Determination of bulk viscosity of liquid water via pulse duration measurements in stimulated Brillouin scattering

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We present a method by which to determine the bulk viscosity of water from pulse duration measurements of stimulated Brillouin scattering (SBS). Beginning from a common model of Brillouin scattering, the bulk viscosity is shown to play an important role in Brillouin linewidth determination. Pulse durations of SBS back-reflected optical pulses are measured over the temperature range of 5–40 °C. SBS linewidths are determined via Fourier transformation of the time-domain results, and the bulk viscosity of water is measured and derived from the obtained values. Our results show that the proposed method for measurement of pulse durations is an effective approach for determining bulk viscosity. The method can be easily extended to determine bulk viscosities of other Newtonian liquids.

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Water plays a fundamental role in many physical situations from geology to biology; hence, significant efforts through both theoretical and experimental studies have been exerted by many researchers to understand the hydrodynamic and thermophysical properties of water[1−4]. The physical properties of water, such as its compressibility (force field) and dynamic (shear) and bulk viscosity, have recently been studied[5−7]. The dynamic or shear viscosity, which is a kinetic property that reflects only the translational molecular motion of a liquid, is useful for parametrizing the force field. The dynamic or shear viscosity of a liquid can be determined easily by experiments or obtained from the CRC Handbook of Chemistry and Physics[8]. In this letter, we focus on the relaxation of both rotational and vibrational degrees of molecular freedom, also referred to as the bulk viscosity, in water.

Shear viscosity can be routinely measured using commercial rheological instrumentation. By contrast, bulk viscosity is not a well-established parameter, and the subject has received little experimental attention despite being defined in hydrodynamic equations (e.g., the Navier–Stokes equation) for compressible liquids. Experimental methods for measuring the bulk viscosity are scarce.

Two methods for measuring bulk viscosity are acoustic spectroscopy[9] and Brillouin spectroscopy[10]. Acoustic spectroscopy generally employs equations of motion for sound propagation in some compressible liquids based on the Navier–Stokes and continuity equations. After thorough analysis, the bulk viscosity and velocity of sound can be expressed by[11]

$$\eta_{\mathrm{B}} = \left[\frac{2\alpha\rho\upsilon^2}{\omega^2} + \frac{4\eta_{\mathrm{S}}}{3} + \frac{(\gamma-1)\,k}{C_{\mathrm{p}}} \right], \qquad (1)$$

where η_{B} denotes the bulk viscosity of fluid, α is the attenuation of the acoustic wave, ρ is the medium density, υ is the sound speed, ω is the angular frequency, η_{S} is the shear viscosity, γ is the ratio of specific heats, C_{p} is the specific heat at constant pressure, and k is the thermal conductivity. A plot of acoustic attenuation as a function of frequency and viscosity shows an asymptotic function curve. Assuming that all of the other parameters are known, the bulk viscosity can be determined by measuring the attenuation and sound speed. Theoretically, the plot of linewidth of spontaneous Brillouin scattering as a function of viscosity and other thermodynamic parameters shows a Lorentz-shaped curve that can be described by[10]

$$\Gamma_B = \frac{1}{\rho} \left[\frac{4}{3}\eta_{\mathrm{S}} + \eta_{\mathrm{B}} + \frac{k}{C_{\mathrm{p}}}\left(\gamma - 1\right) \right] \left[\frac{4n\pi}{\lambda}\sin(\theta/2) \right]^2, \ (2)$$

where n is the refractive index of liquid, λ is the wavelength of incident light, and θ is the scattering angle. Thus, bulk viscosity may be expected to play an important role in the Brillouin linewidth. Equation (2) also provides an alternative method for measuring the bulk viscosity of liquids.

Each of the methods described above has its own advantages and disadvantages in terms of the medium being measured and the measuring circumstances. Acoustic spectroscopy is usually implemented in megahertz frequencies, whereas Brillouin scattering is typically performed at gigahertz frequencies. In this letter, we build upon the foundation laid by previous studies and present a novel and convenient method for measuring the bulk viscosity of water. In this novel method, the bulk viscosity of water is measured by analyzing the pulse duration of stimulated Brillouin scattering (SBS) using laboratory experiment over the temperature range of 5–40 °C.

