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Photoelastic and surface-corrugation contributions to Brillouin scattering from an opaque crystal

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We present a theory of the Brillouin scattering of light from long-wavelength acoustical phonons in an opaque crystal which takes into account the effects associated with a finite skin depth. The theory incorporates the photoelastic back scattering of the penetrating light, the effect of the thermal elastic strain induced surface corrugation on the reflected light, as well as the interference between the two scattering processes. We relate the scattering cross section to the spectral density of the elastic-strain fluctuations in the materials, which we construct from the relevant classical elastic Green's functions for a semi-infinite elastic medium bounded by a stress-free surface. We present representative computed line shapes and compare the relative strengths of the different contributions to the spectra for various angles of incidence.

I. INTRODUCTION

Any theory of the inelastic scattering of light from elementary excitations in opaque solids must take explicit account of the boundary or boundaries through which the light enters and leaves the solid. This is due both to the refraction of the light as it enters and leaves the solid and to its attenuation with increasing distance into the solid.

The experimental observation by Parker *et al.*[1] of inelastic scattering of light from the optical vibration modes of long wavelength in the polyatomic metals Zn, Mg, Bi, and $AuAl_2$ stimulated further interest, both theoretical and experimental, in the inelastic scattering of light from opaque solids. A theory of this effect was presented shortly after,[2] and was soon followed by a theory of the inelastic scattering of light from the long-wavelength acoustical vibration modes of metals and opaque semiconductors.[3] At about the same time experimental results for the inelastic scattering of light from long-wavelength acoustical modes in silicon and germanium were presented by Sandercock,[4] together with theoretical considerations concerning such scattering processes. Subsequent theoretical and experimental work on this problem has been carried out by Dresselhaus and Pine,[5] by Dervisch and Loudon,[6] and by Dil and Brody.[7] Recently, Loudon[8] has pointed out that the approximation of the dynamical Green's function for a semi-infinite crystal used by Bennett *et al.*[3] to simplify the calculation of the scattering cross section led to a distortion of the resulting line shape. In this paper Loudon has further studied the scattering at normal incidence for a crystal slab of finite thickness on the basis of a Green's-function approach. In a recent paper by the present authors[9] a theory of the inelastic scattering of light from the long-wavelength acoustical modes in a semi-infinite elastic medium, assumed to be isotropic, was presented. This work, like that of Loudon,[8] is based on the dynamical Green's-function tensor for a semi-infinite, isotropic elastic medium, bounded by a plane, stress-free surface, which has been determined recently.[10] It is not limited to normal incidence, and no simplifying approximation is made concerning the Green's tensor, of the kind employed earlier by Bennett *et al.*[3] in calculations of line shapes.

All of the work cited above dealing with the inelastic scattering of light by long-wavelength acoustical phonons in opaque solids has invoked the usual mechanism governing the scattering process, namely, the modulation of the dielectric tensor of the solid by the long-wavelength acoustical vibrational modes, which can be treated as spatially and temporally varying strains in the solid, the elasto-optical effect. However, it has been shown recently by Mishra and Bray[11] that the dominant mechanism involved in the inelastic scattering of light from acoustoelectrically amplified long-wavelength transverse bulk acoustical waves in GaAs and CdS is reflection from the ripples produced on the surface of the solid as the acoustical wave passes through it.

In the present paper, we present a theory of the inelastic scattering of light from long-wavelength, thermal equilibrium, acoustical phonons, in an opaque solid which incorporates the contributions to the scattering cross section from scattering via the elasto-optical effect, reflection by surface ripples, and the interference between these two scattering mechanisms. Calculations of the scattering cross section are carried out for varying angles of incidence and for polarizations of the incident light parallel and perpendicular to the plane of incidence, as well as for several scattering angles, and the relative importance of the two primary scattering mechanisms is determined as a function of these experimental conditions. Because the scattering is due to long-wavelength acoustical phonons (and to Rayleigh surface waves as well), the Fourier transforms with respect to time of the strain and displacement correlation functions, and the mixed strain-displacement correlation functions, which enter the expression for the scattering cross section, can all be expressed in terms of the dynamical Green's tensor for a semi-infinite elastic medium bounded by a planar stress-free surface. The elements of this tensor have been obtained recently for an isotropic medium[10] and for an hexagonal medium whose basal plane is parallel to the surface.[12] The scattering of the electromagnetic radiation is studied in first Born approximation with the aid of the electromagnetic Green's functions used in our earlier work[2,3] and developed further in more recent work.[13] The numerical calculations of the scattering cross section are carried out for aluminum, with the approximation that it can be treated as an isotropic elastic medium, and the elements of the photoelastic tensor required are obtained from the work of Bennett *et al.*[3]

When the work reported here was nearly completed, two papers dealing with the theory of the scattering of light from surface ripples due to long-wavelength, thermal equilibrium, acoustical phonons in an opaque solid appeared in print.[14,15] In the first of these, Dervisch and Loudon[14] have shown that the contribution to the cross section for scattering from liquid metals arising from the reflection by surface ripples dominates that from the conventional elasto-optical effect. In the second, Loudon[15] reaches the same conclusion for the scattering from opaque solids generally. In these two papers the scattering by surface ripples is treated independently from the scattering by the elasto-optical effect, but the conclusions reached are qualitatively and quantitatively in agreement with those presented here.

II. FORMAL DERIVATION OF THE SCATTERING EFFICIENCY

We consider the scattering of a plane wave of electromagnetic radiation incident upon the surface of an opaque medium. We assume the amplitude of the electric field inside the medium is attenuated as we move deeper into the material such that we may neglect any scattering from the back surface. Hence it is sufficient to treat the material, in the absence of any thermal fluctuations, as if it filled the entire half space $x_3 < 0$, with its surface in the x_1-x_2 plane.

The scattering geometry we consider is illustrated in Fig. 1. The wave vector of the incident radiation $\vec{k}_0$ is in the x_1-x_3 plane and makes an angle θ_I with the normal to the interface. The angles θ_s and φ_s specify the direction of the scattered wave.

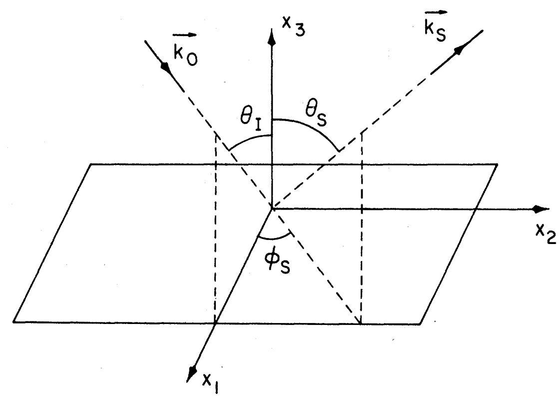

FIG. 1. Scattering geometry considered in this paper.

From Maxwell's equations, we obtain the vector differential equation governing the electric field, in the absence of any macroscopic currents flowing in the medium, to be

$$\vec{\nabla}\times[\vec{\nabla}\times\vec{E}(\vec{x},t)] = -\frac{1}{c^2}\frac{\partial^2}{\partial t^2}\vec{D}(\vec{x},t). \tag{2.1}$$

The electric displacement vector is given by

$$D_i(\vec{x},t) = \sum_j \int dt'\epsilon_{ij}(\vec{x}|t,t')E_j(\vec{x},t'), \tag{2.2}$$

and we write the dielectric tensor as

$$\epsilon_{ij}(\vec{x}|t,t') = \int \frac{d\omega}{2\pi} e^{-i\omega(t-t')}\epsilon_{ij}(\omega|\vec{x},t). \tag{2.3}$$

In writing Eq. (2.2) we have assumed a (spatially) local relationship between $\vec{D}(\vec{x},t)$ and $\vec{E}(\vec{x},t')$. Due to atomic displacements in the medium, the boundary defining the interface between the crystal and vacuum is given by

$$x_3 = u_3(\vec{x}_\parallel 0, t), \tag{2.4}$$

where $\vec{u}(\vec{x}_\parallel x_3, t)$ is the time-dependent elastic displacement at the point $\vec{x} = \vec{x}_\parallel + x_3\hat{x}_3$, $\vec{x}_\parallel = x_1\hat{x}_1 + x_2\hat{x}_2$ being the component of the position vector parallel to the surface of the crystal. Consequently, the dielectric tensor for the vacuum-crystal system is given by

$$\epsilon_{ij}(\omega|\vec{x},t) = \Theta(x_3 - u_3(\vec{x}_\parallel 0,t))\delta_{ij} + \Theta(u_3(\vec{x}_\parallel 0,t) - x_3)\epsilon_{ij}^{(<)}(\omega|\vec{x},t), \tag{2.5}$$

where $\epsilon_{ij}^{(<)}(\omega|\vec{x},t)$ denotes the dielectric tensor inside the material. We may write

$$\epsilon_{ij}^{(<)}(\omega|\vec{x},t) = \epsilon_0^{(<)}(\omega)\delta_{ij} + \delta\epsilon_{ij}^{(1)}(\omega|\vec{x},t), \tag{2.6a}$$

where

$$\epsilon_0^{(<)}(\omega)=\epsilon_1(\omega)+i\epsilon_2(\omega) \tag{2.6b}$$

is the complex dielectric function of the medium in the absence of any strain fluctuations, which we have taken to be isotropic. The modulation $\delta\epsilon_{ij}^{(1)}(\omega|\vec{x},t)$ arises from the photoelastic coupling to the elastic fluctuations,

$$\delta\epsilon_{ij}^{(1)}(\omega|\vec{x},t)=\sum_{kl}k_{ijkl}(\omega)u_{kl}(\vec{x},t)\,. \tag{2.7a}$$

here $k_{ijkl}(\omega)$ are the photoelastic coefficients related to Pockels' elasto-optical coefficients through the relation

$$k_{ijkl}(\omega)=\sum_{mn}\epsilon_{0_{im}}^{(<)}(\omega)p_{mnkl}\epsilon_{0_{nj}}^{(<)}\,, \tag{2.7b}$$

and $u_{ij}(\vec{x},t)$ are the elastic displacement gradients,

$$u_{ij}(\vec{x},t)=\partial u_i(\vec{x},t)/\partial x_j\,. \tag{2.7c}$$

After some manipulation we may write the dielectric tensor of the vacuum-crystal system Eq. (2.5) in the form

$$\epsilon_{ij}(\omega|\vec{x},t)=\epsilon_0(\omega|x_3)\delta_{ij}+\delta\epsilon_{ij}^{(1)}(\omega|\vec{x},t)\Theta(-x_3)+\delta\epsilon_{ij}^{(2)}(\omega|\vec{x},t)\,. \tag{2.8a}$$

Here $\epsilon_0(\omega|x_3)$ is the dielectric function for the vacuum-crystal system in the absence of any fluctuations,

$$\epsilon_0(\omega|x_3)=\Theta(x_3)+\epsilon_0^{(<)}(\omega)\Theta(-x_3)\,. \tag{2.8b}$$

The photoelastic modulation of the dielectric tensor inside the medium $\delta\epsilon_{ij}^{(1)}(\omega|\vec{x},t)$ has been defined in Eq. (2.7a). The second modulation, $\delta\epsilon_{ij}^{(2)}(\omega|\vec{x},t)$, which arises from the presence of surface corrugation, is given by

$$\delta\epsilon_{ij}^{(2)}(\omega|\vec{x},t)=[\epsilon_{ij}^{(<)}(\vec{\omega}|x,t)-\delta_{ij}]U(\vec{x};t)\,, \tag{2.8c}$$

where

$$U(\vec{x};t)=\Theta(u_3(\vec{x}_{\parallel}0,t)-x_3)-\Theta(-x_3)\,. \tag{2.8d}$$

With the use of Eqs. (2.3) and (2.8) the differential equation obeyed by the electric field Eq. (2.1) may be rewritten as

$$\sum_j\left(\frac{\partial^2}{\partial x_i\partial x_j}-\delta_{ij}\nabla^2\right)E_j(\vec{x},t)+\sum_j\frac{\delta_{ij}}{c^2}\frac{\partial^2}{\partial t^2}\int dt'\int\frac{d\Omega}{2\pi}\epsilon_0(\Omega|x_3)e^{-i\Omega(t-t')}E_j(\vec{x},t')$$
$$=-\frac{1}{c^2}\sum_j\frac{\partial^2}{\partial t^2}\int dt'\int\frac{d\Omega}{2\pi}e^{-i\Omega(t-t')}[\Theta(-x_3)\delta\epsilon_{ij}^{(1)}(\omega|\vec{x},t)+\delta\epsilon_{ij}^{(2)}(\omega|\vec{x},t)]E_j(\vec{x},t')\,. \tag{2.9}$$

We define a Green's function $G_{\alpha\beta}(\vec{x},\vec{x}';t-t')$ as the solution of

$$\sum_\beta\left(\frac{\partial^2}{\partial x_\alpha\partial x_\beta}-\delta_{\alpha\beta}\nabla^2\right)G_{\beta\gamma}(\vec{x},\vec{x}';t-t')+\sum_\beta\frac{\delta_{ij}}{c^2}\frac{\partial^2}{\partial t^2}\int dt''\int\frac{d\Omega}{2\pi}\epsilon_0(\Omega|x_3)e^{-i\Omega(t-t'')}G_{\beta\gamma}(\vec{x},\vec{x}';t''-t')$$
$$=-4\pi\delta_{\alpha\gamma}\delta(\vec{x}-\vec{x}')\delta(t-t')\,, \tag{2.10}$$

subject to boundary conditions at the plane $x_3=0$ which ensure the continuity of the appropriate electric and magnetic field components across it, as well as outgoing wave conditions as $x_3\to-\infty$.

With the help of this Green's function Eq. (2.9) may be transformed into an integral equation,

$$E_\alpha(\vec{x},t)=E_\alpha^{(0)}(\vec{x},t)+\frac{1}{4\pi c^2}\sum_{\beta\gamma}\int d^3x'\int dt'G_{\alpha\beta}(\vec{x},\vec{x}';t-t')$$
$$\times\frac{\partial^2}{\partial t'^2}\int dt''\int\frac{d\Omega}{2\pi}e^{-i\Omega(t'-t'')}[\Theta(-x_3')\delta\epsilon_{\beta\gamma}^{(1)}(\Omega|\vec{x}'t')$$
$$+\delta\epsilon_{\beta\gamma}^{(2)}(\Omega|\vec{x}'t')]E_\gamma(\vec{x}',t'')\,,] \tag{2.11}$$

where $\vec{E}^{(0)}(\vec{x},t)$ is the solution of the homogeneous equation and corresponds to the electric field of the wave specularly reflected from the flat surface in the absence of fluctuations.

