ARCHIVES OF ACOUSTICS
26, 3, 255–261 (2001)

COHERENT STATES IN BRILLOUIN LIGHT SCATTERING

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The present article describes inelastic light scattering or the theory of photon acoustic-phonon interactions to which the coherent state formalism can be applied; it belongs partly to quantum acoustics. Such quantum calculations lead to the same results as the classical ones, and what is important, they are not in full agreement with the results of measurements. The article provides some formulae useful for comparison of the optical signals intensity in Brillouin light scattering experiments.

1. Introduction

The inelastic scattering of photons by acoustic phonons, is still not described in depth as a quantum phenomenon in condensed matter physics. In typical areas of research the classical theory of elasticity as well as classical electrodynamics is widely applied. One of the most promising quantum type formalism of performing this task is the coherent state method worked out by R.J. Glauber and J.R. Klauder [1–4]. The formalism has been applied with great success in different areas of physics, especially in quantum optics. The formalism's value results from the fact that it describes the physical reality as a bridge between the quantum and classical phenomenon.

2. Basic elements of the quantum-like approach to the Brillouin scattering

As a basic and most important quantity in the inelastic light scattering theory, the spectral distribution of the electric field in a scattered electromagnetic wave should be calculated. This function is directly connected with quantum effects, where the scattering process between photon and phonon dominates.

From this point of view the principle substitutions required to perform quantum-like calculations will be introduced. The amplitude and time dependence for the acoustic waves can be written as follows [5]

$$|\mathbf{u}|^2 \exp\left[i\omega_{\mathrm{phon}(\mu)}\tau\right] \to \left\langle \left\| \left(n' \| a^{+(\mu)} | n \right) \right\| \right\rangle \exp\left[i\omega_{\mathrm{phon}(\mu)}\tau\right], \tag{1}$$

$$|\mathbf{u}|^2 \exp\left[-i\omega_{\mathrm{phon}(\mu)}\tau\right] \;\rightarrow\; \left\langle\left\|\left(n'\|a^{(\mu)}|n\right)\right\|\right\rangle \exp\left[-i\omega_{\mathrm{phon}(\mu)}\tau\right], \tag{2}$$

$$\mathbf{u}(\mathbf{K}) \;\rightarrow\; a(\mathbf{K}), \tag{3}$$

where the acoustic waves displacements $\mathbf{u}(\mathbf{K})$, as a function of the acoustic wave vector $\mathbf{K}$, are replaced by the creation operator a^+ and the annihilation operator a, and the acoustic wave amplitudes are replaced by the average expected values of the a^+ and a operators in a representation of a number of particle states $|n\rangle$. The time parameter of the phenomenon is the time of correlation τ. It appears in the autocorrelation function for the electric field of the scattered light $\langle \mathbf{E}'(\mathbf{K}, t+\tau) \cdot \mathbf{E}'^{*}(\mathbf{K}, t)\rangle$. Both the amplitudes (1, 2) can be expressed by the average number of phonons (bosons) being in a state of frequency ω_{phon}, in the mode μ (longitudinal or transverse of the first or second kind), in the medium of density ρ and of the total volume V, namely

$$\left\langle\left|(n'|a^{+(\mu)}|n\right|^2\right\rangle = \frac{V}{(2\pi)^3} \cdot \frac{\hbar\omega_{\mathrm{phon}(\mu)}\left(\langle n_\mu\rangle + 1\right)}{2\rho\omega^2_{\mathrm{phon}(\mu)}}, \tag{4}$$

$$\left\langle\left|(n'|a^{(\mu)}|n\right|^2\right\rangle = \frac{V}{(2\pi)^3} \cdot \frac{\hbar\omega_{\mathrm{phon}(\mu)}\left(\langle n_\mu\rangle\right)}{2\rho\omega^2_{\mathrm{phon}(\mu)}}, \tag{5}$$

that is controlled by the Bose-Einstein distribution

$$\langle n_\mu\rangle = \frac{1}{\exp\left[\dfrac{\hbar\omega_{\mathrm{phon}(\mu)}}{kT} - 1\right]}. \tag{6}$$

