Structural relaxation in the wave-vector dependence of the longitudinal rigidity modulus

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Abstract: Brillouin microscopy recently attracted much attention for being a promising tool for all-optical label-free determination of mechanical properties of biological samples. Before its widespread utilization for biomedical applications, numbers of nuances related with this technique need to be recognized. In this article, we discuss the process of structural relaxation, the phenomena not commonly addressed by the emerging bio-Brillouin community, and its effect on longitudinal rigidity modulus. Using a model aqueous polymer mixture, we show how scattering measurements performed on the same specimen using different experimental geometries can lead to different results.

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1. Introduction

Spontaneous Brillouin scattering is a process by which light is inelastically scattered on propagating excitations thermally induced within the material [1,2]. This phenomena for decades was providing important information related mostly to excitations in various condensed matter systems. A decade ago the Brillouin spectrometer was for the first time connected to confocal microscope [3,4] presenting ability to acquire spectra fast enough and with spatial resolution adequate for mechanical mapping of biological samples [5]. The possibility of all-optical, non-invasive, contactless and label-free determination of the mechanical properties with diffraction limited spatial resolution was enough to attract attention of the new community interested in utilization of this technique for biomedical and biophotonic applications.

Although the number of biomedical applications of Brillouin micro-spectroscopy is rapidly growing, still problems exists that needs to be resolved before widespread use of this technique in biomechanics. The well recognized difficulties concern: 1) the actual design of the Brillouin spectrometer appropriate for fast mechanical mapping of biological material; 2) translation of the parameter directly measured in experiment (i.e. the Brillouin frequency shift) to the value of mechanical parameter of interest (i.e. longitudinal modulus); 3) the interrelation between the parameters being determined with Brillouin spectroscopy and the mechanical quantities obtained with other experimental techniques.

In spite of complications in the interpretation of mechanical property measured with Brillouin microscopy, the discussion of the result obtained in many biological samples is conducted under simplifying assumption that the real value of longitudinal modulus, $M'$, is a static property dependent only on the structure (local order) of the material. Moreover, it is believed that the unique value of $M'$ can always be determined, regardless of the experimental configuration, and in particular its scattering geometry. However, for every liquid system, there is always inevitable dynamical process associated with continuous reorganization of its local structure known for the condensed matter physicists as the structural relaxation. Because the biological samples can be often described as concentrated multi-constituent aqueous mixtures (cell’s interior in particular), this phenomenon should also be present in such systems.
The usual interpretation of the longitudinal rigidity modulus, \( M' \), presented in many recent articles devoted to biomedical applications of Brillouin microscopy, is based on the assumption of “simple liquid”, meaning that all dynamical processes of the liquid proceed on a timescale much faster than the period of acoustic oscillation. In this scenario, the sound velocity (and so the rigidity modulus) is in fact just a single-value parameter. However, when the relaxation process is active in the Brillouin spectroscopy frequency window, this simple approximation does not hold anymore and the sound velocity become a function of frequency (and so the function of the scattering wave-vector). As a consequence, the existence of structural relaxation process distorts measured Brillouin spectra [1,2,6,7] and affects the Brillouin measurables. In such a case, it is very likely that the influence of liquid’s dynamics on the measured value of rigidity modulus may be as important as heterogeneity of its structure.

To demonstrate the general effects related with the existence of the structural relaxation phenomena we have measured temperature dependencies of the Brillouin shift for a model relaxing liquid using different scattering geometries. In the conditions where the sound velocity dispersion occurs, the values of longitudinal rigidity modulus, determined for each selected scattering wave-vector, are different. Because the value of the scattering wave-vector is defined by the laser wavelength and the scattering angle, this information should be very important when interpreting and comparing the results obtained using Brillouin microspectroscopy. For biomedical applications of this technique the deeper tissue penetration requires the use of lasers with different (longer) wavelengths, whereas sufficient spatial resolution is achieved using microscope objectives with significant acceptance angles.

