

Uncertainty on Brillouin scattering measurements on bulk materials using a power laser

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ABSTRACT

Through this paper, we describe the method leading to the estimation of the uncertainty. We aim to give an estimation of the uncertainty on the frequency peak by Brillouin Scattering Stimulation. It corresponds to the speed of phonons inside a material excited by a 532 nm wavelength laser. The guideline follows the Guide to the Expression of Uncertainty in Measurement and its estimation is of 0.26% on the Brillouin frequency peak at 15.70 GHz for polymethyl methacrylate (PMMA).

KEYWORDS

Brillouin Scattering Stimulation, Brillouin spectroscopy, uncertainty, uncertainty analysis, tandem Fabry–Pérot interferometer, uncertainty analysis

1. INTRODUCTION

Brillouin light scattering (BLS) is gradually gaining popularity in various industrial applications and in laboratories. This work is about BLS and focus on the discussion about the associated uncertainty. BLS is the inelastic scattering of light through sound waves. BLS is then a good way to study the elastic properties of materials. It is a non-contact, non-destructive method, and relatively easy to implement with appropriate means. We propose here to come back to the main steps, which allow having specific instrumentation in order to estimate the speed of propagation of phononic waves in materials. Of course, we realize that there are necessary aspects of theoretical knowledge. It is not a question here of going back to all the theories in order to understand the matter. This would require going too deeply in a systematical description. To better frame the subject that we are discussing here, we remind about the main contributions to detect sound waves, by brothers Curie [1], and inelastic light scattering in materials thanks to their excitation by sound waves with Brillouin [2]. The whole instrumental aspect has importance as modern benches for Brillouin diffusion need the appropriate technology. Among other necessities, there are interferometry technologies. In addition, such a Brillouin scattering bench needs conventional optical components, and primarily a sufficiently powerful laser. Stimulated Brillouin scattering is a nonlinear process.

It can also occur in optical fibers. It manifests itself concretely through the creation of a backward propagating Stokes wave, which carries most of the input power, once the one reaches Brillouin threshold [3]. The magnetic properties of materials are via their magnetic excitations - magnons, thanks to Brillouin scattering. As with phonons, magnons concern surface and bulk excitations [4]. Indeed, Brillouin inelastic light scattering spectroscopy is widely used for the study of phonons but also magnons in materials. This technique has become an essential tool [5, 6]. It is of course complementary to inelastic light scattering Raman spectroscopy [7]. Kojima shows that those techniques have become essential for studying materials science [8]. Grimsditch and Ramdas have made precise measurements with Brillouin scattering in the early seventies on Diamond [9]. It is also useful to recall the differences between Brillouin scattering and another well-known technique, Raman spectroscopy. The latter type of spectroscopy is used to determine the chemical composition and molecular structure of the transmission medium, while Brillouin scattering can be used to measure the elastic behavior of a material. A more systematic method was implemented based on a tandem Fabry–Perot interferometer by Sandercock [10] in 1970 and then by Lindsay, Anderson and Sandercock [11], and Dil et al [12]. Hillebrands [13] and Scarponi et al [14] improved corresponding instrumentation techniques. Our objective is to assess with a consistent metrological approach the uncertainties of BLS. This paper provides an experimental part and the determination of the uncertainties associated with the determination of peaks corresponding to the shift between the frequency of the signal refracted by a material and to the laser serving as an interrogation signal. The knowledge of this value, as well as the parameters of the studied materials, can also provide the value of the speed of the corresponding phononic waves, when intrinsic characteristic data of the evaluated materials are known. To lead the discussion on the uncertainty associated with the BLS, we rely on the standards of metrology.

2. WHAT WE EXPECT TO PERFORM?

The analysis mainly consists in a method of detection of the refracted light emitted by a material under test, i. e. the Device under Test (DUT). This material can be isotropic or anisotropic. One of the key of the measure is the Tandem Fabry-Perot double interferometer. Detected peaks are shifted from the wavelength of the laser. Those offset frequencies depend on the properties of the material of the DUT. This paper aims to lead an estimation of the uncertainty obtained on the frequency shift that can lead later to parameters of the material like phase velocity of transverse and longitudinal waves, deduced from BLS. Estimating the uncertainty requires knowing the contribution of the different fixed parameters like the optical index, the wavelength, the diffusion angle, the density of the material, and the longitudinal and shear modulus, but especially fluctuation of the source, mechanical stability of the setup, and environmental parameters in the room. For this uncertainty estimation, we use a similar method like in optics and microwaves based on the requirement delivered by the Bureau International des Poids et Mesures (BIPM) in the guide "Evaluation of measurement data – Guide to the expression of uncertainty in measurement (GUM)" [15]. We will focus in this part on the principle of 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 [11, 12]. The general principle is to send the signal generated by the laser focusing it on the part of the sample that we want to characterize. The photons arrive in the material or in the thin layer and interact with the lattice or more generally with the material.

