Temperature-dependent bulk viscosity of nitrogen gas determined from spontaneous Rayleigh–Brillouin scattering

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Received January 30, 2013; accepted February 19, 2013;
posted February 26, 2013 (Doc. ID 184551); published March 25, 2013

Values for the bulk viscosity η_b of molecular nitrogen gas (N_2) were derived from spontaneous Rayleigh–Brillouin scattering at ultraviolet wavelengths ($\lambda = 366.8$ nm) and at a 90° scattering angle. Analysis of the scattering profiles yields values showing a linear increasing trend, ranging from $\eta_b = 0.7 \times 10^{-5}$ to 2.0×10^{-5} kg · m^{-1} · s^{-1} in the temperature interval from 255 to 340 K. The present values, pertaining to hypersound acoustics at frequencies in the gigahertz domain, are found to be in agreement with results from acoustic attenuation experiments in N_2 performed at megahertz frequencies.
OCIS codes: 010.0010, 290.5830, 290.5840, 290.5870.

The concept of bulk viscosity, η_b, also referred to as volume viscosity, is part of a thermodynamic description of gases as a transport coefficient in addition to the shear viscosity η_s [1,2]. Bulk viscosity results from collisional energy exchange between the translational and internal (rotational and vibrational) degrees of freedom in fluids. The value of η_b of gases can be measured via sound absorption, but only a limited number of studies have been reported [3,4]. Furthermore, such measurements yield values for η_b related to acoustic frequencies in the megahertz range, while bulk viscosity is regarded as a frequency-dependent parameter [5], resulting from competition between the internal relaxation time of molecules and the period of acoustic waves. Therefore, the values measured at megahertz frequencies should not be directly applicable to much higher frequencies such as in light-scattering experiments, where the (hypersound) acoustic waves are in the gigahertz domain. For example, Pan *et al.* found that bulk viscosity for CO_2 in their coherent Rayleigh–Brillouin scattering (CRBS) experiment is 1000 times smaller than the sound absorption value [6]. They suggested that values of bulk viscosity at high frequencies could be derived by comparing the light-scattering profiles of gases to accurate models developed by Boley *et al.* [7] and Tenti *et al.* [8], given that in these models the only unknown parameter is η_b.

In this Letter, we present measurements of spontaneous Rayleigh–Brillouin scattering (SRBS) profiles of N_2 in a temperature range of 255–340 K and a pressure range of 850–3400 mbar. The measured scattering profiles are compared to the so-called Tenti S6 model [8], which is generally considered the most accurate model to describe the Rayleigh–Brillouin (RB) scattering profile [9]. Implicit in the model is that the Brillouin side peaks to the

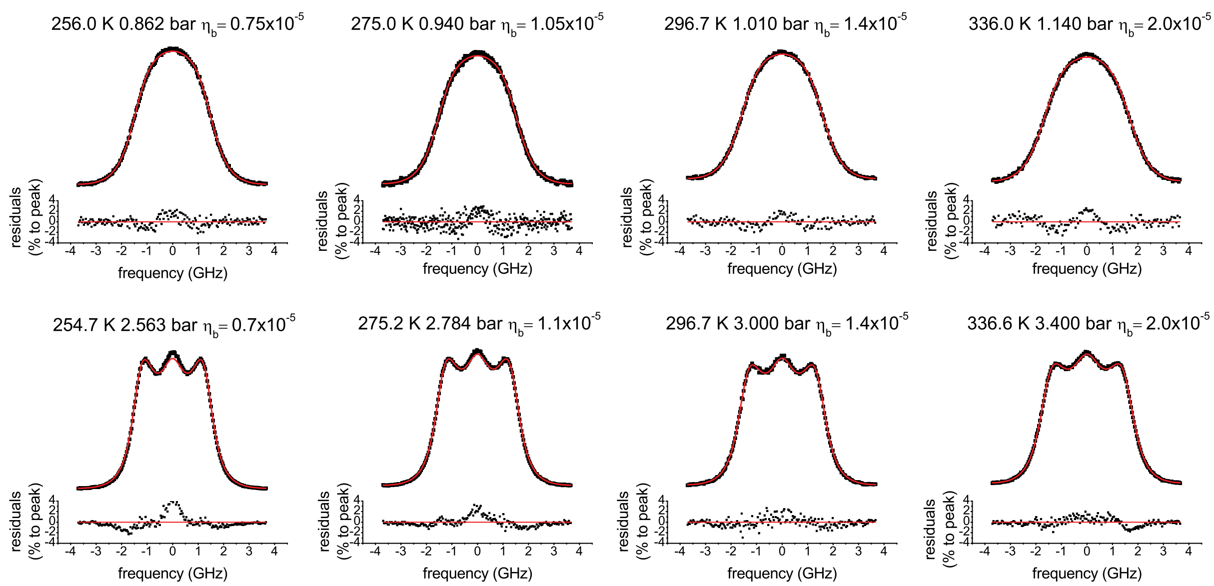

Fig. 1. (Color online) RB scattering profiles (black dots) as measured for various (p, T) pressure–temperature combinations as specified. A comparison is made with calculations via the Tenti S6 model (red lines), convolved for the instrument width of 232 MHz and for values of the bulk viscosity η_b, deduced from the profiles recorded at 3 bar. Residuals between the measurements and the calculations are given underneath.

