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Comment on Keller's theory of Brillouin scattering

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Keller has published a treatment of Brillouin scattering inside an anisotropic crystal with application to off-axis phonons in CdS, whereas the present authors have provided a treatment valid for a crystal of any symmetry and level of anisotropy, any direction of input light, scattered light, and sound (consistent with phase matching) and to either polarization of the light beams and any type of sound wave. Moreover, our treatment includes solid angle and demagnification corrections for an arbitrarily oriented exit crystal surface. Since the treatment of Keller does not use the dyadic Green's function appropriate to an anisotropic medium, his results are in disagreement with ours after we have omitted the surface corrections from our theory.

Keller[1] has written an interesting paper on Brillouin scattering of off-axis phonons in CdS combining work on phase matching kinematics in anisotropic media[2] with the Brillouin scattering theory of Hamaguchi.[3] The first sentence of his abstract reads "Incorporating the rotational contribution to the direct photoelastic effect and the angular deviation of the Poynting vector from the wave vector of the diffracted light, a Brillouin scattering theory, valid for a general anisotropic scattering kinematics in a hexagonal crystal is derived." Keller also acknowledges the present authors's general theory of Brillouin scattering in anisotropic media[4] which is based on a Green's function for electromagnetic propagation in an anisotropic medium[5] and which includes the very important boundary corrections needed for comparison with experiment.[4-8] The reader may assume that if our results are specialized to the hexagonal case, they will agree with Keller's results inside the crystal, and that our principal contribution is the correction associated with the boundary (i. e., solid angle and demagnification corrections associated with observing the light outside the crystal rather than inside). This conclusion is false, in the sense, that if there is appreciable anisotropy, Keller's results[1] (as well as Hamaguchi's[3]) are in significant disagreement with ours, even inside the crystal. We believe ours to be correct. This belief is based not only on an examination of the two theories but also on a successful comparison of our theory with experiment[4] on the highly anisotropic crystal calcite. In that work 36 independent measurements were consistently fitted to nine photoelastic tensor components. Furthermore, the measured value of the antisymmetric component agreed within 5% with theory.[4] This level of accuracy is believed to apply to all components measured. Since this level is small compared to anisotropy corrections, which are at issue here, we believe the agreement supports the calculated anisotropy effects of our theory.

To facilitate comparison with Keller we rewrite our result for the scattered power per unit flux inside the crystal in a form that relates it to the corresponding quantities outside as well as to the scattering efficiency R:

$$\frac{P_{\text{ins}}^{\text{scat}}}{P_{\text{ins}}^{\text{inc}}/A}=\frac{P_{\text{out}}^{\text{scat}}/T^{\text{exit}}}{P_{\text{out}}^{\text{inc}}\,T^{\text{ent}}/A}=RV^{S}d\Omega_{\text{in}}^{r}\,. \tag{1}$$

Here $P_{\text{ins}}^{\text{inc}}$, the incident(inc) power inside(ins) the crystal, differs from the incident power outside(out) the crystal $P_{\text{out}}^{\text{inc}}$ by the entrance transmission coefficient T^{ent}. The incident beam inside the crystal has a cross-sectional area A. A transmission correction T^{exit} also relates the scattered power outside the crystal $P_{\text{out}}^{\text{scat}}$ to the scattered power inside $P_{\text{ins}}^{\text{scat}}$. The scattering cross-section [the right-hand side of Eq. (1)] is proportional to the accepted solid angle $d\Omega_{\text{in}}^{r}$ of ray vectors inside the crystal, and the source volume V^{S} from which light is accepted by the detector field stop. Thus the scattering efficiency R, an intrinsic property of the crystal and the scattering process, has the dimensions of a reciprocal length (R is the scattered power per unit incident power, per unit solid angle, per unit path length).

Since Keller does not discuss the relation between $d\Omega_{\text{in}}^{r}$, V^{S} and the detection optics, we must compare our expression for R with his corresponding expression (denoted σ_B). Equation (4.6) of our Ref. 6, when compared with Eq. (1) yields the expression

$$R=\left(\frac{\omega}{c}\right)^{4}\frac{n^{\phi}}{8\pi^{2}n^{\theta}\cos\delta^{\phi}\cos\delta^{\theta}}\,\frac{J}{(k^{\phi})^{2}K^{\phi}}\,, \tag{2}$$