The schematic diagram considered for measuring the bulk viscosity of water is shown in Fig. 1. An injection-seeded Nd: YAG laser (Continuum Power Precision Plus) working at 532 nm after frequency doubling is used as a light source. The pulse duration of the laser is 7 ns, the repetition rate is 10 Hz, the laser divergence angle is 0.45 mrad, and the beam has a Gaussian spatial profile with $1/e^2$ diameter of 12 mm. Linewidths of 90 MHz and 30 GHz are obtained when the seeder is switched on and off, respectively. The output energy of the laser can be controlled by adjusting the delay time between the oscillator and the amplifier.
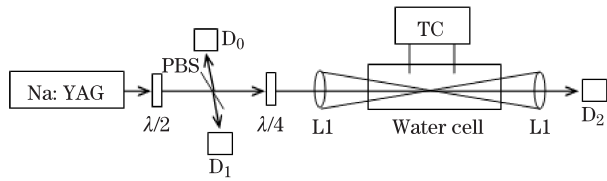
Fig. 1. Optical setup for measuring the bulk viscosity of water. $\lambda/2$ and $\lambda/4$ denote a half-wave plate and a quarter-wave plate, respectively; PBS is the polarization beam splitter; D_0 is the photodetector; D_1 and D_2 are wattmeters; L1 and L2 are lenses; TC is the temperature controller

The polarization of the output laser is oriented vertically but becomes horizontal after passing through a half-wave plate (quartz, zero order). A small fraction is then split from the horizontal polarization by the polarizer and detected by detector D_1 (FieldMate, Coherent Inc.) to monitor the energy of the incident laser. The transmitted laser beam is circularly polarized after passing through the quarter-wave plate (quartz, zero order) and then focused by L1 and L2 at different focus points in a vitreous cistern. Distilled water is used as the liquid sample in this experiment. The water is circulated through a temperature controller that adjusts the temperature from 5–40 °C with 0.02 °C precision. Near the focus point, SBS is excited once the intensity of the incident laser is beyond the threshold intensity. Given that SBS propagates only backward because of the phase conjugation effect, SBS changes the polarization orientation from circular to vertical after passing through the quarter-wave plate. Most of the energy of SBS is reflected by the polarizer and then projected onto the photodetector D_0 (Silicon PIN Detector ET-2000, Electro-Optics Technology, Inc.). The spectra of the pulse durations of SBS are obtained and analyzed using a digital storage oscilloscope at 1 GHz bandwidth (InfiniiVision DSO7104A, Agilent Technologies). Finally, the residual light is collected by the detector D_2 (Molectron PM 500A).

Detailed theories of bulk viscosity are reported elsewhere[12,13]. In this letter, we only outline the results of these developments with the necessary equations for data analysis. According to Ref. [10], the bulk viscosity can be presented conveniently in terms of the linewidth equation of spontaneous Brillouin,

$$\eta_B = \frac{\Gamma_B \lambda^2 \rho}{16\pi^2 n^2(S,T,\lambda)} - \frac{4}{3}\eta_S. \tag{3}$$

The value of shear viscosity η_S can be obtained from the CRC Handbook of Chemistry and Physics. The refractive index of water in terms of salinity (S), temperature (T), and wavelength of incident light (λ) is given by an empirical relation[14]

$$n(S,T,\lambda) = n_0 + (n_1 + n_2 T + n_3 T^2) S +$$
$$n_4 T^2 + \frac{n_5 + n_6 S + n_7 T}{\lambda} + \frac{n_8}{\lambda^2} + \frac{n_9}{\lambda^3}. \tag{4}$$

In our experiments, the salinity of water was 0‰ temperature varied from 5 to 40 °C, and the wavelength of incident light was 532 nm.

The linewidth of spontaneous Brillouin scattering is Γ_B (circular frequency). Generally, frequency analysis of conventional spontaneous Brillouin scattering requires

a high-precision interferometer to separate the Brillouin signals from elastic scattering (Rayleigh scattering). The signal intensity of spontaneous Brillouin is extremely weak and may bring about extra errors during bulk viscosity measurement. This study aims to measure the bulk viscosity of water by determining the time domain spectroscopy of SBS. The determination process is performed as follows.

