kappa'})^{1/2}} V_\beta(\kappa'|j) = \omega_j^2 V_\alpha(\kappa|j), \qquad j = 1, 2, \ldots, 3r \tag{2.35}$$

where

$$\sum_{\kappa\alpha} V_\alpha(\kappa|j) V_\alpha(\kappa|j') = \delta_{jj'}, \tag{2.36a}$$

$$\sum_{j} V_\alpha(\kappa|j) V_\beta(\kappa'|j) = \delta_{\kappa\kappa'} \delta_{\alpha\beta}, \tag{2.36b}$$

we can write $C^{(0)-1}_{\alpha\beta}(\kappa\kappa'|\omega^2)$ as

$$C^{(0)-1}_{\alpha\beta}(\kappa\kappa'|\omega^2) = \sum_{j=1}^{3r} \frac{V_\alpha(\kappa|j) V_\beta(\kappa'|j)}{\omega^2 - \omega_j^2}. \tag{2.37}$$

When we substitute this result into Eqs. (2.34a) and (2.34b), we obtain for $\chi^{(0i)}_{\lambda\mu}(\omega)$

$$\chi^{(0i)}_{\lambda\mu}(\omega) = -\frac{1}{v_a} \sum_{j=1}^{3r} \frac{M_\lambda(j) M_\mu(j)}{\omega^2 - \omega_j^2}, \tag{2.38}$$

and for the ionic contribution to the photoelastic constants,

$$k^{(i)}_{\lambda\mu\rho\sigma}(\omega) = \frac{4\pi}{v_a} \sum_{j=1}^{3r} \frac{M_\lambda(j) M_\mu(j)}{\omega^2 - \omega_j^2} \delta_{\rho\sigma} - \frac{4\pi}{v_a} \sum_{j=1}^{3r} \frac{M_{\lambda\rho\sigma}(j) M_\mu(j)}{\omega^2 - \omega_j^2} - \frac{4\pi}{v_a} \sum_{j=1}^{3r} \frac{M_\lambda(j) M_{\mu\rho\sigma}(j)}{\omega^2 - \omega_j^2}$$
$$- \frac{4\pi}{v_a} \sum_{j=1}^{3r} \sum_{j'=1}^{3r} \frac{M_\lambda(j)}{\omega^2 - \omega_j^2} f_{\rho\sigma}(jj') \frac{M_\mu(j')}{\omega^2 - \omega_{j'}^2}, \tag{2.39}$$

where

$$M_\lambda(j) = \sum_{\kappa\alpha} \frac{M_{\lambda\alpha}(\kappa) V_\alpha(\kappa|j)}{(M_\kappa)^{1/2}}, \tag{2.40a}$$

$$M_{\lambda\rho\sigma}(j) = \sum_{\kappa\alpha} \frac{M_{\lambda\alpha\rho\sigma}(\kappa) V_\alpha(\kappa|j)}{(M_\kappa)^{1/2}}, \tag{2.40b}$$

$$f_{\rho\sigma}(jj') = \sum_{\kappa\alpha} \sum_{\kappa'\beta} V_\alpha(\kappa|j) \frac{\hat{f}^{(1)}_{\alpha\beta\rho\sigma}(\kappa\kappa')}{(M_\kappa M_{\kappa'})^{1/2}} V_\beta(\kappa'|j'). \tag{2.40c}$$

Because the coefficients $M_{\lambda\alpha}(\kappa)$ and $M_{\lambda\alpha\rho\sigma}(\kappa)$ obey the conditions

$$\sum_\kappa M_{\lambda\alpha}(\kappa) = 0, \tag{2.41a}$$

$$\sum_\kappa M_{\lambda\alpha\rho\sigma}(\kappa) = 0, \tag{2.41b}$$

which are consequences of infinitesimal translational invariance, and because $V_\alpha(\kappa|j)/(M_\kappa)^{1/2}$ is independent of κ when j refers to one of the three acoustic branches ($j = 1, 2, 3$), the coefficients $M_\lambda(j)$ and $M_{\lambda\rho\sigma}(j)$ vanish for $j = 1, 2, 3$. Thus, in fact, only the optical branches of the phonon spectrum contribute to the sums on j and on j' on the right-hand side of Eq. (2.39).

The photoelastic constant $k_{\lambda\mu\rho\sigma}$ is the sum of the expressions given by Eqs. (2.29) and (2.39).

In a recent note, Nelson and Lax[5] have pointed out that, contrary to the traditional formulation of the photoelastic effect, the photoelastic tensor $k_{\lambda\mu\rho\sigma}$ is not required to be symmetric in the indices $\rho\sigma$, although it must be symmetric in the indices λ and μ. In addition, they have shown that the part of $k_{\lambda\mu\rho\sigma}$ which is antisymmetric in ρ and σ can be expressed simply in terms of the dielectric tensor of the undeformed crystal. We conclude this section by showing how these results of Nelson and Lax are contained in the results given by Eqs. (2.29) and (2.39).

In the notation of this paper the result of Lax and Nelson is that the part of $k_{\lambda\mu\rho\sigma}$ which is antisymmetric in ρ and σ is given by

$$k_{\lambda\mu[\rho\sigma]} = 2\pi(\delta_{\rho\mu}\chi^{(0)}_{\lambda\sigma} + \delta_{\lambda\rho}\chi^{(0)}_{\sigma\mu} - \delta_{\mu\sigma}\chi^{(0)}_{\lambda\rho} - \delta_{\lambda\sigma}\chi^{(0)}_{\rho\mu}), \tag{2.42}$$

where $\chi^{(0)}_{\alpha\beta} = \chi^{(0e)}_{\alpha\beta} + \chi^{(0i)}_{\alpha\beta}$ is the dielectric susceptibility of the unstrained crystal. The brackets around the indices ρ and σ denote that the corresponding tensor is antisymmetric in these indices. We now show that the results given by Eqs. (2.29) and (2.39) yield precisely Eq. (2.42).

For this purpose we need four results, which are consequences of the transformation properties of the harmonic and cubic anharmonic force constants, second-order dipole-moment coefficients, and first-order polarizabilities when the crystal is subjected to an infinitesimal rigid-body rotation[6]:

$$\sum_{l'\kappa'} \Phi_{\alpha\beta}(l\kappa; l'\kappa') x_\gamma(l'\kappa') - \sum_{l'\kappa'} \Phi_{\alpha\gamma}(l\kappa; l'\kappa') x_\beta(l'\kappa') = 0, \tag{2.43a}$$

$$\sum_{l''\kappa''} \Phi_{\alpha\beta\gamma}(l\kappa; l'\kappa'; l''\kappa'') x_\delta(l''\kappa'') - \sum_{l''\kappa''} \Phi_{\alpha\beta\delta}(l\kappa; l'\kappa'; l''\kappa'') x_\gamma(l''\kappa'')$$
$$= \delta_{\beta\gamma}\Phi_{\alpha\delta}(l\kappa; l'\kappa') - \delta_{\beta\delta}\Phi_{\alpha\gamma}(l\kappa; l'\kappa') + \delta_{\alpha\gamma}\Phi_{\delta\beta}(l\kappa; l'\kappa') - \delta_{\alpha\delta}\Phi_{\gamma\beta}(l\kappa; l'\kappa'), \tag{2.43b}$$

$$\sum_{l'\kappa'} M_{\mu\alpha\beta}(l\kappa;l'\kappa')\,x_\gamma(l'\kappa') - \sum_{l'\kappa'} M_{\mu\alpha\gamma}(l\kappa;l'\kappa')\,x_\beta(l'\kappa') = \delta_{\alpha\beta}M_{\mu\gamma}(\kappa) - \delta_{\alpha\gamma}M_{\mu\beta}(\kappa) + \delta_{\mu\beta}M_{\gamma\alpha}(\kappa) - \delta_{\mu\gamma}M_{\beta\alpha}(\kappa)\,, \tag{2.43c}$$

$$\sum_{l\kappa} P_{\mu\nu\alpha}(l\kappa)\,x_\beta(l\kappa) - \sum_{l\kappa} P_{\mu\nu\beta}(l\kappa)\,x_\alpha(l\kappa) = \delta_{\nu\alpha}P^{(0)}_{\mu\beta} - \delta_{\nu\beta}P_{\mu\alpha} + \delta_{\mu\alpha}P^{(0)}_{\beta\nu} - \delta_{\mu\beta}P^{(0)}_{\alpha\nu}\,. \tag{2.43d}$$

We also note from Eq. (2.23) that because $\Phi_{\alpha\beta}(l\kappa;l'\kappa')$ is a function of the cell indices l and l' only through their difference, the coefficient $G_{\alpha\beta\gamma}(l\kappa)$ in fact is independent of the index l. Consequently, in what follows we will denote this coefficient by $G_{\alpha\beta\gamma}(\kappa)$. From Eq. (2.43a) it follows that $G_{\alpha\beta\gamma}(\kappa)$ is symmetric in β and γ.

We now consider the electronic contribution to the photoelastic constant $k_{\lambda\mu\rho\sigma}$. From the explicit expression for it, Eq. (2.29), we see that the first and third terms are symmetric in ρ and σ: Only the second term can contribute to the antisymmetric part of $k^{(i)}_{\lambda\mu\rho\sigma}$. Using Eq. (2.43d) we can can write the antisymmetric part of $k^{(e)}_{\lambda\mu\rho\sigma}$ as

$$k^{(e)}_{\lambda\mu[\rho\sigma]} = \frac{2\pi}{V_0}\Big[\sum_{l\kappa} P_{\lambda\mu\rho}(l\kappa)\,x_\sigma(l\kappa) - \sum_{l\kappa} P_{\lambda\mu\sigma}(l\kappa)\,x_\rho(l\kappa)\Big]$$

$$= \frac{2\pi}{V_0}\left(\delta_{\mu\rho}P^{(0)}_{\lambda\sigma} - \delta_{\mu\sigma}P^{(0)}_{\lambda\rho} + \delta_{\lambda\rho}P^{(0)}_{\sigma\mu} - \delta_{\lambda\sigma}P^{(0)}_{\rho\mu}\right)$$

$$= 2\pi\left(\delta_{\rho\mu}\chi^{(0e)}_{\lambda\sigma} + \delta_{\lambda\rho}\chi^{(0e)}_{\sigma\mu} - \delta_{\mu\sigma}\chi^{(0e)}_{\lambda\rho} - \delta_{\lambda\sigma}\chi^{(0e)}_{\rho\mu}\right)\,, \tag{2.44}$$

where we have used Eq. (2.28a) in going from the second to the third line of this equation. Thus, Eq. (2.44) for the electronic contribution to $k_{\lambda\mu[\rho\sigma]}$ has precisely the form of Eq. (2.42).

We now turn to the ionic contribution to $k_{\lambda\mu\rho\sigma}$. From Eq. (2.39) we see that the contribution to $k_{\lambda\mu\rho\sigma}$ which is antisymmetric in ρ and σ can only come from the second through fourth terms on the right-hand side, and has its origin in the contributions to the tensors $M_{\lambda\rho\sigma}(j)$ and $f_{\rho\sigma}(jj')$ which are antisymmetric in ρ and σ.

