Temperature Retrieval Error in Rayleigh-Brillouin Scattering using Tenti's S6 Kinetic Model

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Abstract. The Rayleigh-Brillouin spectrum of light scattered by gas density fluctuations contains information about the gas temperature. This information can be retrieved by comparing experimentally measured spectra with theoretical line shapes determined from the linearized Boltzmann equation. However, the linearized Boltzmann equation is difficult to solve so Tenti's S6 kinetic model has been widely used for several decades because of its simplicity. In this paper, the linearized Boltzmann equation is solved by the efficient fast spectral method, and the temperature retrieval error associated with the Tenti's S6 model is systematically investigated for both spontaneous and coherent Rayleigh-Brillouin scattering, for different gas rarefaction and intermolecular potentials. Our results indicate useful calibrations for laser technologies that use Rayleigh-Brillouin scattering to profile for gas temperatures with high accuracy.

Introduction

Advances in laser technologies have paved the way for non-intrusive measurement of gas properties, which is essential in applications like remote atmospheric temperature profiling. However, due to the low Raman scattering cross-section, current methods based on rotational Raman scattering are still not powerful enough to measure daytime atmospheric temperature. Rayleigh-Brillouin scattering (RBS), the scattering of light from gas density fluctuations, mitigates this difficulty because of its large scattering cross-section. It has become an important tool for non-intrusive measurement of gas properties. For instance, spontaneous RBS (where the gas density changes due to spontaneous thermal fluctuations) is used to retrieve daytime atmospheric temperatures [1]. The coherent RBS (where the variation of gas density is driven by external optical potentials) is a promising technique that enables detection of nanoparticles and large molecules with high spatial-temporal resolution [2]; it also has the potential to be used for dynamic measurement of the gas properties in transient environments such as high-speed flows and combustion [3].

The process of extracting information from the gas involves the comparison of experimental and theoretical RBS spectral profiles, where the accuracy of the resulting gas properties depends significantly on the reliability of the experimental and analytical data. Improvements in experimental techniques have been made recently [4, 5, 3, 6], however there is a problem in producing accurate theoretical spectral profiles.

Theoretically, fluctuation in gas density is described by the linearized Boltzmann equations (LBE), and the RBS spectrum depends critically on the Knudsen number Kn (the ratio of the molecular mean free path to the scattering wavelength). When Kn is small, the gas is in the hydrodynamic regime and its density fluctuations can be simply described by the Navier-Stokes-Fourier equations. On the contrary, when Kn approaches infinity, the gas is collisionless and the RBS spectrum is Gaussian, with the full width at half maximum proportional to the mean thermal speed of the gas molecules [7]. In the intermediate region of the Knudsen number, however, analytical expression for the RBS spectrum is difficult to find, and the LBE must be solved numerically to obtain accurate RBS spectra. Due to the complicated nature of the collision integral of the LBE (i.e. a five-fold integral), the Tenti's S6 kinetic model has been widely used and its line shapes are regarded as the most accurate [8, 9, 10, 11]. However, previous experiments using spontaneous RBS have shown that He, Ne and Ar have 3% difference in their spectra even when the Knudsen numbers are the same [12]. Recently, the Tenti's S6 model was used to retrieve gas temperature by comparing its

spectrum with the experimental one and a maximum 5.2% difference in temperature was found when using coherent RBS [13]. Whether the temperature retrieval error (TRE) is caused by the Tenti's S6 kinetic model or experimental measurement remains unclear.

Note that the S6 model is only derived for the Maxwell molecular model, where the shear viscosity of gas is proportional to the temperature. This may not be accurate for real gases, e.g. the shear viscosity of air at standard temperature and pressure is proportional to the temperature raised to the power 0.74. Our recent numerical solutions of the LBE showed that different intermolecular potentials result in quite different coherent RBS spectra at the same Knudsen number [14, 15], and using the S6 model to extract the gas bulk viscosity from the experimental data could lead to an error of more than 100% [15]. Here we systematically investigate the TRE associated with the Tenti's S6 model, and we focus specifically on the influence of intermolecular potentials on the TRE.