We first consider the following integral involving $\delta\epsilon^{(2)}$ in Eq. (2.11),

$$\sum_{\beta\gamma}\int d^3x'\,G_{\alpha\beta}(\vec{x},\vec{x}';t-t')\delta\epsilon_{\beta\gamma}^{(2)}(\Omega|\vec{x}'t')E_\gamma(\vec{x}',t'')$$
$$=\sum_{\beta\gamma}\int d^3x'G_{\alpha\beta}(\vec{x},\vec{x}';t-t')[\epsilon_{\beta\gamma}^{(<)}(\Omega|\vec{x}'t')-\delta_{\beta\gamma}]\times U(\vec{x}';t')E_\gamma(\vec{x}',t'')\,. \tag{2.12}$$

From the definition of the function $U(\vec{x}';t')$ in Eq. (2.8d), we note that when $u_3(\vec{x}_\parallel'0,t')>0$,

$$U(\vec{x}';t)=\begin{cases}1 & \text{if } 0<x_3<u_3(\vec{x}_\parallel 0,t')\,,\\ 0 & \text{otherwise},\end{cases} \tag{2.13a}$$

and when $u_3(\vec{x}_\parallel'0,t')<0$,

$$U(\vec{x}';t)=\begin{cases}1 & \text{if } u_3(\vec{x}_\parallel'0,t')<x_3<0\,,\\ 0 & \text{otherwise.}\end{cases} \tag{2.13b}$$

Thus, we have from Eq. (2.12)

$$\begin{aligned}\sum_{\beta\gamma}\int d^3x' G_{\alpha\beta}(\vec{x},\vec{x}';t-t')\delta\epsilon^{(2)}_{\beta\gamma}(\Omega|\vec{x}',t')E_\gamma(\vec{x}',t'')
&=\sum_{\beta\gamma}\int d^2x_\parallel'\Theta(u_3(\vec{x}_\parallel'0,t'))\int_0^{u_3(\vec{x}_\parallel'0,t')}dx_3' G_{\alpha\beta}(\vec{x},\vec{x}';t-t')\,[\epsilon^{(<)}_{\beta\gamma}(\Omega|\vec{x}'t')-\delta_{\beta\gamma}]E_\gamma(\vec{x}'t'')\\
&+\sum_{\beta\gamma}\int d^2x_\parallel'\Theta(-u_3(\vec{x}_\parallel'0,t'))\int_{u_3(\vec{x}_\parallel'0',t)}^{0}dx_3' G_{\alpha\beta}(\vec{x},\vec{x}';t-t')\,[\epsilon^{(<)}_{\beta\gamma}(\Omega|\vec{x}'t')-\delta_{\beta\gamma}]E_\gamma(\vec{x}'t'')\,.\end{aligned} \tag{2.14}$$

Since $u_3(\vec{x}_\parallel'0,t')$, the surface ripple amplitude, is small we retain only terms up to the lowest order in this amplitude on the right-hand side of Eq. (2.14). We do this by replacing the integrands in the integrals over x_3' by their values at $x_3'=0$. In doing so, we take account of the fact that the discontinuity of $G_{\alpha\beta}$ is across the flat surface, $x_3=0$, while that of the electric field $\vec{E}$ is across the rippled surface, $x_3=u_3(\vec{x}_\parallel 0,t')$. Thus up to terms linear in $u_3(\vec{x}_\parallel 0,t')$, Eq. (2.14) reduces to

$$\begin{aligned}\sum_{\beta\gamma}\int d^3x' G_{\alpha\beta}(\vec{x},\vec{x}';t-t')\delta\epsilon^{(2)}_{\beta\gamma}(\Omega|\vec{x}'t')E_\gamma(\vec{x}',t'')
&=[\epsilon_0^{(<)}(\Omega)-1]\sum_{\beta}\int d^2x_\parallel'[\Theta(u_3(\vec{x}_\parallel'0,t'))G_{\alpha\beta}(\vec{x},\vec{x}_\parallel'0+;t-t')E_\beta(\vec{x}_\parallel'0-;t'')\\
&+\Theta(-u_3(\vec{x}_\parallel'0,t'))G_{\alpha\beta}(\vec{x},\vec{x}_\parallel'0-;t-t')E_\beta(\vec{x}_\parallel'0+;t'')]u_3(\vec{x}_\parallel'0,t')\,.\end{aligned} \tag{2.15}$$

Here 0+ and 0− denote, respectively, whether the limit $x_3=0$ is approached from above or from below. Using the above result we may write Eq. (2.11) in the form

$$\begin{aligned}E_\alpha(\vec{x},t)&=E_\alpha^{(0)}(\vec{x},t)+\frac{1}{4\pi c^2}\sum_{\beta\gamma}\int d^2x_\parallel'\int_{-\infty}^{0}dx_3'\int dt' G_{\alpha\beta}(\vec{x},\vec{x}';t-t')\\
&\times\frac{\partial^2}{\partial t'^2}\int dt''\int\frac{d\Omega}{2\pi}e^{-i\Omega(t'-t'')}\delta\epsilon^{(1)}_{\beta\gamma}(\Omega|\vec{x}'t')E_\gamma(\vec{x}',t'')\\
&+\frac{1}{4\pi c^2}\sum_{\beta}\int d^2x_\parallel''\int dt'\int dt''\int\frac{d\Omega}{2\pi}[\epsilon_0^{(<)}(\Omega)-1]\\
&\times\Big(G_{\alpha\beta}(\vec{x},\vec{x}_\parallel'0+;t-t')\frac{\partial^2}{\partial t'^2}\Theta(u_3(\vec{x}_\parallel'0,t'))E_\beta(\vec{x}_\parallel'0-;t'')\\
&+G_{\alpha\beta}(\vec{x},\vec{x}_\parallel'0-;t-t')\frac{\partial^2}{\partial t'^2}\Theta(-u_3(\vec{x}_\parallel'0,t'))E_\beta(\vec{x}_\parallel'0+;t'')\Big)u_3(\vec{x}_\parallel'0,t')\,.\end{aligned} \tag{2.16}$$

We are interested in obtaining an expression for the scattered field in the first Born approximation. Thus, we replace the exact field $\vec{E}(\vec{x},t)$ in the integrands on the right-hand side of Eq. (2.16) by the solution to the homogeneous equation, $\vec{E}^{(0)}(\vec{x},t)$.

The Green's function $G_{\alpha\beta}(\vec{x},\vec{x}';t-t')$ defined by Eq. (2.10) has been evaluated by Maradudin and Mills.[13] From their results, we observe that

$$G_{\alpha\beta}(\vec{x},\vec{x}_\parallel'0+;t-t')E_\beta^{(0)}(\vec{x}_\parallel'0-;t'')=G_{\alpha\beta}(\vec{x},\vec{x}_\parallel'0-;t-t')E_\beta^{(0)}(\vec{x}_\parallel'0+;t'')\,. \tag{2.17}$$

With the help of Eq. (2.17) and the identity $\Theta(x)+\Theta(-x)\equiv 1$, the expression for the scattered field in the first Born approximation becomes

$$E_\alpha^{(s)}(\vec{x},t)=E_\alpha^{(1)}(\vec{x},t)+E_\alpha^{(2)}(\vec{x},t)\,, \tag{2.18}$$

where

$$\begin{aligned}E_\alpha^{(1)}(\vec{x},t)&=\frac{1}{4\pi c^2}\sum_{\beta\gamma}\int d^2x_\parallel'\int_{-\infty}^{0}dx_3'\int dt' G_{\alpha\beta}(\vec{x},\vec{x}';t-t')\frac{\partial^2}{\partial t'^2}\int dt''\int\frac{d\Omega}{2\pi}e^{-i\Omega(t'-t'')}\delta\epsilon^{(1)}_{\beta\gamma}\\
&\times(\Omega|\vec{x}'t')E_\gamma^{(0)}(\vec{x},t'')\,,\end{aligned} \tag{2.19a}$$

and

$$E_\alpha^{(2)}(\bar{x},t)=\frac{1}{4\pi c^2}\sum_\beta \int d^2x'_\parallel \int dt' G_{\alpha\beta}(\bar{x},\bar{x}'_\parallel 0+;t-t')\frac{\partial^2}{\partial t'^2}\int dt'' \int \frac{d\Omega}{2\pi}[\epsilon_0^{(<)}(\Omega)-1]e^{-i\Omega(t'-t'')} \times u_3(\bar{x}'_\parallel 0,t')E_\beta^{(0)}(\bar{x}'_\parallel 0-;t'') . \qquad (2.19b)$$

$\vec{E}^{(1)}$ is the contribution to the scattered field due to photoelastic scattering and $\vec{E}^{(2)}$ is the contribution due to scattering from surface corrugations.

In obtaining the result for the scattered electric field given by Eqs. (2.18) and (2.19), in particular in deriving Eq. (2.19b) for the amplitude of the field scattered by the surface corrugations, we have, in fact, used the Green's-function approach developed by Agarwal[16] for the study of the scattering of electromagnetic radiation from a rough surface. In the present case the displacement amplitude $u_3(\bar{x}_\parallel 0,t)$ plays the same role as the surface roughness profile function in Agarwal's formulation of the rough surface problem. The scattered field $\vec{E}^{(1)}(\bar{x},t)$ due to the photoelastic effect is a first-order field (in the deformation parameters), is independent of the surface corrugation, and satisfies the Maxwell boundary conditions across the plane surface $x_3=0$. As a result, the amount by which it fails to satisfy the boundary conditions at the corrugated surface is of at least second order, involving products of the deformation parameters and the amplitude of the surface corrugation. The zero-order field $\vec{E}^{(0)}(\bar{x},t)$ and the scattered field $\vec{E}^{(2)}(\bar{x},t)$ due to scattering from the corrugated surface then combine to satisfy the boundary conditions at the corrugated surface to first order in $u_3(\bar{x}_\parallel 0,t)$ just as in Agarwal's theory. For additional references which bear on the equivalence of the result obtained by Agarwal with those obtained by other approaches, the reader is directed to the papers by Kröger and Kretschmann,[17] Marvin *et al.*,[18] Mills,[19] and Elson.[20] From Ref. 13 we have the expression for $E_\alpha^{(0)}(\bar{x},t)$ for $x_3<0$:

$$E_\alpha^{(0)}(\bar{x},t)=e^{i\vec{k}_\parallel^{(0)}\cdot\vec{x}_\parallel-i\omega_0 t}e^{ik_{3;I}^{(i)}x_3}\hat{E}_\alpha^{(0)}(\vec{k}^{(0)}) , \qquad (2.20a)$$

where

$$\hat{E}_\alpha^{(0)}(\vec{k}^{(0)})=\sum_\lambda \Gamma_\alpha^\lambda(\vec{k}^{(0)})E_\lambda^{(I)} . \qquad (2.20b)$$

Here $\vec{k}^{(0)}=k_\parallel^{(0)}\hat{x}_1+k_3^{(0)}\hat{x}_3$ is the wave vector of the incident light in the vacuum, ω_0 the frequency of the incident, light and

$$k_{3;I}^{(i)}=-[\epsilon_0^{(i)}(\omega_0)\omega_0^2/c^2-k_\parallel^{(0)2}]^{1/2} , \ \mathrm{Im}(k_{3;I}^{(i)})<0 , \qquad (2.21)$$

is the x_3 component of the incident wave vector inside the medium. $E_\lambda^{(I)}$ is the amplitude of the incident electric field of polarization λ (parallel or perpendicular to the plane of incidence) and

$$\vec{\Gamma}^\perp(\vec{k}^{(0)})=\frac{2k_3^{(0)}}{k_3^{(0)}-k_{3;I}^{(i)}}\hat{x}_2 , \qquad (2.22a)$$

$$\vec{\Gamma}^\parallel(\vec{k}^{(0)})=2\frac{k_{3;I}^{(i)}\hat{x}_1-k_\parallel^{(0)}\hat{x}_3}{k_{3;I}^{(i)}-\epsilon_0^{(i)}(\omega_0)k_3^{(0)}} . \qquad (2.22b)$$

Substituting Eq. (2.20a) into Eqs. (2.19a) and (2.19b), and performing the integration over t'', we get

$$E_\alpha^{(1)}(\bar{x},t)=\frac{1}{4\pi c^2}\sum_{\beta\gamma}\int d^2x'_\parallel\int_{-\infty}^0 dx'_3\int dt' G_{\alpha\beta}(\bar{x},\bar{x}';t-t')e^{i\vec{k}_\parallel^{(0)}\cdot\vec{x}'_\parallel+ik_{3;I}^{(i)}x'_3}\frac{\partial^2}{\partial t'^2} \times [e^{-i\omega_0 t'}\delta\epsilon_{\beta\gamma}^{(1)}(\omega_0|\bar{x}'t')]\hat{E}_\gamma^{(0)}(\vec{k}^{(0)}) , \qquad (2.23a)$$

and

$$E_\alpha^{(2)}(\bar{x},t)=\frac{1}{4\pi c^2}\sum_\beta\int d^2x'_\parallel\int dt' G_{\alpha\beta}(\vec{x},\vec{x}'_\parallel 0+;t-t')e^{i\vec{k}_\parallel^{(0)}\cdot\vec{x}'_\parallel}[\epsilon_0^{(<)}(\omega_0)-1]\frac{\partial^2}{\partial t'^2} \times[e^{-i\omega_0 t'}u_3(\bar{x}'_\parallel 0,t')]\hat{E}_\beta^{(0)}(\vec{k}^{(0)}) . \qquad (2.23b)$$

In these equations, we observe that due to the smallness of the acoustical-phonon frequencies compared to ω_0, the frequency of the incident light, we may ignore the time derivatives of $\delta\epsilon_{\beta\gamma}^{(1)}(\omega_0|\bar{x}'t')$ and $u_3(\bar{x}'_\parallel 0,t')$ compared to the derivatives of $e^{-i\omega_0 t'}$. Thus, we have

$$E_\alpha^{(1)}(\bar{x},t)=\frac{-\omega_0^2}{4\pi c^2}\sum_{\beta\gamma}\int d^2x'_\parallel\int_{-\infty}^0 dx'_3\int dt' G_{\alpha\beta}(\bar{x},\bar{x}';t-t')e^{i\vec{k}_\parallel^{(0)}\cdot\vec{x}'_\parallel+ik_{3;I}^{(i)}x'_3-i\omega_0 t'} \times\delta\epsilon_{\beta\gamma}^{(1)}(\omega_0|\bar{x}'t')\hat{E}_\gamma^{(0)}(\vec{k}^{(0)}) , \qquad (2.24a)$$

and

$$E_\alpha^{(2)}(\vec{x},t)=\frac{-\omega_0^2[\epsilon_0^{(<)}(\omega_0)-1]}{4\pi c^2}\sum_\beta\int d^2x'_\parallel\int dt' G_{\alpha\beta}(\vec{x},\vec{x}'_\parallel 0+;t-t')\,e^{i\vec{k}_\parallel^{(0)}\cdot\vec{x}'_\parallel}e^{-i\omega_0 t'}u_3(\vec{x}'_\parallel,t')\hat{E}_\beta^{(0)}(\vec{k}^{(0)})\,. \tag{2.24b}$$

The translation invariance of the system in the x_1-x_2 plane allows a partial Fourier transformation of the Green's functions,

$$G_{\alpha\beta}(\vec{x},\vec{x}';t-t')=\int\frac{d^2k_\parallel}{(2\pi)^2}\int\frac{d\Omega}{2\pi}\,e^{i\vec{k}_\parallel\cdot(\vec{x}_\parallel-\vec{x}'_\parallel)-i\Omega(t-t')}\,\hat{G}_{\alpha\beta}(\vec{k}_\parallel\Omega;x_3x'_3)\,. \tag{2.25}$$

One has[13] for $x_3>0$

$$\hat{G}_{\alpha\beta}(\vec{k}_\parallel\Omega;x_3 0+=(4\pi c^2/\Omega^2)e^{ik_3x_3}g_{\alpha\beta}^{(+)}(\vec{k}_\parallel\Omega) \tag{2.26a}$$

with

$$g_{ij}^{(+)}(\vec{k}_\parallel\Omega)=\sum_{kl}S_{ki}(\vec{k}_\parallel)S_{lj}(\vec{k}_\parallel)\tilde{g}_{kl}(\vec{k}_\parallel\Omega|+)\,, \tag{2.26b}$$

$$\tilde{g}_{kl}(\vec{k}_\parallel\Omega|+)=\frac{i}{k_3^{(i)}-\epsilon(\Omega)k_3}\begin{pmatrix}-k_3^{(i)}k_3 & 0 & -k_\parallel k_3\epsilon(\Omega)\\ 0 & \dfrac{\Omega^2}{c^2}\dfrac{k_3^{(i)}-\epsilon(\Omega)k_3}{k_3^{(i)}-k_3} & 0\\ k_\parallel k_3^{(i)} & 0 & \epsilon(\Omega)k_\parallel^2\end{pmatrix}. \tag{2.26c}$$

Here $S_{ij}(\vec{k}_\parallel)$ is the matrix which performs a rotation in the x_1-x_2 plane to align $\vec{k}_\parallel$ along the x_1 direction,

$$S_{ij}(\vec{k}_\parallel)=\frac{1}{k_\parallel}\begin{pmatrix}k_1 & k_2 & 0\\ -k_2 & k_1 & 0\\ 0 & 0 & k_\parallel\end{pmatrix}. \tag{2.27}$$

The wave-vector components k_3 and $k_3^{(i)}$ are defined by

$$k_3=\begin{cases}(\Omega^2/c^2-k_\parallel^2)^{1/2}\,, & \dfrac{\Omega^2}{c^2}>k_\parallel^2 \quad (2.28a)\\[2ex] i\left(k_\parallel^2-\dfrac{\Omega^2}{c^2}\right)^{1/2}\,, & \dfrac{\Omega_2}{c^2}<k_\parallel^2\,, \quad (2.28b)\end{cases}$$

$$k_3^{(i)}=-\left[\epsilon(\Omega)\,\frac{\Omega^2}{c^2}-k_\parallel^2\right]^{1/2}\,,\quad \mathrm{Im}(k_3^{(i)})<0\,. \tag{2.28c}$$