0146-9592/13/071110-03$15.00/0

central Rayleigh peak in the scattering profile are shifted by [10]

$$\Omega_B = \pm 2n\omega \frac{v}{c} \sin \frac{\theta}{2}, \quad (1)$$

where n and v are the index of refraction and the sound velocity in the gas, and ω and θ are the angular frequency of the light and the scattering angle. The Brillouin side peaks exhibit a profile, associated with the damping of acoustic waves, and dependent on the thermodynamic properties of the gaseous medium as well as the light-scattering parameters, yielding a Lorentzian profile of full width at half-maximum:

$$\varGamma_B = \frac{1}{\rho v^2}\left[\frac{4}{3}\eta_s + \eta_b + \frac{\kappa}{C_p}(\gamma - 1)\right]\Omega_B^2, \quad (2)$$

where ρ is the density, κ is the thermal conductivity, and $\gamma = C_p/C_v$. The code implementing the Tenti model (version S6) was based on that of Pan *et al.* [11], and was used for previous studies on spontaneous and coherent RB scattering in gases [12,13]. This method via the Tenti model must be followed for extracting η_b in gases where the central Rayleigh peak overlays the Brillouin side peak, unlike for liquids where the Brillouin features are fully isolated and η_b can be determined directly by measuring the width $\varGamma_B$ [14].

Details of the experimental setup and methods for measuring high signal-to-noise RB scattering profiles have been reported in [15]. The profiles are recorded for scattering at $\theta = 90°$ induced by an effective intracavity circulating power of 5 W at $\lambda = 366.8$ nm, via a plano-concave Fabry–Perot interferometer with an instrument linewidth of 232 MHz. For each measurement, the scattering cell is initially charged to one of the designated pressures, namely 1 or 3 bar, at room temperature, followed by sealing the cell and then setting the temperature to one of the designated values: 255, 275, 297, or 336 K. The actual pressure of each measurement thus differs from the initial pressure, while the number density of the gas molecules remains the same. The actual pressure is derived via the ideal gas law.

Scattering profiles of N_2 at eight different (p, T) pressure–temperature combinations are shown as black dots in Fig. 1. Since the effect of η_b is most significant at the highest pressures, where the Brillouin side peaks become pronounced (see Fig. 1), the data recorded with an initial pressure of 3 bar are used for determining η_b.

Figure 2 elucidates the method for extracting a value for η_b in the comparison of the Tenti S6 model with the RB profiles for the specific recording of an RB profile in N_2 under conditions $T = 336.6$ K and $p = 3.40$ bar. Figure 2(a) shows the measurement (black dots) and the modeled scattering profiles for three different values of bulk viscosity, and for values of the N_2 transport coefficients as obtained from the literature (listed in Table 1). For the dimensionless internal specific heat capacity of internal degrees of freedom c_{int}, a value of 1 is used throughout. Residuals between the measurement and the three modeled scattering profiles are shown in Fig. 2(b). Figure 2(c) shows a χ^2 calculation as a function of bulk viscosity employed in the Tenti S6 model.

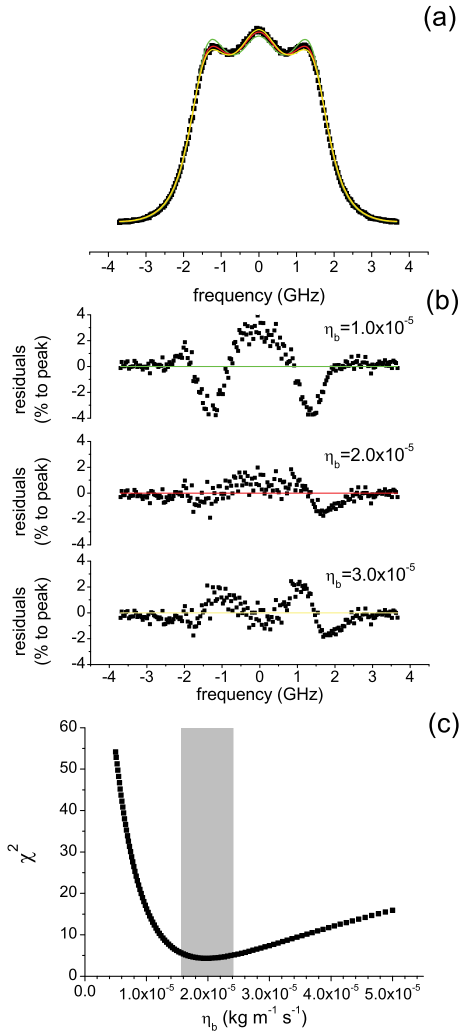

Fig. 2. (Color online) (a) Experimental RB scattering profile in N_2 for 3.40 bar and 336.6 K (black dots), and convolved Tenti S6 calculations for bulk viscosity being 1.0×10^{-5} (green line), 2.0×10^{-5} (red line), and 3.0×10^{-5} (yellow line) $\text{kg} \cdot m^{-1} \cdot \text{s}^{-1}$, respectively. (b) Residuals between measured and calculated scattering profiles for these three values of bulk viscosity. (c) Plot of χ^2 as a function of bulk viscosity. The optimized value of bulk viscosity is found at the minimum of χ^2, with the gray area indicating the estimated statistical error, calculated according to procedures discussed in [12,18].