Light helps to create phonons. These phonons propagate with speeds that may be different depending on whether the mode is transverse or longitudinal. It depends on the nature of the material, as it can be isotropic or anisotropic. The phonons in turn create light, which are shifted in frequency relatively to the wavelength of the laser. The BLS precisely consists in analysis of the refracted light emitted by a material [11, 13]. Tandem Fabry–Perot interferometer produces peaks shifted from the frequency of the laser to characteristic frequencies depending on the material. Figure 1 gives the typical set-up used for the measurement, showing the typical setup (a) and a picture of the system (b). We calibrated the bench with part of the laser signal, used as the bench reference. Inside the commercial bench developed by the Swiss company "The time Stable", the light goes with six passages through two different interferometers. Each pair of mirrors is very precisely aligned during the calibration procedure.

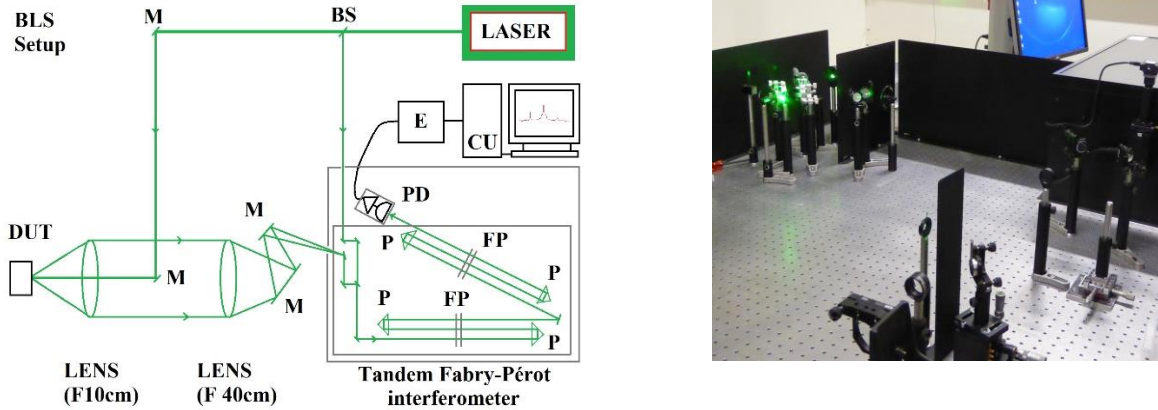


Fig. 1. (a): Typical setup for BLS. JRS TFP2 is a commercial Tandem Fabry-Pérot interferometer. BLS: Brillouin Light Scattering. DUT: device under test. M: mirror. FP: Fabry-Pérot. P: prism. PD: photodetector. E: electronics. CU: computer unit. (b): The commercial Tandem Fabry-Pérot interferometer is inside the box on the right side of this picture.

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. Each of the two cavities consists in a pair of parallel mirrors. Tandem interferometer produces two series of absorption peaks with respect to a flat noisy intensity level. We then obtain a curve providing the number of absorbed photons versus frequency. We have measured the ν_B for PMMA as an example of an isotropic material. The measured Brillouin frequency shift is $\nu_B = 15.70$ GHz (longitudinal acoustic mode), with a Brillouin linewidth of 324 MHz. The measured spectrum for PMMA (backward scattering) is given in Figure 3. Based on the measured frequency shift ν_B , the phase velocity of longitudinal waves in the test material can be derived as,

$$\nu_B = \frac{2nv}{\lambda_0} \sin\left(\frac{\theta}{2}\right)$$

With $\lambda=532\text{nm}$, $n=1.4953$, $c_{11}=9$ GPa, $r=1.19 \times 10^3 \text{ kg/m}^3$, $Q=180^\circ$, $v_L=(c_{11}/r)^{-1/2}=2792.9$ m/s, we can check that $\nu_B=15.70$ GHz, the results given by the system is adequate.