For $x_3>0$ and $x'_3<0$, the region of interest for $E_\alpha^{(1)}(\vec{x},t)$, we have

$$\hat{G}_{\alpha\beta}(\vec{k}_\parallel\Omega;x_3x'_3)=\frac{4\pi c^2}{\Omega^2}\,e^{ik_3x_3}e^{ik_3^{(i)}x'_3}g_{\alpha\beta}(\vec{k}_\parallel,\Omega) \tag{2.29a}$$

with

$$g_{ij}(\vec{k}_\parallel,\Omega)=\sum_{kl}S_{ki}(\vec{k}_\parallel)S_{lj}(\vec{k}_\parallel)\,\tilde{g}_{kl}(\vec{k}_\parallel\Omega)\,, \tag{2.29b}$$

$$\tilde{g}_{kl}(\vec{k}_\parallel\Omega)=\frac{i}{k_3^{(i)}-\epsilon(\Omega)k_3}\begin{pmatrix}-k_3^{(i)}k_3 & 0 & -k_\parallel k_3\\ 0 & \dfrac{\Omega^2}{c^2}\dfrac{k_3^{(i)}-\epsilon(\Omega)k_3}{k_3^{(i)}-k_3} & 0\\ k_\parallel k_3^{(i)} & 0 & k_\parallel^2\end{pmatrix}. \tag{2.29c}$$

Using Eqs. (2.25)–(2.29), we get

$$E_\alpha^{(1)}(\vec{x},t)=-\omega_0^2\sum_{\beta\gamma}\int d^2x'_\parallel\int_{-\infty}^0 dx'_3\int dt'\int\frac{d^2k_\parallel}{(2\pi)^2}\int\frac{d\Omega}{2\pi}\,e^{i\vec{k}\cdot\vec{x}-i\Omega t}\,\frac{1}{\Omega^2}\,e^{i(\vec{k}_\parallel^{(0)}-\vec{k}_\parallel)\cdot\vec{x}'_\parallel}$$

$$\times e^{i(k_3^{(i)}+k_{3;I}^{(i)})x'_3}e^{-i(\omega_0-\Omega)t'}g_{\alpha\beta}(\vec{k}_\parallel,\Omega)\,\delta\epsilon_{\beta\gamma}^{(1)}(\omega_0|\vec{x}'t')\hat{E}_\gamma^{(0)}(\vec{k}^{(0)})\,, \tag{2.30a}$$

$$E_\alpha^{(2)}(\vec{x},t) = -\omega_0^2[\epsilon_0^{(<)}(\omega_0)-1]\sum_\beta \int d^2x_\parallel' \int dt' \int \frac{d^2k_\parallel}{(2\pi)^2}\int\frac{d\Omega}{2\pi}\frac{1}{\Omega^2}\, e^{i\vec{k}\cdot\vec{x}-i\Omega t} \times e^{i(\vec{k}_\parallel^{(0)}-\vec{k}_\parallel)\cdot\vec{x}_\parallel'} e^{-i(\omega_0-\Omega)t'} g_{\alpha\beta}^{(+)}(\vec{k}_\parallel\Omega)\,\hat{E}_\beta^{(0)}(\vec{k}^{(0)})\,u_3(\vec{x}_\parallel' 0;t') . \tag{2.30b}$$

If, recalling Eq. (2.7), we set

$$h_{\alpha\mu\nu}(\vec{k}_\parallel,\Omega) \equiv \sum_{\beta\gamma} g_{\alpha\beta}(\vec{k}_\parallel,\Omega)k_{\beta\gamma\mu\nu}(\omega_0)\,\hat{E}_\gamma^{(0)}(\vec{k}^{(0)}) , \tag{2.31a}$$

in Eq. (2.30a) we obtain

$$E_\alpha^{(1)}(\vec{x},t) = -\omega_0^2\sum_{\mu\nu}\int d^2x_\parallel' \int_{-\infty}^0 dx_3' \int dt' \int\frac{d^2k_\parallel}{(2\pi)^2}\int\frac{d\Omega}{2\pi}\, e^{i\vec{k}\cdot\vec{x}-i\Omega t}\frac{1}{\Omega^2}\, e^{i(\vec{k}_\parallel^{(0)}-\vec{k}_\parallel)\cdot\vec{x}_\parallel'} e^{i(k_3^{(i)}+k_{3;l}^{(i)})x_3'} e^{-i(\omega_0-\Omega)t'} \times h_{\alpha\mu\nu}(\vec{k}_\parallel,\Omega)\,u_{\mu\nu}(\vec{x}',t') . \tag{2.31b}$$

Invoking Maxwell's equations, we may obtain the magnetic field of the scattered wave, $\vec{H}^{(s)}(\vec{x},t)$ in terms of the electric field $\vec{E}^{(s)}(\vec{x},t)$ defined by Eqs. (2.18), (2.30b), and (2.31b). The magnetic field may also be written in the form

$$\vec{H}^{(s)}(\vec{x},t) = \vec{H}^{(1)}(\vec{x},t) + \vec{H}^{(2)}(\vec{x},t) , \tag{2.32}$$

where the first term arises from the photoelastic scattering, while the second term arises from the surface-ripple mechanism of scattering. The time-averaged rate of energy flow in the scattered wave is given by the real part of the complex Poynting vector

$$\vec{S}(\vec{x},t) = \frac{c}{8\pi}[\vec{E}^{(s)}(\vec{x},t)]^* \times \vec{H}^{(s)}(\vec{x},t) . \tag{2.33}$$

In view of Eqs. (2.19a) and (2.32), we may write

$$\vec{S}(\vec{x},t) = \sum_{i=1}^2\sum_{j=1}^2 \vec{S}^{(ij)}(\vec{x},t) , \tag{2.34a}$$

where

$$\vec{S}^{(ij)}(\vec{x},t) = \frac{c}{8\pi}[\vec{E}^{(i)}(\vec{x},t)]^* \times \vec{H}^{(j)}(\vec{x},t) . \tag{2.34b}$$

After some straightforward algebra using Eqs. (2.30b) and (2.31b), one has

$$S_\alpha^{(ij)}(\vec{x},t) = \frac{c^2\omega_0^4}{8\pi}\int\frac{d\Omega}{2\pi}\int\frac{d^2q_\parallel}{(2\pi)^2}\frac{1}{\omega_s^5}\, k_\alpha^{(s)} S_{ij}(\vec{q}_\parallel\Omega) , \tag{2.35a}$$

with

$$S_{11}(\vec{q}_\parallel\Omega) = \sum_{\mu\nu}\sum_{\mu'\nu'}\sum_\beta [h_{\beta\mu\nu}(\vec{k}_\parallel^{(s)},\omega_s)]^* h_{\beta\mu'\nu'}(\vec{k}_\parallel^{(s)},\omega_s) \times \int_{-\infty}^0 dx_3' \int_{-\infty}^0 dx_3''\, e^{-i\kappa_3^* x_3'} e^{i\kappa_3 x_3''}\hat{\mathfrak{D}}_{\mu\mu'}^{\nu\nu'}(\vec{q}_\parallel\Omega|x_3'x_3'') , \tag{2.35b}$$

$$S_{12}(\vec{q}_\parallel\Omega) = [\epsilon_0^{(<)}(\omega_0)-1]\sum_{\mu\nu}\sum_\beta [h_{\beta\mu\nu}(\vec{k}_\parallel^{(s)},\omega_s)]^* \sum_\lambda g_{\beta\lambda}^{(+)}(k_\parallel^{(s)}\omega_s) \times \hat{E}_\lambda^{(0)}(\vec{k}_\parallel^{(0)})\int_{-\infty}^0 dx_3'\, e^{-i\kappa_3^* x_3'}\hat{\mathfrak{D}}_{\mu 3}^{\nu}(\vec{q}_\parallel\Omega|x_3' 0) , \tag{2.35c}$$

$$S_{21}(\vec{q}_\parallel\Omega) = [\epsilon_0^{(<)}(\omega_0)^*-1]\sum_\beta\sum_\lambda [g_{\beta\lambda}^{(+)}(\vec{k}_\parallel^{(s)}\omega_s)]^*[\hat{E}_\lambda^{(0)}(\vec{k}^{(0)})]^* \times\sum_{\mu\nu} h_{\beta\mu\nu}(\vec{k}_\parallel^{(s)}\omega_s)\int_{-\infty}^0 dx_3''\, e^{i\kappa_3 x_3''}\hat{\mathfrak{D}}_{3\mu}^{\nu}(\vec{q}_\parallel\Omega|0x_3'') , \tag{2.35d}$$

and

$$S_{22}(\vec{q}_\parallel\Omega) = |\epsilon_0^{(<)}(\omega_0)-1|^2\sum_\beta\sum_\lambda\sum_{\lambda'} g_{\beta\lambda}^{(+)}(k_\parallel^{(s)}\omega_s)[\hat{E}_\lambda^{(0)}(\vec{k}^{(0)})]^* \times g_{\beta\lambda'}^{(+)}(\vec{k}_\parallel^{(s)}\omega_s)\hat{E}_\lambda^{(0)}(\vec{k}^{(0)})\hat{\mathfrak{D}}_{33}(\vec{q}_\parallel\Omega|00) . \tag{2.35e}$$

In the above equations, we have introduced the quantities

$$\omega_s = \omega_0 - \Omega\,, \tag{2.36a}$$

$$\vec{k}_\parallel^{(s)} = \vec{k}_\parallel^{(0)} - \vec{q}_\parallel\,, \tag{2.36b}$$

$$k_3^{(s)} = \begin{cases} [\omega_s^2/c^2 - (k_\parallel^{(s)})^2]^{1/2}\,, & \dfrac{\omega_s^2}{c^2} > (k_\parallel^{(s)})^2 \\ i[(k_\parallel^{(s)})^2 - \omega_s^2/c^2]^{1/2}\,, & (k_\parallel^{(s)})^2 > \omega_s^2/c^2\,, \end{cases} \tag{2.36c}$$

and

$$\kappa_3 = k_{3;I}^{(i)} + k_{3;s}^{(i)}\,, \tag{2.36d}$$

with

$$k_{3;s}^{(i)} = -\left(\epsilon(\omega_s)\,\frac{\omega_s^2}{c^2} - (k_\parallel^{(s)})^2\right)^{1/2}, \quad \mathrm{Im}(k_{3;s}^{(i)}) < 0\,. \tag{2.36e}$$

Furthermore, the quantities $\hat{\mathfrak{D}}^{\beta\nu}_{\alpha\mu}(\vec{q}_\parallel\Omega\,|\,x_3x_3')$ appearing in Eqs. (2.35b)–(2.35e) are related to the displacement gradient and displacement correlation functions according to

$$\langle u_{\alpha\beta}(\vec{x},t)u_{\mu\nu}(\vec{x}',t')\rangle = \int\frac{d^2q_\parallel}{(2\pi)^2}\int\frac{d\Omega}{2\pi}\,e^{i\vec{q}_\parallel\cdot(\vec{x}_\parallel-\vec{x}_\parallel')} \times e^{-i\Omega(t'-t'')}\hat{\mathfrak{D}}^{\beta\nu}_{\alpha\mu}(\vec{q}_\parallel\Omega\,|\,x_3x_3')\,, \tag{2.37a}$$

$$\langle u_{\alpha\beta}(\vec{x},t)u_3(\vec{x}_\parallel'0,t')\rangle = \int\frac{d^2q_\parallel}{(2\pi)^2}\int\frac{d\Omega}{2\pi}\,e^{i\vec{q}_\parallel\cdot(\vec{x}_\parallel-\vec{x}_\parallel')} \times e^{-i\Omega(t'-t'')}\hat{\mathfrak{D}}^{\beta}_{\alpha3}(\vec{q}_\parallel\Omega\,|\,x_3 0)\,, \tag{2.37b}$$

$$\langle u_3(\vec{x}_\parallel 0,t)u_{\alpha\beta}(\vec{x}',t')\rangle = \int\frac{d^2q_\parallel}{(2\pi)^2}\int\frac{d\Omega}{(2\pi)}\,e^{i\vec{q}_\parallel\cdot(\vec{x}_\parallel-\vec{x}_\parallel')} \times e^{-i\Omega(t'-t'')}\hat{\mathfrak{D}}_{3\alpha}^{\;\;\beta}(\vec{q}_\parallel\Omega\,|\,0x_3')\,, \tag{2.37c}$$

and

$$\langle u_3(\vec{x}_\parallel 0,t)u_3(\vec{x}_\parallel'0,t')\rangle = \int\frac{d^2q_\parallel}{(2\pi)^2}\int\frac{d\Omega}{2\pi}\,e^{i\vec{q}_\parallel\cdot(\vec{x}_\parallel-\vec{x}_\parallel')} \times e^{-i\Omega(t'-t'')}\hat{\mathfrak{D}}_{33}(\vec{q}_\parallel\Omega\,|\,00)\,. \tag{2.37d}$$

Note that the Fourier transforms of all the above correlation functions may be obtained from the Fourier transforms of the displacement correlation functions,

$$\langle u_\alpha(\vec{x},t)u_\beta(\vec{x}',t)\rangle = \int\frac{d^2q_\parallel}{(2\pi)^2}\int\frac{d\Omega}{2\pi}\,e^{i\vec{q}_\parallel\cdot(\vec{x}_\parallel-\vec{x}_\parallel')} \times e^{-i\Omega(t'-t'')}\hat{\mathfrak{D}}_{\alpha\beta}(\vec{q}_\parallel\Omega\,|\,x_3x_3')\,, \tag{2.38}$$

through the relationships

$$\hat{\mathfrak{D}}^{\beta\nu}_{\alpha\mu}(q_\parallel\Omega\,|\,x_3x_3') = \left(iq_\beta(1-\delta_{\beta3}) + \delta_{\beta3}\,\frac{\partial}{\partial x_3}\right) \times\left(-iq_\nu(1-\delta_{\nu3}) + \delta_{\nu3}\,\frac{\partial}{\partial x_3'}\right) \times\hat{\mathfrak{D}}_{\alpha\mu}(q_\parallel\Omega\,|\,x_3x_3')\,, \tag{2.39a}$$

$$\hat{\mathfrak{D}}^{\beta}_{\alpha3}(\vec{q}_\parallel\Omega\,|\,x_3 0) = \left(iq_\beta(1-\delta_{\beta3}) + \delta_{\beta3}\,\frac{\partial}{\partial x_3}\right) \times\hat{\mathfrak{D}}_{\alpha3}(\vec{q}_\parallel\Omega\,|\,x_3 0)\,, \tag{2.39b}$$

and

$$\hat{\mathfrak{D}}_{3\alpha}^{\;\;\beta}(\vec{q}_\parallel\Omega\,|\,0x_3') = \left(-iq_\beta(1-\delta_{\beta3}) + \delta_{\beta3}\,\frac{\partial}{\partial x_3'}\right) \times\hat{\mathfrak{D}}_{3\alpha}(\vec{q}_\parallel\Omega\,|\,0x_3')\,. \tag{2.39c}$$

In Appendix A we show that the displacement correlation functions are in turn related to the classical elastic dynamical Green's functions $D_{\alpha\beta}(\vec{q}_\parallel\Omega\,|\,x_3x_3')$ through

$$\hat{\mathfrak{D}}_{\alpha\beta}(\vec{q}_\parallel\Omega\,|\,x_3x_3') = \frac{i\hbar\,[n(\Omega)+1]}{\rho} \times\{D_{\alpha\beta}(\vec{q}_\parallel,\Omega+i0\,|\,x_3x_3') - [D_{\beta\alpha}(\vec{q}_\parallel,\Omega+i0\,|\,x_3'x_3)]^*\}\,. \tag{2.40}$$

Here $n(\Omega)$ is the Bose-Einstein occupation factor and ρ is the mass density of the crystal.

Now from Eqs. (2.34a) and (2.35a) it is a simple matter to show that the contribution to the average rate of energy flow per unit area into the solid angle $d\Omega_s$ about the direction (θ_s,φ_s) due to modes with a given $\vec{q}_\parallel$, from radiation in the interval $(\omega_s,\omega_s+d\omega_s)$ is given by

$$S_{ij}(\theta_s,\varphi_s)\,d\Omega_s\,d\omega_s = \frac{1}{8\pi c}\,\frac{\cos\theta_s}{8\pi^3}\,\frac{\omega_0^4}{\omega_s^2}\,S_{ij}(q_\parallel\Omega)\,. \tag{2.41}$$

Here, and in what follows, $\Omega = \omega_0 - \omega_s$.