We also show how the existence of structural relaxation makes any attempt of using Brillouin spectroscopy/microscopy for determination of sample refractive index, basing on the measurement on different wave-vectors, generally unsafe.

2. Brillouin scattering and relaxation in liquids

Brillouin scattering is an inelastic process by which light scatters on propagating acoustic waves thermally generated within a material. The wave angular frequency, \( \omega_B \), (read from the experimental spectrum) and its wavelength, \( \Lambda \), (controlled by the experimental geometry) are related through the dispersion relation \( \omega_B = c_B q \), where \( c_B \) is the sound velocity and \( q = 2\pi/\Lambda = (4\pi n/\lambda_0) \sin(\theta/2) \) is scattering wave-vector (\( n, \theta \) and \( \lambda_0 \) are the refractive index, the scattering angle and the wavelength of incident light, respectively). From the sound velocity and the medium density, \( \rho \), the value of longitudinal rigidity modulus (the real part of the complex longitudinal modulus) can be determined, \( M' = \rho c_B^2 \). This quantity is of prime interest for most of the biomedical application as it is relatively easy to extract from experimental spectrum and provide access to mechanical properties of the medium.

As follows from the dispersion relation, the frequency of the wave changes linearly with its wave-vector as long as the sound velocity stays constant. This, however, is not true in general. During the passage of the acoustic wave, the pressure is locally changing and the sample is constantly driven out of equilibrium. The return towards equilibrium requires changing the local volume and, therefore, liquid’s molecules must move: the local structure of the liquid has to rearrange. This process, known as the structural relaxation, appears in any liquid material: from atomic liquids to polymer melts, from single phase to complex multicomponent systems. Depending on the system and its conditions the structural relaxation is characterized by a representative timescale, \( \tau \). For low viscosity liquids, this molecular restructuring takes place on the fraction of picoseconds. For waves of ultrasonic (MHz) or hypersonic (GHz) frequencies the period of pressure oscillations is much longer than the structural relaxation time, \( \omega_B \tau \ll 1 \). Consequently, the liquid’s structure has enough time to adapt to the constantly changing thermodynamic state and during the passage of the wave the liquid structure remains in quasi-equilibrium, or relaxed, conditions. Here, the sound velocity and longitudinal modulus take appropriate relaxed values: \( c_0 \) and \( M'_0 \) respectively. On the
contrary, if the acoustic frequency is much faster than the rate of structural relaxation ($\omega_B \tau \gg 1$), the molecular reorganization will not keep up with the changing pressure. There will be no time for the structure to relax toward equilibrium in the short period of acoustic perturbation and the conditions are called unrelaxed. This situation is common for high viscosity liquids or glasses where structural relaxation time can be very high. Now the wave will propagate in “dynamically frozen” system of rigidity, $M'_\infty$ (and so with velocity, $c_\infty$) higher than this observed for relaxed situation. The transition from relaxed to unrelaxed condition occurs when the frequency of the perturbation and the frequency of structural relaxation equates ($\omega_B \tau \approx 1$). In this case, the sound velocity will strongly depend on the wave frequency. As follows from dispersion relation, in the Brillouin scattering experiment, the wave of a given frequency, $\omega_B$, is observed for a specific value of a wave-vector $q$. As a result, the values of mechanical parameters obtained using different wave-vectors, that is using different scattering angles or different laser wavelengths, will be in general different. At sufficiently broad $q$-range, the value of longitudinal modulus can change from $M'_0$ to $M'_\infty$.