We first measure the pulse duration of SBS over the temperature range from 5 to 40 °C. Figure 2 shows the temperature dependence of the pulse durations of SBS. For each temperature, 10 temporal waveforms were recorded and mean values of the pulse duration are presented with error bars. The pulse duration of SBS has a limited width and is much shorter at low temperatures than at relatively higher temperatures. Velchev et al.[15] analyzed and modeled the pulse compression by SBS in the fully transient regime ($\tau \ll \tau_p$). When $\tau \gg \tau_p$, SBS shows approximately steady-state conditions, and the limit of pulse compression is set by the phonon lifetime (τ_p). The phonon lifetime can be expressed as $\tau_p = 1/\Gamma_B = \lambda^2/4\pi^2\eta$[16], where η is the viscosity of the material and λ is the wavelength of incident light. The phonon lifetime is inversely proportional to the viscosity of the material. For water, the variation of its temperature has a significant effect on its viscosity. As temperature increases, the viscosity of water decreases, which results in an increase in τ_p[10]. The short phonon lifetime boosts energy transfer from incident light to SBS; therefore, the pulse compression effect is more evident. By contrast, a long phonon lifetime is detrimental to energy transfer, and the compression effect weakens. The variational regularity of the pulse duration with temperature may be represented by

$$P(T) = 1.2909\,[1 - \exp(-9.8682T)] + 0.7941$$
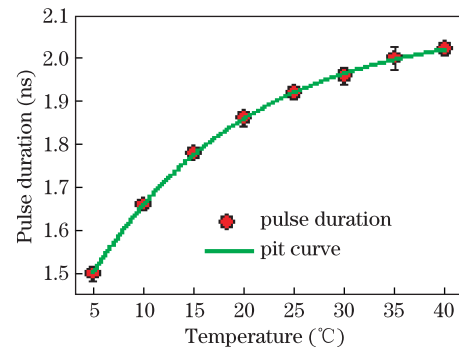$$\cdot\,[1 - \exp(-0.063T)]. \tag{5}$$

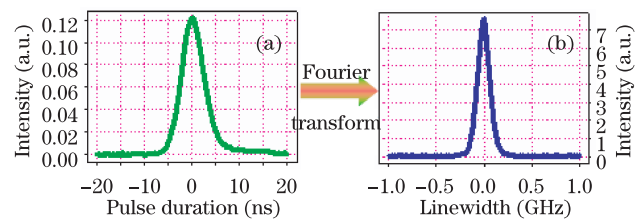Fig. 2. Changes in pulse duration of SBS as a function of temperature.

Fig. 3. Fourier transform of the pulse duration of the incident laser.

The pulse duration is closely related to the linewidth obtained by SBS measurement because the pulse width and linewidth satisfy each other's Fourier transform. That is, the change in pulse duration provides an expression of the change in linewidth. Figure 3 presents a Fourier transform of the pulse duration of the incident laser, and the transformed linewidth shows a Lorentz-shaped profile. The calculated linewidth determined through Fourier transformation of the pulse duration is approximately 79 MHz. A discrepancy may be observed between the linewidth calculated through Fourier transformation and the parameterized linewidth determined at 90 MHz. This discrepancy may be explained by the imperfect Gaussian-shaped temporal profile of the laser, which may introduce calculation differences after Fourier transformation. Variations introduced by the use of the oscilloscope during the recording process of pulse durations may also contribute to the differences in linewidths observed. Fortunately, this difference may be limited by calibrating the calculated linewidth.

Theoretically, if a pulse duration has a stable Fourier transform limited width and operates at a single frequency, the corresponding frequency bandwidth is also limited. Figure 4 shows the measured temporal profile and transformed linewidth of the SBS pulse at different temperatures. The results shown in Figs. 4(a) and (b) represent Gaussian-shaped temporal profiles, whereas those shown in Figs. 4(c) and (d) show transformed linewidths with Lorentz-shaped profiles corresponding to the pulse duration at the same temperature. Pulse compression of the time domain corresponds to a frequency extension.