3. UNCERTAINTY ESTIMATION

In this section, we aim to lead an estimation of the uncertainty on the frequency shift induced by BLS. Before going into more details in how we may estimate the uncertainty, it is useful to think about the approach in its determination. In the scientific community, it is important to underline that a debate exists as to whether there is a true value. Thomas von Clarmann et al offer the benefit of a critical discussion on the error concept versus the uncertainty concept [16]. Jong Wha Lee et al [17] compare the realist view of measurement and uncertainty versus the instrumentalist view of measurement when quantities are not natural attributes of the world that exist independently of the human perception. They show that a clear understanding of the two views is critical for understanding the guide "Evaluation of measurement data – Guide to the expression of uncertainty in measurement (GUM)" [15]. Estimating the uncertainty requires the knowledge of the contribution of the different fixed parameters, such as the optical index, the wavelength, the diffusion angle, the density of the material, and the longitudinal and shear modulus, but especially fluctuation of the source, mechanical stability of the setup, and environmental parameters in the room. From the equation given in the previous part, we see that the phase velocity of the transversal or longitudinal waves linearly depends on v_B (the Brillouin frequency), n (the optical index), λ (the wavelength), Q (the diffusion angle). The estimation of uncertainty follow the modern way of performing it [18]. For this uncertainty estimation, we use a similar method like in optics [19 – 21] and microwaves [22, 23] based on the requirement delivered by GUM. Frequency references of the 5 MHz or 10 MHz type possibly ensures the traceability of the BLS method to national standards [24, 25]. One contribution is due to the laser beam and the axis of displacement, which are not exactly parallel [26, 27]. Another contribution to this term is called Abbe error [28, 29]. We base all our estimation on results published in reference [30], which is more detailed.

4. CONCLUSION

The Brillouin spectroscopy is a non-intrusive measurement method for bulk materials and thin films. A scanning 6-pass TFPI has been described for BLS measurement. Following the GUM, and with reference [30], we have made detailed analysis and estimation of the uncertainties in the Brillouin frequency shift measurement, which is related to the speed of propagation of phononic waves in bulk materials. The expanded relative uncertainty in measured Brillouin frequency shift is estimated to be 0.26% (coverage factor $k=2$), which corresponds to an expanded uncertainty of 41 MHz for the measured frequency shift of 15.70 GHz in testing PMMA.

REFERENCES

- [1] Curie J., Curie P., "Développement par compression de l'électricité polaire dans les cristaux hémihédres à faces inclinées," Bulletin de Minéralogie 4-4, 90-93 (1880). DOI:10.3406/bulmi.1880.1564
- [2] Brillouin L., "Diffusion de la lumière et des rayons X par un corps transparent homogène. Influence de l'agitation thermique," Annales de Physique 17, 88–122 (1922). DOI:10.1051/anphys/192209170088
- [3] Govind P. Agrawal. Chapter 9 - Stimulated Brillouin scattering. Nonlinear Fiber Optics (Sixth edition) 2019, 355-399. DOI:10.1016/B978-0-12-817042-7.00016-6
- [4] Blachowicz T., Grimsditch M., "Scattering, Inelastic: Brillouin," Encyclopedia of Condensed Matter Physics 199-205 (2005). DOI:10.1016/B0-12-369401-9/00643-4
- [5] Mandelstam L., "Light scattering by inhomogeneous media," Zh. Russ. Fiz. Khim. Ova. 58, 381 (1926).
- [6] Kargar F., Balandin A.A., "Advances in Brillouin–Mandelstam light-scattering spectroscopy," Nat. Photon. 15, 720–731 (2021). DOI:10.1038/s41566-021-00836-5
- [7] Raman, C. V., Krishnan, K. S., "A new type of secondary radiation," Nature 121, 501–502 (1928). DOI:10.1038/121501C0

