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Accuracy of Brillouin frequencies for material characterization by light scattering

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ABSTRACT

We describe the method that allows us to give an estimation of the uncertainty on the frequency peak by Brillouin Light Scattering. It is the speed of phonons in a material excited by a visible green 532 nm wavelength laser. We follow the Guide to the Expression of Uncertainty in Measurement to obtain a first estimation of the speed of the phonons at $\pm 5\%$ at 2σ .

KEYWORDS

Brillouin Light Scattering; Brillouin spectroscopy; uncertainty; uncertainty analysis.

1. INTRODUCTION

Brillouin scattering is the inelastic scattering of light by acoustic waves from a medium. This effect was demonstrated by Léon Brillouin one hundred years ago [1]. This effect is used to understand the properties of materials [2]. The laser beam with high power may experience Brillouin scattering due to acoustic vibrations in the medium, usually in the opposite direction of the incident beam, a phenomenon known as stimulated Brillouin scattering. We use a commercial bench based on the J. R. Sandercock multipass tandem interferometer design. What interests us more particularly is to gain confidence to know if the precision given by the bench can lead to a more precise determination of the order of magnitude of the uncertainty which is associated with the result obtained in terms of deviation. to the optical carrier. We are interested in the determination of the uncertainty based on a modern approach by following the recommendations of the "Guide to the expression of uncertainty in measurement" (GUM) established by the "International Bureau of Weights and Measures" (BIPM). We propose to present a poster at the conference to give indications on the expected values of the uncertainty terms [3] which one can give on a result of frequency shift corresponding to the determination of a speed of propagation of phononic waves through or on the surface of the material to be characterized [4, 5].

We have already started to think about the level of uncertainty given when performing Brillouin scattering measurements. This has been the subject of some previous work [6, 7]. What came out mainly

from this work? It must be said that we started to estimate the uncertainty by looking at the measurement results obtained on different materials.

Estimating the uncertainty requires to know contributions of the various fixed parameters such as optical indexes, wavelengths, diffusion angles, density of the material, and longitudinal and shear modulus. Some other parameters like fluctuation of the source, mechanical stability of the setup, and environmental parameters in the room, can also have their contribution to enlarge the uncertainty.

For this uncertainty estimation, we use a similar method, like in optics and microwaves [7 – 13] based on the requirement delivered by the guide "Evaluation of measurement data – Guide to the expression of uncertainty in measurement (GUM)" [14].

2. WHAT WE EXPECT TO PERFORM?

To start with, we will remind you how the Brillouin diffusion bench works. We will recall the principles. Next, we will describe how these principles are implemented. It is important to describe the path of the optical beam through the bench and to understand what important information we are trying to obtain.

To understand how the bench works, it is better to have a look at some reference papers on the subject [15 – 17] and also inspired by works on uncertainty estimations [18].

First, we give an outline, before describing everything in a little more detail. We have a sample to measure, to characterize. What makes us interested? What are we looking for as information? It is a question of being able to determine the speed of propagation of phononic waves inside or at the surface of a given material. To be able to access this information on this speed, it is necessary to use a fairly powerful laser beam which will interact with the material. This laser beam will generate a phononic propagation wave in or on the surface of the material, and this wave will recreate an optical wave when it leaves the material. This will be an optical signal beam at one wavelength offset from the wavelength of the incident laser beam. This optical frequency shift precisely contains the information on the speed of propagation of phononic waves in or on the surface of the material illuminated by the incident laser signal. We have therefore given the broad outlines of the general principle which makes it possible to go back to the information on the speed of propagation. One notes that these speeds can perfectly be different according to the direction of propagation, because there are different modes involved, like modes compared to shearing, or compression, for example. At this stage of the explanation, it is not necessary to go into this level of detail. We remain on the principle that a signal is refracted and that it will have to be analyzed in relation to the incident signal. So we see different aspects emerging. We will need to recover this refracted signal, then compare it with the incident signal. The laser emitted signal is sent to the analysis bench, while most of the signal power is used to interrogate the material to be characterized so that it reacts by emitting a refracted signal, shifted in optical frequency.