The antisymmetric part of $k^{(i)}_{\lambda\mu\rho\sigma}$ is therefore given by

$$k^{(i)}_{\lambda\mu[\rho\sigma]} = -\frac{4\pi}{v_a}\sum_{j=1}^{3r}\frac{M_{\lambda[\rho\sigma]}(j)\,M_\mu(j)}{\omega^2-\omega_j^2} - \frac{4\pi}{v_a}\sum_{j=1}^{3r}\frac{M_\lambda(j)\,M_{\mu[\rho\sigma]}(j)}{\omega^2-\omega_j^2} - \frac{4\pi}{v_a}\sum_{j=1}^{3r}\sum_{j'=1}^{3r}\frac{M_\lambda(j)}{\omega^2-\omega_j^2}\,f_{[\rho\sigma]}(jj')\frac{M_\mu(j')}{\omega^2-\omega_j^2}\,. \tag{2.45}$$

If we combine Eqs. (2.24′), (2.40b), and (2.43c), we find that

$$M_{\lambda[\rho\sigma]}(j) = \frac{1}{2}\left(\delta_{\lambda\rho}M_\sigma(j) - \delta_{\lambda\sigma}M_\rho(j) + \sum_\kappa \frac{V_\rho(\kappa|j)}{(M_\kappa)^{1/2}}M_{\lambda\sigma}(\kappa) - \sum_\kappa \frac{V_\sigma(\kappa|j)}{(M_\kappa)^{1/2}}M_{\lambda\rho}(\kappa)\right)\,. \tag{2.46}$$

Similarly, if we combine Eqs. (2.24′), (2.40c), (2.43b), and use Eq. (2.35), we obtain the result that

$$f_{[\rho\sigma]}(jj') = \tfrac{1}{2}(\omega_j^2 - \omega_{j'}^2)\left[V_\sigma(\kappa|j)\,V_\rho(\kappa|j') - V_\rho(\kappa|j)\,V_\sigma(\kappa|j')\right]\,. \tag{2.47}$$

We now decompose the last term on the right-hand side of Eq. (2.45) by partial fractions, and substitute into the resulting equation Eqs. (2.46) and (2.47). Grouping terms appropriately, we can write the result as

$$\begin{aligned}
k^{(i)}_{\lambda\mu[\rho\sigma]} = {}& -\frac{2\pi}{v_a}\sum_{j=1}^{3r}\left(\delta_{\rho\mu}\frac{M_\lambda(j)M_\sigma(j)}{\omega^2-\omega_j^2} + \delta_{\lambda\rho}\frac{M_\sigma(j)M_\mu(j)}{\omega^2-\omega_j^2} - \delta_{\mu\sigma}\frac{M_\lambda(j)M_\rho(j)}{\omega^2-\omega_j^2} - \delta_{\lambda\sigma}\frac{M_\rho(j)M_\mu(j)}{\omega^2-\omega_j^2}\right) - \frac{2\pi}{v_a}\sum_\kappa\sum_{j=1}^{3r}\frac{M_\mu(j)}{\omega^2-\omega_j^2}\\
&\times\frac{V_\rho(\kappa|j)}{(M_\kappa)^{1/2}}M_{\lambda\sigma}(\kappa) + \frac{2\pi}{v_a}\sum_\kappa\sum_{j=1}^{3r}\frac{M_\mu(j)}{\omega^2-\omega_j^2}\frac{V_\sigma(\kappa|j)}{(M_\kappa)^{1/2}}M_{\lambda\rho}(\kappa) - \frac{2\pi}{v_a}\sum_\kappa\sum_{j=1}^{3r}\frac{M_\lambda(j)}{\omega^2-\omega_j^2}\frac{V_\rho(\kappa|j)}{(M_\kappa)^{1/2}}M_{\mu\sigma}(\kappa) + \frac{2\pi}{v_a}\sum_\kappa\sum_{j=1}^{3r}\frac{M_\lambda(j)}{\omega^2-\omega_j^2}\\
&\times\frac{V_\sigma(\kappa|j)}{(M_\kappa)^{1/2}}M_{\mu\rho}(\kappa) + \frac{2\pi}{v_a}\sum_\kappa\sum_{j'=1}^{3r}\frac{M_\mu(j')}{\omega^2-\omega_{j'}^2}V_\rho(\kappa|j')\sum_{j=1}^{3r}M_\lambda(j)\,V_\sigma(\kappa|j) - \frac{2\pi}{v_a}\sum_\kappa\sum_{j'=1}^{3r}\frac{M_\mu(j')}{\omega^2-\omega_{j'}^2}V_\sigma(\kappa|j')\sum_{j=1}^{3r}M_\lambda(j)\,V_\rho(\kappa|j)\\
&+\frac{2\pi}{v_a}\sum_\kappa\sum_{j=1}^{3r}\frac{M_\lambda(j)}{\omega^2-\omega_j^2}V_\rho(\kappa|j)\sum_{j'=1}^{3r}M_\mu(j')\,V_\sigma(\kappa|j') - \frac{2\pi}{v_a}\sum_\kappa\sum_{j'=1}^{3r}\frac{M_\lambda(j)}{\omega^2-\omega_j^2}V_\sigma(\kappa|j)\sum_{j'=1}^{3r}M_\mu(j')\,V_\rho(\kappa|j')\,.
\end{aligned} \tag{2.48}$$

If we now use the result that

$$\sum_{j=1}^{3r} M_\lambda(j)\,V_\sigma(\kappa|j) = M_{\lambda\sigma}(\kappa)/(M_\kappa)^{1/2}\,, \tag{2.49}$$

which follows from Eqs. (2.36b) and (2.40b), we find that the last eight terms on the right-hand side cancel in pairs. We are therefore left with the result that

$$k_{\lambda\mu[\rho\sigma]} = 2\pi(\delta_{\rho\mu}\,\chi^{(0i)}_{\lambda\sigma} + \delta_{\lambda\rho}\,\chi^{(0i)}_{\sigma\mu} - \delta_{\mu\sigma}\,\chi^{(0i)}_{\lambda\rho} - \delta_{\lambda\sigma}\,\chi^{(0i)}_{\rho\mu}), \tag{2.50}$$

where we have used Eq. (2.38). Adding Eqs. (2.44) and (2.50), we obtain finally Eq. (2.42), as asserted.

We note that for cubic crystals for which $\chi_{\lambda\mu}(\omega)$ is isotropic, $\chi_{\lambda\mu}(\omega) = \delta_{\lambda\mu}\chi(\omega)$, $k_{\lambda\mu[\rho\sigma]}$ vanishes, and the photoelastic tensor $k_{\lambda\mu\rho\sigma}$ is symmetric in ρ and σ.

III. PHOTOELASTIC CONSTANTS OF CUBIC DIATOMIC CRYSTALS

The results of Sec. II are general, and apply to an arbitrary crystal. In this section we specialize these results to cubic crystals containing two ions in a primitive unit cell. The results obtained here therefore apply to crystals of the rocksalt, cesium chloride, diamond, and zinc-blende structures. However, it should be pointed out that in the case of diamond, which is a homopolar crystal, unlike the others mentioned, the only contribution to the photoelastic constants is the electronic contribution. As long as spatial dispersion effects are neglected, as is the case in the present work, there is no ionic contribution to the photoelastic constants. These results will be used in Sec. IV as the basis for estimates of the ionic contribution to the photoelastic constants of crystals of the rocksalt and zinc-blende structures.

In what follows we will label the two ions in a primitive unit cell by $\kappa = +, -$.

We first consider the ionic contribution to the photoelastic constants. It follows from infinitesimal translational invariance that quite generally

$$\sum_{\kappa} \hat{f}_{\alpha\beta}(\kappa\kappa') = 0 = \sum_{\kappa'} \hat{f}_{\alpha\beta}(\kappa\kappa') \tag{3.1}$$

and

$$\sum_{\kappa} \hat{M}_{\alpha\beta}(\kappa) = 0 . \tag{3.2}$$

Consequently, for the diatomic crystals with which we are concerned we have that

$$\hat{f}_{\alpha\beta}(\kappa\kappa') = \hat{f}_{\alpha\beta}\,(\mathrm{sgn}\kappa)\,(\mathrm{sgn}\kappa'), \tag{3.3}$$

$$\hat{M}_{\alpha\beta}(\kappa) = \hat{M}_{\alpha\beta}\,(\mathrm{sgn}\kappa), \tag{3.4}$$

where $\mathrm{sgn}\kappa = \pm 1$ for $\kappa = \pm$, respectively.

The cubic symmetry of each crystal structure yields the results that[7]

$$\sum_{l'} \Phi_{\alpha\beta}(l\kappa; l'\kappa') = \delta_{\alpha\beta}(\mathrm{sgn}\kappa)(\mathrm{sgn}\kappa')\,\mu\omega_T^2 \tag{3.5}$$

and

$$M_{\lambda\alpha}(\kappa) = \delta_{\lambda\alpha}\,(\mathrm{sgn}\kappa)\,e_T^* . \tag{3.6}$$

In these expressions, μ is the reduced mass of the two ions in a primitive unit cell, ω_T is the infinite wavelength transverse optical-mode frequency, and e_T^* is the transverse effective charge. Combining Eq. (3.5) with Eqs. (2.16) and (2.30), we obtain

$$C^{(0)}_{\alpha\beta}(\kappa\kappa'|\,\omega^2) = \delta_{\kappa\kappa'}\,\delta_{\alpha\beta}\omega^2 - \frac{\delta_{\alpha\beta}(\mathrm{sgn}\kappa)\,(\mathrm{sgn}\kappa')\,\mu\omega_T^2}{(M_\kappa M_{\kappa'})^{1/2}} . \tag{3.7}$$

The inverse of $C^{(0)}_{\alpha\beta}(\kappa\kappa'|\,\omega^2)$ is found to be given by

$$C^{(0)-1}_{\alpha\beta}(\kappa\kappa'|\,\omega^2) = \frac{(M_\kappa M_{\kappa'})^{1/2}\,\delta_{\alpha\beta}}{M_c\,\omega^2} + \mu\,\frac{(\mathrm{sgn}\kappa)\,(\mathrm{sgn}\kappa')}{(M_\kappa M_{\kappa'})^{1/2}}\,\frac{\delta_{\alpha\beta}}{\omega^2 - \omega_T^2}, \tag{3.8}$$

where $M_c = M_+ + M_-$.

We substitute Eqs. (3.6) and (3.8) into Eq. (2.34a) and obtain the result that

$$\chi^{(0i)}_{\lambda\mu}(\omega) = \delta_{\lambda\mu}\,\frac{(e_T^*)^2}{\mu v_a}\,\frac{1}{\omega_T^2 - \omega^2} . \tag{3.9}$$

We now use Eqs. (2.14), (3.3), and (3.4) to write

$$\sum_{l'''} \Phi_{\gamma\delta\rho\sigma}(l''\kappa''; l'''\kappa''') = \hat{f}^{(1)}_{\gamma\delta\rho\sigma}(\kappa''\kappa''') = (\mathrm{sgn}\kappa'')\,(\mathrm{sgn}\kappa''')\hat{f}^{(1)}_{\gamma\delta\rho\sigma}, \tag{3.10}$$

where $\hat{f}^{(1)}_{\gamma\delta\rho\sigma} = \hat{f}^{(1)}_{\gamma\delta\rho\sigma}(++)$ and

$$M_{\lambda\mu\rho\sigma}(\kappa) = (\mathrm{sgn}\kappa)\,M_{\lambda\mu\rho\sigma} , \tag{3.11}$$

where $M_{\lambda\mu\rho\sigma} = M_{\lambda\mu\rho\sigma}(+)$.