From this result we now proceed to obtain the scattering efficiencies. Using the definitions Eq. (2.20b) and Eq. (2.31a), we write

$$\sum_\beta\,[h_{\beta\mu\nu}(\vec{k}_\parallel^{(s)},\omega_s)]^*h_{\beta\mu'\nu'}(\vec{k}_\parallel^{(s)},\omega_s) = \sum_{\xi,\eta=\parallel,\perp} T_{\xi\eta}^{(11)}(\mu\nu;\mu'\nu')E_\xi^{(I)}E_\eta^{(I)}\,, \tag{2.42a}$$

where

$$T_{\xi\eta}^{(11)}(\mu\nu;\mu'\nu') = \sum_{\rho\sigma}\sum_{\gamma\delta} m_{\rho\sigma}^{(11)}(\vec{k}_\parallel^{(s)}\omega_s)[k_{\rho\gamma\mu\nu}(\omega_0)]^* \times k_{\sigma\delta\mu'\nu'}(\omega_0)[\Gamma_\gamma^\xi(\vec{k}^{(0)})]^*\Gamma_\delta^\eta(\vec{k}^{(0)})\,, \tag{2.42b}$$

and

$$m_{\rho\sigma}^{(11)}(\vec{k}_{\parallel}^{(s)}\omega_s)=\sum_{\tau}\,[g_{\tau\rho}(\vec{k}_{\parallel}^{(s)}\omega_s)]^* g_{\tau\sigma}(\vec{k}_{\parallel}^{(s)}\omega_s)\,. \tag{2.42c}$$

Also,

$$[\epsilon_0^{(<)}(\omega_0)-1]\sum_{\beta\lambda}\,[h_{\beta\mu\nu}(\vec{k}_{\parallel}^{(s)}\omega_s)]^*\; g_{\beta\lambda}^{(+)}(\vec{k}_{\parallel}^{(s)}\omega_s)\hat{E}_{\lambda}^{(0)}(\vec{k}^{(0)}) = \sum_{\xi,\,\eta=\parallel,\perp} T_{\xi\eta}^{(12)}(\mu\nu)E_{\xi}^{(I)}E_{\eta}^{(I)}\,, \tag{2.43a}$$

$$T_{\xi\eta}^{(12)}(\mu\nu)=\sum_{\rho\sigma}\sum_{\lambda} m_{\rho\sigma}^{(12)}(\vec{k}_{\parallel}^{(s)}\omega_s)[k_{\rho\lambda\mu\nu}(\omega_0)]^* \times[\Gamma_{\lambda}^{\xi}(\vec{k}^{(0)})]^*\Gamma_{\sigma}^{\eta}(\vec{k}^{(0)})\,, \tag{2.43b}$$

$$m_{\rho\sigma}^{(12)}=[\epsilon_0^{(<)}(\omega_0)-1]\sum_{\tau}\,[g_{\tau\rho}(\vec{k}_{\parallel}^{(s)}\omega_s)]^*\, g_{\tau\sigma}^{(+)}(k_{\parallel}^{(s)}\omega_s)\,. \tag{2.43c}$$

Similarly we have two more sets,

$$\{[\epsilon_0^{(<)}(\omega_0)]^*-1\}\sum_{\beta\lambda}\,[g_{\beta\lambda}^{(+)}(\vec{k}_{\parallel}^{(s)}\omega_s)]^* \times[\hat{E}_{\lambda}^{(0)}(\vec{k}^{(0)})]^* h_{\beta\mu\nu}(\vec{k}_{\parallel}^{(s)}\omega_s) = \sum_{\xi,\,\eta=\parallel,\perp} T_{\xi\eta}^{(21)}(\mu\nu)E_{\xi}^{(I)}E_{\eta}^{(I)}\,, \tag{2.44a}$$

$$T_{\xi\eta}^{(21)}(\mu\nu)=\sum_{\rho\sigma}\sum_{\lambda} m_{\rho\sigma}^{(21)}(\vec{k}_{\parallel}^{(s)}\omega_s)k_{\sigma\lambda\mu\nu}(\omega_0) \times[\Gamma_{\rho}^{\xi}(\vec{k}^{(0)})]^*\Gamma_{\lambda}^{\eta}(\vec{k}^{(0)})\,, \tag{2.44b}$$

$$m_{\rho\sigma}^{(21)}(\vec{k}_{\parallel}^{(s)}\omega_s)=[\epsilon_0^{(<)}(\omega_0)^*-1]\sum_{\tau}\,[g_{\tau\rho}^{(+)}(\vec{k}_{\parallel}^{(s)}\omega_s)]^* \times g_{\tau\sigma}(\vec{k}_{\parallel}^{(s)}\omega_s)\,, \tag{2.44c}$$

$$|\epsilon_0^{(<)}(\omega_0)-1|^2\sum_{\beta}\sum_{\lambda}\sum_{\lambda'}\,[g_{\beta\lambda}^{(+)}(\vec{k}_{\parallel}^{(s)}\omega_s)]^* g_{\beta\lambda'}^{(+)}(\vec{k}_{\parallel}^{(s)}\omega_s)[\hat{E}_{\lambda}^{(0)}(\vec{k}^{(0)})]^*\hat{E}_{\lambda'}^{(0)}(\vec{k}^{(0)})=\sum_{\xi,\,\eta=\parallel,\perp} T_{\xi\eta}^{(22)}E_{\xi}^{(I)}E_{\eta}^{(I)}\,, \tag{2.45a}$$

$$T_{\xi\eta}^{(22)}=|\epsilon_0^{(<)}(\omega_0)-1|^2\sum_{\rho\sigma} m_{\rho\sigma}^{(22)}(\vec{k}_{\parallel}^{(s)}\omega_s) \times[\Gamma_{\rho}^{\xi}(\vec{k}^{(0)})]^*\Gamma_{\sigma}^{\eta}(\vec{k}^{(0)})\,, \tag{2.45b}$$

and

$$m_{\rho\sigma}^{(22)}(\vec{k}_{\parallel}^{(s)}\omega_s)=\sum_{\tau}\,[g_{\tau\rho}^{(+)}(\vec{k}_{\parallel}^{(s)}\omega_s)]^* g_{\tau\sigma}^{(+)}(\vec{k}_{\parallel}^{(s)}\omega_s)\,. \tag{2.45c}$$

If we choose the incident radiation to be polarized either parallel or perpendicular to the plane of incidence, from Eqs. (2.34)–(2.45) we obtain for the scattering efficiency for polarization η (= $\parallel$ or $\perp$),

$$\frac{d^2 Z_\eta}{d\Omega_s d\omega_s}=\frac{\omega_0^4\cos\theta_s}{c^2\omega_s^2} \times\frac{\hbar[n(\Omega)+1]}{8\pi^3\rho}\,\mathrm{Re}\,\sum_{i,j=1}^{2}\left(A_{ij}^{(\eta)}(\vec{q}_{\parallel}\Omega)\right), \tag{2.46}$$

where

$$A_{11}^{(\eta)}(\vec{q}_{\parallel}\Omega)=i\sum_{\alpha\beta}\sum_{\mu\nu} T_{\eta\eta}^{(11)}(\alpha\beta;\mu\nu) \times\int_{-\infty}^{0}dx_3\int_{-\infty}^{0}dx_3'e^{-i\kappa_3^* x_3} \times e^{i\kappa_3 x_3'}\mathfrak{D}_{\alpha\mu}^{\beta\nu}(\vec{q}_{\parallel}\Omega\,|\,x_3x_3') \tag{2,47a}$$

$$A_{12}^{(\eta)}(\vec{q}_{\parallel}\Omega)=i\sum_{\alpha\beta} T_{\eta\eta}^{(12)}(\alpha\beta) \times\int_{-\infty}^{0}dx_3 e^{-i\kappa_3^* x_3}\mathfrak{D}_{\alpha 3}^{\beta}(\vec{q}_{\parallel}\Omega\,|\,x_3 0)\,, \tag{2.47b}$$

$$A_{21}^{(\eta)}(\vec{q}_{\parallel}\Omega)=i\sum_{\alpha\beta} T_{\eta\eta}^{(21)}(\alpha\beta) \times\int_{-\infty}^{0}dx_3'e^{i\kappa_3 x_3'}\mathfrak{D}_{3\alpha}^{\;\beta}(\vec{q}_{\parallel}\Omega\,|\,0x_3)\,, \tag{2.47c}$$

and

$$A_{22}^{(\eta)}(\vec{q}_{\parallel}\Omega)=iT_{\eta\eta}^{(22)}\mathfrak{D}_{33}(\vec{q}_{\parallel}\Omega\,|\,00)\,. \tag{2.47d}$$

In the above we have introduced

$$\mathfrak{D}_{\alpha\beta}(\vec{q}_{\parallel}\Omega\,|\,x_3x_3')=\frac{\rho}{i\hbar[n(\Omega)+1]}\,\hat{\mathfrak{D}}_{\alpha\beta}(\vec{q}_{\parallel}\Omega\,|\,x_3x_3')\,. \tag{2.48}$$

From Eqs. (2.48) and (2.40), we note that the quantities $\mathfrak{D}_{\alpha\beta}$ are related to the elastic Green's functions $D_{\alpha\beta}$ through

$$\mathfrak{D}_{\alpha\beta}(\vec{q}_{\parallel}\Omega\,|\,x_3x_3')=\{D_{\alpha\beta}(\vec{q}_{\parallel}\Omega+i0\,|\,x_3x_3') - [D_{\beta\alpha}(\vec{q}_{\parallel}\Omega+i0\,|\,x_3'x_3)]^*\}\,. \tag{2.49}$$

This completes the formal derivation of the scattering efficiency in terms of the classical elastic Green's functions.

III. BRILLOUIN SPECTRUM FOR A CRYSTAL WITH ELASTIC ISOTROPY

The discussion in Sec. II has been quite general. In this section we obtain explicit expressions for the scattering efficiencies assuming photoelastic coefficients appropriate to a cubic medium and elastic isotropy.

With this assumption the photoelastic tensor assumes the following form

$$k_{\alpha\beta\mu\nu}=k_{12}\delta_{\alpha\beta}\delta_{\mu\nu}+k_{44}(\delta_{\alpha\mu}\delta_{\beta\nu}+\delta_{\alpha\nu}\delta_{\beta\mu})+(k_{11}-k_{12}-2k_{44})\delta_{\alpha\beta}\delta_{\mu\nu}\delta_{\alpha\mu}. \tag{3.1}$$

Using this form for the photoelastic tensor in Eqs. (2.42b), (2.43b), and (2.44b), we may write Eqs. (2.47a)–(2.47c) as

$$A_{11}^{(\eta)}(\vec{q}_{\parallel}\Omega)=i\int_{-\infty}^{0}dx_3\int_{-\infty}^{0}dx_3'e^{-i\kappa_3^*x_3}e^{i\kappa_3x_3'}\times\sum_{i=1}^{3}\sum_{j\geq i}\Bigg[\left(-\alpha_{ij}^{(11)}(\eta)+\beta_{ij}^{(11)}(\eta)\frac{\partial^2}{\partial x_3\partial x_3'}-i\chi_{ij}^{(11)}(\eta)\frac{\partial}{\partial x_3}+i\lambda_{ij}^{(11)}(\eta)\frac{\partial}{\partial x_3'}\right)\mathfrak{D}_{ij}(\vec{q}_{\parallel}\Omega\,|\,x_3x_3')$$
$$+\left(-\alpha_{ij}^{(11)}(\eta)^*+\beta_{ij}^{(11)}(\eta)^*\frac{\partial^2}{\partial x_3\partial x_3'}+i\chi_{ij}^{(11)}(\eta)^*\frac{\partial}{\partial x_3'}-i\lambda_{ij}(\eta)^*\frac{\partial}{\partial x_3}\right)\mathfrak{D}_{ji}(\vec{q}_{\parallel}\Omega\,|\,x_3x_3')\Bigg]. \tag{3.2a}$$

$$\tilde{A}_{12}^{(\eta)}(\vec{q}_{\parallel}\Omega)=A_{12}^{(\eta)}(\vec{q}_{\parallel}\Omega)+A_{21}^{(\eta)}(\vec{q}_{\parallel}\Omega)$$
$$=i\Bigg[\int_{-\infty}^{0}dx_3e^{-i\kappa_3^*x_3}\sum_{j=1}^{3}\left(i\alpha_{j3}^{(12)}(\eta)+\beta_{j3}^{(12)}(\eta)\frac{\partial}{\partial x_3}\right)\mathfrak{D}_{j3}(x_3,0)$$
$$+\int_{-\infty}^{0}dx_3e^{i\kappa_3x_3}\sum_{j=1}^{3}\left(-i\alpha_{j3}^{(12)}(\eta)^*+\beta_{j3}^{(12)}(\eta)^*\frac{\partial}{\partial x_3}\right)\mathfrak{D}_{3j}(0,x_3)\Bigg]. \tag{3.2b}$$

Explicit expressions for the coefficients $\alpha_{ij}^{(11)}$, $\beta_{ij}^{(11)}$, and $\alpha_j^{(12)}$ are written out in Appendix B.

The relation between the correlation functions $\mathfrak{D}_{ij}$ appearing in Eq. (3.2) and the dynamical elastic Green's functions D_{ij} is established in Appendix A and displayed in Eq. (2.49). Using this relation, we may note that the integrals in Eq. (3.2) have the form

$$\int_{-\infty}^{0}dx_3\int_{-\infty}^{0}dx_3'e^{-i\kappa_3^*x_3}e^{i\kappa_3x_3'}\times[A\mathfrak{D}_{ij}(x_3,x_3')\pm A^*\mathfrak{D}_{ji}(x_3,x_3')]$$
$$=\begin{cases}2i\,\mathrm{Im}\\ 2\,\mathrm{Re}\end{cases}\Bigg[\int_{-\infty}^{0}dx_3\int_{-\infty}^{0}dx_3'\,e^{-i\kappa_3^*x_3}e^{i\kappa_3x_3'}\times[AD_{ij}(x_3,x_3')\pm A^*D_{ji}(x_3,x_3')]\Bigg]. \tag{3.3}$$

A similar relation holds for the integrals involving the derivatives with respect to x_3 or x_3'.

