An uncertainty wave-vector diagram approach for Brillouin light scattering on bulk phonons

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1. Introduction

Brillouin scattering measures changes of photon frequencies scattered in the annihilation or creation processes by phonons lying at the beginning of the first Brillouin zone. The method can be applied for small samples of arbitrary dimension. Experimentally observed optical signals in Brillouin scattering experiments possess a characteristic pattern, where we see a set of strong lines from elastic scattering, the Rayleigh lines, and very weak Brillouin peaks resulting from inelastic creation (Stokes lines) and annihilation (anti-Stokes lines) of acoustic phonons by photons.[1-4]

Observed recently anomaly in Brillouin scattering experiments for the SLAO oxide type crystal, which shows very high background, but which does not suppress strong stokes and anti-stokes signals. It can be explained by the uncertainty wave-vector diagram approach similar to the fuzzy diagram approach introduced by A. Korpel[5] in strong acousto-optical interactions. The presented approach uses wave vectors of incident light, scattered light, and acoustic waves, which express the quasi-momentum conservation principle, and the new uncertainty wave vector resulting from multi-phonon processes.

2. Experiment

The equipment used in measurements included: a single-mode argon-ion laser, a single-pass pressure-scanned Fabry-Perot interferometer,[6] and the single photon counting unit produced by Photon Inc. for low-level intensity light detection. All the measurements have been carried out for configurations where the angle between incident and scattered light was equal to $\pi/2$. The polarization of incident light and scattered light were perpendicular. The signals from the photomultiplier along with pressure were detected at one second intervals. The typical duration of measurement was one hour. A time resolution equal to 1 second generates a frequency error, of measured Brillouin signals. equal to about 0.04 GHz, when one full spectral range (FSR) equal to 37.50 GHz, is scanned during 1 hour.[6]

The obtained results enabled us to draw some hypotheses presented in conclusions.

3. An uncertainty wave-vector diagram approach

It is well known that the wave-vector diagram plays an important role in the explanation of photon-phonon interaction of different types. It explains the Bragg-angle condition and the Doppler shift of scattered light in typical acousto-optic measurements.[7] It describes also the geometry of interaction in Brillouin light scattering experiments, where three wave vectors express quasi-momentum conservation principle.[8]

Because of the absorption origin of the phenomenon – the effect strongly depends on the light wave-length applied, with a minimum for the dark blue region - the introduction of uncertainty in the wave vectors of light (Fig. 1a) similar to Korpel's fuzzy-diagram approach for strong acousto-optical interaction can explain observed high level background.[5, 7]

The model introduces an energy conversion coefficient dependent on the light wavelength applied. Light energy passing through a sample can be converted into other types of excitations. This conversion phenomenon is detected by a lens, what means that optical

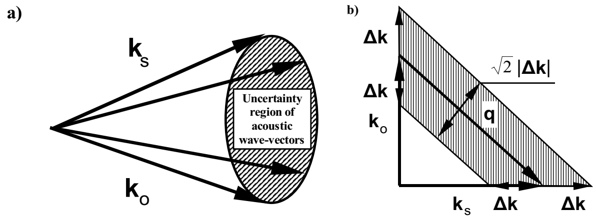

Fig. 1. Uncertainty wave-diagram approach which introduces uncertainties in wave-vectors of scattering process (a), and the model of scattering process constructed from wave vectors of incident light k_0, scattered light k_s, and acoustic wave q, which expresses the quasi-momentum conservation principle, and the new uncertainty wave vector $\mathbf{\Delta k}$ resulting from absorption and conversion of light energy into acoustic GHz modes – the 90^o scattering geometry (b).

information is detected from a region of the lens focal spot, on the order of micrometers. Light conversion is governed by a familiar formula of the *exp(-x/a)* type, where a is the energy absorption (conversion) coefficient. It allows as to introduce an uncertainty wave vector associated with incident and scattered light in the following way

$$|\mathbf{\Delta k}| = \frac{2\pi}{a(\lambda)}, \tag{1}$$

and to modify the classical wave diagram by the uncertainty wave-vector diagram (Fig. 1b). In this way, the model introduces uncertainty in the observed acoustic frequencies Δf by the obvious relation

$$\sqrt{2}|\mathbf{\Delta k}| = \frac{2\pi\,\Delta f}{\mathrm{v}}, \tag{2}$$

where v is the speed of acoustic wave. Simple calculation shows, under assumption that absorption takes place at region comparable with light wave-length (a parameter), that the Δf can be estimated from the following formula

$$\Delta f = \sqrt{2}\,\frac{\mathrm{v}}{a}, \tag{3}$$

which gives, for example, a value equal to about 12 GHz for the observed frequency of the acoustic wave equal to 23.1 GHz for the experiment with the 488 nm wave-length.

Because of the uncertainty background for the phenomenon, the following equation can be introduced

$$|\mathbf{\Delta k}| \sim h, \tag{4}$$

where the h should be associated with the observed depth of the background. As a consequence, a ratio of background amplitudes should be expressed as a ratio of appropriate coefficients in the following way

$$\frac{h_1(\lambda_1)}{h_2(\lambda_2)} = \frac{a_2(\lambda_2)}{a_1(\lambda_1)}, \tag{5}$$

In the current state of our knowledge, it is difficult to define precisely the physical sense of the *a(λ)* coefficient. As will be shown below more easily to interpretation are values of uncertainty wave-vectors.