To the best of our knowledge, there is no detailed microscopic study of the GHz dynamical behavior of any real biological system (i.e. cell’s region) and the above described behavior cannot be illustrated on a real example. For this reason, in Fig. 1, we present dispersion plot for a hypothetical relaxing system to visualize the general behavior. In the absence of values of parameters characterizing any true biological system, we used constant limiting sound velocities close to those of bulk water [8] ($c_0 = 1500\text{m/s}$ and $c_\infty = 3000\text{m/s}$) and calculated the behavior of the system for three arbitrary values of the structural relaxation time (3, 10 and 30ps). It is obvious that for such system, examined at given $q$-vector (fixed scattering geometry), the value of measured sound velocity (or rigidity modulus) will strongly depend on the system dynamics. Similar result will be obtained when the system characterized by a given relaxation time is examined in experiments differing in $q$-value. Therefore, it should be clear that interpretation of the measured $M'$ value, only on the basis of the local order of the system, without considering its dynamics, could be at least unsafe.

One could argue that above a priori values of $c_0$, $c_\infty$ and $\tau$ will surely change when the system much more complex then bulk water will be investigated. That’s is obviously true. However, the details of the system are less important for a general picture we wish to draw. That is, as long as the system exhibits structural dynamics with mean relaxation time fulfilling the $\omega_B \tau = 1$ condition, the general behavior presented in Fig. 1, and so the resulting consequences, will remain valid. Which structural parameters controls the relaxation time in true biological systems is a question which should be answered in future dedicated experiments.

![Fig. 1. Longitudinal acoustic dispersion plot for a relaxing liquid. a) Transition from a relaxed into the unrelaxed branch occurs at certain range of wave-vectors which depends on the actual time of the structural relaxation; b) Corresponding wave-vector dependence of the hypersonic velocity (and so the rigidity modulus $M'$).](image-url)
3. Experimental

The Polyethylene glycol of molar weigh 300g/mol (PEG300) was obtained from Fluka Chemie GmbH and was used without any further purification procedure. Doubly distilled and deionized water was used in order to prepare a concentrated binary mixture. The volume fraction of the polymer, \( \phi = 0.6 \), was set close to the eutectic composition for this system [9]. This characteristic concentration (corresponding to full hydration of the polymer chain) allow aqueous mixtures of short PEG molecules to be cooled down to the glass transition temperature without any phase separation [9,10].

The temperature dependence of the refractive index, \( n_{\text{Pulfrich}}(T) \), was measured in 273 – 323K temperature range using Pulfrich refractometer. In this limited range the \( n_{\text{Pulfrich}}(T) \) dependence behaves linearly (Fig. 3(b)) justifying linear extrapolation for a broader temperature range (although nonlinear behavior is not excluded). The temperature dependence of the density was determined using Lorenz-Lorentz relation assuming additivity of molar refraction and its independence on temperature [11].

During the Brillouin Scattering experiment, the vertically polarized line (\( \lambda_0 = 532 \text{ nm} \)) of a Coherent DPSS 532 laser, working at a mean power of about 80 mW, was used as the probe. The scattered radiation was analyzed using a Sandercock-type (3 + 3)-pass Tandem Fabry-Perot interferometer (TFPI) with free spectral range of 18.8 GHz. The working finesse, estimated by the line-width of the elastic line, was about 90.

For temperature control the sample was kept in liquid nitrogen cooled thermostat. Thermocouple joint was dipped inside the liquid near the laser beam and the sample temperature was read to an accuracy of 0.1 K. The measurements were performed in a 170-370K temperature range using a temperature scanning method. The spectra were registered continuously during the temperature change and quasi-equilibrium condition was ensured applying constant ramp of 1K/min. About 2 spectra per 1K were registered.

The experiment was performed using 90A/90R double scattering geometry illustrated in Fig. 2(a). The sample plate was oriented at 45° respect to the incoming laser beam. The laser beam was focused on the sample using the lens L1 (focal length 100mm) and after passing the sample was directed (through the lens L2) to the mirror M. After reflecting from the mirror, the laser beam is again focused on the sample with lens L3 (focal length 100mm). Notice, that mirror M allows the light to irradiate the sample from two opposite directions. Because of the plate-shape of the sample and its tilted orientation, the incoming laser beam is refracted entering the sample. As a results, the inner scattering angle is different from 90° and depend on the direction of incident laser beam. The light scattered in both utilized geometries was collected using lens L3 (focal length 150mm) at the 90° angle (the outer scattering angle) with respect to the incoming light. The detailed illustration of the incident and scattered wave-vectors directions realized in 90A and 90R situations is presented in Figs. 2(b) and 2(c).