Now, in this formalism, the spectral distribution function for Brillouin scattering can be expressed as follows [5],

$$S(\mathbf{K}, \omega') = \sum_\mu \left\{ \frac{\mathrm{const} \cdot \hbar\omega_{\mathrm{phon}(\mu)}\Gamma_\mu}{\omega^2_{\mathrm{phon}(\mu)}} \left[\frac{\langle n_\mu\rangle}{\left[\omega' - (\omega_0 + \omega_{\mathrm{phon}(\mu)})\right]^2 + \Gamma^2_\mu} + \frac{\langle n_\mu\rangle + 1}{\left[\omega' - (\omega_0 - \omega_{\mathrm{phon}(\mu)})\right]^2 + \Gamma^2_\mu} \right] \right\} \Bigg/ 2\sum_\mu \frac{\mathrm{const}}{\omega_{\mathrm{phon}(\mu)}} \hbar\omega_{\mathrm{phon}(\mu)} \left(2\langle n_\mu\rangle + 1\right), \tag{7}$$

where ω_0 is the incident light angular frequency. The above formulae contains quantum parameters. For example, the reciprocal of Γ_μ can be interpreted as the life-time of a phonon from the branch μ. However, it is known that above calculations began from a classical point of view, and were generalized by artificial substitutions. The next section reviews some definitions of the coherent state formalism because the a^+ and a operators and the number of quasi-particles $\langle n\rangle$ belong to such a theory.

3. Definition of coherent states

Coherent states are applied in the description of physical processes where bosons, especially phonons, are involved. The coherent state $|\alpha\rangle$ can be defined as a normalized eigen-state of the annihilation operator [1]. The equation describing this situation can be written as follows

$$a|\alpha = \alpha|\alpha\rangle, \tag{8}$$

where a is the annihilation operator and α is its eigen-value. The α symbol might be interpreted as a number of phonons participating in the light scattering process.

Consider any optional coherent state [1]

$$|\alpha\rangle = \exp\left[-\frac{1}{2}|\alpha|^2\right]\sum_{n=0}^{\infty}\frac{\alpha^n}{\sqrt{n!}}|n\rangle, \tag{9}$$

and calculate its expected value in a representation of the coherent state basis of the number of particles operator $N = a^+a$. Acting on both the left and right sides of the above equation by the $\langle n|$ state we obtain

$$\langle n|\alpha\rangle = \exp\left[-\frac{1}{2}|\alpha|^2\right]\frac{\alpha^n}{\sqrt{n!}}. \tag{10}$$

Then both sides can be raised to the second power to calculate the probability density

$$|\langle n|\alpha\rangle|^2 = \exp\left[-|\alpha|^2\right]\frac{(\alpha^2)n}{n!}. \tag{11}$$

The above formula is the Poisson distribution, where the $|\alpha|^2$ value measures the average number of states occupied by a coherent state $|\alpha\rangle$.

The physical sense of the above definitions can be clearly recognized by the fact that the average number of quasi-particles occupying any optional state, the $\langle n\rangle$ quantity, is governed by the well known Bose-Einstein distribution

$$\langle n\rangle = \frac{1}{\exp\left[\frac{\hbar\omega_{\text{phon}}}{kT}\right]-1}, \tag{12}$$

where ω_{phon} is the frequency of a state (phonon), and T is the temperature of the medium. In the next considerations, the classical calculations will be modified by the appropriate exchanges between the kT energy and the $\hbar\omega(\langle n\rangle + 1/2)$ quantum quantity.

4. Calculations of the scattering efficiency. The ratio of two inelastically scattered optical signals

In this section three important formulae will be provided. Let us define the ratio of the square of the electric field in the scattered light to the square of the electric field in the incident light (scattering efficiency) in the following form

$$A = \frac{\langle|\mathbf{E}'|^2\rangle}{\langle|\mathbf{E}_0|^2\rangle}. \tag{13}$$

Next, taking into account the above we can derive the ratio of the A_{quant} quantity calculated from the quantum point of view to the A_{class} calculated from the classical point of view, namely

$$\frac{A_{\text{quant}}}{A_{\text{class}}} = \frac{\hbar\omega\left(\langle n\rangle + \dfrac{1}{2}\right)}{kT}. \tag{14}$$

At room temperature this ratio is, as a very good approximation, equal to one.