We substitute the above results as well as Eqs. (3.6) and (3.8) into Eq. (2.34b) and find that the ionic contribution to the photoelastic constants is given by the expression

$$k^{(i)}_{\lambda\mu\rho\sigma}(\omega) = \frac{a_{\lambda\mu\rho\sigma}}{\omega_T^2 - \omega^2} + \frac{b_{\lambda\mu\rho\sigma}}{(\omega_T^2 - \omega^2)^2} , \tag{3.12}$$

where

$$a_{\lambda\mu\rho\sigma} = -\frac{4\pi(e_T^*)^2}{\mu v_a}\left(\delta_{\lambda\mu}\,\delta_{\rho\sigma} - \frac{M_{\lambda\mu\rho\sigma} + M_{\mu\lambda\rho\sigma}}{e_T^*}\right), \tag{3.13}$$

$$b_{\lambda\mu\rho\sigma} = -\frac{4\pi(e_T^*)^2}{\mu v_a}\,\frac{\hat{f}^{(1)}_{\lambda\mu\rho\sigma}}{\mu} . \tag{3.14}$$

It is worth pointing out that for crystals of the kind considered in this section[8]

$$\frac{4\pi(e_T^*)^2}{\mu v_a} = \epsilon_\infty(\omega_L^2 - \omega_T^2) = \omega_T^2(\epsilon_0 - \epsilon_\infty) , \tag{3.15}$$

where ω_L is the frequency of the longitudinal optical mode of infinite wavelength and ϵ_0 and ϵ_∞ are the static and optical frequency dielectric constants, respectively.

Each of the fourth-rank tensors $\hat{f}^{(1)}_{\lambda\mu\rho\sigma}$ and $M_{\lambda\mu\rho\sigma}$ is invariant under the operations of the point group of the crystal, viz., T_d in the case of crystals of the zinc-blende structure, and O_h in the case of crystals of the rocksalt, cesium chloride, and diamond structures. An arbitrary fourth-rank tensor $\overleftrightarrow{t}$, possessing no intrinsic symmetry, which is invariant under T_d and O_h has four independent nonzero components, which are obtained by cyclic permutations of the indices from t_{xxxx}, t_{xxyy}, t_{xyxy}, and t_{xyyx}. However, the tensor $\hat{f}^{(1)}_{\lambda\mu\rho\sigma}$ is required to be symmetric in the indices λ and μ, due to the symmetry of the atomic force constants

$$\hat{\Phi}_{\alpha\beta}(l\kappa; l'\kappa') = \hat{\Phi}_{\beta\alpha}(l'\kappa'; l\kappa) .$$

Consequently, this tensor has only three independent nonzero components,

$$\hat{f}^{(1)}_{xxxx} , \quad \hat{f}^{(1)}_{xxyy} , \quad \hat{f}^{(1)}_{xyxy} , \tag{3.16}$$

and, in fact, possesses the symmetry properties of the second-order elastic constants belonging to the crystal classes T_d and O_h. The tensor $M_{\lambda\mu\rho\sigma}$, on the other hand, has no intrinsic symmetry in general. However, for crystals of the zinc-blende and rocksalt structures (in general, for crystals whose transverse effective-charge tensors either vanish or are multiples of the unit matrix) infinitesimal rotational invariance, as expressed by Eq. (2.43c), requires that $M_{\lambda\mu\rho\sigma}$ be symmetric in ρ and σ. Thus, for such crystals $M_{\lambda\mu\rho\sigma}$ also has only three independent nonzero components,

$$M_{xxxx} , \quad M_{xxyy} , \quad M_{xyxy} ,$$

and also possesses the symmetry properties of the second-order elastic constants.

By similar arguments the electronic contribution to the photoelastic constants of crystals of the rocksalt and zinc-blende structures can be shown to have only three independent nonzero components, viz., $k^{(e)}_{xxxx}$, $k^{(e)}_{xxyy}$, and $k^{(e)}_{xyxy}$, and to be symmetric in both the first and second pairs of subscripts.

In Sec. IV, we will obtain estimates for the quantities appearing in Eqs. (2.29) and (3.12).

IV. PHOTOELASTIC CONSTANTS OF KCl AND GaAs

In this section we use the results of Sec. III together with simple lattice-dynamical models to obtain estimates of the photoelastic constants of a typical alkali halide crystal of the rocksalt structure, KCl, and of a typical III-V semiconductor of the zinc-blende structure, GaAs. These results will be used in Sec. V to estimate the scattering efficiency for the ionic Brillouin effect for these two crystals.

A. Electronic Contribution to Photoelastic Constants

The electronic contribution to the photoelastic constants is the only contribution at frequencies of the incident light much greater than the lattice dispersion frequencies $\{\omega_j\}$ defined by Eq. (2.35), but below the frequencies of the electronic transitions in a crystal. It is this contribution which is obtained from conventional measurements carried out with the frequency of the incident light in the visible range. Consequently, instead of trying to devise a microscopic model for the electronic polarizability of a crystal, and evaluating the electronic contribution to the photoelastic constants from Eq. (2.29), we will obtain this contribution from experiment. (For an attempt at constructing a microscopic model of the electronic polarizability of a crystal, the reader is referred to the paper by Maradudin and Burstein.[9])

What are usually tabulated in the literature, however, are not the photoelastic constants, but related quantities, the Pockels elasto-optic constants $\{p_{\lambda\mu\rho\sigma}\}$. The latter relate the change in the dielectric permittivity tensor ϵ^{-1} to the deformation parameters $\{\epsilon_{\alpha\beta}\}$,

$$\delta(\epsilon^{-1})_{\lambda\mu} = \sum_{\rho\sigma} p_{\lambda\mu\rho\sigma} \epsilon_{\rho\sigma} . \tag{4.1}$$

The relation between the photoelastic constants and the elasto-optic constants is[10]

$$k_{\lambda\mu\rho\sigma} = -\sum_{\gamma\delta} (\overleftrightarrow{\epsilon})_{\lambda\gamma} p_{\gamma\delta\rho\sigma} (\overleftrightarrow{\epsilon})_{\delta\mu} , \tag{4.2}$$

where ϵ is the dielectric tensor at the frequency of the incident light. In the case in which we are interested, p is the electronic contribution to the photoelastic constants and $\overleftrightarrow{\epsilon}$ is the optical-frequency dielectric tensor. For cubic crystals the dielectric tensor is equivalent to a scalar, and Eq. (4.2) becomes

$$k_{\lambda\mu\rho\sigma} = -\epsilon^2 p_{\lambda\mu\rho\sigma} . \tag{4.3}$$

In addition, there are only three independent nonzero components of the tensors $k_{\lambda\mu\rho\sigma}$ and $p_{\lambda\mu\rho\sigma}$ for such crystals, as we have seen at the end of Sec. III. In the contracted Voigt notation these are

$$\begin{aligned} k_{xxxx} &= k_{11} , \quad & p_{xxxx} &= p_{11} , \\ k_{xxyy} &= k_{12} , \quad & p_{xxyy} &= p_{12} , \\ k_{xyxy} &= k_{44} , \quad & p_{xyxy} &= p_{44} . \end{aligned} \tag{4.4}$$

In Table I we present values of the $\{p_{ij}\}$ for KCl and GaAs, the wavelength λ of the light used in their determination, the optical frequency dielectric constants for these two crystals, and finally, the values of the electronic contributions to the photoelastic constants, the $\{k^{(e)}_{ij}\}$, derived from these data by the use of Eqs. (4.3) and (4.4).

We now turn to the ionic contribution to the photoelastic constants. Since the physics underlying the dynamical models is different for alkali halide crystals from what it is for the predominantly covalently bonded III-V semiconductor compounds, we will take up these two crystal types in turn.

TABLE I. The electronic contributions to the photoelastic constants of KCl and GaAs.

	KCl	GaAs
p_{11}	0.182[a]	−0.165[b]
p_{12}	0.134[a]	−0.140[b]
p_{44}	−0.026[a]	−0.072[b]
λ	435.8 nm	1150 nm
ϵ_∞	2.13[c]	10.9[c]
$k_{11}^{(e)}$	−0.826	19.60
$k_{12}^{(e)}$	−0.608	16.63
$k_{44}^{(e)}$	0.118	8.55

[a] K. S. Iyengar, Nature 176, 1119 (1955).

[b] R. W. Dixon, J. Appl. Phys. 38, 5149 (1967).

[c] E. Burstein, in Ref. 17, Table I, p. 296, Table II, p. 297.

B. Ionic Contribution to Photoelastic Constants of Alkali Halide Crystal

The short-range force constant $\Phi_{\alpha\beta}(l\kappa; l'\kappa')$ entering the present theory is the sum of the force constant derived from the short-range repulsive interaction $\varphi(r)$ between ions arising from the overlap of their electronic charge distributions, and the force constant associated with the Lorentz field in the crystal.[11] It is a good approximation to assume that only the former potential contributes significantly to the anharmonic force constants,[12] and we make this assumption here.

In the case of an alkali halide crystal, $\varphi(r)$ is usually assumed to have the Born–Mayer form

$$\varphi(r) = \lambda e^{-r/\rho}, \tag{4.5}$$

and is assumed to act only between nearest-neighbor ions. The values of the constants λ and ρ for the alkali halide crystals of the rocksalt and cesium chloride structures can be found, for example, in the book by Born and Huang.[13]

The cubic anharmonic force constants $\{\Phi_{\alpha\beta\gamma}(l\kappa; l'\kappa'; l''\kappa'')\}$ obtained from a nearest-neighbor central-force potential such as $\varphi(r)$ can be written in the form

$$\begin{aligned}\Phi_{\alpha\beta\gamma}(l\kappa; l\kappa; l'\kappa') &= \Phi_{\alpha\beta\gamma}(l\kappa; l'\kappa'; l\kappa) = \Phi_{\alpha\beta\gamma}(l'\kappa'; l\kappa; l\kappa)\\ &= -\Phi_{\alpha\beta\gamma}(l'\kappa'; l'\kappa'; l\kappa) = -\Phi_{\alpha\beta\gamma}(l'\kappa'; l\kappa; l'\kappa')\\ &= -\Phi_{\alpha\beta\gamma}(l\kappa; l'\kappa'; l'\kappa') = -\varphi_{\alpha\beta\gamma}(l\kappa; l'\kappa'),\ (l\kappa) \neq (l'\kappa')\end{aligned} \tag{4.6a}$$

$$\Phi_{\alpha\beta\gamma}(l\kappa; l\kappa; l\kappa) = \sum_{l'\kappa'(\neq l\kappa)} \varphi_{\alpha\beta\gamma}(l\kappa; l'\kappa'), \tag{4.6b}$$

where

$$\varphi_{\alpha\beta\gamma}(l\kappa; l'\kappa') = [A x_\alpha x_\beta x_\gamma + B(x_\alpha \delta_{\beta\gamma} + x_\beta \delta_{\gamma\alpha} + x_\gamma \delta_{\alpha\beta})]_{\vec{r}=\vec{x}(l\kappa; l'\kappa')} \tag{4.6c}$$

with

$$A = \frac{1}{r_0^3}\left(\varphi'''(r_0) - \frac{3}{r_0}\varphi''(r_0) + \frac{3}{r_0^2}\varphi'(r_0)\right), \tag{4.6d}$$

$$B = \frac{1}{r_0^2}\left(\varphi''(r_0) - \frac{1}{r_0}\varphi'(r_0)\right). \tag{4.6e}$$

Here r_0 is the nearest-neighbor separation.