A plot of the experimental and calculated data is shown in Fig. 5 to illustrate the relationship between the pulse duration and the linewidth obtained through Fourier transformation. At low temperatures, the compressing effect of pulse duration or the extension effect of the linewidth of SBS is more significant than that at high temperatures; this variation is probably due to the change in viscosity of the medium[16,17].
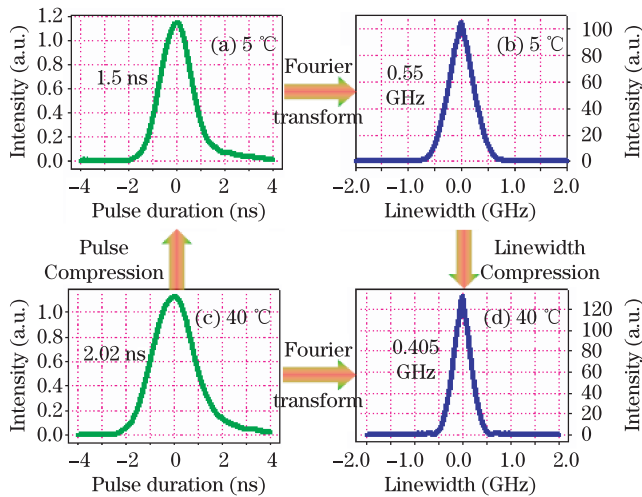

Fig. 4. Correlation between the pulse duration and linewidth of SBS at different temperatures. (a) and (c) show the pulse durations of SBS at 5 and 40 °C, respectively. (b) and (d) show the transformational linewidths of SBS at 5 and 40 °C, respectively.
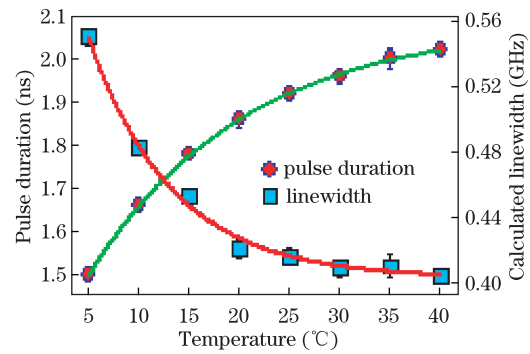

Fig. 5. Pulse duration and calculated linewidth of SBS as functions of temperature.
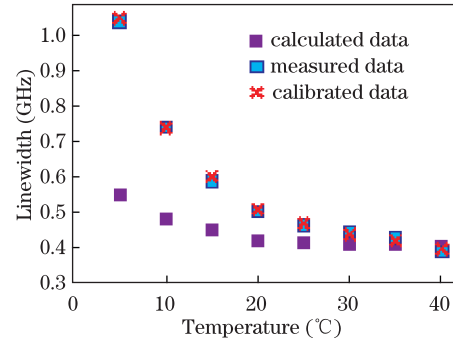

Fig. 6. Data calibration for the linewidth calculated through Fourier transformation of the pulse duration using directly measured data.

The points described above are based on a hypothesis that the pulse has an ideal Gaussian-shaped temporal profile. Thus, a method by which to compensate for the difference between the calculated linewidth and its actual value must be determined. The Fabry–Perot (F-P) etalon is commonly used to analyze the spectrum. In the current study, we employ a F-P etalon with free spectral range of 20.1 GHz and finesse of $F \geqslant 300$ to form the spectrum, after which an ICCD camera (PI-MAX 1003) with pixel size of 12 μm records and analyzes the spectrum. The linewidth of SBS can be measured accurately using this method. The linewidth obtained by Fourier transformation was calibrated using the ICCD-measured standard data resource (Fig. 6). The calibration formula profile is

$$C(T) = 1.1512 \exp(-0.1558T) + 0.5421 \exp(-0.0078T). \tag{6}$$

The calibrated equation can then be written as

$$\Gamma_{\mathrm{C}}(T) = \Gamma_{\mathrm{FT}}(T) * C(T), \tag{7}$$

where $\Gamma_{\mathrm{C}}(T)$ denotes the calibrated linewidth and $\Gamma_{\mathrm{FT}}(T) = \mathcal{F}[P(T)]$ denotes the linewidth calculated from Fourier transformation of the pulse duration.

