Correction of the Brillouin-scattering cross section in CdS for the Gaussian curvature of the $\omega(\vec{k})$ surface

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A Green's-function theory of Brillouin scattering in anisotropic, nonconducting crystals has been published by Nelson, Lazay, and Lax. Using the stationary-phase method they have obtained the dependence of the scattered far field on the principal curvatures of the $\omega(\vec{k})$ surface. The dipole approximation used by the present author to calculate scattering cross sections in nonconducting, hexagonal crystals involves properties that depend only on the first derivatives of $\omega(\vec{k})$. In this work we compare the two methods and give an explicit expression for the correction factor valid for an arbitrary optical anisotropy. The correction for scattering from T_1 phonons in a plane containing the optic axis of CdS is discussed.

In a recent comment[1] on my original work on Brillouin-scattering cross sections in CdS,[2] Lax and Nelson have pointed out, correctly, that the cross section in anisotropic crystals in general depends on the Gaussian curvature of the $\omega(\vec{k})$ surface. Thus the present addenda to my original work[2] has been made as a consequence of the Lax-Nelson paper.[1]

Let us consider the inelastic scattering of the incident electric field $\vec{E}_i = \vec{E}^\theta(\vec{k}^\theta)\exp[i(\vec{k}^\theta\cdot\vec{r}-\omega_i t)]$ in the eigenstate θ, $\vec{k}^\theta$ by the fluctuation in the dielectric tensor $\delta\overleftrightarrow{\epsilon} = \delta\overleftrightarrow{\epsilon}^\mu(\vec{K})\exp[i(\vec{K}\cdot\vec{r}-\Omega t)]$ produced by the acoustic-phonon mode $\mu, \vec{K}$.

In the linear dipole approximation the scattered electric field at a point $\vec{R}$ in the far field is given by (mks units)

$$\vec{E}_{\text{dip}}(\vec{R},t) = \frac{1}{4\pi\epsilon_0 R}\left(\frac{\omega_d}{c}\right)^2 \hat{R}\times(\hat{R}\times[\delta\vec{P}]), \qquad (1)$$

where the square bracket around the nonlinear driving polarization $\delta\vec{P}$ denotes retarded value. The Poynting vector of the scattered field is parallel to the unit vector $\hat{R}=\vec{R}/R$ in the direction of observation. The angular frequency (ω_d) of the diffracted light is given by $\omega \equiv \omega_d = \omega_i \pm \Omega \approx \omega_i$.

In nonconducting crystals the unit field eigenvectors $\hat{e}^\varphi$ ($\varphi = 1, 2$) associated with a given Poynting vector are real (linear polarization of eigenmodes) and form together with $\hat{R}$ an orthonormal set. Using the expansion $\hat{R}\times(\hat{R}\times\vec{A}) = \sum_{\varphi=1,2}(\hat{e}^\varphi\cdot\vec{A})\hat{e}^\varphi$, the time-independent part of the electric field can be written

$$\vec{E}_{\text{dip}}(\vec{R},\omega) = \frac{1}{4\pi\epsilon_0 R}\left(\frac{\omega}{c}\right)^2 \sum_{\varphi=1,2}[\hat{e}^\varphi\cdot\delta\vec{P}(\omega)]\hat{e}^\varphi e^{i\vec{k}^\varphi\cdot\vec{R}}, \qquad (2)$$

where the amplitude of the driving polarization is given by $\delta\vec{P} = \epsilon_0\delta\overleftrightarrow{\epsilon}_r^\mu(\vec{K})\cdot\vec{E}^\theta$. The diffracted wave vector is determined by the selection rule $\vec{k}^\varphi = \vec{k}^\theta \pm \vec{K}^\mu$ (phase-matched scattering).

The scattering cross section between the light polarization states θ, φ defined as $\sigma_{\text{dip}}^{\theta,\varphi} = |\vec{S}_d^\varphi| R^2/|\vec{S}_i^\theta|$ can now be obtained from the magnitude of the Poynting vectors

$$|\vec{S}_d^\varphi| = \tfrac{1}{2}|\mathrm{Re}[(\vec{E}_{\text{dip}}^\varphi)^* \times (\vec{\nabla}\times\vec{E}_{\text{dip}}^\varphi)/i\omega\mu_0]|$$

and

$$|\vec{S}_i^\theta| = \tfrac{1}{2}\epsilon_0 c n^\theta |\vec{E}^\theta|^2 \cos\delta^\theta$$

of the diffracted and incident beams. Using the relations

$$k^\varphi = (\omega/c)n^\varphi$$

and

$$|\vec{k}^\varphi(\hat{e}^\varphi\cdot\hat{e}^\varphi) - \hat{e}^\varphi(\hat{e}^\varphi\cdot\vec{k}^\varphi)| = (\omega/c)n^\varphi\cos\delta^\varphi,$$

one finds

$$\sigma_{\text{dip}}^{\theta,\varphi} = \left(\frac{1}{4\pi\epsilon_0}\right)^2\left(\frac{\omega}{c}\right)^4 |\hat{e}^\varphi\cdot\delta\overleftrightarrow{\epsilon}^\mu(\vec{K})\cdot\hat{e}^\theta|^2 \times \frac{n^\varphi\cos\delta^\varphi}{n^\theta\cos\delta^\theta}. \qquad (3)$$