The gas kinetic theory

In gas kinetic theory, the microscopic system state is described by the velocity distribution function (VDF), and all macroscopic quantities, such as density and temperature, are velocity moments of the VDF. For spontaneous RBS in monatomic gases, the evolution of the VDF $h(\mathbf{x}, \mathbf{v}, t)$ is governed by the following LBE:

$$\frac{\partial h}{\partial t} + \mathbf{v} \cdot \frac{\partial h}{\partial \mathbf{x}} = \mathcal{L}_g(h) - \nu_{eq}(\mathbf{v})h, \quad (1)$$

where $\mathcal{L}_g(h) = \int \int B[f_{eq}(\mathbf{v}')h(\mathbf{v}') + h(\mathbf{v}')f_{eq}(\mathbf{v}_*) - f_{eq}(\mathbf{v})h(\mathbf{v}_*)]d\Omega d\mathbf{v}_*$ is the gain part of the linearized Boltzmann collision operator, and $\nu_{eq}(\mathbf{v}) = \int \int Bf_{eq}(\mathbf{v}_*)d\Omega d\mathbf{v}_*$ is the equilibrium collision frequency [16], with $f_{eq}(\mathbf{v}) = \pi^{-3/2}e^{-|\mathbf{v}|^2}$ being the global equilibrium VDF. Note that $\mathbf{v}$ and $\mathbf{v}_*$ are the molecular velocities of the first and second molecule during a binary collision, and Ω is the solid angle. The molecular velocity $\mathbf{v}$, the spatial coordinate $\mathbf{x}$, and the time have been normalized by the most probable velocity $v_m = \sqrt{2k_B T_0/m}$, the effective wavelength λ , and λ/v_m , respectively, where k_B is the Boltzmann constant, T_0 is the equilibrium temperature of the gas, and m is the molecular mass.

The Boltzmann collision kernel B is determined by the intermolecular potential that affects the gas transport properties. Here we consider inverse power-law potentials, where the gas shear viscosity μ is proportional to T^ω , with ω being the viscosity index whose values for typical gases can be found in [17]. Specifically, a Maxwell gas has $\omega = 1$ and a hard-sphere gas has $\omega = 0.5$, while nitrogen at standard temperature and pressure has $\omega = 0.74$. We choose $B = 5|v - v_*|^{2(\omega-1)} \sin^{1/2-\omega} \theta / 2^{15/2-2\omega} \Gamma^2\left(\frac{9-2\omega}{4}\right) Kn$, where θ is the deflection angle after the binary collision, Γ is the gamma function, and the Knudsen number is [16]

$$Kn = \frac{\mu(T_0)}{P_0 \lambda} \sqrt{\frac{\pi k_B T_0}{2m}}, \quad (2)$$

with P_0 and T_0 being the equilibrium pressure and temperature, respectively. The Knudsen number is related to the y parameter frequently used in RBS experiments and Tenti's S6 model: $Kn = (4\sqrt{\pi}y)^{-1}$.

The initial condition in spontaneous RBS is a density impulse $h(\mathbf{x}, \mathbf{v}, 0) = f_{eq}(\mathbf{v})\delta(\mathbf{x})$ [18]. Taking the Laplace-Fourier transform of Eq. (1) in the temporal-spatial domain to find the RBS spectra, we obtain

$$2\pi i(f_s - \nu_x)\hat{h} = f_{eq}(\mathbf{v}) + \mathcal{L}_g(\hat{h}) - \nu\hat{h}. \quad (3)$$