This procedure of optimizing η_b was applied to the RB scattering measurements for initial pressure of 3 bar N_2. The resulting values for η_b and their uncertainties are plotted in Fig. 3, combined with values from the literature. Prangsma *et al.* [4] determined bulk viscosities for N_2 using sound absorption measurements in the

Table 1. Transport Coefficients used for Modeling the RB Profiles of N_2[a]

T (K)	η_s $(\text{kg} \cdot \text{m}^{-1} \cdot \text{s}^{-1})$	κ $(\text{W} \cdot \text{K}^{-1} \cdot \text{m}^{-1})$	η_b $(\text{kg} \cdot \text{m}^{-1} \cdot \text{s}^{-1})$
254.7	1.57×10^{-5}	2.28×10^{-2}	0.7×10^{-5}
275.2	1.67×10^{-5}	2.44×10^{-2}	1.1×10^{-5}
296.7	1.76×10^{-5}	2.52×10^{-2}	1.4×10^{-5}
336.6	1.95×10^{-5}	2.88×10^{-2}	2.0×10^{-5}

[a]Values for η_s and κ are calculated according to the Sutherland formula in [16], and η_b from the present experiment.

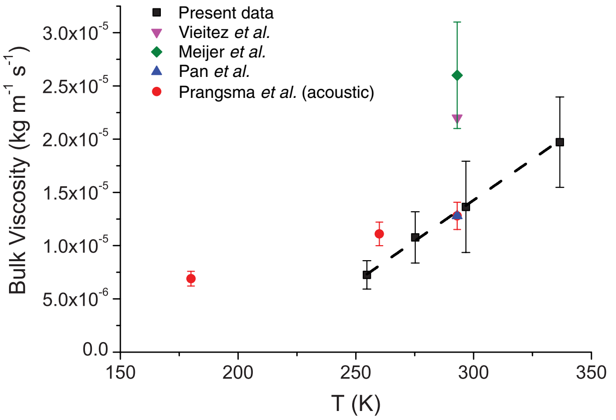

Fig. 3. (Color online) Comparison of bulk viscosity measured from different experiments. Note that the result of Pan *et al.* [11] overlays a data point by Prangsma *et al.* [4]. Data of Vieitez *et al.* [12] and Meijer *et al.* [18] also included.

temperature range $T = 70 \sim 300$ K. The experiment investigated a wide range of acoustic-frequency-to-pressure ratios, but all in the megahertz domain. Pan *et al.* [11] used the value from Prangsma *et al.* [4] and found good agreement between their CRBS profile and a calculation using the Tenti model (the S7 variant) [7], suggesting that the value of bulk viscosity for N_2 obtained at megahertz frequencies is also valid for the gigahertz range. Cornella *et al.* [17] successfully modeled CRBS profiles in N_2 assuming an η_b/η_s ratio of 0.73 from [4], valid at room temperature, and extrapolated this to 500 K. Values previously obtained by Vieitez *et al.* [12] using SRBS at 3 bar N_2 slightly deviate; however, no uncertainty was specified, and if a similar uncertainty is assumed as in the present study, agreement within combined 1σ follows. Meijer *et al.* [18] using CRBS (at 532 nm) at 5 bar N_2 deduce an even larger value, but still agreement within 2σ results.

The present experimental results for η_b in the temperature interval 254–337 K, shown as black dots in Fig. 3 show a linear dependence with temperature, roughly similar to that in [4]. While the data of [4] extend to temperatures as low as 180 K, and the present data extend to 337 K, for the overlapping range 250–300 K good agreement is found. It is assumed that for dilute gases, the bulk viscosity is independent of pressure, similar to shear viscosity and thermal conductivity [16]. The RB profiles recorded for 1 bar N_2 gas, shown in the upper panels of Fig. 1, are modeled with the $\eta_b(T)$ values obtained for 3 bar, also yielding good agreement. While the shear viscosity η_s is known to exhibit a linear temperature dependence in the window 254–337 K [16], the ratio η_b/η_s grows from 0.46 to 1.01 for the present data. This behavior may be related to the freezing out of internal degrees of freedom at lower temperatures.

A general conclusion is drawn that for pure nitrogen (N_2) gas, the bulk viscosity at acoustic frequencies in the megahertz regime is the same as that for hypersound frequencies in the gigahertz regime. This result is surprising in view of the results in carbon dioxide (CO_2) gas, where differences by orders of magnitude were found [6].

This work has been supported by the European Space Agency contract no. 21369 under the supervision of Anne Grete Straume and Olivier Le Rille. The code for computing the Tenti S6 model was obtained from Xingguo Pan and modified by Willem van de Water.

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