Our experimental setup involves a simple-cell SBS process similar to that in Fig. 1(a) described in the work of Schiemann *et al.*[18]. This setup achieves efficient pulse compression at low input energies. At high input energies, the temporal profile of SBS deteriorates and does not show a Gaussian profile; in this case, the Fourier-transform method is unsuitable. In our experiment, the

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Table 1. Measured Bulk Viscosities and Parameters used in the Calculations

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Temperature T (°C)	Pulse Duration P (ns)	Linewidth Γ_{FT} (GHz)	Shear Viscosity η_{S} ($\times 10^{-3}$ Pa.s)	Bulk Viscosity η_B ($\times 10^{-3}$ Pa.s)
5	1.50	0.550	1.5141	7.8386
10	1.66	0.482	1.3028	4.7152
15	1.78	0.453	1.1362	3.4811
20	1.86	0.421	1.0018	2.7529
25	1,92	0.416	0.8917	2.4535
30	1.96	0.409	0.8002	2.311
35	2.00	0.410	0.7232	2.23325
40	2.02	0.404	0.6577	2.1237

intensity of the incident beam was set to a value slightly higher than that of the SBS threshold so that the SBS profile presents a Gaussian line-shape. In addition, after the geometry of the experimental setup and the incident energy are fixed, the pulse duration is determined only by varying the temperature of water.

To determine the bulk viscosity using Eq. (3), determination of the relationship between the linewidth of SBS and the spontaneous is necessary because the calculated bulk viscosity of water (Eq. (3)) is based on the linewidth of spontaneous Brillouin scattering. However, the linewidth we calculated from Fourier transformation of the pulse duration describes SBS. Therefore, converting the linewidth of SBS to that of spontaneous Brillouin scattering is necessary. According to Ref. [10], the mapping relationship between the SBS and spontaneous Brillouin scattering linewidths can be expressed as

$$\Gamma_{\mathrm{B}}(T) = [1.0894 + 0.5065 \exp(-0.0985T)] \cdot 2\pi \Gamma_{\mathrm{SBS}}(T), \tag{8}$$

where $\Gamma_{\mathrm{SBS}}(T) = \Gamma_{\mathrm{C}}(T) = \Gamma_{\mathrm{FT}}(T) * C(T)$ is the linewidth of SBS. Substituting Eqs. (6) and (8) into Eq. (3), we obtain the bulk viscosity equation as a function of SBS linewidth in the following form:

$$\eta_{\mathrm{B}} = \frac{A(T)\Gamma_{\mathrm{FT}}(T)\lambda^2 \rho}{16\pi^2 n^2(S, T, \lambda)} - \frac{4}{3}\eta_{\mathrm{S}}, \tag{9}$$

where $A(T)$ is an empirical coefficient of temperature dependence.

$$A(T) = 7.88\exp(-0.1558T) + 3.714\exp(-0.0078T)$$
$$+ 3.664\exp(-0.249T) + 1.728\exp(-0.106T). \tag{10}$$

The bulk viscosity of water can be calculated using Eq. (9). Table 1 shows the calculated linewidth and bulk viscosity obtained at each temperature considered.

For comparison, we also present the results of two other known methods used for determining the bulk viscosity of water at room temperature. Dukhin *et al.*[9] reported a bulk viscosity value of 2.436×10^{-3} Pa·s at 25 °C for distilled water, which was measured using acoustic attenuation method. Using the same method, Holmes *et al.*[11] obtained a value of 2.469×10^{-3} Pa·s. Xu *et al.*[10] also reported a water bulk viscosity value of 2.337×10^{-3}
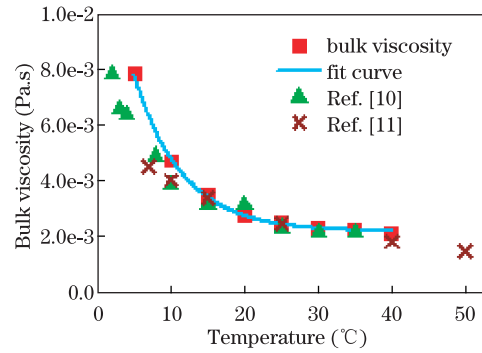

Fig. 7. Temperature dependence of bulk viscosity of water.

