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THE EXACT EQUATION FOR BRILLOUIN SHIFTS

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Résumé. — L'équation exacte des déplacements dans l'effet Brillouin est représentée par :

$$\frac{\Delta \nu}{\nu_i} = \frac{\beta_o^2 - \beta_s \beta_i \cos \theta \pm \sqrt{(\beta_o - \beta_i)^2 + 4 \beta_o \beta_i \sin^2 \frac{\theta}{2} - \beta_o \beta_i \sin^2 \theta}}{1 - \beta_o^2}$$

$$\approx \beta_o^2 - \beta_s \beta_i \cos \theta \pm \sqrt{(\beta_o - \beta_i)^2 + 4 \beta_o \beta_i \sin^2 \frac{\theta}{2}}.$$ 

$\nu_i$ est la fréquence de l'onde incidente, $\theta$ l'angle de diffusion ; $\beta_o = n_o v_o / c$, $\beta_i = n_i v_i / c$ où $n_o$ et $n_i$ sont les indices de réfraction pour l'onde incidente et l'onde diffusée, $c$ la vitesse de la lumière dans le vide, et $v_o$ la vitesse de l'onde acoustique responsable de la diffusion.

Les deux faits les plus importants sont l'effet d'anisotropie dans les cristaux birefringents, déjà discuté par l'auteur et l'effet dû au terme quadratique.

Dans le cas des cristaux isotropes et des liquides, on a :

$$\frac{\Delta \nu}{\nu_i} = \pm 2 \beta_i \sin \frac{\theta}{2} + 2 \beta_i \sin^2 \frac{\theta}{2} \left(1 + \frac{n_i}{n_i} \frac{dn}{d\nu}\right).$$

Le terme quadratique conduit à des déplacements inégaux pour les composantes Stokes et anti-Stokes, correspondant à l'effet Doppler relativiste, plus un petit terme de dispersion.

Pour la diffusion arrière dans le diamant, l'effet Brillouin quadratique pourrait être déterminé expérimentalement ; les déplacements prédits pour les composantes longitudinalles suivant la direction [111] valent, pour $\lambda = 2537 \text{ Å}$

$\Delta \nu_{AS} = +12.79887 \text{ cm}^{-1}$

$\Delta \nu_{S} = -12.79370 \text{ cm}^{-1}.$

Abstract. — The exact equation for the Brillouin shifts is given by

$$\frac{\Delta \nu}{\nu_i} = \frac{\beta_o^2 - \beta_s \beta_i \cos \theta \pm \sqrt{(\beta_o - \beta_i)^2 + 4 \beta_o \beta_i \sin^2 \frac{\theta}{2} - \beta_o \beta_i \sin^2 \theta}}{1 - \beta_o^2}$$

$$\approx \beta_o^2 - \beta_s \beta_i \cos \theta \pm \sqrt{(\beta_o - \beta_i)^2 + 4 \beta_o \beta_i \sin^2 \frac{\theta}{2}}.$$ 

where $\nu_i$ is the frequency of the incident light, $\theta$ the scattering angle, $n_o$ and $n_i$ are the refractive indices for the incident and the scattered waves, $v_o$ the velocity of light in vacuum, $v_i$ the velocity of the acoustic wave responsible for the scattering ; $\beta_o = n_o v_o / c$, $\beta_i = n_i v_i / c.$ The two main additional features are the anisotropic effect for birefringent crystals discussed earlier by the author [3] and the quadratic term. In the case of isotropic crystals or liquids.

$$\frac{\Delta \nu}{\nu_i} = \pm 2 \beta_i \sin \frac{\theta}{2} + 2 \beta_i \sin^2 \frac{\theta}{2} \left(1 + \frac{n_i}{n_i} \frac{dn}{d\nu}\right).$$

The quadratic term leads to unequal displacements of anti-Stokes and Stokes components corresponding to the relativistic Doppler effect but with a small dispersive term.