For crystals of the rocksalt structure, in which every ion is at a center of inversion symmetry, the third-rank tensor $G_{\alpha\beta\gamma}(\kappa)$ vanishes identically for each value of κ. Consequently, the only nonzero contribution to the tensor $\hat{f}^{(1)}_{\alpha\beta\gamma\delta}(\kappa\kappa')$ defined by Eq. (2.30c) is

$$\hat{f}^{(1a)}_{\alpha\beta\gamma\delta}(\kappa\kappa') = \sum_{l'} \sum_{l''\kappa''} \Phi_{\alpha\beta\gamma}(l\kappa; l'\kappa'; l''\kappa'')\, x_\delta(l''\kappa''). \tag{4.7}$$

We can use the infinitesimal translation invariance condition

$$\sum_{l''\kappa''} \Phi_{\alpha\beta\gamma}(l\kappa; l'\kappa'; l''\kappa'') = 0 \tag{4.8}$$

to rewrite $\hat{f}^{(1a)}_{\alpha\beta\gamma\delta}(\kappa\kappa')$ in a form better suited for its evaluation,

$$\hat{f}^{(1a)}_{\alpha\beta\gamma\delta}(\kappa\kappa') = -\sum_{l'} \sum_{l''\kappa''} \Phi_{\alpha\beta\gamma}(l\kappa; l'\kappa'; l''\kappa'') \times x_\delta(l'\kappa'; l''\kappa''). \tag{4.9}$$

If we consider the tensor $\hat{f}^{(1a)}_{\alpha\beta\gamma\delta}(++) \equiv \hat{f}^{(1a)}_{\alpha\beta\gamma\delta}$, then from Eqs. (4.6), (4.9), and our assumption of nearest-neighbor (nn) repulsive interactions we find that this tensor is given by

$$\begin{aligned}\hat{f}^{(1a)}_{\alpha\beta\gamma\delta} &= \sum_{l'}^{\mathrm{nn}} \varphi_{\alpha\beta\gamma}(l+; l'-)\, x_\delta(l+; l'-) = \sum_{l'}^{\mathrm{nn}} [A x_\alpha x_\beta x_\gamma x_\delta + B(x_\alpha x_\delta \delta_{\beta\gamma} + x_\beta x_\delta \delta_{\gamma\alpha} + x_\gamma x_\delta \delta_{\alpha\beta})]_{\vec{r}=\vec{x}(l+; l'-)}\\ &= A \sum_{l'}^{\mathrm{nn}} x_\alpha x_\beta x_\gamma x_\delta \Big|_{\vec{r}=\vec{x}(l+; l-)} + 2Br_0^2(\delta_{\alpha\delta}\delta_{\beta\gamma} + \delta_{\beta\delta}\delta_{\gamma\alpha} + \delta_{\gamma\delta}\delta_{\alpha\beta}).\end{aligned} \tag{4.10}$$

Consequently, we obtain the results that

$$\hat{f}^{(1a)}_{xxxx} = 2Ar_0^4 + 6Br_0^2, \tag{4.11a}$$

$$\hat{f}^{(1a)}_{xxyy} = \hat{f}^{(1a)}_{xyxy} = 2Br_0^2. \tag{4.11b}$$

For the particular choice of $\varphi(r)$ given by Eq. (4.5) it is the case that

$$r_0^4 A = -\frac{\lambda e^{-r_0/\rho}}{r_0^2}\left[\left(\frac{r_0}{\rho}\right)^3 + 3\left(\frac{r_0}{\rho}\right)^2 + 3\left(\frac{r_0}{\rho}\right)\right], \tag{4.12a}$$

$$r_0^2 B = \frac{\lambda e^{-r_0/\rho}}{r_0^2}\left[\left(\frac{r_0}{\rho}\right)^2 + \left(\frac{r_0}{\rho}\right)\right]. \tag{4.12b}$$

In Table II the values of r_0, ρ, and λ for KCl are presented. The results for $f^{(1a)}_{\alpha\beta\gamma\delta}$ obtained on substituting these values into Eqs. (4.11) and (4.12) are also presented in Table II.

We now turn to the determination of the coefficients $\{M_{\lambda\mu\rho\sigma}\}$. If we assume that the effective, or local, field acting on an ion is the sum of the macroscopic field $\vec{E}$ and the Lorentz field $(\frac{4}{3}\pi)\vec{P}$, where $\vec{P}$ is the polarization, we can formally expand the dipole moment of the crystal in powers of the ionic displacements and the components of the macroscopic field as

$$M_\mu = \sum_{l\kappa\alpha} M'_{\mu\alpha}(l\kappa)\, u_\alpha(l\kappa) + \tfrac{1}{2}\sum_{l\kappa\alpha}\ \sum_{l'\kappa'\beta} M'_{\mu\alpha\beta}(l\kappa; l'\kappa')\, u_\alpha(l\kappa)\, u_\beta(l'\kappa') + \cdots + \sum_\nu \Big[P'_{\mu\nu} + \sum_{l\kappa\alpha} P'_{\mu\nu\alpha}(l\kappa) + \cdots \Big]\Big[E_\nu + \tfrac{4}{3}\pi P_\nu\Big]. \tag{4.13}$$

The coefficient $M'_{\mu\alpha}(l\kappa)$ is the Szigeti effective-charge tensor[14] for the ion $(l\kappa)$, while $P'_{\mu\nu}$ is the sum of the atomic polarizability tensors for all the positive and negative ions in the crystal. If we use the fact that the polarization P_ν is

$$P_\nu = M_\nu / V_0\,, \tag{4.14}$$

where V_0 is the volume of the (undeformed) crystal, together with the fact that $P'_{\mu\nu\alpha}(l\kappa)$ vanishes identically for crystals of the rocksalt structure because every ion is at a center of inversion, and that for such crystals the tensor $P'_{\mu\nu}$ is a multiple of the unit matrix,

$$P'_{\mu\nu} = \delta_{\mu\nu} P'\,, \tag{4.15}$$

we can solve Eq. (4.13) for M_μ to obtain

$$M_\mu = \sum_{l\kappa\alpha} M_{\mu\alpha}(l\kappa)\, u_\alpha(l\kappa) + \tfrac{1}{2}\sum_{l\kappa\alpha}\ \sum_{l'\kappa'\beta} M_{\mu\alpha\beta}(l\kappa; l'\kappa') \times u_\alpha(l\kappa)\, u_\beta(l'\kappa') + \cdots + \sum_\nu (P_{\mu\nu} + \cdots)\, E_\nu\,. \tag{4.16}$$

The coefficients in this expansion are those appearing in Eq. (2.1). They are related to the coefficients in Eq. (4.13) by

$$M_{\mu\alpha}(l\kappa) = S^{(0)} M'_{\mu\alpha}(l\kappa)\,, \tag{4.17a}$$

$$M_{\mu\alpha\beta}(l\kappa; l'\kappa') = S^{(0)} M'_{\mu\alpha\beta}(l\kappa; l'\kappa')\,, \tag{4.17b}$$

$$P_{\mu\nu} = S^{(0)} P'_{\mu\nu}\,, \tag{4.17c}$$

where

$$S^{(0)} = \left(1 - \frac{4\pi P'}{3V_0}\right)^{-1} = \frac{\epsilon_\infty + 2}{3}\,. \tag{4.17d}$$

The last equation follows from the Clausius–Mosotti relation[15]

$$\frac{4\pi}{3}\frac{P'}{V_0} = \frac{\epsilon_\infty - 1}{\epsilon_\infty + 2}\,. \tag{4.18}$$

To obtain the coefficients $\{M'_{\mu\alpha}(l\kappa)\}$ and $\{M'_{\mu\alpha\beta}(l\kappa; l'\kappa')\}$, we assume that in the absence of the effective field the dipole moment of the crystal is the sum of two contributions: that arising from the displacement of the static charge on each ion, and that arising from the distortions of the charge distributions on the ions due to the changes in their relative separations accompanying the displacements of the ions from their equilibrium positions. It seems to be a good approximation to assume that the static charge on a positive (negative) ion is $+e$ $(-e)$ for alkali halide crystals, where e is the magnitude of the electronic charge.

We consequently express the crystal dipole moment in the absence of an effective field as

$$M_\mu = \sum_{l\kappa} e\,(\mathrm{sgn}\,\kappa)\, u_\mu(l\kappa) + \tfrac{1}{2}\sum_{l\kappa}\ \sum_{l'\kappa'}{}' \xi(\kappa\kappa')\, \frac{x_\mu(l\kappa; l'\kappa') + u_\mu(l\kappa; l'\kappa')}{|\vec{x}(l\kappa; l'\kappa') + \vec{u}(l\kappa; l'\kappa')|} \times m(|\vec{x}(l\kappa; l'\kappa') + \vec{u}(l\kappa; l'\kappa')|)\,. \tag{4.19a}$$

In this expression the second sum is over all nearest-neighbor pairs of ions, the prime excludes the terms with $(l\kappa) = (l'\kappa')$, and

$$\begin{aligned} \xi(\kappa\kappa') &= +1 \quad \text{if } \kappa = +, \quad \kappa' = - \\ &= -1 \quad \text{if } \kappa = -, \quad \kappa' = + \\ &= 0 \quad \text{otherwise.} \end{aligned} \tag{4.19b}$$

The physical content of the second term on the right-hand side of Eq. (4.19a) is that the contribution to the dipole moment of a crystal arising from the deformability of the ions is the sum of dipole moments associated with each nearest-neighbor bond. The distortion dipole moment $m(r)$ between a positive and a negative ion is a function of their separation r, and its sign is chosen to be positive if the moment is directed from the negative to the positive ion, and negative if the reverse is the case.

When Eq. (4.19a) is expanded in powers of the ionic displacements,

$$M_\mu = \sum_{l\kappa\alpha} M'_{\mu\alpha}(l\kappa) u_\alpha(l\kappa) + \tfrac{1}{2}\sum_{l\kappa\alpha}\ \sum_{l'\kappa'\beta} M'_{\mu\alpha\beta}(l\kappa; l'\kappa')$$

TABLE II. Values of the physical constants and derived quantities entering the calculation of the ionic contributions to the photoelastic constants of KCl.

	KCl
r_0	3.139 Å[a]
ρ	0.324 Å[a]
λ	3.63×10^{-9} erg[a]
ϵ_0	4.85[b]
ϵ_∞	2.13[b]
ω_T	142 cm^{-1}[b]
ω_L	214 cm^{-1}[b]
e_S^*	$0.81e$[b]
e_T^*	$1.1151e$
μ	3.0595×10^{-23} g
C_{11}	4.06×10^{11} dyn/cm^2[c]
C_{12}	0.67×10^{11} dyn/cm^2[c]
C_{44}	0.629×10^{11} dyn/cm^2[c]
m_0	1.9928×10^2
M_{xxxx}	$2.8638e_T^*$
M_{xxyy}	$-0.3261e_T^*$
M_{xyxy}	$-0.3261e_T^*$
$\hat{f}^{(1)}_{xxxx}$	-4.1545×10^5 erg/cm^2
$\hat{f}^{(1)}_{xxyy}$	0.47308×10^5 erg/cm^2
$\hat{f}^{(1)}_{xyxy}$	0.47308×10^5 erg/cm^2
a_{xxxx}	$12.859\omega_T^2$
a_{xxyy}	$-4.494\omega_T^2$
a_{xyxy}	$-1.774\omega_T^2$
b_{xxxx}	$51.626\omega_T^4$
b_{xxyy}	$-5.8787\omega_T^4$
b_{xyxy}	$-5.8787\omega_T^4$

[a] M. Born and K. Huang, *Dynamical Theory of Crystal Lattices* (Clarendon, Oxford, England, 1954), Table 9, p. 26.