where ω is the (angular) frequency of the scattered light, c is the velocity of light in vacuum, θ refers to the incident beam (in the crystal), ϕ refers to the scattered beam, n^{θ} and n^{ϕ} are the corresponding indices of refraction, δ^{θ} (or δ^{ϕ}) is the small angle between ray and wave vector for the incident beam (or scattered beam), k^ϕ is the propagation constant of the scattered beam, and K^ϕ is the Gaussian curvature of the $\omega(\vec{k})$ surface at the propagation vector $\vec{k}^\phi$. All the above mentioned factors enter because of the way in which light propagates in an anisotropic crystal according to our Green's-function formalisms.[5] The nonlinear interaction that yields the scattering is contained in the factor,[6]

$$J \equiv \frac{\left| \vec{e}^\phi \cdot \int_{V^S} e^{-i\vec{k}^\phi\cdot\vec{r}}\, \vec{P}^{\mathrm{NL}}(\vec{r},\omega)\, d^3r \right|^2}{2\epsilon_o^2 \left| E^\theta \right|^2 V^S} , \qquad (3)$$

where $\vec{P}^{\mathrm{NL}}$ is the nonlinear polarization induced by the incident electric field $\vec{E}^\theta$, $\vec{e}^\phi$ is the unit electric field vector of the scattered beam and ϵ_o is the free space permittivity. The evaluation of J for Raman scattering by polaritons is given in Ref. 6, whereas its evaluation for Brillouin scattering is given in Ref. 4. For the Brillouin case

$$J = (kT/4\rho \mathrm{v}^2)\ (n^\phi n^\theta)^4 (\cos\delta^\phi \cos\delta^\theta)^2 F , \qquad (4)$$

where

$$F \equiv \left| d_i^\phi p_{ijkl}^{\mathrm{eff}} d_j^\theta b_k a_l \right|^2 , \qquad (5)$$

$\vec{d}^\theta$ (or $\vec{d}^\phi$) is the unit electric displacement vector for the incident (or scattered) light beam, $\vec{b}$ is the unit sound wave displacement vector, and $\vec{a}$ is the unit sound wave propagation vector. The Pockels tensor p_{ijkl} carries the superscript eff to remind us that the indirect effects[9-11] and the direct rotational effect[12,13,10] are included. The conventional factors k, T, ρ and v are Boltzmann's constant, the absolute temperature, the crystal density, and sound velocity, respectively.

Equations (2)-(5) may be combined to yield the Brillouin scattering efficiency in the form

$$R = \frac{(\omega/c)^4}{32\pi^2} \frac{kT}{\rho v^2} r_{\mathrm{LN}} , \qquad (6)$$

where the dimensionless angular-dependent factor r_{LN} is given by

$$r_{\mathrm{LN}} = \frac{(n^\phi)^5 (n^\theta)^3 \cos\delta^\phi \cos\delta^\theta}{(k^\phi)^2 K^\phi} F . \qquad (7)$$

Equation (6) is the result for Stokes or anti-Stokes Brillouin scattering for phonons such that $\hbar\omega << kT$ at thermal equilibrium. For the nonequilibrium and/or quantum case Stokes scattering should be corrected by a factor $(\hbar\omega/kT)(n+1)$, and the anti-Stokes by $(\hbar\omega/kT)n$, where n is the actual number of phonons in the mode. In the equilibrium case with $n = [\exp(\hbar\omega/kT)-1]^{-1}$ these factors are close to the unity for typical frequencies and temperatures in ordinary Brillouin scattering experiments. To facilitate the application of Eq. (7) to the uniaxial case considered by Keller, we note that for the extraordinary ray in a uniaxial crystal[5]

$$(k^\phi)^2 K^\phi = \frac{(\det \overleftrightarrow{\kappa})\, (\vec{s}\cdot\overleftrightarrow{\kappa}\cdot\vec{s})}{(\vec{s}\cdot\overleftrightarrow{\kappa}^2\cdot\vec{s})^2} , \qquad (8)$$

$$\cos\delta^\phi = (\vec{s}\cdot\overleftrightarrow{\kappa}\cdot\vec{s})/(\vec{s}\cdot\overleftrightarrow{\kappa}^2\cdot\vec{s})^{1/2} , \qquad (9)$$

where $\vec{s} = \vec{k}^\phi/k^\phi$ is a unit vector in the direction of the scattered wave vector and $\overleftrightarrow{\kappa}$ is the dielectric tensor.