In the case of backward scattering in diamond the quadratic Brillouin effect can be determined experimentally ; the expected shifts for longitudinal components along the [111] direction are for $\lambda = 2537$, $\Delta \nu_{AS} = +12.79887 \text{ cm}^{-1}$, $\Delta \nu_{S} = -12.79370 \text{ cm}^{-1}.$

Introduction. — Several years ago a systematic investigation of Brillouin scattering in crystals was undertaken by Krishnan and the author [1-5] using 2 537 Å resonance radiation of mercury. In this connection, the author [3] generalized the Brillouin equation to birefringent crystals and showed that, in general, there would be 12 pairs of Brillouin components instead of 3 as in the case of isotropic crystals. Interest in this field has been revived recently, by the discovery of the laser and the stimulated Brillouin scattering [6].

In the usual theory, the small change in wavelength of the scattered radiation is implicitly ignored in the equation for conservation of momentum. It was considered worthwhile to take into account this second order effect in view of high precision

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attainable with a laser source. The exact equation for Brillouin shifts is, therefore, derived and applied to the case of diamond.

**Derivation of the exact equation.** — After cancelling out $h$ in the elastic interaction of photons and acoustic phonons, the conservation laws of energy and momentum yield

$$v_s - v_i = \pm v_e$$

$$\frac{I}{\lambda_s} - \frac{I}{\lambda_i} = \frac{I}{\lambda_e}$$

where the subscript $i$ denotes the incident, $s$ the scattered photon and $e$ the effective phonon responsible for scattering, $v$ is the frequency and $\lambda$ the wavelength.

If $n$ is the refractive index, $v_e$ is the velocity of the phonon and $c$ the velocity of light, then

$$\frac{n_s v_e - n_i v_i}{c} = \frac{v_o}{v_e}$$

putting

$$\frac{n_s v_e}{c} = \beta_s, \quad \frac{n_i v_i}{c} = \beta_i$$

$$\beta_s v_s - \beta_i v_i = v_o$$

Squaring (4) to convert it into a scalar equation

$$\beta^2_s v_s^2 - 2\beta_s \beta_i \cos \theta v_s v_i + \beta^2_i v_i^2 = v_o^2$$

where $\theta$ is the angle of scattering.

Combining equations (1) and (5)

$$\frac{(1 - \beta^2_s) v_s^2 - 2(1 - \beta_s \beta_i \cos \theta) v_s v_i + (1 - \beta^2_i) v_i^2}{1 - \beta^2_i} = 0$$

This is the exact equation for Brillouin shifts.

Ignoring terms to order higher than $\beta^2$,

$$\frac{\Delta v}{v} \approx \beta^2_s - \beta_s \beta_i \cos \theta \pm \sqrt{4\beta_s^2 \beta_i^2 \sin^2 \frac{\theta}{2} + (\beta_s - \beta_i)^2}$$

Leaving out the second order term in this equation leads to that developed by the author [4] earlier for birefringent crystals

$$\frac{\Delta v}{v} = \pm \sqrt{4\beta_s^2 \beta_i^2 \sin^2 \frac{\theta}{2} + (\beta_s - \beta_i)^2}$$

If further, one sets $\beta_s = \beta_i = \beta$ then the usual Brillouin equation results.

$$\frac{\Delta v}{v} = \pm 2\beta \sin \frac{\theta}{2} = \pm \frac{2n}{c} n \sin \frac{\theta}{2}$$

In the usual derivation this simplification is usually made in equation (4).

**Discussion of the exact equation.** — In the case of an isotropic medium ($\beta_s = \beta_i$) is a second order term arising from the dispersion of the medium. Let $\beta_s - \beta_i = \Delta \beta$ and $n_s - n_i = \Delta n$

$$\frac{\Delta v}{v} \approx \beta^2_i - \beta^2_i \cos \theta \pm 2\beta_i \sin \frac{\theta}{2} \sqrt{1 + \frac{\Delta \beta}{\beta_i}}$$

$$\approx \pm 2\beta_i \sin \frac{\theta}{2} + 2\beta_i \sin^2 \frac{\theta}{2} \pm \Delta \beta \sin \frac{\theta}{2}$$