[b] E. Burstein, in Ref. 17, Table I, p. 296.

[c] G. B. Benedek and K. Fritsch, Phys. Rev. 149, 647 (1966).

$$\times\, u_\alpha(l\kappa)\, u_\beta(l'\kappa') + \cdots, \tag{4.20}$$

we find that the coefficients are given by

$$M'_{\mu\alpha}(l\kappa) = \delta_{\mu\alpha}\ (\text{sgn}\kappa)\, e + \tfrac{1}{2}\sum_{l'\kappa'}^{\text{nn}} \left[\xi(\kappa\kappa') - \xi(\kappa'\kappa)\right] \times f_{\mu\alpha}(l\kappa; l'\kappa'), \tag{4.21a}$$

$$M'_{\mu\alpha\beta}(l\kappa; l'\kappa') = -\tfrac{1}{2}\left[\xi(\kappa\kappa') - \xi(\kappa'\kappa)\right] f_{\mu\alpha\beta}(l\kappa; l'\kappa'), \quad (l\kappa) \neq (l'\kappa') \tag{4.21b}$$

$$M'_{\mu\alpha\beta}(l\kappa; l\kappa) = \tfrac{1}{2}\sum_{l'\kappa'(\neq l\kappa)}^{\text{nn}}{}' \left[\xi(\kappa\kappa') - \xi(\kappa'\kappa)\right] f_{\mu\alpha\beta}(l\kappa; l'\kappa'), \tag{4.21c}$$

where

$$f_{\mu\alpha}(l\kappa; l'\kappa') = \left[\frac{x_\mu x_\alpha}{r_0^2}\left(m'(r_0) - \frac{m(r_0)}{r_0}\right) + \delta_{\mu\alpha}\frac{m(r_0)}{r_0}\right]_{\vec{r}=\vec{x}(l\kappa; l'\kappa')}, \tag{4.22}$$

$$f_{\mu\alpha\beta}(l\kappa; l'\kappa') = \left[M x_\mu x_\alpha x_\beta + N(x_\mu\delta_{\alpha\beta} + x_\alpha\delta_{\mu\beta} + x_\beta\delta_{\mu\alpha})\right]_{\vec{r}=\vec{x}(l\kappa; l'\kappa')}, \tag{4.23a}$$

with

$$M = \frac{m''(r_0)}{r_0^3} - \frac{3m'(r_0)}{r_0^4} + \frac{3m(r_0)}{r_0^5}, \tag{4.23b}$$

$$N = \frac{m'(r_0)}{r_0^2} - \frac{m(r_0)}{r_0^3}. \tag{4.23c}$$

For the distortion dipole moment $m(r)$ we assume the expression

$$m(r) = m_0 e r_0 e^{-r/\rho}. \tag{4.24}$$

The parameter ρ appearing in this expression is the same as that entering the short-range repulsive potential $\varphi(r)$, Eq. (4.5), because the physical origin of the distortion dipole moment and the short-range repulsive interaction between ions is the same, namely, the overlap of the electronic charge distributions on neighboring ions.

For crystals of the rocksalt structure, Eq. (4.21a) simplifies to

$$M'_{\mu\alpha}(l\kappa) \equiv M'_{\mu\alpha}(\kappa) = \delta_{\mu\alpha}\ (\text{sgn}\kappa)\{e + 2[m'(r_0) + 2m(r_0)/r_0]\}. \tag{4.25}$$

Consequently, the Szigeti effective charge is given by

$$e_S^* = e + 2[m'(r_0) + 2m(r_0)/r_0] \tag{4.26a}$$

$$= e[1 - 2m_0 e^{-r_0/\rho}((r_0/\rho) - 2)], \tag{4.26b}$$

where the second form obtains when the expression for $m(r)$ given by Eq. (4.24) is used. Equation (4.26), together with experimental values of the Szigeti effective charge serves to determine the coefficient m_0 entering the right-hand side of Eq. (4.24). The value of e_S^* for KCl and the value of m_0 obtained from it are presented in Table II.

For crystals of the rocksalt structure the only nonzero contribution to the tensor $M_{\alpha\beta\gamma\delta}(\kappa)$ defined by Eq. (2.25′) is

$$M^{(a)}_{\alpha\beta\gamma\delta}(\kappa) = \sum_{l'\kappa'} M_{\alpha\beta\gamma}(l\kappa; l'\kappa')\, x_\delta(l'\kappa'). \tag{4.27}$$

We can use the infinitesimal translation invariance condition

$$\sum_{l'\kappa'} M_{\alpha\beta\gamma}(l\kappa;l'\kappa')=0 \tag{4.28}$$

to rewrite $M^{(a)}_{\alpha\beta\gamma\delta}(\kappa)$ in a more convenient form as

$$M^{(a)}_{\alpha\beta\gamma\delta}(\kappa)=-\sum_{l'\kappa'} M_{\alpha\beta\gamma}(l\kappa;l'\kappa')\,x_\delta(l\kappa;l'\kappa') . \tag{4.29}$$

If we consider the tensor $M^{(a)}_{\alpha\beta\gamma\delta}(+)\equiv M^{(a)}_{\alpha\beta\gamma\delta}$, then from Eqs. (4.17), (4.21), and (4.22) we find that this tensor is given by

$$M^{(a)}_{\alpha\beta\gamma\delta}=\tfrac{1}{3}(\epsilon_\infty+2)\Big[M\sum_{l'}^{\mathrm{nn}} x_\alpha x_\beta x_\gamma x_\delta\Big|_{\vec{x}=\vec{x}(l+;l'-)} + 2Nr_0^2(\delta_{\alpha\delta}\,\delta_{\beta\gamma}+\delta_{\alpha\gamma}\,\delta_{\beta\delta}+\delta_{\alpha\beta}\,\delta_{\gamma\delta})\Big] . \tag{4.30}$$

Consequently, we obtain the results that

$$M^{(a)}_{xxxx}=\tfrac{1}{3}(\epsilon_\infty+2)(2Mr_0^4+6Nr_0^2) , \tag{4.31a}$$

$$M^{(a)}_{xxyy}=M^{(a)}_{xyxy}=\tfrac{1}{3}(\epsilon_\infty+2)\,2Nr_0^2 . \tag{4.13b}$$

For the particular choice of $m(r)$ given by Eq. (4.24), it is the case that

$$r_0^4M=e\,m_0\,e^{-r_0/\rho}\,[(r_0/\rho)^2+3(r_0/\rho)+3] , \tag{4.32a}$$

$$r_0^2N=-em_0\,e^{-r_0/\rho}[(r_0/\rho)+1] . \tag{4.32b}$$

In Table II the values of $M^{(a)}_{\alpha\beta\gamma\delta}$ for KCl obtained in this fashion are presented.

Finally, if we rewrite the expressions for the coefficients $a_{\lambda\mu\rho\sigma}$ and $b_{\lambda\mu\rho\sigma}$ given by Eqs. (3.13) and (3.14) in the forms

$$a_{\lambda\mu\rho\sigma}=-(\epsilon_0-\epsilon_\infty)(\delta_{\lambda\mu}\,\delta_{\rho\sigma}-2M_{\lambda\mu\rho\sigma}/e_T^*)\,\omega_T^2 , \tag{4.33a}$$

$$b_{\lambda\mu\rho\sigma}=-(\epsilon_0-\epsilon_\infty)(\hat{f}^{(1)}_{\lambda\mu\rho\sigma}/\mu\omega_T^2)\,\omega_T^4 , \tag{4.33b}$$

the results of this section yield the values of these coefficients for KCl given in Table II. Combining the latter results with the results given in Table I, we obtain finally the following expressions for the components of the photoelastic tensor of KCl, including the ionic contribution:

$$k_{11}(\omega)=-0.826+\frac{12.859\omega_T^2}{\omega_T^2-\omega^2}+\frac{51.626\omega_T^4}{(\omega_T^2-\omega^2)^2} , \tag{4.34a}$$

$$k_{12}(\omega)=-0.608-\frac{4.494\omega_T^2}{\omega_T^2-\omega^2}-\frac{5.8787\omega_T^4}{(\omega_T^2-\omega^2)^2} , \tag{4.34b}$$

$$k_{44}(\omega)=0.118-\frac{1.774\omega_T^2}{\omega_T^2-\omega^2}-\frac{5.8787\omega_T^4}{(\omega_T^2-\omega^2)^2} . \tag{4.34c}$$

C. Ionic Contribution to Photoelastic Constants of III-V Compound Semiconductor of Zinc-Blende Structure

The model used to describe alkali halide crystals has to be modified substantially before it can be applied to III-V compound semiconductors of the zinc-blende structure. In the first place, there is evidence that the static charges of the atoms constituting such crystals are zero, i.e., that the atoms are neutral.[16] In the second place, the effective electric field acting on any ion is the macroscopic field: The Lorentz field vanishes.[17] Consequently, the existence of a nonzero transverse effective charge on the atoms in such crystals is due entirely to the deformation of their electronic charge distributions as their relative separations change due to their displacements from the equilibrium positions.

In the absence of a Lorentz field, we assume that the short-range force constants $\{\Phi_{\alpha\beta}(l\kappa;l'\kappa')\}$ entering the present theory are derived from a nearest-neighbor interatomic potential which we write in the form

$$\varphi(r)=a\frac{r_0^m}{r^m}-b\frac{r_0^n}{r^n} , \quad m>n . \tag{4.35}$$

The first term describes the repulsion between the atoms due to the overlap of their charge distributions, while the second term describes the attractive force which stabilizes the static lattice. A more realistic model would undoubtedly include interactions between more distant neighbors, as well as noncentral angle-bending forces to represent more accurately the covalent character of the bonding in these crystals. However, the present simple model should yield results for the photoelastic constants which have the correct order of magnitude, even if the details of the dynamical properties of these crystals are described incorrectly.

The equilibrium condition

$$\varphi'(r_0)=0=-ma/r_0+nb/r_0 \tag{4.36}$$

yields the relation

$$b=(m/n)\,a . \tag{4.37}$$

The compressibility is given by

$$1/\beta=\tfrac{4}{9}(1/v_a)\,am(m-n) , \tag{4.38}$$

while the transverse-optical mode frequency ω_T is

$$\mu\omega_T^2=\tfrac{4}{3}(a/r_0^2)\,m(m-n) . \tag{4.39}$$

The relations (4.38) and (4.39) are not independent, however. Their quotient predicts the relation

$$\frac{16}{\sqrt{3}}\,\frac{r_0/\beta}{\mu\omega_T^2}=1 \tag{4.40}$$

for our model, where we have used the fact that the volume of a primitive unit cell for a crystal of the zinc-blende structure is

$$v_a=\tfrac{1}{4}a_0^3=(16/3\sqrt{3})\,r_0^3 . \tag{4.41}$$

If we substitute the values of r_0, β, μ, ω_T appropriate to GaAs into the left-hand side of Eq. (4.40) (see Table III), we find that

$$\frac{16}{\sqrt{3}}\,\frac{r_0/\beta}{\mu\,\omega_T^2}=1.08\,, \tag{4.42}$$

which is in satisfactory agreement with Eq. (4.40), particularly in view of the simplicity of our model.