The magnitudes of the exchanged wave-vectors in 90A and 90R geometries are given by [12]:

\[
q_{90A} = 2\pi \sqrt{\frac{2}{\lambda_0}}, \quad q_{90R} = 2\pi \sqrt{\frac{4n^2 - 2}{\lambda_0}}. \tag{1}
\]

Notice that in 90A geometry \( q_{90A} \) is independent of the refractive index.

In addition to double-scattering experiment, the measurements using usual back-scattering geometry was also performed allowing to probe highest possible (for given laser wavelength) scattering wave-vector \( q_{\text{BS}} \).

Comparing the sound velocity obtained with a 90A geometry (for which the scattering wave-vector is independent on refractive index) with the velocity observed for any other scattering geometry, the opto-acoustical dispersion function (D-function) may be defined. If no relaxation process is present in the frequency range analyzed then D-function takes the value exactly equal to the refractive index of the sample. In the present case of 90A/90R realization, the function can be written as [12,13]:

\[
\text{D-function} = \frac{n_{\text{Pulfrich}}(T)}{n_{\text{BS}}(T)}. \tag{2}
\]
\[ D_{90A}(T) = n = \frac{1}{2} \left[ 1 + \left( \frac{\omega_{B_{90A}}(T)}{\omega_{B_{90A}}(T)} \right)^2 \right] \] (2)

Fig. 2. The double 90A/90R scattering geometry used in this study. a) Scheme of the experimental setup: L1, L2, L3 – lenses, M – mirror, S – sample; b) inner scattering for 90A geometry; c) inner scattering for 90R geometry; d) Experimental Brillouin spectrum for PEG300/H2O mixture at four different temperatures. The solid lines are the fit results obtained with Eq. (3).

With the scattering geometry utilized, the spectrum registered consists of two peaks corresponding to the light scattered on two longitudinal phonons differing in a magnitude of wave-vector. One of the main advantages of the double-scattering experimental realization is that the scattered signal comes from the same scattering volume and is registered at exactly the same temperature. As shown in Fig. 2(d), the two peaks are always well separated in frequency scale which allow the estimation of their position with high precision.

In order to obtain the frequency shifts of both Brillouin peaks seen in Fig. 2(d) (\(\omega_{90A}\) and \(\omega_{90R}\)), the spectra acquired at each temperature were fitted with the sum of two Brillouin contributions using a proper hydrodynamic expression [1,2] (instead of simple Lorentzian approximation) convoluted with the instrumental resolution function

\[
I(\omega) = \frac{A_c}{\omega^2 + \Gamma_C^2} + \sum_{i=90A,90R} \left\{ \frac{A_{B_{i}} \Gamma_{B_{i}}}{(\omega + \omega_{B_{i}})^2 + \Gamma_{B_{i}}^2} + \frac{A_{B_{i}} \Gamma_{B_{i}}}{(\omega - \omega_{B_{i}})^2 + \Gamma_{B_{i}}^2} \right\} + \frac{1}{\omega_{B_{i}}} \left[ A_c \Gamma_C + A_b \Gamma_B \right] \left[ \frac{\omega + \omega_{B_{i}}}{(\omega + \omega_{B_{i}})^2 + \Gamma_{B_{i}}^2} - \frac{\omega - \omega_{B_{i}}}{(\omega - \omega_{B_{i}})^2 + \Gamma_{B_{i}}^2} \right].
\] (3)