Pa·s. All of these values are consistent with our results (2.453×10^{-3} Pa·s). In Fig. 7, we plot the bulk viscosities measured in the current study as well as those reported by several other studies. Our results concur well with those of other studies in which the bulk viscosity of water was measured using different techniques.

Finally, we present an empirical formula [Eq. (11)] that shows the temperature dependence of the bulk viscosity of water over a certain temperature range.

$$\eta_{\mathrm{B}} = 0.0022 + 0.0121\exp(-0.1522T). \tag{11}$$

The maximum relative error between the calculated results from Eq. (11) and the data shown in Table 1 is $<5\%$, which is well within the range of measurement uncertainty. The temperature dependence of the viscosity of nearly all pure and multicomponent liquids can be adequately expressed over a limited range of temperature using the empirical model $\eta = A\exp(-BT)$ (A and B are empirical constants), as observed by Reynolds in 1886[19]. Our empirical formula follows the same asymptotic behavior with coefficients A=0.0121 and B=0.1522.

The uncertainty of measurement $\sigma_{\Gamma_{\mathrm{FT}}}$ of the calculated linewidth depends on the uncertainty of measurement σ_{P} of the pulse duration. The uncertainty can be estimated as

$$\sigma_{\Gamma_{\mathrm{FT}}} = \frac{\partial \Gamma_{\mathrm{FT}}}{\partial P}\sigma_{\mathrm{P}}, \tag{12}$$

$$\sigma_{\mathrm{P}} = \sqrt{\sigma_{\mathrm{PA}}^2 + \sigma_{\mathrm{PB}}^2}, \tag{13}$$

where $\sigma_{\mathrm{P}A} = \sqrt{\sum\limits_{i=1}^{10}\left(P_i - \overline{P}\right)^2 \Big/ 9}$ and $\sigma_{\mathrm{P}B} = \Delta_{\mathrm{OSC}}/\sqrt{3}$ are type A and type B uncertainties, respectively; P_i and $\overline{P}$ denote the measured and average pulse durations, respectively; Δ_{OSC} is the intrinsic error of the oscilloscope, which is equal to 100×10^{-12} s. The value of σ_{P} is then calculated to be 58×10^{-12} s. Upon further calculation, the uncertainty of measurement $\sigma_{\Gamma_{\mathrm{FT}}}$ shows a limit of 7.46–30.79 MHz.

From Eq. (9), the uncertainty of measurement $\Delta\eta_{\mathrm{B}}$ of the bulk viscosity can be expressed as

$$\Delta\eta_{\mathrm{B}} = \frac{A(T)\lambda^2\rho}{16\pi^2 n^2\left(S,T,\lambda\right)}\sigma_{\Gamma_{\mathrm{FT}}}. \tag{14}$$

To estimate the relative error of the proposed method, the following values were chosen: $\lambda = 532$ nm, $\rho = 1\,000$ kg/m^3, $n = 1.33$. When the uncertainty of the calculated linewidth is 7.46 MHz, the maximum uncertainty of the bulk viscosity is 2.088×10^{-5}. Given that the bulk viscosity is in the 10^{-3} order of magnitude, the relative error of the measurement is $<1\%$. When the uncertainty of $\sigma_{\Gamma_{\mathrm{FT}}}$ is 30.79 MHz, the maximum uncertainty of the bulk viscosity is 2.892×10^{-4} and the relative error of bulk viscosity remains $<5\%$.

In conclusion, an experimental method for determining the bulk viscosity of water by measuring the pulse duration of SBS is proposed. First, we measure the pulse duration of SBS in water over the temperature range of 5–40 °C and observe that the temperature dependence of pulse durations presents an exponential increase. Next, the linewidth of SBS is calculated through Fourier transformation of the pulse duration and then calibrated using the measured data obtained from the F-P etalon and ICCD experiment. Third, the bulk viscosity equation is obtained as a function of linewidth of SBS. Finally, the bulk viscosity of water is determined, and an empirical equation is presented. The measured results are consistent with those reported by other studies. Our results demonstrate that Fourier transformation of the pulse duration of SBS can be readily and generally used to measure the bulk viscosity of liquids accurately.

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