Keller's equation (27) can be rewritten in the form of Eq. (6), above, with r_{LN} replaced by

$$r_K = a \left[\vec{s}\times(\vec{s}\times\vec{\xi})\right] \cdot \overleftrightarrow{\kappa} \cdot \left[\vec{s}\times(\vec{s}\times\vec{\xi})\right] , \qquad (10)$$

with

$$a \equiv \frac{n^\theta}{n^\phi(\vec{e}^\theta\cdot\overleftrightarrow{\kappa}\cdot\vec{e}^\theta)} = \frac{1}{n^\phi n^\theta \cos^2\delta^\theta} , \qquad (11)$$

where $\vec{e}^\theta$ is a unit vector in the direction of the incident electric field in the crystal. From Keller's definition, his Eq. (18), the vector[14]

$$\begin{aligned} \xi_i &= -\chi_{ijkl} e_j^\theta b_k a_l / \kappa_{11}^2 \\ &= \frac{(n^\theta)^2 \cos\delta^\theta}{2\kappa_{11}^2} \kappa_{im} p_{mnkl}^{\mathrm{eff}} b_k a_l d_n^\theta , \end{aligned} \qquad (12)$$

where χ_{ijkl} is the complete photoelastic susceptibility.

Although there are numerous identities relating the vectors $\vec{e}^\theta$, $\vec{e}^\phi$, $\vec{s}$ and the direction $\vec{t} = \vec{\nabla}\omega(\vec{k})/|\vec{\nabla}\omega(\vec{k})|$ of the ray vector, these involve properties that depend only on the first derivatives of $\omega(\vec{k})$. Since r_{LN} involves the Gaussian curvature K^ϕ, which involves the second derivatives of $\omega(\vec{k})$, and Keller's result, r_K, does not, it is not possible to transform Keller's result into ours. This difference arises because Keller does not use the correct Green's function for an anisotropic crystal[5] that involves the Gaussian curvature. In short, a correct expression for light scattering that applies to observation in a particular direction *inside* an optically anisotropic medium *must* contain the Gaussian curvature of $\omega(\vec{k})$ at the value of $\vec{k}$ corresponding to the observed light.

Keller's work is also limited by the fact that he has not calculated the separate powers in the two components of scattered polarization. This is evident since his answer does not depend on the unit scattered electric field vector $\vec{e}^\phi$.

To demonstrate the disagreement with Keller more specifically, we evaluate Eq. (7) for the case of T_1 phonons in a hexagonal crystal and quote Keller's answer for the same result. For the case considered by Keller, the phonon propagates in the x,z plane

$$\vec{a} = (\sin\theta,\ 0,\ \cos\theta) , \qquad (13)$$

but has its displacement vector perpendicular to that plane

$$\vec{b} = (0,\ 1,\ 0)\ . \tag{14}$$

The incident light is ordinary with its unit electric displacement vector given by

$$\vec{d}^{\theta} = (0,\ 1,\ 0)\ . \tag{15}$$

The scattered light has its propagation vector in the direction

$$\vec{s} = (-\cos\beta,\ 0,\ \sin\beta). \tag{16}$$

If the scattered light is ordinary, Eq. (7) yields a vanishing result because F=0. For scattering from ordinary into extraordinary polarization, Eq. (7) yields

$$r_{\mathrm{LN}} = \kappa_{11}^{4}\mu^{3/2}\ \frac{(\cos^2\beta+\mu^2\sin^2\beta)^{3/2}}{(\cos^2\beta+\mu\sin^2\beta)^{5/2}}$$

$$\times\ (p_{1221}\sin\beta\ \sin\theta + p_{3223}\cos\beta\ \cos\theta)^2\ , \tag{17}$$

where $\mu = \kappa_{33}/\kappa_{11}$.

Instead of evaluating r_K using Eq. (10) we shall instead use Keller's equation (32), his final answer for this geometry. His result is

$$r_K = \kappa_{11}^{4}\ (\cos^2\beta+\mu\ \sin^2\beta)^{1/2}/\mu^{1/2}$$

$$\times\ (p_{1221}\sin\theta\ \sin\beta + \mu p_{3223}\cos\theta\ \cos\beta)^2\ . \tag{18}$$

Equations (17) and (18) are clearly different. Although the difference is small in CdS, it can be of significance in materials of greater anisotropy, such as calcite, and Keller's remarks lead the reader to believe his results have wider applicability.

Keller dismisses our work[4] with the statement, "Unfortunately, the above theories are too complicated to be applied to elastic waves with arbitrary polarization and propagation directions." We claim, however, that Eq. (7) is not only correct but is as easy to apply as Eq. (10).