In Eq. (3), the first quasi-static (unshifted) term, with the width \(\Gamma_C\), accounts for relaxation contribution, subsequent two terms describe the Brillouin doublet composed of two lines symmetrically shifted at \(\omega_{B_{i}}\), with the width (half width at half maximum) \(\Gamma_{B_{i}}\), whereas the last two terms ensure for the preservation of the first moment sum rule and only affect the symmetry of the Brillouin doublet. Quantities \(A_c\) and \(A_B\) are the amplitudes of the central contribution and of the Brillouin lines, respectively [1,2]. It is worth to note, that expression of the form of Eq. (3) was historically derived for “simple” (atomic) liquids and then...
generalized for molecular liquids (to include internal relaxation). However its usability applies also to more complex situation. Because the description of the Brillouin scattering phenomenon combines thermodynamics and hydrodynamics considerations, Eq. (3) can be adopted to any system fulfilling hydrodynamic approximation, that is as long as it can be considered as homogenous on the spatial-scale of acoustic wave. Successful description of the experimental spectra acquired for different biological materials, studied recently with Brillouin microscopy [5,14,15], confirm that hydrodynamic description applies not only to atomic or simple molecular liquids, but also to much more complex systems, including biological ones.

4. Results and discussion

The values of Brillouin shifts obtained from fitting the experimental spectra were recalculated to corresponding rigidity moduli. The temperature dependencies of $M_{90A}$ and $M_{90R}$, together with the $M_{BS}$ obtained with usual back-scattering geometry, are presented in Fig. 3(a). This plot shows the characteristic behavior observed in many relaxing systems. At high temperatures, where the relaxation is fast ($\omega_B \tau << 1$ condition holds), the measured rigidity modulus probes its relaxed value ($M^0$). At intermediate temperatures $\omega_B \tau = 1$ conditions is fulfilled and the inflection of $M'(T)$ is observed. At lowest temperatures the liquids dynamics slows and for $\omega_B \tau >> 1$ the rigidity modulus approaches unrelaxed value ($M^{\infty}$). The striking feature of Fig. 3(a) is that rigidity moduli, corresponding to different scattering wave-vectors, do not follow the same path. It is clear that all the moduli values converge only for the highest and the lowest temperatures examined, i.e. when the structural relaxation time is either much faster or much slower than the period of the acoustic perturbation. For intermediate temperatures, the $\omega_B \tau = 1$ conditions is fulfilled and, consistently with what was written earlier, $M'(\omega)$ or $M'(q)$ is never a constant. It is clear that the same system can be characterized by different value of rigidity modulus even if its objective properties have not changed. For this to happen it is enough to observe the sample in different scattering geometry. As an example of this situation, in the inset of Fig. 3(a), we present the $M'(q)$ dependence for arbitrary temperature $T = 298K$. In addition to a simple indication of the presence of the relaxation process ($M'(q) \neq \text{const}$), the $M'(q)$ dependence can be also used to estimate the value of relaxation time, which in this case is about $\tau = 6$ ps.

Fig. 3. Outcome of the temperature Brillouin scattering experiment performed on PEG300/Water mixture at different values of wave-vectors. a) Rigidity modulus obtained for 90A, 90R and back-scattering (BS) geometry; inset: $q$-dependence of the $M$ obtained for $T = 298K$ reproduced assuming mean relaxation time of 6ps (solid line). b) Temperature behavior of the $D_{90R}$ function compared with the experimental refractive index (nPulfrich). Inset: comparison between $D_{90R}$ and refractive index for bulk Water.
From the $\omega_{90A}$ and $\omega_{90R}$ values, we calculated the $D_{90R}$ function which should be exactly equal to the refractive index. This property of double-scattering geometry is very attractive as in principle it allows to determine the value of refractive index from the Brillouin measurements (also with microscopic spatial resolution). In Fig. 3(b), we show that this is not true in general. It is clear that for PEG300/Water system, the value of $D_{90R}$ function correspond to the refractive index only for the limited temperature ranges, specifically when the relaxation process is outside of the frequency window of the Brillouin light scattering method. This indicates that the Brillouin method can be used for refractive index measurements only for very high viscosity systems (glasses) or very-low viscous liquids. This last case is tested for the pure water (inset of Fig. 3(b)). Because the structural relaxation for this liquid is much faster than the period of mechanical agitation [16], the value of $D_{90R}$ function registered for broad temperature range is a good measure of the true value of refractive index.