Here f_s is the frequency difference (shift) normalized by the characteristic frequency $\sqrt{2k_B T_0/m}/\lambda$, and a hat denotes the Laplace-Fourier transform of the corresponding quantity. Note that $\hat{h}$ can be solved in the following iterative manner:

$$\hat{h}^{j+1}(\mathbf{v}) = \frac{f_{eq}(\mathbf{v}) + \mathcal{L}_g(\hat{h}^j)}{2\pi i(f_s - \nu_x) + \nu_{eq}(\mathbf{v})}, \quad (4)$$

where j is the iteration step, and $\mathcal{L}_g(\hat{h})$ and $\nu_{eq}(\mathbf{v})$ can be solved by the fast spectral method effectively and accurately [16]. The spontaneous RBS spectrum is given by

$$S(Kn, \omega, f_s) = \int Re(\hat{h})d\mathbf{v}, \quad (5)$$

Figure 1. (Color online) Spectra of spontaneous and coherent RBS when $Kn = 0.07$ from the LBE (lines) and Tenti's S6 kinetic model (circles). Solid lines: hard-sphere gas with $\omega = 0.5$; dashed lines: $\omega = 0.74$; dash-dotted lines: Maxwell gas with $\omega = 1$. The area below each curve are normalized to 1.

where Re represents the real part of a complex number.

Given the Knudsen number Kn , viscosity index ω , and frequency shift f_s , the iteration is terminated when the difference in S between two consecutive iteration steps is less than 10^{-8} .

In coherent RBS experiments, gas molecules are under optical confinement induced by the interference of two pump laser beams. The beam intensities are so small that the spectrum of the scattered light can be obtained by solving the following LBE:

$$\frac{\partial h}{\partial t} + \mathbf{v} \cdot \frac{\partial h}{\partial \mathbf{x}} - \cos(x - f_s t) v_x f_{eq}(\mathbf{v}) = \mathcal{L}_g(h) - v_{eq}(\mathbf{v})h. \quad (6)$$

We take the Fourier transform of Eq. (6) in the temporal-spatial domain, and solve the resultant equation in an iterative manner. Given the frequency difference f_s , Knudsen number Kn , and viscosity index ω , we have

$$\tilde{h}^{j+1}(\mathbf{v}) = \frac{v_x f_{eq}(\mathbf{v}) + \mathcal{L}_g(\tilde{h}^j)}{2\pi i(f_s - v_x) + v_{eq}(\mathbf{v})}, \quad (7)$$

where $\tilde{h}$ is the spatial-temporal Fourier transform of h .

Since in coherent RBS the spectrum is proportional to the square of the density variations, the corresponding RBS spectrum is

$$S(Kn, \omega, f_s) = \left| \int \tilde{h} d\mathbf{v} \right|^2. \quad (8)$$

To obtain spontaneous and coherent RBS spectra of polyatomic gases, we solve the linearized Boltzmann equation by the method proposed by the authors [15].

Numerical results

To analyze the TRE associated with the Tenti's S6 kinetic model, we compare the spectra obtained from the Tenti's S6 model with those from the LBE. Note that the Matlab code for computing the Tenti's S6 model, which has been verified for both the spontaneous and coherent RBS, can be downloaded freely from internet. We first calculate the

Figure 2. (Color online) The variation of the deviation function $Dev = \sum [S_{LBE}(Kn, \omega, f_s, T_0) - S_{Tenti}(Kn, \omega, f_s, T_r)]^2$ as a function of T_t/T_0 when $Kn = 0.07$. The retrieved temperature is the temperature at which the value of Dev is smallest. Left: SRBS; Right: CRBS. Solid lines: hard-sphere gas with $\omega = 0.5$; dashed lines: $\omega = 0.74$; dash-dotted lines: Maxwell gas with $\omega = 1$. The area below each curve are normalized to 1.