In typical inelastic light scattering experiments of the Brillouin type it is possible to observe for specific experimental configurations, simultaneously, two optical signals resulting from scattering on the two transverse or quasi-transverse acoustic waves of hypersonic frequency. Very often the scattered signals are very close when the acoustic frequencies are close. The ratio of intensities from these signals contains information on the acoustic wave frequencies and the wave-vectors. We can describe in a similar way this quantity from both the quantum and classical points of view. The classical formula is as follows

$$\frac{I_1}{I_2} \approx \left(\frac{K_1\omega_2}{K_2\omega_1}\right)^2, \tag{15}$$

where the ratio is proportional to the square of frequencies and to the wave-vectors ratio of the two acoustic waves. The same formula calculated from the quantum point of view is equal to

$$\begin{aligned}\frac{I_1}{I_2} &\approx \left(\frac{K_1}{K_2}\right)^2 \cdot \frac{\omega_2}{\omega_1} \cdot \frac{\langle n_1\rangle + \dfrac{1}{2}}{\langle n_2\rangle + \dfrac{1}{2}} \\ &= \left(\frac{K_1}{K_2}\right)^2 \cdot \frac{\omega_2}{\omega_1} \cdot \frac{\exp\left[\dfrac{\hbar\omega_1}{kT}\right] + 1}{\exp\left[\dfrac{\hbar\omega_1}{kT}\right] - 1} \cdot \frac{\exp\left[\dfrac{\hbar\omega_2}{kT}\right] - 1}{\exp\left[\dfrac{\hbar\omega_2}{kT}\right] + 1},\end{aligned} \tag{16}$$

where the dependence on temperature is clearly evident.

5. Example of a theoretical and experimental comparison

As an example lets us consider experimental data in that we deal with the two acoustic wave frequencies measured by the Brillouin light scattering method [6–9] (Fig. 1) in the $LiNbO_3$ crystal. The values of the ratio of intensities from experiment compared with those calculated from Eqs. (15) and (16), are given in Table 1. Experiment and calculations have been made for different angles between light wave vectors and are valid for room temperature.

However, other theoretical results based on the theory of elasticity and the classical elasto-optic interactions provide for the case where the angle between light wave vectors was equal to 90° an additional and unfortunately quite different value 0.14 [6].

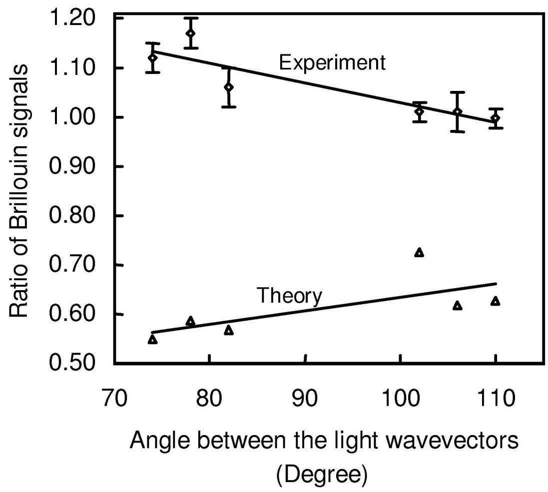

Fig. 1. The ratio of the Brillouin spectrum signals from the $LiNbO_3$ crystal. The equipment used in the measurements included a single-mode argon ion laser working at 514.5 nm wavelength with a power equal to about 100 mW and using the single-pass Fabry–Perot interferometer. For the low-level intensity light, the detection by the photon counting method was used.

Table 1. Comparison of the measured values of ratio of signals with theoretical calculations in the $LiNbO_3$ crystal.

Angle (deg.)	First wave frequency (GHz)	Second wave frequency (GHz)	Ratio of signals Experiment	Ratio of signals Theory
74	28.08 ± 0.23	20.81 ± 0.17	1.12 ± 0.02	0.55
78	28.66 ± 0.21	21.96 ± 0.19	1.17 ± 0.02	0.59
82	29.96 ± 0.30	22.58 ± 0.27	1.06 ± 0.04	0.57
102	27.47 ± 0.21	23.40 ± 0.19	1.01 ± 0.01	0.73
106	28.32 ± 0.24	22.25 ± 0.19	1.01 ± 0.03	0.62
110	29.30 ± 0.52	23.20 ± 1.11	1.00 ± 0.01	0.63