From Eq. (4.38) we obtain the coefficient a as

$$a=(9v_a/4\beta)[1/m(m-n)]. \tag{4.43}$$

To define this model completely we (somewhat arbitrarily) set

$$m=9,\quad n=\tfrac{9}{2}\,. \tag{4.44}$$

The values of a and b obtained in this way are presented in Table III.

We now specify the zinc-blende structure by postulating that the origin of coordinates is occupied by a positive (e.g., Ga) ion, while the four nearest neighbors at $\frac{1}{4}a_0\,(1,1,1)$, $\frac{1}{2}a_0\,(-1,-1,1)$, $\frac{1}{4}a_0\,(1,-1,-1)$, and $\frac{1}{4}a_0\,(-1,1,-1)$ are negative (e.g., As) ions.

We begin the determination of the ionic contribution to the photoelastic constants of a typical III-V compound of the zinc-blende structure, GaAs, by determining the tensor $G_{\alpha\beta\gamma}(l\kappa)\equiv G_{\alpha\beta\gamma}(\kappa)$. From considerations of symmetry and translational invariance it follows that $G_{\alpha\beta\gamma}(\kappa)$ has only one independent nonzero component, and can be written in the form

$$G_{\alpha\beta\gamma}(\kappa)=\left|\epsilon_{\alpha\beta\gamma}\right|(\mathrm{sgn}\kappa)\,G\,, \tag{4.45a}$$

where

$$G\equiv G_{xyz}(+)\,, \tag{4.45b}$$

and $\epsilon_{\alpha\beta\gamma}$ is the Levi-Civita tensor. From Eqs. (2.23) and (4.45) we see that we are thus led to evaluate the expression

$$\begin{aligned}G&=\sum_{l'\kappa'}\Phi_{xy}(l+;l'\kappa')\,x_z(l+;l'\kappa')=-\sum_{l'}^{\mathrm{nn}}\varphi_{xy}(l+;l'-)\,x_z(l+;l'-)\\&=-\frac{1}{r_0^2}\left(\varphi''(r_0)-\frac{1}{r_0}\,\varphi'(r_0)\right)\sum_{l'}^{\mathrm{nn}}x_x(l+;l'-)\,x_y(l+;l'-)\,x_z(l+;l'-)=\frac{4r_0}{3\sqrt{3}}\left(\varphi''(r_0)-\frac{1}{r_0}\,\varphi'(r_0)\right)=\frac{54}{\sqrt{3}}\,\frac{a}{r_0}\,.\end{aligned} \tag{4.46}$$

The value of G obtained in this way is presented in Table III.

We can now obtain the elements of the tensor $\hat{f}^{(1)}_{\alpha\beta\gamma\delta}(\kappa\kappa')$. For crystals of the zinc-blende structure we see from Eq. (2.30) that this tensor is the sum of two contributions, $\hat{f}^{(1a)}_{\alpha\beta\gamma\delta}(\kappa\kappa')+\hat{f}^{(1b)}_{\alpha\beta\gamma\delta}(\kappa\kappa')$, of which the first has been discussed in Sec. IV B. The result for $\hat{f}^{(1a)}_{\alpha\beta\gamma\delta}\equiv\hat{f}^{(1a)}_{\alpha\beta\gamma\delta}(++)$ given by Eqs. (4.6) and (4.10) in the case of crystals of the zinc-blende structure becomes

$$\hat{f}^{(1a)}_{xxxx}=\tfrac{4}{9}Ar_0^4+4Br_0^2\,, \tag{4.47a}$$

$$\hat{f}^{(1a)}_{xxyy}=\hat{f}^{(1a)}_{xyxy}=\tfrac{4}{9}Ar_0^4+\tfrac{4}{3}Br_0^2\,, \tag{4.47b}$$

where, for the present model,

$$r_0^4A=-(3159/4)\,(a/r_0^2)\,, \tag{4.48a}$$

$$r_0^2B=\tfrac{81}{2}\,a/r_0^2\,. \tag{4.48b}$$

The second contribution to $\hat{f}^{(1)}_{\alpha\beta\gamma\delta}$, viz., $\hat{f}^{(1b)}_{\alpha\beta\gamma\delta}$, is given by

$$\hat{f}^{(1b)}_{\alpha\beta\gamma\delta}=\sum_{l'}\sum_{l''\kappa''}\sum_{l'''\kappa'''}\sum_{\mu\nu}\Phi_{\alpha\beta\mu}(l+;l'+;l''\kappa'')\times\Gamma_{\mu\nu}(l''\kappa'';l'''\kappa''')\,G_{\nu\gamma\delta}(l'''\kappa''')\,. \tag{4.49}$$

For crystals of the zinc-blende structure we have the result that[9]

$$\sum_{l'''}\Gamma_{\lambda\mu}(l''\kappa'';l'''\kappa''')=\delta_{\lambda\mu}\,(\mathrm{sgn}\kappa'')\,(\mathrm{sgn}\kappa''')\,\frac{\mu}{M_{\kappa''}M_{\kappa''}}\,\frac{1}{\omega_T^2}\,. \tag{4.50}$$

When Eqs. (4.45) and (4.50) are substituted into Eq. (4.49), we find that $\hat{f}^{(1b)}_{\alpha\beta\gamma\delta}$ is given by

$$\hat{f}^{(1b)}_{\alpha\beta\gamma\delta}=-\frac{G}{\mu\omega_T^2}\sum_{l'l''}\sum_{\mu}\left|\epsilon_{\mu\gamma\delta}\right|\Phi_{\alpha\beta\mu}(l+;l'+;l''-)\,. \tag{4.51}$$

It follows from the T_d symmetry at each site of the zinc-blende structure, and from infinitesimal translational invariance, that

$$\sum_{l'l''}\Phi_{\alpha\beta\mu}(l\kappa;l'\kappa';l''\kappa'')=\left|\epsilon_{\alpha\beta\mu}\right|\,(\mathrm{sgn}\kappa)\,(\mathrm{sgn}\kappa')\,(\mathrm{sgn}\kappa'')f\,. \tag{4.52}$$

Thus, we have that

$$\hat{f}^{(1b)}_{\alpha\beta\gamma\delta}=\frac{Gf}{\mu\omega_T^2}\sum_{\mu}\left|\epsilon_{\alpha\beta\gamma}\right|\left|\epsilon_{\mu\gamma\delta}\right|\,. \tag{4.53}$$

The independent elements of this tensor are

$$\hat{f}^{(1b)}_{xxxx}=0\,, \tag{4.54a}$$

$$\hat{f}^{(1b)}_{xxyy}=0\,, \tag{4.54b}$$

$$\hat{f}^{(1b)}_{xyxy}=Gf/\mu\omega_T^2\,. \tag{4.54c}$$

We see from Eq. (4.52) that f is given by

$$f=-\sum_{l'l''}\Phi_{xyz}(l+;\ l'+;\ l''-)$$

$$=\sum_{l'}^{\text{nn}}{}'\varphi_{xyz}(l+;\ l'-)$$

$$=A\sum_{l'}^{\text{nn}}{}'x_x(l+;\ l'-)x_y(l+;\ l'-)x_z(l+;\ l'-)$$

$$=-\frac{4}{3\sqrt{3}}\,r_0^3A=\frac{1053}{\sqrt{3}}\,\frac{a}{r_0^3}\,. \tag{4.55}$$

Values of $\hat{f}^{(1)}_{xxxx}$, $\hat{f}^{(1)}_{xxyy}$, and $\hat{f}^{(1)}_{xyxy}$ calculated for GaAs are presented in Table III.

If the static charge on an atom of a III-V semiconductor compound of the zinc-blende structure vanishes, and the effective field acting on an atom is the macroscopic field, we find from the results of Sec. IV B that

$$M_{\mu\alpha}(l\kappa)=\tfrac{1}{2}\sum_{l'\kappa'}^{\text{nn}}{}'[\xi(\kappa\kappa')-\xi(\kappa'\kappa)]f_{\mu\alpha}(l\kappa;\ l'\kappa')\,, \tag{4.56}$$

$$M_{\mu\alpha\beta}(l\kappa;\ l'\kappa')=-\tfrac{1}{2}[\xi(\kappa\kappa')-\xi(\kappa'\kappa)]\,f_{\mu\alpha\beta}(l\kappa;l'\kappa')\qquad (l\kappa)\neq(l'\kappa')\,, \tag{4.57a}$$

$$M_{\mu\alpha\beta}(l\kappa;\ l\kappa)=\tfrac{1}{2}\sum_{l'\kappa'}^{\text{nn}}{}'[\xi(\kappa\kappa')-\xi(\kappa'\kappa)]f_{\mu\alpha\beta}(l\kappa;\ l'\kappa')\,, \tag{4.57b}$$

where $M_{\mu\alpha}(l\kappa)$ is the transverse effective-charge tensor, and $f_{\mu\alpha}(l\kappa;\ l'\kappa')$ and $f_{\mu\alpha\beta}(l\kappa;\ l'\kappa')$ are given by Eqs. (4.22) and (4.23).

For crystals of the zinc-blende structure

$$M_{\mu\alpha}(+)\equiv M_{\mu\alpha}(l+)=\delta_{\mu\alpha}\tfrac{4}{3}[m'(r_0)+2m(r_0)/r_0]$$

$$=\delta_{\mu\alpha}e_T^*\,. \tag{4.58}$$

We choose for $m(r)$, in this case,

$$m(r)=m_0er_0^{10}/r^9\,, \tag{4.59}$$

which has the same functional dependence on r as the short-range repulsive potential, and find that

$$e_T^*=-\tfrac{28}{3}m_0e\,. \tag{4.60}$$

The results of Bennett and Maradudin[18] show that the sign of the transverse effective charge is positive for Ga atoms in GaAs. This result, together with the magnitude of e_T^* obtained from experiment, enables us to obtain m_0 from Eq. (4.60). The value so obtained for GaAs is presented in Table III.

The coefficients $M_{\alpha\beta\gamma\delta}$ for crystals of the zinc-blende structure are the sums of two contributions, $M^{(a)}_{\alpha\beta\gamma\delta}+M^{(b)}_{\alpha\beta\gamma\delta}$, the former of which has been discussed in Sec. IV B. In the present case we find for the independent nonzero components of $M^{(a)}_{\alpha\beta\gamma\delta}$,

$$M^{(a)}_{xxxx}=\tfrac{4}{9}r_0^4M+4r_0^2N\,, \tag{4.61a}$$

$$M^{(a)}_{xxyy}=M^{(a)}_{xyxy}=\tfrac{4}{9}r_0^4M+\tfrac{4}{3}r_0^2N\,, \tag{4.61b}$$

where

$$r_0^4M=120m_0e\,, \tag{4.62a}$$

TABLE III. Values of the physical constants and derived quantities entering the calculation of the ionic contributions to the photoelastic constants of GaAs.