Figure 3. (Color online) The relative temperature retrieval (TRE) error of the Tenti's S6 model for spontaneous RBS in monatomic gases. Insets, the relative error in the spectrum between the LBE and Tenti's S6 model at the same temperature, i.e. $(S_{LBE} - S_{Tenti})/\max(S_{Tenti})$. Solid lines: hard-sphere gas with $\omega = 0.5$; dashed lines: $\omega = 0.74$; dash-dotted lines: Maxwell gas with $\omega = 1$.

RBS line shape $S_{\text{LBE}}(Kn, \omega, f_s, T_0)$ from the LBE at a reference temperature T_0 ; some of them are shown in Fig. 1. We then calculate the RBS line shapes $S_{\text{Tenti}}(Kn, \omega, f_s, T_t)$ from the S6 model at various discrete values of T_t around T_0 . Finally, the temperature is retrieved when $Dev = \sum [S_{\text{LBE}}(Kn, \omega, f_s, T_0) - S_{\text{Tenti}}(Kn, \omega, f_s, T_r)]^2$ reaches a minimum value, see the detailed process in Fig. 2. The TRE is $T_r - T_0$ and the relative TRE is $(T_r - T_0)/T_0$.

The relative TRE in spontaneous RBS in a monatomic gas is shown in Fig. 3. The TRE approaches zero at very small Knudsen number and very large Knudsen number. This is because at small Kn the LBE and the S6 kinetic model both lead to the same Navier-Stokes-Fourier equations, while at very large Kn the collision terms of the LBE and the S6 kinetic model vanish so the LBE and the S6 kinetic model are the same. Depending on the Knudsen number, the S6 model could overpredict or underpredict the temperature. For example, for a hard-sphere gas, the Tenti's S6 model overpredicts the temperature when $Kn < 0.3$ and underpredicts the temperature otherwise. The maximum TRE is at $Kn \approx 0.1$, at which the maximum relative TRE error is about 0.2% for a Maxwell gas and 2.2% for a hard-sphere gas. This means for a gas of temperature 300 K, using the Tenti's S6 model to retrieve the temperature produces an error of less than 0.6 K for a Maxwell gas and 6.6 K for a hard-sphere gas. For a gas with viscosity index $\omega = 0.74$, i.e. nitrogen at standard temperature and pressure, the error lies between the Maxwell and hard-sphere gas cases, and the maximum relative TRE is about 1.2%. From this comparison of three different viscosity indices, we conclude that the smaller the viscosity index, the larger the TRE. The fact that a Maxwell gas has the lowest TRE is that the Tenti's S6 model is specifically derived for Maxwell gases.

Figure 4. The relative temperature retrieval (TRE) error of the Tenti's S6 model for coherent RBS in monatomic gases. Insets, the relative error in the spectrum between the LBE and Tenti's S6 model at the same temperature, i.e. $(S_{\text{LBE}} - S_{\text{Tenti}})/\max(S_{\text{Tenti}})$. Solid lines: hard-sphere gas with $\omega = 0.5$; dashed lines: $\omega = 0.74$; dash-dotted lines: Maxwell gas with $\omega = 1$.

The relative TRE in coherent RBS in a monatomic gas is shown in Fig. 4. Here unlike the spontaneous RBS, the Tenti's S6 model always overpredicts the temperature: as Kn increases, the relative TRE first increases and then decreases, and for $Kn > 0.6$ the TRE is roughly proportional to $1/Kn$. As with the spontaneous RBS, the smaller the viscosity index, the larger the TRE. However, here the TRE associated with the Tenti's S6 model is much larger than that in the spontaneous case. The maximum relative TRE reaches nearly 5%, which is close to the experimental error reported in Ref. [13]. If the reference temperature is 300 K, then the maximum TRE for a hard-sphere gas, a gas with $\omega = 0.74$, and a Maxwell gas would be 14.3 K, 9.7 K, and 6.7 K, respectively.

The TRE in both spontaneous and coherent RBS in polyatomic gases at the reference temperature 300 K is shown in Fig. 5, where the magnitude and trend are similar to that for monatomic gases. Interestingly, the ratio of the bulk viscosity to the shear viscosity does not affect the TRE in spontaneous RBS, but does influence the TRE of coherent RBS.