Some insight into presented phenomena is obtained from results of measurements done with the full spectral range (FSR) of an interferometer equal to 75 GHz (Fig. 2). The measurements done with the laser power equal to 135 mW for every laser line, show new surprising features. Despite the clear Brillouin peaks not masked by the background, the 457.9 nm case manifest itself in a symmetrical and wide curvilinear background superimposed on typical Brillouin spectrum. Now the idea of uncertainty wave-vector diagram introduced intuitively on Fig. 1 can be explained physically by the two-phonon scattering processes. Examples of such processes

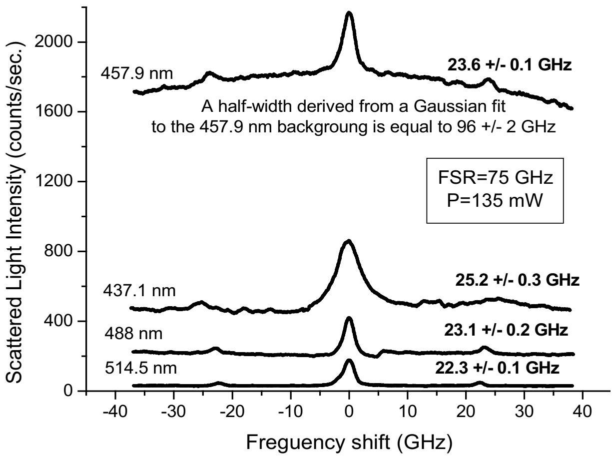

Fig. 2. Brillouin spectrum for different laser wavelengths, for the FSR equal to 75 GHz.

are annihilation of two phonons, creation of two phonons, and creation-annihilation phenomenon. In every case when photons can exchange energy with two and more phonons, then energy conservation and quasi-momentum principles do not restrict additional degrees of freedom of a process. This is why we deal with continuous energy distribution superimposed on single-phonon interactions. In other words in a multi-phonon process the length of phonon wave-vector can not be controlled fully by the experiment geometry (Fig. 3).

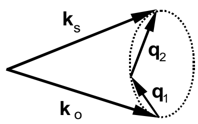

Fig. 3. The physical origin of the uncertainty wave-vectors can be explained through multi-phonon processes – the present diagram shows the scattering with the two acoustic phonons.

Derived from a Gaussian fit, the width of the dark-blue background on the Fig. 2, is equal to 96 GHz. It allows us to use again formula (3) to calculate directly values of the *a* parameter. Table 1 provides the values calculated from Eq. 5 and derived from observed Brillouin spectra.

Tab. 1. Values of the *a* parameter, the uncertainty wave-vectors, uncertainty wave-vectors related to incident light wave-vectors, and uncertainties in frequency, for different light wavelengths; 437.1 nm, 457.9 nm, 488 nm, 514.5 nm.

λ(nm)	*a* (nm)	$\lvert\mathbf{\Delta k}\rvert$ (m^{-1})	$\lvert\mathbf{\Delta k}\rvert / \lvert\mathbf{k}\rvert$	Δf (GHz)
437.1	217	$2.9 \cdot 10^7$	1	25
457.9	58	$1.1 \cdot 10^8$	4.1	96
488.0	484	$1.3 \cdot 10^7$	0.5	12
514.5	2823	$2.2 \cdot 10^6$	0.09	2

The obtained values of the uncertainty wave-vectors are formally reasonable. Especially, the value of the 488 nm wave-length is comparable to the *a* parameter, which means that the uncertainty wave-vector is comparable with the light wave vector.

4. Conclusions

The whole phenomenon represents an uncertainty-type principle associated with photon-phonon interactions. Significantly, similar measurements done with the SLGO crystal ($SrLaGaO_4$) of the same tetragonal (4/mmm) symmetry as the SLAO ($SrLaAlO_4$) crystal did not give these unexpected results. In both cases, measurements were done for the same scattering configuration where all wave-vectors have been placed in the (110) crystallographic plane, and the other experimental conditions were the same.

The fuzzy wave-vector diagram approach is strictly related to acousto-optic interactions between light and artificial ultrasonic field. The uncertainty wave-vector diagram is related to the above approach in the general geometrical meaning. Presented model allows us to estimate the order of uncertainty in frequencies measured in Brillouin scattering experiments. The approach gives qualitative insight into phenomenon, but it is probable that phenomenon related to the 2-phonons scattering is responsible for observed large background noise.

The value of the energy conversion parameter measured for one Brillouin spectrum and calculated for others applied light-colors shows this aspect of the presented phenomenon where physical conservation principles do not restrict internal conversions of different types of energies. Similarities to the presented phenomenon have been observed in rutile, where a background with narrow (1.1 GHz) and broad (330 GHz) components were measured.[9] In the paper it was shown that backgrounds result from two-phonon difference Raman processes. The narrow component resulted from a single acoustic phonon branch could not scatter light with a frequency shift larger that the Brillouin component so cannot account for the broad component. On the other hand the broad component was explained by two-phonon difference Raman processes from different phonon branches near the zone boundary. It seems that the provided model is able to explain current results, especially in the interpretation of the half-width of background.

From the frequency domain point of view, the absorption-like channel in a scattering process should be interpreted as a light-energy conversion into GHz acoustic modes forming the background where some specific values of frequencies are seen as Rayleigh peaks and others as Brillouin components. The energy distribution between background and elastic/inelastic channels is a function of the light wavelength. All of the above results appear at the strong absorption origin of the phenomenon.

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