The cross section in Eq. (3) can be expressed in terms of the unit electric displacement vectors $\hat{d}^\theta, \hat{d}^\varphi$ which satisfy the relation $\overleftrightarrow{\epsilon}_r\cdot\hat{e} = n^2\cos\delta\,\hat{d}$, and in terms of the effective photoelastic tensor $\overleftrightarrow{p}^{\text{eff}}$, which accounts for the rotational effect and the indirect photoelastic effect. By means of the well-known relation $K^2|u^\mu(\vec{K})|^2 = \hbar\Omega N_K^\mu/2\rho(V_p^\mu)^2$ between the Fourier amplitude of the atomic displacement $u^\mu(\vec{K})$ and the occupation number $n_K^\mu = N_K^\mu$ or $N_K^\mu - 1$, one obtains in usual notation

$$\sigma_{\text{dip}}^{\theta,\varphi} = \frac{\pi^2}{\lambda_0^4}|\hat{d}^\varphi\cdot\overleftrightarrow{p}^{\text{eff}}\cdot\hat{\pi}^\mu\hat{\kappa}\cdot\hat{d}^\theta|^2 \times (n^\theta)^3(n^\varphi)^5\cos\delta^\theta\cos^3\delta^\varphi\,\frac{\hbar\Omega N_K^\mu}{2\rho(V_p^\mu)^2}. \qquad (4)$$

In Ref. 2 the scattering cross section was, somewhat inconveniently, defined as the scattering efficiency per unit solid angle around the scattered-wave vector $\vec{k}^{\varphi}$ and per unit length along the incident-wave vector $\vec{k}^{\theta}$. Furthermore, separate powers in the two components of the scattered polarization were not calculated, and $\hat{R}\times(\hat{R}\times\vec{\xi}^{\mu})$ were wrongly replaced by $\hat{k}^{\varphi}\times(\hat{k}^{\varphi}\times\vec{\xi}^{\mu})$, $\hat{k}^{\varphi}=\vec{k}^{\varphi}/k^{\varphi}$ [Eqs. (3) and (21) of Ref. 2].

Using the dyadic Green's function appropriate to anisotropic media Nelson *et al.*[3,4] have obtained a general expression for the scattering cross section which involves the Gaussian curvature χ^{φ} of the $\omega(\vec{k})$ surface at the propagation vector $\vec{k}^{\varphi}$. The Green's-function analysis shows that the factor

$$r_1=(n^{\theta})^3(n^{\varphi})^5\cos\delta^{\theta}\cos^3\delta^{\varphi}\,, \tag{5}$$

obtained in the dipole approximation, should be replaced by

$$r_2=(n^{\theta})^3(n^{\varphi})^5\cos\delta^{\theta}\cos\delta^{\varphi}/(k^{\varphi})^2\chi^{\varphi}\,. \tag{6}$$

It follows from Eqs. (5) and (6) that the correction factor g^{φ} is given by

$$g^{\varphi}=\frac{\sigma_G^{\theta,\varphi}}{\sigma_{\rm dip}^{\theta,\varphi}}=\frac{1}{(\omega/c)^2\chi^{\varphi}(n^{\varphi})^2\cos^2\delta^{\varphi}}\,. \tag{7}$$

Note that the optical anisotropy contained in $|\hat{d}^{\varphi}\cdot\vec{p}^{\rm eff}\cdot\pi^{\mu}\hat{k}\cdot\hat{d}^{\theta}|^2$ is the same for the two models.

If one neglects the angular deviation of the group velocity from the phase velocity, but retain an index of refraction appropriate to the direction of propagation,[5] Eq. (5) is reduced to $r_0=(n^{\theta})^3(n^{\varphi})^5$.

As an example of relevance to Ref. 2 we consider the scattering from T_1 phonons in the xz plane of a uniaxial crystal. Since the T_1 mode is piezoelectrically inactive, the scattering is due to the direct photoelastic effect only. The selection rule shows that an ordinarily polarized incident beam is changed into an extraordinarily polarized scattered beam and vice versa. For an extraordinary ray in a uniaxial crystal one has[6]

$$(k)^2\chi=(\det\vec{\epsilon}_r)(\hat{k}\cdot\vec{\epsilon}_r\cdot\hat{k})/(\hat{k}\cdot\vec{\epsilon}_r^{\,2}\cdot\hat{k})^2$$

and

$$\cos\delta=(\hat{k}\cdot\vec{\epsilon}_r\cdot\hat{k})/(\hat{k}\cdot\vec{\epsilon}_r^{\,2}\cdot\hat{k})^{1/2}\,,$$

where $\hat{k}=\vec{k}/k$. If the incident and scattered lights have their propagation vectors in the directions $\hat{k}^{\theta}=(\cos\alpha,0,\sin\alpha)$ and $\hat{k}^{\varphi}=(\cos\beta,0,\sin\beta)$, the appropriate refractive indices for scattering from ordinary into extraordinary polarization [case (i)] are $n_i=n_0$ and $n_d=n_en_0(n_0^2\cos^2\beta+n_e^2\sin^2\beta)^{-1/2}$; and from extraordinary into ordinary polarization [case (ii)] $n_i=n_en_0(n_0^2\cos^2\alpha+n_e^2\sin^2\alpha)^{-1/2}$ and $n_d=n_0$.