The exact dynamical elastic Green's functions for an isotropic semi-infinite medium bounded by a stress-free planar surface have been determined recently by Maradudin and Mills.[10] We list these in Appendix C, modified to apply to our geometry. These functions may be split into two parts

$$D_{ij}(\vec{q}_{\parallel}\Omega\,|\,x_3x_3')=D_{ij}^{(B)}(\vec{q}_{\parallel}\Omega\,|\,x_3x_3')+D_{ij}^{(S)}(\vec{q}_{\parallel}\Omega\,|\,x_3x_3'). \tag{3.4}$$

Here $D_{ij}^{(B)}$ is that part of the Green's function that is independent of the presence of the surface and $D_{ij}^{(S)}$ is the part that arises from the presence of the surface. The following symmetry properties of these functions may be noted

$$D_{ii}^{(B)}(x_3,x_3')=D_{ii}^{(B)}(x_3',x_3), \tag{3.5a}$$

$$D_{12}^{(B)}(x_3,x_3')=D_{21}^{(B)}(x_3,x_3')=D_{12}^{(B)}(x_3',x_3), \tag{3.5b}$$

$$D_{13,23}^{(B)}(x_3,x_3')=D_{31,32}^{(B)}(x_3,x_3')=-D_{13,23}^{(B)}(x_3',x_3), \tag{3.5c}$$

$$D_{ii}^{(S)}(x_3,x_3')=D_{ii}^{(S)}(x_3',x_3), \tag{3.6a}$$

$$D_{12}^{(S)}(x_3,x_3')=D_{21}^{(S)}(x_3,x_3')=D_{12}^{(S)}(x_3',x_3), \tag{3.6b}$$

$$D_{13,23}^{(S)}(x_3,x_3')=-D_{31,32}^{(S)}(x_3',x_3). \tag{3.6c}$$

These properties in conjunction with Eqs. (3.4) allow us to obtain expressions for the scattering efficiencies in closed form. In order to write these in compact form, we write the elastic Green's functions in the following forms

$$D_{ii,12}^{(B)}(x_3,x_3')=\frac{-1}{2\Omega^2}\sum_{\mu=l,t}(\hat{D}_{ii,12}^{(B)})_{\mu}e^{-\alpha_\mu|x_3-x_3'|}, \tag{3.7a}$$

$$D_{13,23}^{(B)}(x_3,x_3')=\frac{-1}{2\Omega^2}\sum_{\mu=l,t}(\hat{D}_{13,23}^{(B)})_{\mu}e^{-\alpha_\mu|x_3-x_3'|}\times\mathrm{sgn}(x_3'-x_3), \tag{3.7b}$$

$$D_{ij}^{(S)}(x_3,x_3')=\frac{-1}{2\Omega^2}\frac{1}{r_+}\sum_{\mu=l,t}\sum_{\nu=l,t}(\hat{D}_{ij}^{(S)})_{\mu\nu}\,e^{\alpha_\mu x_3}e^{\alpha_\nu x_3'}. \tag{3.7c}$$

Here the subscripts l and t refer to longitudinal and transverse parts, respectively. The quantities $\hat{D}_{ij}^{(B)}$ and $\hat{D}_{ij}^{(S)}$ may be inferred from the expressions for the D_{ij} given in Appendix C. We recall that

$$\alpha_{l,t} = (q_\parallel^2 - \Omega^2/c_{l,t}^2)^{1/2}, \quad \mathrm{Re}(\alpha_{l,t}) > 0, \tag{3.8a}$$

$$r_+ = \frac{-4\alpha_l\alpha_t q_\parallel^2 + (\alpha_t^2 + q_\parallel^2)^2}{4\alpha_l\alpha_t(\alpha_t^2 + q_\parallel^2)}, \tag{3.8b}$$

where c_l and c_t are the longitudinal and transverse velocities of acoustic waves in the isotropic medium. Using Eqs. (3.3)–(3.6) we may write Eq. (3.2) in the following form

$$A_{11}^{(\eta)}(\vec{q}_\parallel\Omega) = A_{11}^{(\eta,B)}(\vec{q}_\parallel\Omega) + A_{11}^{(\eta,S)}(\vec{q}_\parallel\Omega), \tag{3.9a}$$

$$\begin{aligned}
A_{11}^{(\eta,B)}(\vec{q}_\parallel,\Omega) = \frac{2}{\Omega^2}\sum_{\mu=l,t}\Bigg\{ \mathrm{Im}\Bigg[\Bigg(&\sum_{i=1}^{3}(\hat{D}_{ii}^{(B)})_\mu\{\mathrm{Re}[\alpha_{ii}^{(11)}(\eta)] - \alpha_\mu^2\mathrm{Re}[\beta_{ii}^{(11)}(\eta)]\} \\
&+ (\hat{D}_{12}^{(B)})_\mu\{\mathrm{Re}[\alpha_{12}^{(11)}(\eta)] - \alpha_\mu^2\mathrm{Re}[\beta_{12}^{(11)}(\eta)]\}\Bigg)I_\mu \\
&+ \Bigg(\sum_{i=1}^{2}(\hat{D}_{13}^{(B)})_\mu\{\mathrm{Re}[\alpha_{13}^{(11)}(\eta)] - \alpha_\mu^2\mathrm{Re}[\beta_{i3}^{(11)}(\eta)]\}\Bigg)J_\mu\Bigg] \\
- \mathrm{Re}\Bigg[\Bigg(&\sum_{i=1}^{3}(\hat{D}_{ii}^{(B)})_\mu\{\mathrm{Re}[\chi_{ii}^{(11)}(\eta)] - \mathrm{Re}[\lambda_{ii}^{(11)}(\eta)]\}\alpha_\mu \\
&+ (\hat{D}_{12}^{(B)})_\mu\{\mathrm{Re}[\chi_{12}^{(11)}(\eta)] - \mathrm{Re}[\lambda_{12}^{(11)}(\eta)]\}\alpha_\mu\Bigg)J_\mu \\
&+ \sum_{i=1}^{2}(\hat{D}_{13}^{(B)})_\mu\{\mathrm{Re}[\chi_{i3}^{(11)}(\eta)] - \mathrm{Re}(\lambda_{i3}^{(11)})\}\alpha_\mu I_\mu\Bigg]\Bigg\}.
\end{aligned} \tag{3.9b}$$

$$\begin{aligned}
A_{11}^{(\eta,S)}(\vec{q}_\parallel\Omega) = \frac{2}{\Omega^2}\sum_{\mu=l,t}\sum_{\nu=l,t}\Bigg\{\mathrm{Im}\Bigg[\frac{1}{r_+}\Bigg(&\sum_{i=1}^{3}(\hat{D}_{ii}^{(S)})_{\mu\nu}\{\mathrm{Re}[\alpha_{ii}^{(11)}(\eta)] + \alpha_\mu\alpha_\nu\mathrm{Re}(\beta_{ii}^{(11)})\} \\
&+ (\hat{D}_{12}^{(S)})_{\mu\nu}\{\mathrm{Re}[\alpha_{12}^{(11)}(\eta)] + \alpha_\mu\alpha_\nu\mathrm{Re}[\beta_{12}^{(11)}(\eta)]\} \\
&+ \frac{1}{2}\sum_{i=1}^{2}((\hat{D}_{i3}^{(S)})_{\mu\nu}[\alpha_{i3}^{(11)}(\eta) + \alpha_\mu\alpha_\nu\beta_{i3}^{(11)}(\eta)] \\
&- (\hat{D}_{i3}^{(S)})_{\nu\mu}\{[\alpha_{i3}^{(11)}(\eta)]^* + \alpha_\mu\alpha_\nu[\beta_{i3}^{(11)}(\eta)]^*\})\Bigg)K_\mu L_\nu\Bigg] \\
- \frac{1}{2}\mathrm{Re}\Bigg[\frac{1}{r_+}\Bigg(&\sum_{i=1}^{3}(\hat{D}_{ii}^{(S)})_{\mu\nu}(\{\chi_{ii}^{(11)}(\eta) + [\lambda_{ii}^{(11)}(\eta)]^*\}\alpha_\mu - \{\lambda_{ii}^{(11)}(\eta) + [\chi_{ii}^{(11)}(\eta)]^*\}\alpha_\nu) \\
&+ (\hat{D}_{12}^{(S)})_{\mu\nu}(\{\chi_{12}^{(11)}(\eta) + [\lambda_{12}^{(11)}(\eta)]^*\}\alpha_\mu - \{[\chi_{12}^{(11)}(\eta)]^* + \lambda_{12}^{(11)}(\eta)\}\alpha_\nu) \\
&+ \sum_{j=1}^{2}([\hat{D}_{i3}^{(S)}]_{\mu\nu}[\chi_{i3}^{(11)}(\eta)\alpha_\mu - \lambda_{i3}^{(11)}(\eta)\alpha_\nu] \\
&+ (\hat{D}_{i3}^{(S)})_{\nu\mu}\{[\chi_{i3}^{(11)}(\eta)]^*\alpha_\nu - [\lambda_{i3}^{(11)}(\eta)]^*\alpha_\mu\})\Bigg)K_\mu L_\nu\Bigg]\Bigg\}.
\end{aligned} \tag{3.9c}$$

Here, the integrals I_μ, J_μ, K_μ, and L_μ are given by

$$\begin{aligned}
I_\mu &\equiv \int_{-\infty}^{0} dx_3 \int_{-\infty}^{0} dx_3'\, e^{-i\kappa_3^* x_3} e^{i\kappa_3 x_3'} e^{-\alpha_\mu |x_3 - x_3'|} \\
&= \frac{-2i\alpha_\mu}{(\kappa_3 - \kappa_3^*)(\kappa_3^2 + \alpha_\mu^2)} + \frac{1}{(i\kappa_3 - \alpha_\mu)(\alpha_\mu - i\kappa_3^*)},
\end{aligned} \tag{3.10a}$$

$$\begin{aligned}
J_\mu &\equiv \int_{-\infty}^{0} dx_3 \int_{-\infty}^{0} dx_3'\, e^{-i\kappa_3^* x_3} e^{i\kappa_3 x_3'} \\
&\quad \times e^{-\alpha_\mu |x_3 - x_3'|}\,\mathrm{sgn}(x_3' - x_3) \\
&= \frac{2\kappa_3}{(\kappa_3 - \kappa_3^*)(\kappa_3^2 + \alpha_\mu^2)} + \frac{1}{(i\kappa_3 - \alpha_\mu)(\alpha_\mu - i\kappa_3^*)},
\end{aligned} \tag{3.10b}$$

$$K_\mu \equiv \int_{-\infty}^{0} dx_3\, e^{-i\kappa_3^* x_3} e^{\alpha_\mu x_3} = \frac{1}{\alpha_\mu - i\kappa_3^*}, \tag{3.10c}$$

and

$$L_\mu \equiv \int_{-\infty}^{0} dx_3'\, e^{i\kappa_3^* x_3'} e^{\alpha_\mu x_3'} = \frac{1}{\alpha_\mu + i\kappa_3}. \tag{3.10d}$$

Similarly, Eq. (3.2b) reduces to

$$\tilde{A}_{12}^{(\eta)}(\vec{q}_\parallel\Omega) = \tilde{A}_{12}^{(\eta,B)}(\vec{q}_\parallel\Omega) + \tilde{A}_{12}^{(\eta,s)}(\vec{q}_\parallel\Omega), \tag{3.11a}$$

$$\tilde{A}_{12}^{(\eta,B)}(\vec{q}_{\parallel}\Omega)=\frac{1}{\Omega^2}\sum_{\mu=l,t}\Bigg[\mathrm{Im}\Bigg(\sum_{j=1}^{2}(D_{j3}^{(B)})_{\mu}\alpha_{\mu}\{\beta_{j3}^{(12)}(\eta)K_{\mu}-[\beta_{j3}^{(12)}(\eta)]^{*}L_{\mu}\}$$
$$+(\hat{D}_{33}^{(B)})_{\mu}\alpha_{\mu}\{\beta_{33}^{(12)}(\eta)K_{\mu}+[\beta_{33}^{(12)}(\eta)]^{*}L_{\mu}\}\Bigg)$$
$$+\mathrm{Re}\Bigg(\sum_{j=1}^{2}(\hat{D}_{j3}^{(B)})_{\mu}\{\alpha_{j3}^{(12)}(\eta)K_{\mu}+[\alpha_{j3}^{(12)}(\eta)]^{*}L_{\mu}\}$$
$$+(\hat{D}_{33}^{(B)})_{\mu}\{\alpha_{33}^{(12)}(\eta)K_{\mu}-[\alpha_{33}^{(12)}(\eta)]^{*}L_{\nu}\}\Bigg)\Bigg]\quad, \tag{3.11b}$$

$$\tilde{A}_{12}^{(\eta,s)}(\vec{q}_{\parallel}\Omega)=\frac{1}{\Omega^2}\sum_{\mu=l,t}\sum_{\nu=l,t}\Bigg\{\mathrm{Im}\Bigg[\frac{1}{r_{+}}\Bigg(\sum_{j=1}^{2}\{(\hat{D}_{j3}^{(s)})_{\mu\nu}\beta_{j3}^{(12)}(\eta)\alpha_{\mu}K_{\mu}-(\hat{D}_{j3}^{(s)})_{\nu\mu}[\beta_{j3}^{(12)}(\eta)]^{*}\alpha_{\nu}L_{\nu}\}$$
$$+(\hat{D}_{33}^{(s)})_{\mu\nu}\{\beta_{33}^{(12)}(\eta)\alpha_{\mu}K_{\mu}+[\beta_{33}^{(12)}(\eta)]^{*}\alpha_{\nu}L_{\nu}\}\Bigg)\Bigg]$$
$$+\mathrm{Re}\Bigg[\frac{1}{r_{+}}\Bigg(\sum_{j=1}^{2}\{(\hat{D}_{j3}^{(s)})_{\mu\nu}\alpha_{j3}^{(12)}(\eta)K_{\mu}+(\hat{D}_{j3}^{(s)})_{\nu\mu}[\alpha_{j3}^{(12)}(\eta)]^{*}L_{\nu}\}$$
$$+(\hat{D}_{33}^{(s)})_{\mu\nu}\{\alpha_{33}^{(12)}(\eta)K_{\mu}-[\alpha_{33}^{(12)}(\eta)]^{*}L_{\nu}\}\Bigg)\Bigg]\Bigg\}. \tag{3.11c}$$

These equations, in conjunction with Eqs. (3.9), (2.47d), and (2.46) constitute algebraic expressions for the scattering efficiency for both polarizations $\eta(=\parallel$ or $\perp)$ of the incident light for an isotropic elastic medium.

In view of the complexity of the final expressions for the cross section it is, perhaps, worthwhile to present a brief summary of the derivation and comment on the meaning of the various contributions to the spectral density before we turn to the numerical evaluation of the line shapes.

We have considered two mechanisms having their origin in thermal acoustical phonons that produce modulation in the dielectric tensor of an opaque crystal. The first is the usual photoelastic modulation given by Eq. (2.7). The second contribution arises from the fact that in the presence of acoustical phonons, the surface of the crystal is no longer flat, but is dynamically corrugated. This term may be written as in Eq. (2.8c). We solve for the scattered field in the first Born approximation with the above two dielectric modulations as the source term in Maxwell's equations. The result is presented in Eqs. (2.18), (2.30b), and (2.31b). The scattering cross section is then obtained from the averaged Poynting's vector given in Eqs. (2.34) and (2.35). The necessary correlation functions, Eqs. (2.37a)—(2.37d), are constructed from the classical elastic dynamical Green's functions through Eq. (2.40). In this section we have specialized to the case of an elastically isotropic medium with photoelastic coefficients appropriate to a cubic medium. The necessary elastic Green's function for such an isotropic semi-infinite medium have been reported recently, and are listed in Appendix C.

The imaginary part of A_{22} given in Eq. (2.47d) is the contribution of the surface corrugation mechanism alone to the Brillouin spectrum. For the special case under consideration, one may easily show that

$$\mathrm{Im}A_{22}(q_{\parallel}\Omega)\sim\mathrm{Im}\frac{\alpha_l}{-4\alpha_l\,\alpha_t\,q_{\parallel}^2+(\alpha_t^2+q_{\parallel}^2)^2}, \tag{3.12}$$

where $\alpha_{l,t}$ are defined in Eq. (3.8a). This agrees with the result of Loudon.[15] This expression has a pole at the frequency corresponding to the Rayleigh surface wave, and zeros at $\Omega=c_t q_{\parallel}$ and $\Omega=c_l q_{\parallel}$ (corresponding to the bulk TA and LA frequencies). There is a nonvanishing contribution for $c_t q_{\parallel}<\Omega<c_l q_{\parallel}$ and for $\Omega>c_l q_{\parallel}$.

The purely photoelastic contribution is given by Eq. (3.9). A variety of line shapes are possible[6,8,21] depending upon the opacity of the medium and the scattering geometry. The general features of this contribution are an intense peak at the frequency corresponding to the Rayleigh surface wave and non-Lorentzian structures at the transverse and longitudinal acoustical wave cutoff frequencies ($\Omega=c_t q_{\parallel}$ and $\Omega=c_l q_{\parallel}$, respectively). For less opaque crystals, structures in the spectrum also appear at the bulk acoustical wave frequencies.

The interference terms that arise from the simultaneous presence of two scattering mechanisms are collected in Eq. (3.11). Any complete theory of Brillouin scattering from an opaque crystal should take account of all the above four contributions. In the next section we present the results of a detailed numerical study of the spectrum using the expressions derived in this section.

IV. NUMERICAL STUDY OF REPRESENTATIVE LINE SHAPES AND DISCUSSION

In the previous section we derived expressions for the Brillouin scattering cross sections for an opaque crystal with elastic isotropy. These expressions are simple algebraic equations, albeit quite lengthy. Rather than make approximations in order to obtain simpler expressions for the line shapes we have carried out a detailed numerical study of the spectra as functions of the angle of incidence and polarization of the incident light.