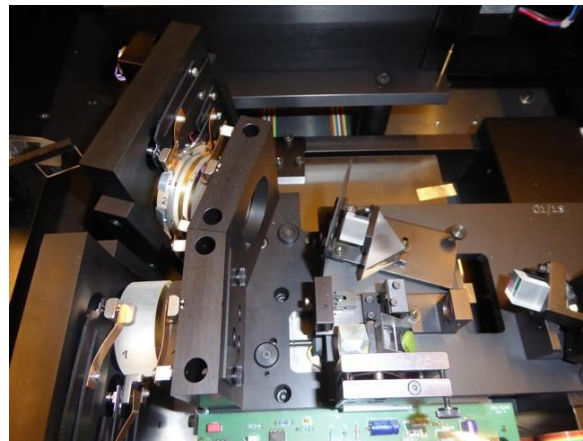


Fig. 1. View of the double Fabry-Pérot interferometer.

The refracted signal is filtered after focusing it on the analysis bench and compare it to the signal emitted by the laser to finally arrive at plotting a frequency curve which represents the offset of the refracted signal with respect to the initial optical carrier which is the signal emitted by the laser. Fig. 1 shows the double Fabry-Pérot interferometer. We already discuss how the bench works [6, 19]. Tandem Fabry-Pérot interferometer produces peaks shifted from the frequency of the laser to characteristic frequencies depending on the material. Figure 1 shows the typical set-up used for the measurement. Brillouin Light Scattering (BLS) using a 532 nm powerful Class 4 laser up to 600 mW is efficient to reveal spin wave or acoustic signals, at frequencies from few Giga Hertz to more than a hundred of Giga Hertz. Fluctuations of refractive index in a medium enables the detection and analysis of laser light scattered, thanks to BLS setup. Once we have fully understood the operation of the Brillouin diffusion bench, it is time to ask ourselves the question of the precision and the truth of the data that we extract from this bench. What is really important? What will have an influence on the data we are trying to extract? When we draw the curve, we notice that there is data that we must enter into the program. Among these data, there is the spacing of the mirrors which form the cavity where several round trips of the optical signal take place, to be analyzed. We use a commercial bench developed by the company "The time Stable". The light goes through six passages through two interferometers. Each pair of mirrors is very precisely aligned during the calibration procedure. The data of the spacing between the mirrors is quite important since it conditions the value of the frequency peaks that will be obtained. And these peaks will then make it possible to determine the desired propagation speed. This parameter of spacing between the parallel plates seems to be important since a bad evaluation or an oversight can falsify the result. You have to trust the bench for this value to be the right one. One method to validate what the bench gives as a value was to find the frequency peaks on known materials. This was done during the installation phase of the measurement bench. This serves as a sort of bench calibration. It is easy to check this type of parameter by measuring the peaks on an isotropic material, Poly(methyl methacrylate) known as PMMA, for example. It is necessary to calibrate accurately the instrument. It is sensitive to mechanical vibrations, temperature and hygrometry. Alignment process requires an alignment of the two cavities. The two cavities each consist of a pair of parallel mirrors. Tandem interferometer produces two series of absorption peaks with respect to a flat noisy intensity level. It makes possible to obtain a curve providing the number of absorbed photons versus frequency.