The second term arises from the fact that the magnitude of the momentum of the scattered photon is different from that of the incident photon ($v_s \neq v_i$) even if dispersion is ignored. The last term is due to dispersion and $\Delta \beta$ is positive for the anti-Stokes component and negative for the Stokes component. Denoting them by $AS$ and $S$ respectively,

$$\frac{\Delta v_{AS}}{v_i} = \frac{\Delta v}{v_i} - \frac{\Delta v}{v_i} = 2\beta_i \sin \frac{\theta}{2} + 2\beta_i \sin^2 \frac{\theta}{2} + |\Delta \beta_i| \sin \frac{\theta}{2}$$

$$\frac{\Delta v_S}{v_i} = \frac{\Delta v}{v_i} = 2\beta_i \sin \frac{\theta}{2} + 2\beta_i \sin^2 \frac{\theta}{2} + |\Delta \beta_i| \sin \frac{\theta}{2}$$

$$1 \frac{\Delta v_{AS} - \Delta v_S}{v_i} = \frac{\Delta v_{AS}}{v_i} - \frac{\Delta v_S}{v_i} = 2\beta_i \sin \frac{\theta}{2} + 2\beta_i \sin^2 \frac{\theta}{2} + 1 |\Delta \beta_i| \sin \frac{\theta}{2}$$

$$\frac{1}{2} \frac{\Delta v_{AS} + \Delta v_S}{v_i} = \frac{\Delta v_{AS}}{v_i} + \frac{\Delta v_S}{v_i} = 2\beta_i \sin \frac{\theta}{2} + 1 |\Delta \beta_i| \sin \frac{\theta}{2}$$

$$\frac{\Delta \beta_i}{n_i} = \frac{\Delta n}{n_i} \frac{\Delta v}{n_i \Delta n} = \frac{1}{n_i \Delta n} \frac{d}{v} = \pm 2\beta_i \sin \frac{\theta}{2} + \frac{1}{n_i \Delta n} \frac{d}{v}$$

$$\frac{\Delta v_{AS} + \Delta v_S}{v_i} = \frac{\Delta v_{AS}}{v_i} + \frac{\Delta v_S}{v_i} = 2\beta_i \sin \frac{\theta}{2} \left(1 + \frac{1}{n_i \Delta n} \frac{d}{v} \right)$$

$$\frac{\Delta v_{AS}}{v_i} = \frac{1}{2} (\Delta v + \Delta v_S).$$
The linear Brillouin shift expressed as the average $\Delta v$ in equation (12) remains the same as in the usual approximation. But the quadratic Brillouin shift given by equation (14) leads to unequal displacements of the anti-Stokes and Stokes components, the former being larger in frequency units. This is more conveniently expressed as a positive shift $\Delta v_Q$ of the average frequency of the two components $v_Q$ with respect to the frequency $v_i$.

Expressing the shifts in wavelength units

$$\frac{\Delta \lambda}{\lambda_i} = \lambda_{AS} - \lambda_S = -2\beta_i \sin \frac{\theta}{2} \left(1 + \frac{\nu_i \Delta n}{n_i \Delta v}\right)$$  \hspace{1cm} (15)

$$\frac{\Delta \lambda Q}{\lambda_i} = \lambda_{AS} + \lambda_S - 2\lambda_i = 2\beta_i \sin^2 \frac{\theta}{2} \left(1 + \frac{\nu_i \Delta n}{n_i \Delta v}\right).$$ \hspace{1cm} (16)

It is remarkable that the average wavelength of the two components $\lambda_Q$ is also larger than $\lambda_i$ so that both $\Delta \lambda Q$ and $\Delta v_Q$ are positive.

**Backward scattering and diamond.** — In the case of backward scattering $\theta = 180^\circ$, the Brillouin shift and the asymmetric quadratic term are both largest. Even the exact equation (6) simplifies to

$$\Delta v / v_i = (\beta_S + \beta_t) / (1 - \beta_i^2) \approx 2\beta_t + \beta_i \left(1 + \frac{\nu_i \Delta n}{n_i \Delta v}\right).$$

$$\Delta v / v_i = 2\beta_t$$ \hspace{1cm} (17)

$$\Delta v_Q / v_i = 2\beta_i \left(1 + \frac{\nu_i \Delta n}{n_i \Delta v}\right).$$ \hspace{1cm} (18)

Since the second order term could only be experimentally measured when $\beta$ is large, diamond is extremely favorable for such a test. On account of the exceptionally large velocity of sound waves in it and its high refractive index, the Brillouin components have been recorded by Krishnan [1] and the author [3].