The question may arise if the results obtained for the mixture studied in this work are representative for any true biological sample. Although our model system is far less complex than real biological systems, the existence of structural relaxation in GHz frequency range was previously indicated in concentrated aqueous mixtures of many biologically important solutes (polymers, salts, proteins) [10,17,18]. It is then justified to assume, that in any aqueous mixture of high enough solute concentration the value of the Brillouin rigidity modulus will be influenced by the system dynamics. This assertion applies also to many biological materials which, perceived on the spatial scale of the acoustic wave length, may be imagined as a concentrated multi-constituent aqueous mixtures. Besides this, the Brillouin spectrum for every system fulfilling the hydrodynamic approximation can be described by the same expression independently on its actual structural complexity. As long as the hydrodynamic approximation holds, the Brillouin spectrum carries information on the collective behavior of the system, where the details of the structural and dynamical contributions are averaged in the function of complex longitudinal modulus. Therefore, the results we presented above are general for every locally uniform fluid and should also be applicable to many biological samples. However, the question of how the structural relaxation affects particular biological systems and what are the structural parameters controlling its dynamics, is still open.

5. Conclusion

In this article we discussed the influence of the structural relaxation on the value of longitudinal rigidity modulus $M'$ determined in Brillouin scattering experiment. We presented the results of the Brillouin scattering measurements performed on an aqueous mixture of PEG polymer in a wide temperature range for different scattering geometries. Although the choice of the sample may seem arbitrary, and the experimental conditions are far from these relevant for biological interests, the conclusions we reached can be very important for successful utilization of the Brillouin microscopy technique for mechanical examination of various biomaterials.

The biological samples are often multicomponent aqueous solutions where proteins, lipids, sugars, salts, etc. are present at different concentration. In this multi-phase situation, the rate of the structural relaxation can be altered through many additional (to temperature) control parameters, like quantity and quality of the system components. It is not hard to imagine a particular biological specimen which is in conditions similar to these reported in our study, that is to say its structural dynamics proceed in the range of several picoseconds. In such a situation, the results of the measurements of the same specimen but using different experimental setups (operating on different values of scattering vector $q$) will not agree. This knowledge should be practical when comparing outcomes of experiments performed with different lasers or microscope objectives as the value of the scattering wave-vector is defined by the laser wavelength and the scattering angle. To avoid inconsistency, it would be
convenient to present the “relaxed” \( \langle M'_0 \rangle \) value of the rigidity modulus. Only then the results from experiments performed using different scattering geometries will coincide and the results may be discussed in terms of mechanical (local order) changes alone. This should make any potential correlation of Brillouin results with other mechanical techniques safer.

As an example of the practical difficulties related to the existence of relaxation process we have presented that any attempt of using Brillouin spectroscopy/microscopy for refractive index measurements of sample with active relaxation process will generally fail.

Finally, we have shown how the Brillouin measurables change when the liquid’s dynamics is changing. However, it is probably more interesting for biologists to ask which structural parameters of particular system are responsible for changing its dynamics. To the best of our knowledge, there is still no research activity focused on this particular issue. Surely, dedicated research program should be started in order to understand to what extent structural relaxation phenomena influences the value of the longitudinal modulus in real biological systems. The presence of the structural relaxation may be perceived as a kind of obstacle, but it should be envisioned as an important piece of additional information which, when properly addressed, will make the Brillouin microscopy even more comprehensive biomedical technique then initially imagined.

Disclosures

The authors declare that there are no conflicts of interest related to this article.

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