We consider the scattering of incident light of wavelength 4880 Å from a crystal of aluminum with a planar surface. The elastic constants of Al at room temperature are[22]

$$c_{11}=1.08\times10^{12}\ \text{dynes/cm}^2,$$
$$c_{12}=0.61\times10^{12}\ \text{dynes/cm}^2,$$
$$c_{44}=0.29\times10^{12}\ \text{dynes/cm}^2. \quad (4.1)$$

These values yield an anisotropy ratio, $2c_{44}/(c_{11}-c_{12})$ of 1.24. Hence the use of elastic Green's functions calculated on the basis of elastic isotropy is reasonable. We take for c_t, the speed of transverse acoustical waves in an isotropic medium, the expression

$$c_t=(c_{44}/\rho)^{1/2}, \quad (4.2)$$

which is the speed of transverse acoustical waves propagating along a (100) direction in a cubic crystal. Using the value of c_{44} given above, and a value for the density ρ of 2.7 g/cm^3,[22] we get

$$c_t=3.22\times10^5\ \text{cm/sec}. \quad (4.3a)$$

For an isotropic solid with a Poisson's ratio $\sigma=\frac{1}{3}$, this gives for the speed of longitudinal acoustical waves,

$$c_l=2c_t. \quad (4.3b)$$

For the speed of surface Rayleigh waves, one obtains[23]

$$c_R=0.933c_t. \quad (4.3c)$$

We use the values of ϵ_0, k_{11}, k_{12}, k_{44} at the frequency corresponding to a wavelength of 4880 Å calculated by Bennett *et al.*[3]:

$$\begin{aligned}\epsilon_0&=-38.1+i\,6.05,\\ k_{11}&=42.5-i\,58.8,\\ k_{12}&=34.9-i13.38,\\ k_{44}&=2.08-i\,9.61.\end{aligned} \quad (4.4)$$

The damping of elastic waves (due to anharmonicity, etc.) has not been incorporated into the elastic Green's functions used in the last section. Hence the spectral density contains sharp singularities at frequencies corresponding to the various excitations of the system. The presence of damping would round-off these sharp features. In order to simulate this effect of damping, we have introduced a small positive imaginary part to the frequency shift Ω in computing the spectral densities.

Since frequency shifts due to acoustic modes are very small, to a good approximation $n(\Omega)+1 \approx k_BT/\hbar\Omega$. In this limit, there is no difference between the Stokes and anti-Stokes spectra. The frequency shifts are measured in dimensionless units $\Omega/c_tq_{\parallel}$, where $q_{\parallel}$ is the magnitude of the wave-vector transfer, parallel to the surface.

In Fig. 2 we show a representative spectrum which includes contributions from the surface-ripple mechanism, the photoelastic modulation and the interference between the two processes. The particular spectrum shown is for $\theta_I=67.5°$, $\theta_s=20.7°$, $\phi_s=0°$ with the incident electric field polarized in the plane of incidence. The general features of the spectrum for ($\vec{q}_{\parallel}\neq0$) are quite insensitive to the angles of incidence and scattering as well as to the polarization of the incident light. The integrated intensity of the spectrum for the case when the incident light is polarized parallel to the plane of incidence is higher than when the

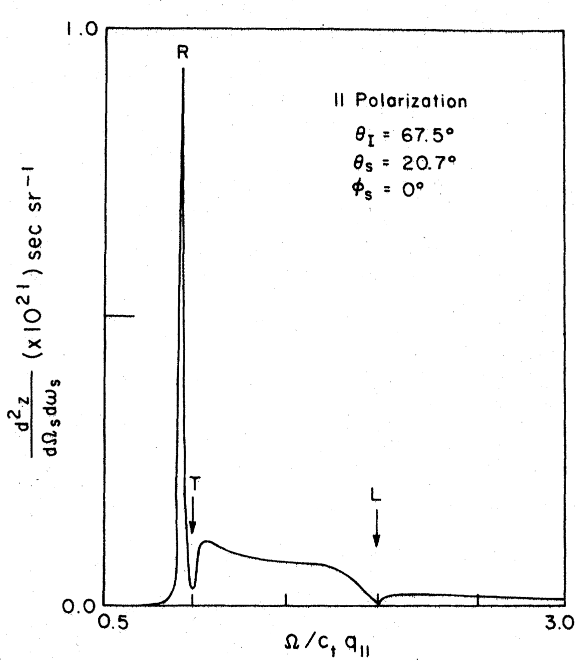

FIG. 2. Representative Brillouin spectrum which includes all contributions to the spectrum.

incident light is polarized perpendicular to the plane of incidence. For a fixed angle of scattering the intensity of the spectrum decreases as the angle of incidence is increased.

The primary features of the spectrum in Fig. 2 to be noted are the Lorentzian peak (R) at the frequency of the Rayleigh waves, the cutoff for propagating transverse acoustic waves (T) and the minimum at the frequency corresponding to that of propagating longitudinal acoustic waves (L).

As indicated by the experiments of Mishra and Bray,[11] and pointed out by Loudon[15] the surface-ripple contribution dominates the spectrum. Indeed, on the scale of Fig. 2 the total spectrum is nearly indistinguishable from the surface-ripple contribution alone. The photoelastic contribution, which is nearly three orders of magnitude smaller than the surface-ripple contribution, is shown in Fig. 3. The incident electric field is polarized perpendicular to the plane of incidence in Fig. (3a). The features to be noted are the Rayleigh peak (R), the "singularity" at the transverse cutoff (T), and the peak at the longitudinal acoustical wave frequency (L). In Fig. 3b the incident light is polarized parallel to the plane of incidence and the "singularity" at the transverse cutoff is absent. These features differ from the photoelastic spectra calculated by Bennett *et al.*[3] due to their neglect of surface contributions to the response function. Our results are in accord with the recent calculations of Loudon.[21]

In Fig. 4 we show the influence of the interference effects of the two scattering mechanisms on the spectra. The solid lines represent the total spectra and the dashed lines give the surface-ripple contributions. As expected, the interference effects are small. They tend to decrease the intensity in the vicinity of the transverse acoustical frequency and enhance the intensity in the vicinity of the longitudinal acoustical frequency. Furthermore, for a fixed angle of scattering, the interference contributions grow as the angle of incidence is increased. We note that the interference effects are much larger than the photoelastic contribution alone.

The primary reason for the dominance of the surface-ripple mechanism in highly opaque crystals is that the cross section for this mechanism is proportional to the reflectivity at the optical frequency of the incident light, which is of the order of unity. On the other hand, the photoelastic contribution depends on the transmission coefficients which are quite small. Hence the interference effects contained in our theory will be more significant in less absorbing materials than the one considered in our numerical study. However, if the optical skin depth for the crystal is

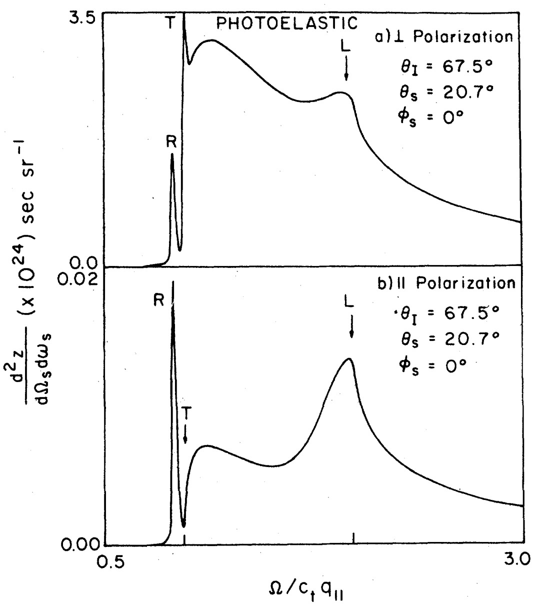

FIG. 3. Photo-elastic contribution to the spectrum when the incident electric field is polarized (a) perpendicular to the plane of incidence, (b) parallel to the plane of incidence.

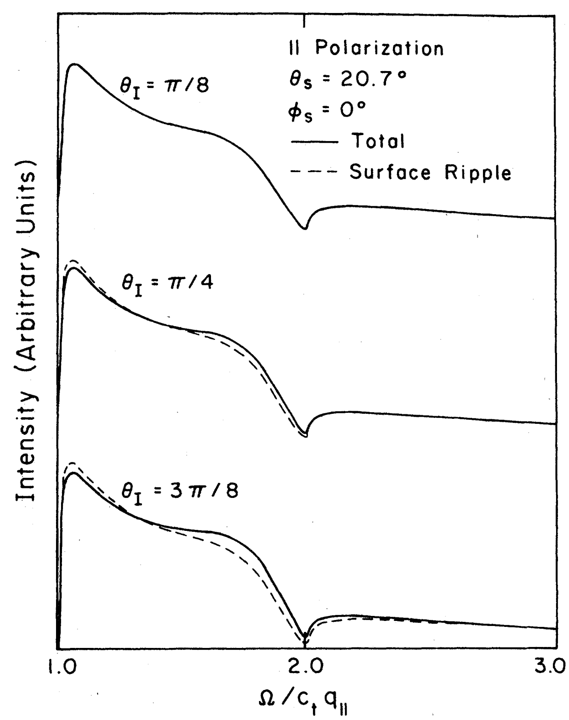

FIG. 4. Interference effect between the two scattering mechanisms as a function of the angle of incidence for a fixed angle of scattering. The solid lines are the total spectra and the dashed lines are the surface-ripple contributions alone.

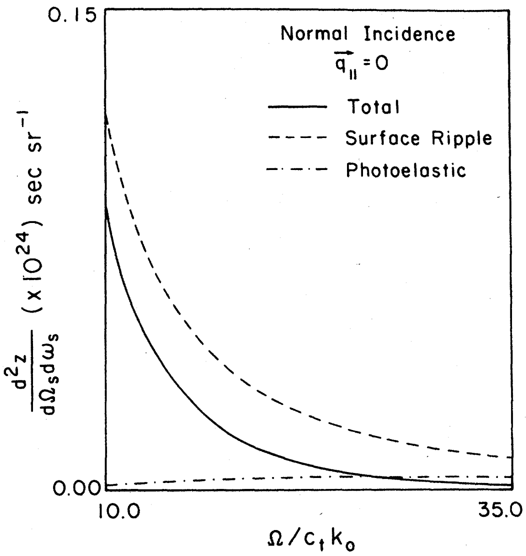

FIG. 5. Brillouin spectrum for normally incident and normally scattered light. The dashed line is the surface-ripple contribution and the dot-dashed line is the photoelastic contribution. The solid line is the total spectrum including interference effects.

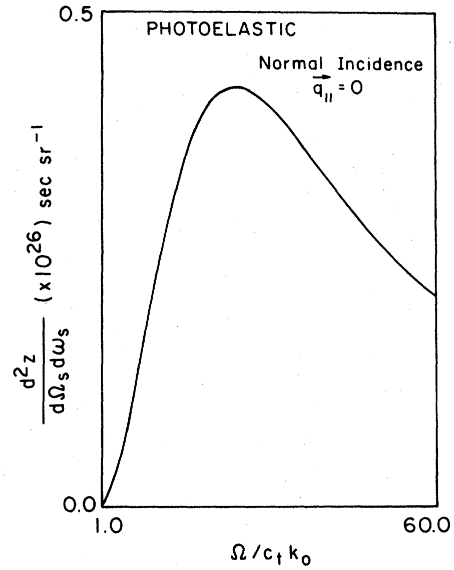

FIG. 6. Photoelastic contribution to the Brillouin spectrum for normally incident and normally scattered light.

comparable to its thickness, one has to account for the reflection of light at the back surface and the semi-infinite treatment is inappropriate.

In Fig. 5, we show the spectrum for normally incident and normally scattered light ($\vec{q}_{\parallel}=0$). The frequency shift is measured in dimensionless units $\Omega/c_t k_0$. The surface-ripple contribution goes as $1/\Omega^2$ and is shown as the dashed line. The photoelastic contribution is shown as the dot-dashed line and the total spectrum, including the interference effects, is shown as the solid line. The photoelastic contribution and the interference effects are not quite as small as in the previous case considered. In Fig. 6, we show the photoelastic contribution on a larger scale. The normal incidence photoelastic contribution has been considered by Dervisch and Loudon[6] and our result is in accord with theirs. The distorted Lorentzian has a peak at $\Omega \approx 2c_l k_0 |\sqrt{\epsilon_0}|$ corresponding to the frequency of the longitudinal acoustic wave. The opacity broadening is evident in the spectrum.

In conclusion, we have presented a comprehensive theory of Brillouin scattering from an opaque crystal with elastic isotropy. The theory takes account of the photoelastic mechanism, the surface-ripple mechanism as well as the interference between these two scattering processes. We find that the interference effects are larger than the photoelastic contribution itself, which is quite small compared to the surface-ripple contribution.

ACKNOWLEDGMENT

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APPENDIX A

We first recall the well-known result[24] that the correlation function

$$\hat{\mathfrak{D}}_{\alpha\beta}(\vec{x},\vec{x}';\omega)=\int_{-\infty}^{\infty} dt\, e^{i\omega t}\langle u_\alpha(\vec{x},t)u_\beta(\vec{x}',0)\rangle \tag{A1}$$

and the Fourier transform of the retarded Green's function

$$\hat{D}^{(R)}_{\alpha\beta}(\vec{x},\vec{x}';\omega)=\int_{-\infty}^{\infty} dt\, e^{i\omega t} \times\{-i\Theta(t)\langle[u_\alpha(\vec{x},t),u_\beta(\vec{x}',0)]\rangle\} \tag{A2}$$

are related through

$$\hat{\mathfrak{D}}_{\alpha\beta}(\vec{x},\vec{x}';\omega)=i(1-e^{-\beta\hbar\omega})^{-1}[\hat{D}^{(R)}_{\alpha\beta}(\vec{x},\vec{x}';\omega+i0) - \hat{D}^{(R)}_{\alpha\beta}(\vec{x},\vec{x}';\omega-i0)]. \tag{A3}$$

Now consider the second derivative of the retarded Green's function with respect to time,

$$\frac{\partial^2}{\partial t^2} D^{(R)}_{\alpha\beta}(\vec{x}, \vec{x}'; t) = \frac{\partial^2}{\partial t^2}\{-i\Theta(t)\langle[u_\alpha(\vec{x}, t), u_\beta(\vec{x}', 0)]\rangle\}$$
$$= -i\delta(t)\langle[\dot{u}_\alpha(\vec{x}, 0), u_\beta(\vec{x}', 0)]\rangle$$
$$-i\Theta(t)\langle[\ddot{u}_\alpha(\vec{x}, t), u_\beta(\vec{x}', 0)]\rangle . \quad (A4)$$

Here $\dot{u}_\alpha$ denotes $\partial u_\alpha/\partial t$ and $\ddot{u}_\alpha$ denotes $\partial^2 u_\alpha/\partial t^2$. The equation of motion satisfied by $\vec{u}(\vec{x}, t)$ is

$$\ddot{u}_\alpha(\vec{x}, t) = \frac{1}{\rho} \sum_{\beta\mu\nu} \frac{\partial}{\partial x_\beta} c_{\alpha\beta\mu\nu}(\vec{x}) \frac{\partial u_\mu(\vec{x}, t)}{\partial x_\nu} , \quad (A5a)$$

where $c_{\alpha\beta\mu\nu}(x)$ are the position dependent elastic coefficients,

$$c_{\alpha\beta\mu\nu}(\vec{x}) = \Theta(-x_3) c_{\alpha\beta\mu\nu} . \quad (A5b)$$

The stress-free boundary conditions are built into the equations of motion through use of position-dependent elastic coefficients.[10] Furthermore,

$$[\rho\dot{u}_\alpha(\vec{x}, 0), u_\beta(\vec{x}, 0) = i\hbar\delta_{\alpha\beta}\delta(\vec{x} - \vec{x}') . \quad (A6)$$

From Eqs. (A4)–(A6) we get the equation obeyed by the retarded Green's function

$$\sum_\mu \left(-\delta_{\alpha\mu}\frac{\partial^2}{\partial t^2} + \frac{1}{\rho}\sum_{\lambda\nu} \frac{\partial}{\partial x_\lambda} c_{\alpha\lambda\mu\nu} \frac{\partial}{\partial x_\nu}\right) D^{(R)}_{\mu\beta}(\vec{x}, \vec{x}'; t)$$
$$= \frac{\hbar}{\rho}\delta(t)\delta_{\alpha\beta}\delta(\vec{x} - \vec{x}') . \quad (A7)$$

Except for the factor $\hbar/\rho$ on the right-hand side, Eq. (A7) is identical to the equation which determines the classical dynamical Green's function $D_{\alpha\beta}(\vec{x}, \vec{x}'; t)$ of an elastic medium.[10] Since the boundary conditions are incorporated into the equations of motion through the use of position-dependent elastic coefficients, it follows from the above observation that

$$D_{\alpha\beta}(\vec{x}, \vec{x}'; t) \equiv (\rho/\hbar) D^{(R)}_{\alpha\beta}(\vec{x}, \vec{x}'; t) . \quad (A8)$$

It now remains to show that

$$\hat{D}_{\alpha\beta}(\vec{x}, \vec{x}'; \omega + i0) = [D_{\beta\alpha}(\vec{x}', \vec{x} | \omega - i0)]^* . \quad (A9)$$

This is most easily established by noting that $\hat{D}_{\alpha\beta}(\vec{x}, \vec{x}'; \omega + i\eta)$ may be expanded in terms of the normal mode eigenfunctions and eigenfrequencies as

$$\hat{D}_{\alpha\beta}(\vec{x}, \vec{x}'; \omega + i\eta) = \sum_s \frac{V^{(s)}_\alpha(\vec{x})[V^{(s)}_\beta(\vec{x}')]^*}{\omega^2 + 2i\omega\eta - \omega_s^2} . \quad (A10)$$

Expressing both the left-hand side and the right-hand side of Eq. (A9) as in Eq. (A10), the identity follows.