	GaAs
r_0	2.448 Å [a]
$1/\beta$	0.755×10^{12} dyn/cm^2
ϵ_0	12.9 [b]
ϵ_∞	10.9 [b]
ω_T	273 cm^{-1} [b]
ω_L	297 cm^{-1} [b]
e_T^*	$2.2e$ [b]
μ	5.9935×10^{-23} g
C_{11}	1.188×10^{12} dyn/cm^2 [c]
C_{12}	0.538×10^{12} dyn/cm^2 [c]
C_{44}	0.594×10^{12} dyn/cm^2 [c]
a	1.8947×10^{-12} erg
b	3.7895×10^{-12} erg
G	24.1303×10^{-4} erg/cm
f	78.5189×10^{12} erg/cm^3
m_0	-0.2357
m	$(4.0431\times10^8)e_T^*$ cm^{-1}
$\hat{f}^{(1a)}_{xxxx}$	-5.9754×10^5 erg/cm^2
$\hat{f}^{(1b)}_{xxxx}$	0
$\hat{f}^{(1a)}_{xxyy}$	-9.3900×10^5 erg/cm^2
$\hat{f}^{(1b)}_{xxyy}$	0
$\hat{f}^{(1a)}_{xyxy}$	-9.3900×10^5 erg/cm^2
$\hat{f}^{(1b)}_{xyxy}$	11.9548×10^5 erg/cm^2
$M^{(a)}_{xxxx}$	$-1.4288e_T^*$
$M^{(b)}_{xxxx}$	0
$M^{(a)}_{xxyy}$	$-4.2858e_T^*$
$M^{(b)}_{xxyy}$	0
$M^{(a)}_{xyxy}$	$-4.2858e_T^*$
$M^{(b)}_{xyxy}$	$6.1557e_T^*$
a_{xxxx}	$-7.7152\omega_T^2$
a_{xxyy}	$-19.1432\omega_T^2$
a_{xyxy}	$7.4796\omega_T^2$
b_{xxxx}	$7.5405\omega_T^4$
b_{xxyy}	$11.8495\omega_T^4$
b_{xyxy}	$-3.2366\omega_T^4$

[a] A. S. Cooper, Acta Cryst. 15, 578 (1962).

[b] E. Burstein, in Ref. 17, Table II, p. 297.

[c] T. E. Bateman, H. J. McSkimin, and J. M. Whelan, J. Appl. Phys. 30, 544 (1959).

$$r_0^2 N = -10 m_0 e \, . \tag{4.62b}$$

The second contribution to $M_{\alpha\beta\gamma\delta}$, $M^{(b)}_{\alpha\beta\gamma\delta}$ is obtained from Eq. (2.25b) as

$$M^{(b)}_{\alpha\beta\gamma\delta} \equiv M^{(b)}_{\alpha\beta\gamma\delta}(+) = \sum_{l'\kappa'} \sum_{l''\kappa''} \sum_{\mu\nu} M_{\alpha\beta\mu}(l+;\, l'\kappa') \times \Gamma_{\mu\nu}(l'\kappa';\, l''\kappa'') G_{\nu\gamma\delta}(l''\kappa'') \, . \tag{4.63}$$

With the aid of Eqs. (4.45) and (4.50) we can rewrite this expression as

$$M^{(b)}_{\alpha\beta\gamma\delta} = -\frac{G}{\mu\omega_T^2} \sum_{l'\mu} |\epsilon_{\mu\gamma\delta}| M_{\alpha\beta\mu}(l+;\, l'-) \, . \tag{4.64}$$

The T_d-site symmetry in the zinc-blende structure, coupled with infinitesimal translational invariance, yields the result that

$$\sum_{l'} M_{\alpha\beta\mu}(l\kappa;\, l'\kappa') = |\epsilon_{\alpha\beta\mu}| (\operatorname{sgn}\kappa)(\operatorname{sgn}\kappa') m \, . \tag{4.65}$$

Thus we have that

$$M^{(b)}_{\alpha\beta\gamma\delta} = (Gm/\mu\omega_T^2) \textstyle\sum_\mu |\epsilon_{\alpha\beta\mu}|\, |\epsilon_{\mu\gamma\delta}| \, , \tag{4.66}$$

so that

$$M^{(b)}_{xxxx} = 0 \, , \tag{4.67a}$$

$$M^{(b)}_{xxyy} = 0 \, , \tag{4.67b}$$

$$M^{(b)}_{xyxy} = Gm/\mu\omega_T^2 \, . \tag{4.67c}$$

From Eq. (4.65) we see that the coefficient m is given by

$$m = -\sum_{l'} M_{xyz}(l+;\, l'-) = -\frac{4}{3\sqrt{3}} r_0^3 M \, . \tag{4.68}$$

The values of M_{xxxx}, M_{xxyy}, and M_{xyxy} calculated in this way are presented in Table III for GaAs. From these values and the values of $\hat{f}^{(1)}_{xxxx}$, $\hat{f}^{(1)}_{xxyy}$, and $\hat{f}^{(1)}_{xyxy}$ obtained above, we calculate the tensors $a_{\lambda\mu\rho\sigma}$ and $b_{\lambda\mu\rho\sigma}$ on the basis of Eqs. (4.33). These results are also presented in Table III. With these results we obtain finally for the photoelastic constants of GaAs

$$k_{11}(\omega) = 19.60 - \frac{7.7152\omega_T^2}{\omega_T^2 - \omega^2} + \frac{7.5405\omega_T^4}{(\omega_T^2 - \omega^2)^2} \, , \tag{4.69a}$$

$$k_{12}(\omega) = 16.63 - \frac{19.1432\omega_T^2}{\omega_T^2 - \omega^2} + \frac{11.8495\omega_T^2}{(\omega_T^2 - \omega^2)^2} \, , \tag{4.69b}$$

$$k_{44}(\omega) = 8.55 + \frac{7.4796\omega_T^2}{\omega_T^2 - \omega^2} - \frac{3.2366\omega_T^4}{(\omega_T^2 - \omega^2)^2} \, . \tag{4.69c}$$

It is interesting to note that our results for GaAs show that the coefficients $\hat{f}^{(1b)}_{xyxy}$ and $M^{(b)}_{xyxy}$, which describe the changes in the harmonic atomic force constants and in the transverse effective charge due to the strain-induced inner displacements of the sublattices, respectively, are of opposite sign to, and are larger in magnitude than, the coefficients $\hat{f}^{(1a)}_{xyxy}$ and $M^{(a)}_{xyxy}$, which describe the changes in these physical quantities due to the homogeneous deformation of each sublattice at zero inner displacement.

V. CALCULATION OF SCATTERING EFFICIENCY

We conclude this paper by utilizing the results obtained in Sec. IV to obtain an estimate of the scattering efficiencies of KCl and GaAs as functions of the frequency of the incident light, when the latter is in the vicinity of ω_T.

We assume that the incident radiation is directed along the x axis and is polarized in the z direction, while the scattered radiation is observed along the y axis and is polarized in (a) the z direction; and (b) the x direction. The unit vectors $\vec{n}_0$ and $\vec{n}$ in these two cases therefore are

$$\begin{aligned} &\text{case (a)} \quad \vec{n}_0 = (0,\ 0,\ 1) \, , \quad \vec{n} = (0,\ 0,\ 1) \, , \\ &\text{case (b)} \quad \vec{n}_0 = (0,\ 0,\ 1) \, , \quad \vec{n} = (1,\ 0,\ 0) \, . \end{aligned} \tag{5.1}$$

For such a 90° scattering geometry the magnitudes of the vectors $\vec{q}$, $\vec{k}_0$, and $\vec{k}_s$ are related by

$$q^2 = k_0^2 + k_s^2 \, , \tag{5.2}$$

while conservation of energy in the scattering process yields the relations

$$\omega_s = \omega_0 \mp \omega_j(\vec{q}) \, , \tag{5.3}$$

where the upper (lower) sign refers to Stokes (anti-Stokes) scattering processes. The dispersion relation for the long-wavelength acoustic modes from which the scattering takes place can be expressed as

$$\omega_j(\vec{q}) = c_j(\hat{q}) q \, , \tag{5.4}$$

where $c_j(\hat{q})$ is the speed of sound for the jth branch of the acoustic spectrum for elastic waves propagating in the direction of the vector $\vec{q}$. Dividing Eq. (5.3) by c, the speed of light, we obtain

$$k_s = k_0 \mp [c_j(\hat{q})/c] q \, . \tag{5.5}$$

Since the speed of elastic waves is typically smaller than the speed of light by four to five orders of magnitude, it follows from Eqs. (5.2)–(5.5) that to a very good approximation

$$k_s = k_0 = k \, . \tag{5.6}$$

Thus we have in the present case that

$$\begin{aligned} &\vec{k}_0 = (k,\ 0,\ 0) \, , \quad \vec{k}_s = (0,\ k,\ 0) \, , \\ &\vec{q} = (-k,\ k,\ 0) \, , \end{aligned} \tag{5.7}$$

so that the scattering vector $\vec{q}$ is in the $[\bar{1}10]$ direction.

The speeds of sound, and the corresponding polarization vectors $\{e_\mu(\vec{q}, j)\}$ for elastic waves propagating in the $[\bar{1}10]$ direction in a cubic crystal are[19]

j	$c_j^2(\hat{q})$	$e_1(\vec{q}j)$	$e_2(\vec{q}j)$	$e_3(\vec{q}j)$
1	c_{44}/ρ	0	0	1
2	$(c_{11} - c_{12})/2\rho$	$1/\sqrt{2}$	$1/\sqrt{2}$	0
3	$(c_{11} + c_{12} + 2c_{44})/2\rho$	$-1/\sqrt{2}$	$1/\sqrt{2}$	0 .

(5.8)

In these expressions c_{11}, c_{12}, and c_{44} are the three independent elastic constants of the crystal. The branches $j = 1$, 2 are clearly the transverse branches, while the branch $j = 3$ is the longitudinal branch.

The quotient $f_j(\vec{q})/\omega_j(\vec{q})$, which enters the expression (1.1) for the scattering efficiency, can be expressed as

$$\frac{f_j(\vec{q})}{\omega_j(\vec{q})} = \frac{1}{c_j(\hat{q})} \sum_{\alpha\gamma\mu\nu} n_\alpha n_{0\gamma} k_{\alpha\gamma\mu\nu} e_\mu(\vec{q}j)\hat{q}_\nu \, . \qquad (5.9)$$

Using the results given by Eqs. (5.1) and (5.7)–(5.9), we obtain the following results for this quotient in cases (a) and (b):

$$\begin{array}{ccc} j & (a) & (b) \\ 1 & 0 & \dfrac{-k_{44}}{[2c_{44}/\rho]^{1/2}} \\ 2 & 0 & 0 \\ 3 & \dfrac{k_{12}}{[(c_{11}+c_{12}+2c_{44})/2\rho]^{1/2}} & 0\, . \end{array} \qquad (5.10)$$

The scattering efficiencies corresponding to cases (a) and (b) are obtained as

$$\text{case (a)}\ \mathcal{S} = L\left(\frac{\omega_0}{c}\right)^4 \frac{k_B T}{16\pi^2} \frac{k_{12}^2}{(c_{11}+c_{12}+2c_{44})}$$

$$\times [\delta(\Omega - \omega_3(\vec{q})) + \delta(\Omega + \omega_3(\vec{q}))] , \qquad (5.11a)$$

$$\text{case (b)}\ \mathcal{S} = L\left(\frac{\omega_0}{c}\right)^4 \frac{k_B T}{16\pi^2} \frac{k_{44}^2}{4c_{44}}$$

$$\times [\delta(\Omega - \omega_1(\vec{q}) + \delta(\Omega + \omega_1(\vec{q}))] . \qquad (5.11b)$$

The coefficients of the δ-function terms in brackets give the integrated scattering efficiency for either Stokes or anti-Stokes scattering. These coefficients are plotted in Figs. 1 and 2 as functions of ω_0 for $\omega_0 \lesssim \omega_T$ and $\omega_0 \gtrsim \omega_T$, for KCl and GaAs, on the basis of the results of Sec. IV for the frequency dependence of the photoelastic constants of these crystals in this frequency range, where the resonant enhancement of the scattering efficiency is largest. It has been assumed that $L = 1$ cm and that $T = 300\,^\circ$K.

