

Time-domain Brillouin scattering in paraxial sound and light beams: Contra-intuitive features

Gusev VE(1)*

(1) Laboratoire d'Acoustique de l'Université du Mans (LAUM), UMR 6613, Institut d'Acoustique – Graduate School (IA-GS), CNRS, Le Mans Université, France

*Corresponding author's email: vitali.goussev@univ-lemans.fr

Background – Time-domain Brillouin scattering (TDBS) is an opto-acousto-optical technique for the evaluation of transparent materials. Via optoacoustic conversion, ultrashort pump laser pulses launch coherent acoustic pulses (CAPs) in the sample. Time-delayed ultrashort probe laser pulses monitor the propagation of the CAPs via the photo-elastic effect, which induces light scattering (Fig.1).

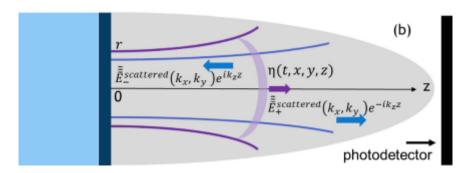


Fig. 1. Schematic presentation of co-focused probe light (in blue) and coherent sound (in violet) fields contributing to TDBS signal detection. Probe light field is due to laser pulses incident from a light transparent medium z > 0 on the sample surface z=0 and the laser pulses reflected from this surface. A CAP $\eta(t, x, y, z)$ is launched in the medium z>0 due the absorption in the sample z < 0 of the pump laser pulses (not presented) co-focused on the surface z=0 with the probe laser pulses. When the pump laser pulses are incident on the sample before the probe laser pulses, then the acousto-optic interaction between the CAP and the initial probe light field creates optical polarization sources of scattered light. TDBS signal results from heterodyne detection of the initially reflected and acoustically scattered probe light collected by the photodetector

A photodetector collects both the acoustically scattered light and the probe light reflected by the sample structure for the heterodyning and reveals the signal periodically oscillating in time, the so-called Brillouin oscillation (BO). The scattered probe light carries information on the acoustical, optical and acousto-optical parameters of the material for the current position of the picosecond CAP. Thus, among other applications, time-domain Brillouin scattering is a technique for three-dimensional imaging at nanoscale [1]. One of the fundamental applications of the TDBS is the evaluation of the absorption of phonons in the GHz–THz frequency range. This is achieved by measuring the decay in time of the BO amplitude. The pioneer research paper [2] contains the estimates of the other physical factors that could cause the attenuation of the BO and should be properly taken into account to reveal acoustic absorption. Among them is diffraction of both CAPs and the probe laser pulses. Particularly, the theoretical analysis in [2] confirms, that the characteristic spatial scale for the influence of the diffraction phenomena is the Rayleigh range, also sometimes called the diffraction length. However, the theory in [2] is for equal radii of the acoustic and light beams, while the acoustic wavelength in the considered backward Brillouin scattering is tightly related to the probe light wavelength in the medium, being twice shorter.



These circumstances make it impossible to disentangle the roles of the acoustical and optical diffraction in the decay of the BO amplitude predicted in [2].

Theoretical method and results – Analytical theory of the TDBS for the diffracting collinear light and sound beams with different radii was developed in paraxial approximation [3]. The theory revealed a general phenomenon independent of the transverse distributions of the acoustic and probe laser fields at z=0: for the detection of the CAP motion, the heterodyning creates a complex spectral sensitivity function in wave vector domain. For the backward Brillouin scattering process, the phase of this function contains a part, which is conjugated to the paraxial phase of the CAP and cancels it. Consequently, but contra-intuitively, the BO amplitude variation with time does not depend on the variations of the CAP amplitude in the diffraction process. The key origin of this phenomenon is the phase sensitive process of the acoustically scattered and the reflected probe light interference. Sharp focusing of CAPs and probe laser pulses could increase lateral spatial resolution of TDBS imaging, but could potentially diminish its depth. However, the theoretical analysis contra-intuitively demonstrates that the depth and spectral resolution of the TDBS imaging, with collinearly propagating paraxial sound and light beams, do not depend on the focusing/diffraction of sound and only due to the variations of the probe light amplitude caused by light focusing/diffraction.

Comparison with the experimental observations – The developed theory contra-intuitively predicts, that, although the amplitude of the acoustically scattered light is proportional to the product of the local acoustical and probe light field amplitudes, the temporal dynamics of the TDBS signal amplitude is independent of the CAP amplitude dynamics caused by the diffraction/focusing of the CAP. This prediction correlates with earlier reported experimental observations [4]. In addition, the theory provides explanations to some existing experimental observations [5], which are different from the earlier suggested.

Conclusions – The developed theory predicts that, as far as the CAPs, photo-generated by the pump lase rpulses, and the probe laser beam are paraxial, the lateral resolution of the imaging could be enhanced by sharper focusing of the pump laser beam without diminishing the imaging depth and spectral resolution. The theory reveals earlier unexpected features in the TDBS that could be useful and advantageous in the applications of the TDBS imaging and microscopy technique for the fundamental and applied research.

Acknowledgments – This research is supported by the Agence Nationale de la Recherche [project <ANR-18-CE42-I2T2M>]

References

- [1] V.E. Gusev, P. Ruello, Advances in applications of time-domain Brillouin scattering for nanoscale imaging, Appl. Phys. Rev. 5 (2018) 031101. https://doi.org/10.1063/1.5017241.
- [2] H.-N. Lin, R.J. Stoner, H.J. Maris, J. Tauc, Phonon attenuation and velocity measurements in transparent materials by picosecond acoustic interferometry, J. Appl. Phys. 69 (1991) 3816. https://doi.org/10.1063/1.348958.
- [3] V.E. Gusev, Contra-intuitive features of time-domain Brillouin scattering in collinear paraxial sound and light beams, Photoacoustics 20, 100205 (2020). https://doi.org/10.1016/j.pacs.2020.100205.
- [4] T. Dehoux, K. Ishikawa, P.H. Otsuka, M. Tomoda, O. Matsuda, M. Fujiwara, S. Takeuchi, I.A. Veres, V.E. Gusev, O.B. Wright, Optical tracking of picosecond coherent phonon pulse focusing inside a sub-micron object, Light: Sci. Appl. 5 (2016) e16082. https://doi.org/10.1038/lsa.2016.82.
- [5] T. Dehoux, N. Tsapis, B. Audoin, Relaxation dynamics in single polymer microcapsules probed with laser-generated GHz acoustic waves, Soft Matter 8 (2012) 2586. https://doi.org/10.1039/C2SM07146K.