3. ESTIMATION OF THE UNCERTAINTY

Once we take the measurement on an unknown material, but the calibration by comparison is done, we can say that we have some confidence in the results obtained. The question which then arises is to determine what will really count for the uncertainty on the value of the peaks for example. The formal method consists in determining the law of diffusion on the data obtained in relation to the data entered into the "system". This is based on how to calculate the uncertainty based on the BIPM recommendation. The phase velocity of the transversal or longitudinal waves linearly depends on u_B (the Brillouin frequency), n (the optical index), λ (the wavelength), Q (the diffusion angle). The fact of knowing which law or which equation gives the results can lead us for example to have terms with a coefficient of correction, or raised to a power, to the square, or to the cube for example. Coefficients can therefore appear in front of each elementary uncertainty term, associated with each different phenomenon. This is something that uncertainty calculators are familiar with, in laboratories accredited for calibrations or tests, for example. We have the same type of approach here, even if there is no legal obligation as in the case of accredited laboratories. This approach makes it possible to really question the validity of the results obtained, and therefore, to ask the question of the weight of each of the contributions of various parameters. We then realize that some will be negligible, while others will be preponderant. Important parameters relate, for example, to the misalignment of the incident optical beam. But we can see that if nothing works, we will not get any result at all. The same applies to the focusing of the beam. After a few meters, the size of the diameter of the beam has widened and it is necessary to refocus it. This is entirely appropriate since the path of the photons is carried out over a few meters in this type of experiment.

We will now recall that the calculation of the uncertainty is based on being able to list and then estimate the value of various terms of elementary uncertainties. We lead our estimation by following the recommendations of the GUM.

We have the uncertainty terms of the statistical type which are grouped in the so-called category A. It is those, which are evaluated by statistical methods such as reproducibility, repeatability. We can point out here the variations of results versus time, or with various operators. There is also a question of finding a good compromise between measuring fast enough (from few minutes to few hours) and increasing the resolution by having enough samples (we generally choose to have 1000 photons).

The other terms are in the category of type B, determined by other means. This category is divided into two sub-groups of uncertainty terms. The first sub-group concerns the so-called connection terms to national standards. The second subgroup includes all the other terms. They can be terms of uncertainties estimated by various methods, including sometimes simply based on documentations, which are in the datasheets of the devices used. In this case, we are obliged to trust the values given by instrument manufacturers. Terms of this category are called B-type and depend on various components and temperature control. Experience with or general knowledge of the behavior and properties of relevant materials and instruments determines them. When there is no traceability or calibration certificates, we refer to manufacturer's specifications, data provided in calibration and other certificates, or uncertainties assigned to reference data taken from handbooks. Quantities, which may be considered as approximations to the corresponding variances, should be characterized in the components in B-type category.

Second family of uncertainties contributions is for those assessed by other means. We find here the contribution of the laser, mainly with its uncertainty on the wavelength. We also find here that the uncertainty relative to the beam diameter of the laser is given by 1.7 ± 0.2 mm. The pointing stability of the laser is less than $2 \mu\text{rad}/^\circ\text{C}$. The beam angle is less than 1 mrad. Other parameters have their contribution such as the specifications of probes i. e. photodetectors, and the influence of the supply voltage alimentation that are well stabilized, the temperature varying less than 0.1°C during measurement, despite it can vary from 0.5 degrees during a whole day.

At that point the estimated uncertainty at a two sigma is still rather large, being in the range of 0.2 GHz for a signal higher than 10 GHz and larger than 0.1 GHz for a signal in the range 2 – 10 GHz. It corresponds to an uncertainty of 2% for the speed of the phonons when the microwave signal is higher than 10 GHz and even only 5% for speed when the microwave signal detected is between 2 and 10 GHz.

Note that the individual contribution of each elementary terms strongly depends on the shape of their diffusion law. For the terms of types A, we logically have shapes according to the Gaussian law. However, this is not always true for other terms. For example, among the terms of type B, some will follow the normal distribution, others the triangular distribution.

Reflexion about BLS is a topic of interest. We can for instance cite a paper about dispersion of the surface phonons studied by BLS [20], recent works in magnetostatic [21] or on optical resonators [22].

5. CONCLUSION

To conclude this paper, we can write that the estimation of the uncertainty on the frequency peaks corresponding to the response of the material to laser illumination must be carried out by applying the recommendations of the *Bureau International des Poids et Mesures* (BIPM), keeping that general principle to find an individual contribution for each parameter. We can ask ourselves if it is relevant, and we can possibly eliminate the contributions if they are negligible. The estimated uncertainty is estimated to be better than $\pm 5\%$ at 2σ . This estimation should be optimized.

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