VI. DISCUSSION

In this paper we have presented a theory of the Brillouin scattering of light by the long-wavelength acoustic modes of a nonmetallic crystal, when the frequency of the incident light is in the infrared. Central to this theory is a theory of the ionic contribution to the photoelastic constants of such crystals. We have presented a lattice-dynamical theory of this contribution for an arbitrary crystal, and have shown that our results possess the general symmetry properties derived recently by Nelson and Lax[3,5] for the components of the photoelastic tensor. The general lattice-dynamical expressions for the photoelastic constants obtained here have been specialized to the cases of crystals of the rocksalt and zinc-blende structures. Simple lattice-dynamical models of such crystals have been used to estimate the ionic contribution to the photoelastic constants of KCl and GaAs, and consequently the scattering efficiency for the Brillouin effect for these crystals for incident light in the infrared.

Our results, presented in Figs. 1 and 2, show that for frequencies of the incident light within 5% of the frequencies of the transverse optical modes of each crystal, the integrated scattering efficiency for Stokes or anti-Stokes scattering exceeds 10^{-10} $\text{cm}^{-1}\,\text{sr}^{-1}$. To compare these scattering efficiencies with those obtained for frequencies of the incident light in the visible, we have calculated the scattering efficiencies for KCl and GaAs taking for ω_0 a value of 20 000 cm^{-1}, which is essentially the frequency of one of the lines of an argon-ion laser, and using for the photoelastic constants only the electronic contribution, given in Table I. The results obtained are

$$\text{KCl:}\ \text{(a)}\ \int_0^\infty \mathcal{S}(\Omega)\,d\Omega = 4.03\times 10^{-8}\ \text{cm}^{-1}\,\text{sr}^{-1} ,$$

$$\text{(b)}\ \int_0^\infty \mathcal{S}(\Omega)\,d\Omega = 3.62\times 10^{-9}\ \text{cm}^{-1}\,\text{sr}^{-1} ;$$

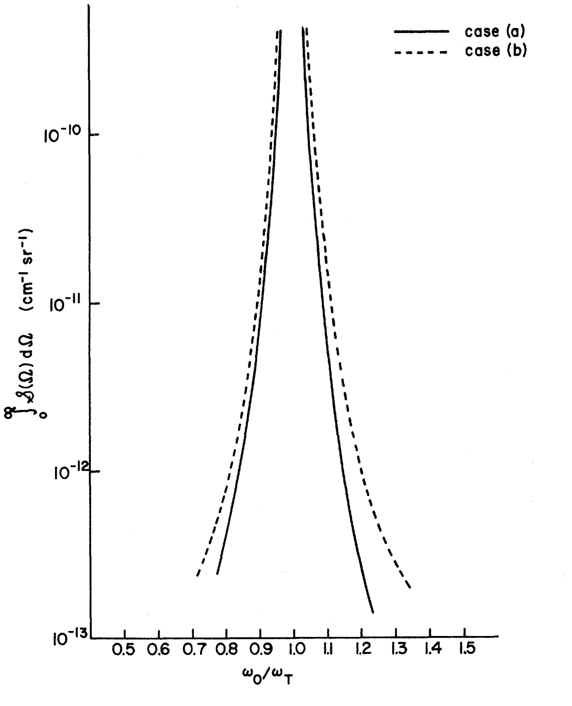

FIG. 1. Dependence of the integrated scattering efficiency on the frequency of the incident light for the ionic Brillouin effect in KCl. Cases (a) and (b) refer to the two scattering geometries defined by Eq. (5.1).

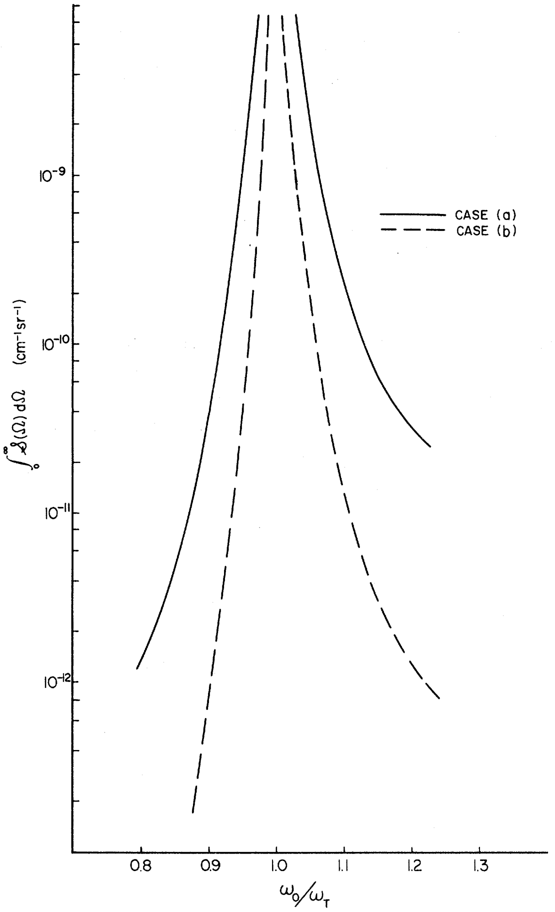

FIG. 2. Dependence of the integrated scattering efficiency on the frequency of the incident light for the ionic Brillouin effect in GaAs. Cases (a) and (b) refer to the two scattering geometries defined by Eq. (5.1).

$$\text{GaAs: (a)} \int_0^\infty S(\Omega)\,d\Omega = 6.2\times 10^{-6}\ \text{cm}^{-1}\,\text{sr}^{-1};$$

$$\text{(b)} \int_0^\infty S(\Omega)\,d\Omega = 2.01\times 10^{-6}\ \text{cm}^{-1}\,\text{sr}^{-1}.$$

From these results we see that by exploiting the resonance in the ionic contribution to the photoelastic constants for frequencies of the incident light close to ω_T it is possible to compensate to a great extent the decrease in scattering efficiency which follows on reducing the frequency of the incident light from the visible range to the infrared, due to the factor of ω_0^4 in the expression for the scattering efficiency. The scattering efficiency for the ionic Brillouin effect is only one to two orders of magnitude smaller than that for the conventional Brillouin effect in KCl, and three to four orders of magnitude smaller in the case of GaAs. A reason for the scattering efficiency being comparatively smaller in the case of GaAs, at least for the scattering geometry of case (b), may be the reduction in the magnitudes of the coefficients a_{xyxy} and b_{xyxy} due to the near cancellation of the contributions from the coefficients $M_{xyxy}^{(a)}$ and $M_{xyxy}^{(b)}$ in the former case, and from the coefficients $\hat{f}_{xyxy}^{(1a)}$ and $\hat{f}_{xyxy}^{(1b)}$ in the latter. In addition, the electronic contributions to the photoelastic constants of GaAs are about an order of magnitude greater than those of KCl, while the ionic contributions are comparable. Thus one has to work with incident frequencies much closer to ω_T in the case of GaAs to obtain the same degree of resonant enhancement of the scattering efficiency than is the case for KCl.

It remains only to discuss whether it is possible in practice to use frequencies of the incident light within 5% of ω_T without absorption becoming large. In the case of GaAs the half-width of the transverse optical phonon at low temperatures ($< 100\,^\circ$K) is less than 0.3 cm^{-1},[20] which is less than 0.1% of ω_T. We are unaware of experimental values for the half-width of the transverse optical phonon in KCl. However, room-temperature data for the half-widths of the transverse optical phonons in NaCl[21] and LiF[22] show that these are of the order of 2 and 4% of ω_T, respectively. These widths are likely to be somewhat smaller at low temperatures, and set the scale for the half-width of the transverse optical phonon in KCl. Consequently, it appears as if it is possible to use incident light of frequencies close enough to ω_T in alkali halide and III-V compound semiconductor crystals to obtain the kind of resonance enhancement of the scattering efficiency predicted here.

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Ionic Raman Effect. III. First- and Second-Order Ionic Raman Effects*

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The modulation of the ionic, rather than the electronic, contribution to the crystal polarizability by the atomic displacements is the dominant mechanism for scattering light when the incident light is in the infrared, and it is the ionic Raman effect that is studied in this paper. The problem considered is the effect of the second-order dipole moment on one- and two-phonon scattering spectra, in addition to the effect of the first-order dipole moment. An expression for the scattering efficiency is obtained and an order-of-magnitude estimate is included for the case of one-phonon scattering and for the case of two-phonon scattering in a crystal that possesses no first-order dipole moment. In the case of a crystal which also possesses a first-order dipole moment, there are additional contributions to the scattering efficiency, but these contributions are discussed qualitatively.

I. INTRODUCTION

The Raman scattering of light by the phonons in a crystal arises from the modulation of both the electronic and ionic contributions to the crystal polarizability by the displacements of the atoms from their equilibrium positions. For incident light in the visible region, the dominant scattering mechanism is the electronic one because the frequency of the incident light is large compared with the transition frequencies between vibrational states of the crystal corresponding to the electronic ground state.[1] However, a previous study[2] indicates that for incident light in the infrared region, the ionic mechanism is the dominant one, and as we now have available infrared lasers which emit at frequencies comparable with the vibrational transition frequencies, the observation of the ionic Raman effect should be possible.

The reason for this is that the scattering efficiency is proportional to the fourth power of the scattered frequency. In going from the region of visible frequencies to the infrared this factor can decrease by as much as five orders of magnitude. The electronic contribution to the Raman tensor is largely independent of frequency for frequencies of the incident (and scattered) light well below the frequency of the lowest electronic transition. Thus the intensity of infrared radiation scattered by the electronic mechanism will be as much as five orders of magnitude lower than the intensity of visible light scattered by this mechanism. However, in the ionic Raman effect, this fourth-power proportionality can be overcome due to the existence of resonances in the Raman tensor for frequencies of the incident light close to the frequencies of the optical vibration modes.

In a recent paper, Wallis and Maradudin[3] considered the contribution to the ionic Raman effect from the first-order dipole moment. In this paper we supplement their results by including the effect of the second-order dipole moment on one-phonon scattering spectra. In addition, we consider the case of two-phonon, or second-order, processes both in crystals which lack a first-order dipole moment and in crystals which possess a first-order dipole moment. We present expressions for the scattering efficiency for one-phonon and two-phonon scattering processes in the case of crystals which lack a first-order dipole moment.

II. EQUATIONS OF MOTION FOR SECOND-ORDER DIPOLE MOMENT

The scattering efficiency per unit solid angle, per unit frequency interval for scattering by the