Figure 5. The TRE of the Tenti's S6 model for spontaneous (first row) and coherent (second row) RBS in diatomic (left column) and nonlinear polyatomic (right column) gases with the viscosity index $\omega = 0.74$, at a reference temperature of 300 K, and different ratio of the bulk viscosity μ_b to the shear viscosity μ .

Conclusions

In this paper, we have systematically investigated the temperature retrieval error associated with Tenti's S6 kinetic model for single-species monatomic and polyatomic gases, with different types of inverse power-law potentials. We have found that the intermolecular potential (i.e. the viscosity index) has a significant effect on the temperature measurements. These results could provide temperature calibration to the Tenti's S6 model, and be useful for future laser technologies that employ Rayleigh-Brillouin scattering to profile gas temperatures with high accuracy.

Acknowledgment

This work is financially supported by the UK's Engineering and Physical Sciences Research Council (EPSRC) under grants EP/M021475/1, EP/L00030X/1, EP/K038621/1, EP/I011927/1, and EP/N016602/1. LW acknowledges the financial support of an ECR International Exchange Award from the Glasgow Research Partnership in Engineering.

References

- [1] B. Witschas, C. Lemmerz, and O. Reitebuch, *Opt. Lett.* **39**, 1972–1975 (2014).
- [2] M. N. Shneider and S. F. Gimelshein, *Appl. Phys. Lett.* **102**, p. 173109 (2013).
- [3] A. Gerakis, M. N. Shneider, and P. F. Barker, *Opt. Letts.* **38**, p. 4449 (2013).
- [4] M. O. Vieitez, E. J. van Duijn, W. Ubachs, B. Witschas, A. Meijer, A. S. de Wijn, N. J. Dam, and W. van de Water, *Phys. Rev. A* **82**, p. 043836 (2010).
- [5] Z. Gu and W. Ubachs, *Opt. Lett.* **38**, p. 1110 (2013).
- [6] Z. Gu, W. Ubachs, W. Marques, and W. van de Water, *Phys. Rev. Lett.* **114**, p. 243902 (2015).
- [7] J. H. Grinstead and P. F. Barker, *Phys. Rev. Lett.* **85**, p. 1222 (2000).
- [8] G. Tenti, C. Boley, and R. Desai, *Can J. Phys.* **52**, p. 285 (1974).
- [9] X. Pan, M. N. Shneider, and R. B. Miles, *Phys. Rev. A* **69**, p. 033814 (2004).
- [10] Y. Ma, F. Fan, K. Liang, H. Li, Y. Yu, and B. Zhou, *J. Opt.* **14**, p. 095703 (2012).
- [11] R. Xia, X. He, T. Wu, X. Zhang, and H. Yu, *Optik* **127**, 7262–7267 (2016).
- [12] V. Ghaem-Maghami and A. D. May, *Phys. Rev. A* **22**, p. 692 (1980).
- [13] J. Gaul and T. Lilly, *Opt. Exp.* **22**, 20117–20129 (2014).
- [14] L. Wu, C. White, T. J. Scanlon, J. M. Reese, and Y. Zhang, *AIP Conf. Proc.* **1628**, p. 648 (2014).
- [15] L. Wu, C. White, T. J. Scanlon, J. M. Reese, and Y. H. Zhang, *J. Fluid Mech.* **763**, 24–50 (2015).
- [16] L. Wu, J. M. Reese, and Y. H. Zhang, *J. Fluid Mech.* **746**, 53–84 (2014).
- [17] S. Chapman and T. Cowling, *The Mathematical Theory of Non-uniform Gases* (Cambridge University Press, 1970).
- [18] A. Sugawara, S. Yip, and L. Sirovich, *Phys. Fluids* **11**, p. 925 (1968).