APPENDIX B

In this Appendix we list the coefficients α, β, χ, and λ appearing in Eqs. (3.2a) and (3.2b).

(i) Incident light polarized perpendicular to the plane of incidence. All symbols have the same meaning as in the text.

$$\alpha^{(11)}_{11}(\perp) = \tfrac{1}{2}|\Gamma^\perp_2|^2[q_1^2 m^{(11)}_{22}|k_{12}|^2 + 2q_1q_2\,\mathrm{Re}(m^{(11)}_{12}k^*_{44}k_{12}) + q_2^2 m^{(11)}_{11}|k_{44}|^2] , \quad (B1a)$$

$$\alpha^{(11)}_{22}(\perp) = \tfrac{1}{2}|\Gamma^\perp_2|^2[q_1^2 m^{(11)}_{11}|k_{44}|^2 + 2q_1q_2\,\mathrm{Re}(m^{(11)}_{12}k^*_{44}k_{11}) + q_2^2 m^{(11)}_{22}|k_{11}|^2] , \quad (B1b)$$

$$\alpha^{(11)}_{33}(\perp) = \tfrac{1}{2}|\Gamma^\perp_2|^2(q_2^2 m^{(11)}_{33}|k_{44}|^2) , \quad (B1c)$$

$$\alpha^{(11)}_{12}(\perp) = |\Gamma^\perp_2|^2(q_1^2 m^{(11)*}_{12}k^*_{12}k_{44} + q_1q_2 m^{(11)}_{22}k^*_{12}k_{11} + q_1q_2 m^{(11)}_{11}|k_{44}|^2 + q_2^2 m^{(11)}_{12}k^*_{44}k_{11}) , \quad (B1d)$$

$$\alpha^{(11)}_{13}(\perp) = |\Gamma^\perp_2|^2(q_1q_2 m^{(11)}_{23}k^*_{12}k_{44} + q_2^2 m^{(11)}_{13}|k_{44}|^2) , \quad (B1e)$$

$$\alpha^{(11)}_{23}(\perp) = |\Gamma^\perp_2|^2(q_1q_2 m^{(11)}_{13}|k_{44}|^2 + q_2^2 m^{(11)}_{23}k^*_{11}k_{44}) . \quad (B1f)$$

$$\beta^{(11)}_{22}(\perp) = \tfrac{1}{2}|\Gamma^\perp_2|^2 m^{(11)}_{33}|k_{44}|^2 , \quad (B2a)$$

$$\beta^{(11)}_{33}(\perp) = \tfrac{1}{2}|\Gamma^\perp_2|^2 m^{(11)}_{22}|k_{12}|^2 , \quad (B2b)$$

$$\beta^{(11)}_{23}(\perp) = |\Gamma^\perp_2|^2 m^{(11)*}_{23}k^*_{44}k_{12} . \quad (B2c)$$

$\beta^{(11)}_{11}(\perp), \beta^{(11)}_{12}(\perp), \beta^{(11)}_{13}(\perp)$ are identically zero.

$$\chi^{(11)}_{22}(\perp) = |\Gamma^\perp_2|^2(q_1 m^{(11)*}_{13}|k_{44}|^2 + q_2 m^{(11)*}_{23}k^*_{44}k_{11}) , \quad (B3a)$$

$$\chi^{(11)}_{23}(\perp) = |\Gamma^\perp_2|^2 q_2 m^{(11)}_{33}|k_{44}|^2 , \quad (B3b)$$

$$\chi^{(11)}_{33}(\perp) = |\Gamma^\perp_2|^2 q_2 m^{(11)}_{23}k^*_{12}k_{44} , \quad (B3c)$$

$$\lambda^{(11)}_{12}(\perp) = |\Gamma^\perp_2|^2(q_1 m^{(11)}_{23}k^*_{12}k_{44} + q_2 m^{(11)}_{13}|k_{44}|^2) , \quad (B3d)$$

$$\lambda^{(11)}_{13}(\perp) = |\Gamma^\perp_2|^2(q_1 m^{(11)}_{22}|k_{12}|^2 + q_2 m^{(11)}_{12}k^*_{44}k_{12}) , \quad (B3e)$$

$$\lambda^{(11)}_{23}(\perp) = |\Gamma^\perp_2|^2(q_1 m^{(11)}_{12}k^*_{44}k_{12} + q_2 m^{(11)}_{22}k^*_{11}k_{12}) . \quad (B3f)$$

The remaining $\chi^{(11)}(\perp)$ and $\lambda^{(11)}(\perp)$ are zero.

$$\alpha^{(12)}_{13}(\perp) = |\Gamma^\perp_2|^2(q_1 m^{(12)}_{22}k^*_{12} + q_2 m^{(12)}_{12}k^*_{44}) , \quad (B4a)$$

$$\alpha^{(12)}_{23}(\perp) = |\Gamma^\perp_2|^2(q_1 m^{(12)}_{12}k^*_{44} + q_2 m^{(12)}_{22}k^*_{11}) , \quad (B4b)$$

$$\alpha^{(12)}_{33}(\perp) = |\Gamma^\perp_2|^2 q_2 m^{(12)}_{32}k^*_{44} , \quad (B4c)$$

$$\beta^{(12)}_{13}(\perp) = 0 , \quad (B4d)$$

$$\beta^{(12)}_{23}(\perp) = m^{(12)}_{32}k^*_{44}|\Gamma^\perp_2|^2 , \quad (B4e)$$

$$\beta^{(12)}_{33}(\perp) = m^{(12)}_{22}k^*_{12}|\Gamma^\perp_2|^2 . \quad (B4f)$$

(ii) Incident light polarized parallel to the plane of incidence.

$$\alpha_{11}^{(11)}(\|) = \tfrac{1}{2}\{|\Gamma_1^{\|}|^2[q_1^2 m_{11}^{(11)}|k_{11}|^2 + 2q_1q_2 \operatorname{Re}(m_{12}^{(11)}k_{11}^* k_{44}) + q_2^2 m_{22}^{(11)}|k_{44}|^2] + |\Gamma_3^{\|}|^2 q_1^2 m_{33}^{(11)}|k_{12}|^2 + 2\operatorname{Re}[\Gamma_1^{\|}\Gamma_3^{\|}(q_1^2 m_{13}^{(11)}k_{11}^* k_{12} + q_1q_2 m_{23}^{(11)}k_{44}^* k_{12})]\}, \tag{B5a}$$

$$\alpha_{12}^{(11)}(\|) = |\Gamma_1^{\|}|^2(q_1^2 m_{12}^{(11)}k_{11}^* k_{44} + q_1q_2 m_{11}^{(11)}k_{11}^* k_{12} + q_1q_2 m_{22}^{(11)}|k_{44}|^2 + q_2^2 m_{12}^{(11)*}k_{44}^* k_{12}) + |\Gamma_3^{\|}|^2 q_1q_2 m_{33}^{(11)}|k_{12}|^2 + \Gamma_1^{\|*}\Gamma_3^{\|}(q_1q_2 m_{13}^{(11)}k_{11}^* k_{12} + q_2^2 m_{23}^{(11)}k_{44}^* k_{12}) + \Gamma_1^{\|}\Gamma_3^{\|*}(q_1^2 m_{23}^{(11)*}k_{12}^* k_{44} + q_1q_2 m_{13}^{(11)*}|k_{12}|^2), \tag{B5b}$$

$$\alpha_{13}^{(11)}(\|) = |\Gamma_1^{\|}|^2(q_1^2 m_{13}^{(11)}k_{11}^* k_{44} + q_1q_2 m_{23}^{(11)}|k_{44}|^2) + |\Gamma_3^{\|}|^2(q_1^2 m_{13}^{(11)*}k_{12}^* k_{44} + q_1q_2 m_{23}^{(11)*}k_{12}^* k_{44}) + \Gamma_1^{\|*}\Gamma_3^{\|}(q_1^2 m_{11}^{(11)}k_{11}^* k_{44} + q_1q_2 m_{12}^{(11)}k_{11}^* k_{44} + q_1q_2 m_{12}^{(11)*}|k_{44}|^2 + q_2^2 m_{22}^{(11)}|k_{44}|^2) + \Gamma_1^{\|}\Gamma_3^{\|*}q_1^2 m_{33}^{(11)}k_{12}^* k_{44}, \tag{B5c}$$

$$\alpha_{22}^{(11)}(\|) = \tfrac{1}{2}\{|\Gamma_1^{\|}|^2[q_1^2 m_{22}^{(11)}|k_{44}|^2 + 2q_1q_2 \operatorname{Re}(m_{12}^{(11)*}k_{44}^* k_{12}) + q_2^2 m_{11}^{(11)}|k_{12}|^2] + |\Gamma_3^{\|}|^2 m_{33}^{(11)}q_2^2|k_{12}|^2 + 2\operatorname{Re}[\Gamma_1^{\|*}\Gamma_3^{\|}(q_1q_2 m_{23}^{(11)}k_{44}^* k_{12} + q_2^2 m_{13}^{(11)}|k_{12}|^2)]\}, \tag{B5d}$$

$$\alpha_{23}^{(11)}(\|) = |\Gamma_1^{\|}|^2(q_1^2 m_{23}^{(11)}|k_{44}|^2 + q_1q_2 m_{13}^{(11)}k_{12}^* k_{44}) + |\Gamma_3^{\|}|^2(q_1q_2 m_{13}^{(11)*}k_{12}^* k_{44} + q_2^2 m_{23}^{(11)*}k_{12}^* k_{44}) + \Gamma_1^{\|*}\Gamma_3^{\|}(q_1^2 m_{12}^{(11)*}|k_{44}|^2 + q_1q_2 m_{22}^{(11)}|k_{44}|^2 + q_1q_2 m_{11}^{(11)}k_{12}^* k_{44} + q_2^2 m_{12}^{(11)}k_{12}^* k_{44}) + \Gamma_1^{\|}\Gamma_3^{\|*}q_1q_2 m_{33}^{(11)}k_{12}^* k_{44}, \tag{B5e}$$

$$\alpha_{33}^{(11)}(\|) = \tfrac{1}{2}\{|\Gamma_1^{\|}|^2 q_1^2 m_{33}^{(11)}|k_{44}|^2 + |\Gamma_3^{\|}|^2[q_1^2 m_{11}^{(11)}|k_{44}|^2 + 2q_1q_2 \operatorname{Re}(m_{12}^{(11)})|k_{44}|^2 + q_2^2 m_{22}^{(11)}|k_{44}|^2] + 2\operatorname{Re}[\Gamma_1^{\|*}\Gamma_3^{\|}(q_1^2 m_{13}^{(11)}|k_{44}|^2 + q_1q_2 m_{23}^{(11)*}|k_{44}|^2)]\}. \tag{B5f}$$

$$\beta_{11}^{(11)}(\|) = \tfrac{1}{2}[|\Gamma_1^{\|}|^2 m_{33}^{(11)}|k_{44}|^2 + |\Gamma_3^{\|}|^2 m_{11}^{(11)}|k_{44}|^2 + 2\operatorname{Re}(\Gamma_1^{\|*}\Gamma_3^{\|}m_{13}^{(11)*})|k_{44}|^2], \tag{B6a}$$

$$\beta_{12}^{(11)}(\|) = |\Gamma_3^{\|}|^2 m_{12}^{(11)}|k_{44}|^2 + \Gamma_1^{\|*}\Gamma_3^{\|}m_{23}^{(11)*}|k_{44}|^2, \tag{B6b}$$

$$\beta_{13}^{(11)}(\|) = |\Gamma_1^{\|}|^2 m_{13}^{(11)*}k_{44}^* k_{12} + |\Gamma_3^{\|}|^2 m_{13}^{(11)}k_{44}^* k_{11} + \Gamma_1^{\|*}\Gamma_3^{\|}m_{33}^{(11)}k_{44}^* k_{11} + \Gamma_1^{\|}\Gamma_3^{\|*}m_{11}^{(11)}k_{44}^* k_{12}, \tag{B6c}$$

$$\beta_{22}^{(11)}(\|) = \tfrac{1}{2}|\Gamma_3^{\|}|^2 m_{22}^{(11)}|k_{44}|^2 \tag{B6d}$$

$$\beta_{23}^{(11)}(\|) = |\Gamma_3^{\|}|^2 m_{23}^{(11)}k_{44}^* k_{11} + \Gamma_1^{\|}\Gamma_3^{\|*}m_{12}^{(11)*}k_{44}^* k_{12}, \tag{B6e}$$

$$\beta_{33}^{(11)}(\|) = \tfrac{1}{2}[|\Gamma_1^{\|}|^2 m_{11}^{(11)}|k_{12}|^2 + |\Gamma_3^{\|}|^2 m_{33}^{(11)}|k_{\|}|^2 + 2\operatorname{Re}(\Gamma_1^{\|*}\Gamma_3^{\|}m_{13}^{(11)}k_{12}^* k_{11})]. \tag{B6f}$$

$$\chi_{11}^{(11)}(\|) = |\Gamma_1^{\|}|^2(q_1 m_{13}^{(11)*}k_{44}^* k_{11} + q_2 m_{23}^{(11)*}|k_{44}|^2) + \Gamma_1^{\|*}\Gamma_3^{\|}q_1 m_{33}^{(11)}k_{44}^* k_{12} + \Gamma_1^{\|}\Gamma_3^{\|*}(q_1 m_{11}^{(11)}k_{44}^* k_{11} + q_2 m_{12}^{(11)}|k_{44}|^2) + |\Gamma_3^{\|}|^2 q_1 m_{13}^{(11)}k_{44}^* k_{12}, \tag{B7a}$$

$$\chi_{12}^{(11)}(\|) = |\Gamma_1^{\|}|^2(q_1 m_{23}^{(11)*}|k_{44}|^2 + q_2 m_{13}^{(11)*}k_{44}^* k_{12}) + \Gamma_1^{\|*}\Gamma_3^{\|}q_2 m_{33}^{(11)}k_{44}^* k_{12} + \Gamma_1^{\|}\Gamma_3^{\|*}(q_1 m_{12}^{(11)}|k_{44}|^2 + q_2 m_{11}^{(11)}k_{44}^* k_{12}) + |\Gamma_3^{\|}|^2 q_2 m_{13}^{(11)}k_{44}^* k_{12}, \tag{B7b}$$

$$\chi_{13}^{(11)}(\|) = |\Gamma_1^{\|}|^2 q_1 m_{33}^{(11)}|k_{44}|^2 + \Gamma_1^{\|*}\Gamma_3^{\|}(q_1 m_{13}^{(11)*}|k_{44}|^2 + q_2 m_{23}^{(11)*}|k_{44}|^2) + \Gamma_1^{\|}\Gamma_3^{\|*}q_1 m_{13}^{(11)}|k_{44}|^2 + |\Gamma_3^{\|}|^2(q_1 m_{11}^{(11)}|k_{44}|^2 + q_2 m_{12}^{(11)}|k_{44}|^2), \tag{B7c}$$