Using the elastic constants of diamond [7], we have calculated in the following table I the expected shifts using equation (15) for the two wavelengths corresponding to the gas laser and Hg 198 resonance radiation:

- $\rho = 3.512 \text{ g/cm}^3$, velocity of longitudinal wave (L) = 18 562 m/s along the [111] direction. Velocity of transverse wave (T) = 12 038 m/s.

One notices that the anti-Stokes and Stokes shift differ appreciably by as much as 5 mK in the most favorable case. This quadratic Brillouin effect is within experimental means in view of modern precisions of 50 $\mu$A or 0.4 mK attainable even in absolute wavelengths [8-9]: further, this effect as measured by $\Delta v_Q$ is different for the transverse and longitudinal components. This permits an accurate measurement even when the incident radiation is absorbed by a resonance filter in the scattered path.

The dispersion term $\frac{\nu_i \Delta n}{n_i \Delta v}$ is practically negligible at 6 328 Å but is appreciable at 2 537 Å and this may also be possibly measured.

**Collimation effects.** — If there is a finite divergence in the incident beam of $\Delta \theta$ and a similar divergence in the scattered beam, then the angle of scattering varies between $180^\circ$ and $180^\circ + \Delta \theta/n$ since the divergence inside the crystal is effective for backward scattering; the first order effect on Brillouin shift is zero. In the second order, the change in shift

$$d[\Delta v] = -2\beta_i \nu_i \frac{(\Delta \theta)^2}{8\pi^2}.$$  \hspace{1cm}

For $\Delta \theta = 1^\circ$ the width amounts to only 0.00008 cm$^{-1}$. Further, this broadening is symmetrical towards $v_i$ and hence the effect of this on $\Delta v_Q$ is zero.

**Comparison with relativistic Doppler effect.** — The Brillouin shift may be interpreted [10] as a Doppler effect arising from reflections of light waves by progressive sound waves. Extending this idea, it is interesting to compare the quadratic Brillouin effect to the asymmetric term in the relativistic Doppler shift [11]. The Doppler shifts of light emitted by an atom moving with a velocity $\pm v$ at an angle $0^\circ$ to the observation direction are given by

$$\frac{\Delta \lambda_B}{\lambda_o} = \frac{\lambda_B - \lambda_o}{\lambda_o} = -\beta + \frac{1}{2} \beta^2$$ \hspace{1cm} (19)

$$\frac{\Delta \lambda_R}{\lambda_o} = \frac{\lambda_R - \lambda_o}{\lambda_o} = \frac{1}{2} \beta^2$$ \hspace{1cm} (20)

$$\frac{\Delta \lambda}{\lambda_o} = \frac{\Delta \lambda_B - \Delta \lambda_R}{2\lambda_o} = -\beta$$ \hspace{1cm} (21)

$$\frac{\Delta \lambda Q}{\lambda_o} = \frac{1}{2} \beta^2$$ \hspace{1cm} (22)

$$\lambda_Q = \frac{1}{2} (\lambda_B + \lambda_R).$$

The subscripts B and R indicate blue and red shifts respectively.

Remembering that in Brillouin effect the "moving mirror" is equivalent to both the source and the observer are moving, $\beta$ in equations (21) and (22) must be replaced by $2\beta_i$. Then these equations agree with (15) and (16) respectively if one sets $\theta = 180^\circ$ in them and ignores the dispersion term in (16). This demonstrates the validity of the analogy and indicates that the observation of the quadratic Brillouin effect in diamond would be...
a test of the special theory of relativity applied to a dispersive medium.

In conclusion the author wishes to express his thanks to Dr. Oksengorn for this kind interest.

Discussion

M. Burstein. — What are the widths of Brillouin components? It would seem to me that in existing data, the width probably arises from the finite solid angle subtended by the lens that collects the scattered radiation.

The use of lasers should enable one to obtain detailed information about the widths of the Brillouin components, including temperature dependence which should in turn lead to information about the scattering of acoustic phonons as a function of frequency, up to kilomegacycle frequencies as a function of temperature for a wide variety of materials, which otherwise could not be studied by kilomegacycle acoustics. Also at higher temperature it should perhaps be possible to observe "higher order Brillouin scattering" involving phonon-phonon interactions.

REFERENCES