This kind of calculations are based on the so-called scattering efficiency R, which measures the cross section divided by the total volume V of a scattered medium, namely [7−9]

$$R = \frac{1}{V} \cdot \frac{d\sigma}{d\Omega}\,. \tag{17}$$

Calculating the ratio of two values of R for two cases of frequencies discussed above, respectively, one obtains [6]

$$R_{1/2} = \frac{\pi^2 k T n_0^8}{2\lambda^4 X_{1/2}} \left[p_{41}\gamma_1 - \left(p_{66} - \frac{r_{22}e_{16}}{\varepsilon_{11}} \right) \gamma_2 - \left(p_{14} - \frac{r_{22}e_{15}}{\varepsilon_{11}} \right) \gamma_3 \right]^2, \tag{18}$$

that is equivalent to the ratios obtained from Eqs. (15) and (16). The meaning of the symbols in the above formula is as follows: k is the Boltzmann constant, T is the temperature of the crystal, λ is the light wavelength, n_0 is the refractive index of the medium, p_{ik}, r_{ik}, and e_{ik} are elements of the elastooptic Pockels tensor, the electrooptic tensor, and the piezoelectric tensor, respectively. $X_{1/2}$ are the eigen-values and γ_i is the eigen-vector element (it describes the state of polarization of the acoustic wave) of a so-called "characteristic matrix" defined as $Q_{ik} = c^{ef}_{ijkl}\chi_j\chi_l$, where the c^{ef}_{ijkl} are elastic constants of the medium and where the unit vector χ informs of the direction of propagation of the acoustic wave. The eigen-values $X_{1/2}$, divided by the density of the medium inform us of the squared velocities of the two quasi-transverse waves.

The disagreement obtained between the experimental data and theory for the cross section can not be explained in the frame of the presented results.

6. Conclusions

In the article chosen theories of inelastic light scattering has been compared with the Brillouin scattering results. In some aspects they are of quantum character, in other moments they are almost classical. By an appropriate substitution, the theory was modified by the use of the average number of phonons (bosons) $\langle n \rangle$ expressed by the Bose–Einstein distribution. However the applied methods do not provide a satisfactory agreement between the experimental results and theoretical predictions, especially if we take into account the relative intensity of the two optical signals inelastically scattered on acoustic waves. The quantity $\langle n \rangle$ is only in one case concerned with the more general theory of coherent states.

Acknowledgement

The author gratefully acknowledges the Polish State Committee for Scientific Research for support under grant No 2 P03B 038 16.

References

[1] R.J. Glauber, *Coherent and incoherent states of the radiation field*, Phys. Rev., **131**, 6, 2766–2788 (1963).

[2] J. Perina, *Quantum statistics of linear and nonlinear optical phenomena*, D. Reidel Publishing Comp., Dordrecht – Boston – Lancaster 1984.

[3] J.R. Klauder and B. Skagerstam, *Coherent states, applications in physics and mathematical physics*, World Scientific Pub. Co. Pte. Ltd., Singapore 1985.

[4] A. Perelomov, *Generalized coherent states and their applications*, Springer-Verlag, Berlin Heildelberg 1986.

[5] G.B. Benedek and K. Fritsch, *Brillouin scattering in cubic crystals*, Phys. Rev., **149**, 2, 647–662 (1966).

[6] T. Błachowicz, *Relative intensity of Brillouin lines resulting from quasi-transverse hypersonic acoustic waves*, [in:] American Institute of Physics Conference Proceedings, R.M. Herman [Ed.], **467**, 341–342, Woodbury, New York 1999.

[7] T. Błachowicz, *Study of the elastic properties of the lithium tantalate crystal by the Brillouin laser light scattering*, Archives of Acoustics, **25**, 1, 23–34 (2000).

[8] T. Błachowicz, *Numerical calculations of the rotational contributions to a scattering coefficient in the piezoelectric* $LiTaO_3$ *crystal*, J. Opt. Soc. Am., **B15**, 10, 2599–2606 (1998).

[9] T. Błachowicz, *Inelastic light scattering on two degenerated hypersonic waves: the switching effect*, Acoust. Lett., **21**, 10, 203–207 (1998).