In case (i), Eqs. (5) and (6) yield

$$r_1=n_0^8\mu^{5/2}\,\frac{(\cos^2\beta+\mu\sin^2\beta)^{1/2}}{(\cos^2\beta+\mu^2\sin^2\beta)^{3/2}} \tag{8}$$

and

$$r_2=n_0^8\mu^{3/2}\,\frac{(\cos^2\beta+\mu^2\sin^2\beta)^{3/2}}{(\cos^2\beta+\mu\sin^2\beta)^{5/2}}\,, \tag{9}$$

where $\mu=(n_e/n_0)^2$. Neglecting the deviation δ^{φ} one gets $r_0=n_0^8\mu^{5/2}(\cos^2\beta+\mu\sin^2\beta)^{5/2}$.

In case (ii), one finds

$$r_1=r_2=n_0^8\mu^{3/2}[(\cos^2\alpha+\mu\sin^2\alpha)\times(\cos^2\alpha+\mu^2\sin^2\alpha)]^{-1/2} \tag{10}$$

as expected since a spherical $\omega(\vec{k})$ surface for the scattered beam implies that $\chi^{\varphi}(k^{\varphi})^2=1$ and $\delta^{\varphi}=0$.

In the theory of Nelson *et al.*[3,4] only the Gaussian curvature of the $\omega(\vec{k})$ surface for the scattered light occurs. This is due to the fact that the incident light normally forms a parallel beam (laser radiation). Neglecting δ^{θ} one obtains $r_0=n_0^8\mu^{3/2}\times(\cos^2\alpha+\sin^2\alpha)^{-3/2}$.

In Fig. 1 is shown the angular variation of r_1 and r_2 in CdS for $\lambda_0=6328$ Å ($n_0=2.460, n_e=2.477$) for the cases (i) and (ii). It is seen that the angular dependence predicted by the dipole approximation is far from the correct variation obtained by the Green's-function formalism [case (i)]. Furthermore, the simple model retaining only the directional dependence of the index of refraction ($\delta^{\theta}=\delta^{\varphi}=0$) gives results close to r_1. (With the vertical scale used in the figure r_0 and r_1 coincide.)

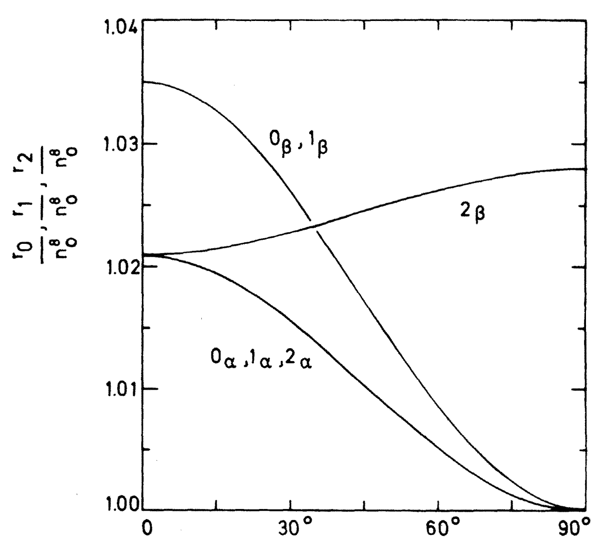

FIG. 1. Angular variation of the anisotropy factors for T_1-phonon scattering in the xz plane of CdS($\lambda_0=6328$ Å) as obtained from a "zero-order" (0_α, 0_β) and a "first-order" (1_α, 1_β) dipole approximation, and from a Green's-function calculation (2_α, 2_β). The incident and scattered photons are denoted by α and β.

This holds in general for weak anisotropic media since $\mu = 1 + \Delta, \Delta \ll 1$ implies that $r_1 = r_0 = n_0^8(1 + \frac{5}{2}\Delta \cos^2\beta)$ [and $r_2 = n_0^8(\frac{1}{2}\Delta)(3 + \sin^2\beta)$] for case (i), and $r_1 = r_0 = n_0^8(1 + \frac{3}{2}\Delta \cos^2\alpha)$ for case (ii) if one calculates to first order in Δ. It is obvious that the correction introduced by the anisotropy factors (r_0, r_1, r_2) is small in weak anisotropic crystals like CdS.

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