- [8] Kojima S., "100th Anniversary of Brillouin Scattering: Impact on Materials Science," *Materials* 15, 3518 (2022). DOI:10.3390/ma15103518
- [9] Grimsditch M. H., Ramdas A. K., "Brillouin scattering in diamond," *Phys. Rev. B* 11, 3139 (1975). DOI:10.1103/PhysRevB.11.3139
- [10] Sandercock J., "Brillouin scattering study of SbSI using a doubled-passed stabilised scanning interferometer," *Opt. Commun.* 2, 73–76 (1970). DOI:10.1016/0030-4018(70)90047-7
- [11] Lindsay S. M., Anderson M. W., Sandercock J. R., "Construction and alignment of a high performance multipass Vernier tandem Fabry–Perot interferometer," *Review of Scientific Instruments* 52(10), 1478-1486 (1981). DOI:10.1063/1.1136479
- [12] Dil J. G., van Hijningen N. C. J. A., van Dorst F., Aarts R. M., "Tandem multipass Fabry-Perot interferometer for Brillouin scattering," *Applied Optics* 20(8), 1374-1381 (1981). DOI:10.1364/AO.20.001374
- [13] Hillebrands B., "Progress in multipass tandem Fabry-Perot interferometry: I. A fully automated, easy to use, self-aligning spectrometer with increased stability and flexibility," *Review of Scientific Instruments* 70(3), 1589-1598 (1999). DOI:10.1063/1.1149637
- [14] Scarponi F., Mattana S., Corezzi S et al, "High-Performance Versatile Setup for Simultaneous Brillouin-Raman Microspectroscopy," *Physical Review X* 7, 031015 (2017). DOI:10.1103/PhysRevX.7.031015
- [15] GUM: Guide to the Expression of Uncertainty in Measurement, fundamental reference document, JCGM100:2008 (GUM 1995 minor corrections): <https://www.bipm.org/en/committees/jc/jcgm/publications>
- [16] Thomas von Clarmann, Steven Compennolle, Frank Hase, "Truth and uncertainty. A critical discussion of the error concept versus the uncertainty concept," *Atmospheric Measurement Techniques* 15, 1145-1157 (2022). DOI:10.5194/amt-15-1145-2022
- [17] Jong Wha Lee, Euijin Hwang, Raghu N. Kacker, "True value, error, and measurement uncertainty: two views," *Accreditation and Quality Assurance* 27, 235-242 (2022). DOI:10.1007/s00769-022-01508-9
- [18] Kacker R.; Sommer K. D.; Kessel R., "Evolution of modern approaches to express uncertainty in measurement. *Metrologia*," 44(6)513–529 (2007). DOI:10.1088/0026-1394/44/6/011
- [19] Salzenstein P., Pavlyuchenko E., Hmima A., Cholley N., Zarubin M., Galliou S., Chembo Y. K., Larger L., "Estimation of the uncertainty for a phase noise optoelectronic metrology system," *Physica Scripta T* 149, 014025 (2012). DOI:10.1088/0031-8949/2012/T149/014025
- [20] Pavlyuchenko E., Salzenstein P., "Application of modern method of calculating uncertainty to microwaves and opto-electronics," *Laser Optics, 2014 International Conference, Saint Petersburg, Russia, June 30 2014–July 4 2014*, 6886449 (2014). DOI:10.1109/LO.2014.6886449
- [21] Salzenstein P., Pavlyuchenko E., "Uncertainty Evaluation on a 10.52 GHz (5 dBm) Optoelectronic Oscillator Phase Noise Performance," *Micromachines* 12, 474 (2021). DOI:10.3390/mi12050474
- [22] Won-Kyu Lee, Dai-Hyuk Yu, Chang Yong Park, Jongchul Mun, "The uncertainty associated with the weighted mean frequency of a phase-stabilized signal with white phase noise," *Metrologia* 47(1), 24–32 (2010). DOI:10.1088/0026-1394/47/1/004
- [23] Salzenstein P., Wu T. Y., "Uncertainty analysis for a phase-detector based phase noise measurement system," *Measurement* 85, 118–123 (2016). DOI:10.1016/j.measurement.2016.02.026
- [24] Salzenstein P., Kuna A., Sojdr L. and Chauvin J., "Significant step in ultra high stability quartz crystal oscillators," *Electronics Letters* 46(21), 1433–1434, (2010). DOI:10.1049/el.2010.1828
- [25] Salzenstein P., Cholley N., Kuna A., Abbé P., Lardet-Vieudrin F., Sojdr L. and Chauvin J., "Distributed amplified ultra-stable signal quartz oscillator based," *Measurement* 45(7), 1937–1939 (2012). DOI:10.1016/j.measurement.2012.03.035
- [26] Howard L., Stone J., Fu J., "Real-time displacement measurements with a Fabry-Perot cavity and a diode laser," *Precision Engineering* 25(4), 321-335 (2001). DOI:10.1016/S0141-6359(01)00086-1
- [27] Joo K.-N., Ellis J. D., Spronck J. W., Munnig Schmidt R. H., "Design of a folded, multi-pass Fabry–Perot cavity for displacement metrology," *Measurement Science and Technology* 20, 107001 (2009). DOI:10.1088/0957-0233/20/10/107001

- [28] Köning R., Flügge J., Bosse H., "A method for the in situ determination of Abbe errors and their correction," *Measurement Science and Technology* 18, 476 (2007). DOI:10.1088/0957-0233/18/2/S21
- [29] Leach R., "Abbe Error/Offset," *CIRP Encyclopedia of Production Engineering* 1–4 (2014). DOI:10.1007/978-3-642-35950-7_16793-1
- [30] Salzenstein P., Wu T. Y., "Uncertainty estimation for the Brillouin frequency shift measurement using a scanning tandem Fabry-Pérot interferometer," *Micromachines* 14(7), 1429 (2023). DOI: 10.3390/mi14071429