$$\chi_{22}^{(11)}(\|) = \Gamma_1^{\|}\Gamma_3^{\|*}(q_1 m_{22}^{(11)}|k_{44}|^2 + q_2 m_{12}^{(11)*}k_{44}^* k_{12}) + |\Gamma_3^{\|}|^2 q_2 m_{23}^{(11)}k_{44}^* k_{12}, \tag{B7d}$$

$$\chi_{23}^{(11)}(\|) = \Gamma_1^{\|}\Gamma_3^{\|*}q_1 m_{23}^{(11)}|k_{44}|^2 + |\Gamma_3^{\|}|^2(q_1 m_{12}^{(11)*}|k_{44}|^2 + q_2 m_{22}^{(11)}|k_{44}|^2), \tag{B7e}$$

$$\chi_{33}^{(11)}(\|) = |\Gamma_1^{\|}|^2 q_1 m_{13}^{(11)}k_{12}^* k_{44} + \Gamma_1^{\|*}\Gamma_3^{\|}(q_1 m_{11}^{(11)}k_{12}^* k_{44} + q_2 m_{12}^{(11)}k_{12}^* k_{44}) + \Gamma_1^{\|}\Gamma_3^{\|*}q_1 m_{33}^{(11)}k_{11}^* k_{44} + |\Gamma_3^{\|}|^2(q_1 m_{13}^{(11)*}k_{11}^* k_{44} + q_2 m_{23}^{(11)*}k_{11}^* k_{44}), \tag{B7f}$$

$$\lambda_{13}^{(11)}(\|) = |\Gamma_1^{\|}|^2(q_1 m_{11}^{(11)}k_{11}^* k_{12} + q_2 m_{12}^{(11)*}k_{44}^* k_{12}) + \Gamma_1^{\|*}\Gamma_3^{\|}(q_1 m_{13}^{(11)}|k_{11}|^2 + q_2 m_{23}^{(11)}k_{44}^* k_{11}) + \Gamma_1^{\|}\Gamma_3^{\|*}q_1 m_{13}^{(11)*}|k_{12}|^2 + |\Gamma_3^{\|}|^2 q_1 m_{33}^{(11)}k_{12}^* k_{11}, \tag{B7g}$$

$$\lambda_{12}^{(11)}(\|) = \Gamma_1^{\|*}\Gamma_3^{\|}(q_1 m_{12}^{(11)}k_{11}^* k_{44} + q_2 m_{22}^{(11)}|k_{44}|^2) + |\Gamma_3^{\|}|^2 q_1 m_{23}^{(11)*}k_{12}^* k_{44}, \tag{B7h}$$

$$\lambda_{23}^{(11)}(\|) = |\Gamma_1^{\|}|^2(q_1 m_{12}^{(11)*}k_{44}^* k_{12} + q_2 m_{11}^{(11)}|k_{12}|^2) + \Gamma_1^{\|*}\Gamma_3^{\|}(q_1 m_{23}^{(11)}k_{44}^* k_{11} + q_2 m_{13}^{(11)}k_{12}^* k_{11}) + \Gamma_1^{\|}\Gamma_3^{\|*}q_2 m_{13}^{(11)*}|k_{12}|^2 + |\Gamma_3^{\|}|^2 q_2 m_{33}^{(11)}k_{12}^* k_{11}. \tag{B7i}$$

$$\alpha_{13}^{(12)}(\|) = |\Gamma_1^{\|}|^2(q_1 m_{11}^{(12)}k_{11}^* + q_2 m_{21}^{(12)}k_{44}^*) + \Gamma_1^{\|*}\Gamma_3^{\|}(q_1 m_{13}^{(12)}k_{11}^* + q_2 m_{23}^{(12)}k_{44}^*) + \Gamma_1^{\|}\Gamma_3^{\|*}q_1 m_{31}^{(12)}k_{12}^* + |\Gamma_3^{\|}|^2 q_1 m_{33}^{(12)}k_{12}^*, \tag{B8a}$$

$$\alpha_{23}^{(12)}(\|) = |\Gamma_1^{\|}|^2(q_1 m_{21}^{(12)}k_{44}^* + q_2 m_{11}^{(12)}k_{12}^*) + \Gamma_1^{\|*}\Gamma_3^{\|}(q_1 m_{23}^{(12)}k_{44}^* + q_2 m_{13}^{(12)}k_{12}^*) + \Gamma_1^{\|}\Gamma_3^{\|*}q_2 m_{31}^{(12)}k_{12}^* + |\Gamma_3^{\|}|^2 m_{33}^{(12)}k_{12}^* q_2, \tag{B8b}$$

$$\alpha_{33}^{(12)}(\|) = |\Gamma_1^{\|}|^2 m_{31}^{(12)} q_1 k_{44}^* + \Gamma_1^{\|*}\Gamma_3^{\|} q_1 m_{33}^{(12)} k_{44}^* + \Gamma_1^{\|}\Gamma_3^{\|*}(q_1 m_{11}^{(12)} k_{44}^* + q_2 m_{21}^{(12)} k_{44}^*) + |\Gamma_3^{\|}|^2 (q_1 m_{13}^{(12)} k_{44}^* + q_2 m_{23}^{(12)} k_{44}^*). \tag{B8c}$$

$$\beta_{13}^{(12)}(\|) = |\Gamma_3^{\|}|^2 m_{31}^{(12)} k_{44}^* + (\Gamma_1^{\|*}\Gamma_3^{\|} m_{33}^{(12)} + \Gamma_1^{\|}\Gamma_3^{\|*} m_{11}^{(12)}) k_{44}^* + |\Gamma_3^{\|}|^2 m_{13}^{(12)} k_{44}^*, \tag{B9a}$$

$$\beta_{23}^{(12)}(\|) = \Gamma_1^{\|}\Gamma_3^{\|*} m_{21}^{(12)} k_{44}^* + |\Gamma_3^{\|}|^2 m_{23}^{(12)} k_{44}^*, \tag{B9b}$$

$$\beta_{33}^{(12)}(\|) = |\Gamma_1^{\|}|^2 m_{11}^{(12)} k_{12}^* + \Gamma_1^{\|*}\Gamma_3^{\|} m_{13}^{(12)} k_{12}^* + \Gamma_1^{\|}\Gamma_3^{\|*} m_{31}^{(12)} k_{11}^* + |\Gamma_3^{\|}|^2 m_{33}^{(12)} k_{11}^*. \tag{B9c}$$

APPENDIX C

We quote below the dynamical Green's functions for an isotropic semi-infinite elastic continuum bounded by a stress-free planar surface derived by Maradudin and Mills.[10] The results have been adopted to suit the geometry of the present work. We introduce the quantities

$$\alpha_{l,t} = \left(q_\|^2 - \frac{(\Omega + i\eta)^2}{c_{l,t}^2}\right)^{1/2}, \quad \mathrm{Re}(\alpha_{l,t}) > 0, \tag{C1}$$

$$r_\pm = \frac{-4\alpha_l\alpha_t q_\|^2 \pm (\alpha_t^2 + q_\|^2)^2}{4\alpha_l\alpha_t(\alpha_t^2 + q_\|^2)}. \tag{C2}$$

Here c_l and c_t are, respectively, the longitudinal and transverse acoustical velocities in the isotropic medium. All other quantities have the same meaning as in the text. We shall suppress the arguments of the Green's functions $D_{ij}(\vec{q}_\|\Omega\,|x_3, x_3')$ for brevity. We write

$$D_{ij} = D_{ij}^{(B)} + D_{ij}^{(S)} \tag{C3}$$

where the notations B and S refer, respectively, to the bulk and the surface terms. Also, let

$$X_\mu = e^{-\alpha_\mu |x_3 - x_3'|}, \tag{C4a}$$

$$Y_{\mu\nu} = e^{\alpha_\mu x_3} e^{\alpha_\nu x_3'}, \tag{C4b}$$

with μ and ν being l or t. We write the wave-vector transfer in the plane, $\vec{q}_\|$, as $q_1\hat{x}_1 + q_2\hat{x}_2$. Then, ,

$$D_{11}^{(B)}(x_3, x_3') = \frac{-1}{2\Omega^2}\left[\frac{q_1^2}{\alpha_l} X_l + \left(\frac{-q_1^2\alpha_t}{q_\|^2} + \frac{q_2^2\Omega^2}{\alpha_t c_t^2 q_\|^2}\right) X_t\right], \tag{C5a}$$

$$D_{12}^{(B)}(x_3, x_3') = D_{21}^{(B)}(x_3, x_3') = \frac{-1}{2\Omega^2}\left[\frac{q_1 q_2}{\alpha_l} X_l - \left(\frac{q_1 q_2\alpha_t}{q_\|^2} + \frac{q_1 q_2\Omega^2}{\alpha_t c_t^2 q_\|^2}\right) X_t\right], \tag{C5b}$$

$$D_{13}^{(B)}(x_3, x_3') = D_{31}^{(B)}(x_3, x_3') = \frac{-1}{2\Omega^2}(-iq_1 X_l + iq_1 X_t)\,\mathrm{sgn}(x_3' - x_3), \tag{C5c}$$

$$D_{22}^{(B)}(x_3, x_3') = \frac{-1}{2\Omega^2}\left[\frac{q_2^2}{\alpha_l} X_l + \left(\frac{-q_2^2\alpha_t}{q_\|^2} + \frac{q_1^2\Omega^2}{\alpha_t c_t^2 q_\|^2}\right) X_t\right], \tag{C5d}$$

$$D_{23}^{(B)}(x_3, x_3') = D_{32}^{(B)}(x_3, x_3') = \frac{-1}{2\Omega^2}(-iq_2 X_l + iq_2 X_t)\,\mathrm{sgn}(x_3' - x_3), \tag{C5e}$$

$$D_{33}^{(B)}(x_3, x_3') = \frac{-1}{2\Omega^2}\left(-\alpha_l X_l + \frac{q_\|^2}{\alpha_t} X_t\right). \tag{C5f}$$

For the surface terms we have

$$D_{11}^{(S)}(x_3, x_3') = \frac{-1}{2\Omega^2 r_+}\left[\frac{q_1^2 r_-}{\alpha_l} Y_{ll} + \frac{q_1^2}{\alpha_l}(Y_{lt} + Y_{tl}) + \left(\frac{q_1^2\alpha_t r_-}{q_\|^2} + \frac{q_2^2\Omega^2 r_+}{\alpha_t c_t^2 q_\|^2}\right) Y_{tt}\right]. \tag{C6a}$$

$$D_{12}^{(S)}(x_3, x_3') = D_{21}^{(S)}(x_3, x_3') = \frac{-1}{2\Omega^2 r_+}\left[\frac{q_1 q_2 r_-}{\alpha_l} Y_{ll} + \frac{q_1 q_2}{\alpha_l}(Y_{lt} + Y_{tl}) + \left(\frac{q_1 q_2\alpha_t r_-}{q_\|^2} - \frac{q_1 q_2\Omega^2 r_+}{\alpha_t c_t^2 q_\|^2}\right) Y_{tt}\right], \tag{C6b}$$

$$D_{13}^{(S)}(x_3, x_3') = -D_{31}^{(S)}(x_3', x_3) = \frac{-1}{2\Omega^2 r_+}\left[iq_1 r_- Y_{ll} + \frac{iq_1 q_\|^2}{\alpha_l\alpha_t} Y_{lt} + iq_1 Y_{tl} + iq_1 r_- Y_{tt}\right], \tag{C6c}$$

$$D_{22}^{(S)}(x_3, x_3') = \frac{-1}{2\Omega^2 r_+}\left[\frac{q_2^2 r_-}{\alpha_l} Y_{ll} + \frac{q_2^2}{\alpha_l}(Y_{lt} + Y_{tl}) + \left(\frac{q_2^2\alpha_t r_-}{q_\|^2} + \frac{q_1^2\Omega^2 r_+}{\alpha_t c_t^2 q_\|^2}\right) Y_{tt}\right], \tag{C6d}$$

$$D_{23}^{(S)}(x_3, x_3') = -D_{32}^{(S)}(x_3', x_3) = \frac{-1}{2\Omega^2 r_+}\left[iq_2 r_- Y_{ll} + \frac{iq_2 q_\|^2}{\alpha_l\alpha_t} Y_{lt} + iq_2 Y_{tl} + iq_2 r_- Y_{tt}\right], \tag{C6e}$$

$$D_{33}^{(S)}(x_3, x_3') = \frac{-1}{2\Omega^2 r_+}\left[\alpha_l r_- Y_{ll} + \frac{q_\|^2}{\alpha_t}(Y_{lt} + Y_{tl}) + \frac{q_\|^2 r_-}{\alpha_t} Y_{tt}\right]. \tag{C6f}$$

We remark that in the limit $q_\| \to 0$, we have $D_{12} = 0 = D_{21}$, $D_{13} = 0 = D_{31}$, $D_{23} = 0 = D_{32}$ and

$$D_{11}(x_3, x_3') \to D_{22}(x_3, x_3') \to \frac{-1}{2\alpha_t c_t^2}(X_t + Y_{tt}) \tag{C7a}$$

and

$$D_{33}(x_3, x_3') \to \frac{-1}{2\Omega^2}(-\alpha_l X_l - \alpha_l Y_{ll}). \tag{C7b}$$

[1]D. W. Feldman, James H. Parker, Jr., and M. Ashkin, Phys. Rev. Lett. 21, 607 (1968); James H. Parker, Jr., D. W. Feldman, and M. Ashkin, in *Light Scattering Spectra of Solids*, edited by George B. Wright (Springer-Verlag, New York, 1969), p. 389.

[2]D. L. Mills, A. A. Maradudin, and E. Burstein, Phys. Rev. Lett. 21, 1178 (1968); in *Light Scattering Spectra of Solids*, edited by George B. Wright (Springer-Verlag, New York, 1969), p. 399; Ann. Phys. (N. Y.) 56, 504 (1970).

[3]B. I. Bennett, A. A. Maradudin, and L. R. Swanson, in *Proceedings of the Second International Conference on Light Scattering in Solids*, edited by M. Balkanski (Flammarion, Paris, 1971), p. 443; Ann. Phys. (N. Y.) 71, 357 (1972).

[4]J. R. Sandercock, Phys. Rev. Lett. 28, 237 (1972); 29, 1735 (1972).

[5]G. Dresselhaus and A. S. Pine, Solid State Commun. 16, 1001 (1975).

[6]A. Dervisch and R. Loudon, J. Phys. C 9, L669 (1976).

[7]J. Dil and E. Brody, Phys. Rev. B 14, 5218 (1976).

[8]R. Loudon, J. Phys. C 11, 403 (1978).

[9]K. R. Subbaswamy and A. A. Maradudin, Indian J. Pure Appl. Phys. 16, 282 (1978). There is a sign error in Eq. (2.16) of this paper which led to the neglect of certain terms in the cross section. This has been corrected in the present work.

[10]A. A. Maradudin and D. L. Mills, Ann. Phys. (N. Y.) 100, 262 (1976).

[11]S. Mishra and R. Bray, Phys. Rev. Lett. 39, 222 (1977).

[12]L. Dobrzynski and A. A. Maradudin, Phys. Rev. B 14, 2200 (1976).

[13]A. A. Maradudin and D. L. Mills, Phys. Rev. B 11, 1392 (1975).

[14]A. Dervisch and R. Loudon (unpublished).

[15]R. Loudon, Phys. Rev. Lett. 40, 581 (1978).

[16]G. S. Agarwal, Phys. Rev. B 14, 846 (1976).

[17]K. Kröger and E. Kretschmann, Phys. Status. Solidi B 76, 515 (1976).

[18]A. Marvin, F. Tiogo, and V. Celli, Phys. Rev. B 11, 2777 (1975).

[19]D. L. Mills, Phys. Rev. B 12, 4036 (1975).

[20]J. M. Elson, Phys. Rev. B 12, 2541 (1975).

[21]R. Loudon (unpublished).

[22]*American Institute of Physics Handbook* (McGraw-Hill, New York, 1972), Sec. 3.

[23]A. A. Maradudin, E. W. Montroll, G. H. Weiss, and I. P. Ipatova, *Theory of Lattice Dynamics in the Harmonic Approximation*, 2nd ed. (Academic, New York, 1971), Chap. IX.

[24]A. A. Abrikosov, L. D. Gorkov, and I. E. Dzyaloshinski, *Methods of Quantum Field Theory in Statistical Physics* (Prentice-Hall, Englewood Cliffs, N. J., 1963), Sec. 28.