Expression (6) for R describes the scattering process inside the crystal. But the angle $d\Omega_{\mathrm{in}}^{r}$ is actually determined from the detector solid angle $d\Omega^D$ by the relation[5]

$$d\Omega_{\mathrm{in}}^{r} = d\Omega^D K^{\phi}\ (\omega/c)^2\ \cos\alpha/\cos\beta\ , \tag{19}$$

where β is the angle of arrival of the scattered light ray to the normal of the exit surface inside the crystal and α is the corresponding departure angle outside the crystal. Similarly, the scattering length $l_S = V_S/A$ inside the crystal is limited by a length l_D associated with the detector field stop according to the relation[5]

$$l_S = Nl_D\cos\beta/(\cos\alpha\ \sin\theta_S)\ , \tag{20}$$

where θ_S is the scattering angle inside the crystal, and N is a correction for noncoplanarity given by[8]

$$N = 1/(\cos\phi\ \cos\phi' - \cos\beta\ \sin\phi\ \sin\phi')\ . \tag{21}$$

Here ϕ' is the angle of tilt between the plane defined by the unscattered laser beam and the normal to the input surface and the plane defined by scattered ray and exit surface normal; ϕ is the angle of tilt between the latter plane and the departure plane defined by the normal to the exit surface and the scattered ray after it has left the crystal. N reduces to unity when the unscattered beam, the scattered wave vector, scattered ray, and exit surface normal are all in the same plane.

Equation (1) can then be written in a form useful for describing any scattering process outside the crystal

$$\frac{P_{\mathrm{out}}^{\mathrm{scat}}}{P_{\mathrm{out}}^{\mathrm{inc}}} = \frac{Rl_D d\Omega^D N K^{\phi} T^{\mathrm{ent}} T^{\mathrm{exit}}}{\sin\theta_S}\ \left(\frac{\omega}{c}\right)^2 \tag{22}$$

or

$$\frac{P_{\mathrm{out}}^{\mathrm{scat}}}{P_{\mathrm{out}}^{\mathrm{inc}}} = \left(\frac{\omega}{c}\right)^4 \frac{Nl_D d\Omega^D}{\sin\theta_S}\ \frac{T^{\mathrm{ent}} T^{\mathrm{exit}} J}{8\pi^2 n^{\phi} n^{\theta}\cos\delta^{\phi}\cos\delta^{\theta}} \tag{23}$$

after Eq. (2) is used to relate R to the intrinsic property J of the scattering mechanism. If the expression, Eq. (4), for J that applies to Brillouin scattering is inserted into Eq. (23), we obtain

$$\frac{P_{\mathrm{out}}^{\mathrm{scat}}}{P_{\mathrm{out}}^{\mathrm{inc}}} = \frac{(\omega/c)^4}{32\pi^2}\ \frac{kT}{\sin\theta_S}\ N T^{\mathrm{ent}} T^{\mathrm{exit}} l_D d\Omega^D$$

$$\times\ \frac{(n^{\phi} n^{\theta})^3\cos\delta^{\phi}\cos\delta^{\theta}}{\rho v^2}\ \left| d_i^{\phi} p_{ijkl}^{\mathrm{eff}} d_j^{\theta} b_k a_l \right|^2 . \tag{24}$$

Equation (24) with $N = 1$ (noncoplanarity neglected) agrees with Eq. (2.35) of Ref. 4. The latter was used to analyze the experimental data for Brillouin scattering in calcite where anisotropy plays a key role. For the calcite measurements, no noncoplanarity correction was needed.

In summary our results inside the crystal differ from those of Keller because we have used the correct Green's function inside an anisotropic medium. Our result Eq. (24), for Brillouin scattering applies to a crystal of arbitrary symmetry and level of anisotropy, to arbitrary directions of the input light, the scattered light and the sound wave, consistent with phase matching, to sound waves of any type, and to either polarization of input and scattered light. Equation (24) also includes the solid angle and demagnification corrections for arbitrary directions of the above mentioned rays to the crystal axes and surface normal (when the noncoplanarity correction factor N is included). These surface corrections are important because measurements are always performed outside the crystal. Contrary to Keller's comment, we believe Eq. (24) is in an easily applicable form, in spite of its great generality.

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[14] The factor of 2 that enters the relation between the photoelastic susceptibility χ_{ijkl} and the Pockels tensor p_{mnkl} arises because χ is conventionally quoted for an ac measurement in which Stokes and anti-Stokes contributions are distinguished, whereas the Pockels tensor is quoted as a static quantity. See Ref. 12, Eq. (V-4.6), or Ref. 11, Eq. (4.17).