A Study of Linear Waves Based on Extended Thermodynamics for Rarefied Polyatomic Gases

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Abstract We study the dispersion relation for sound in rarefied polyatomic gases basing on the recently developed theory of extended thermodynamics (ET) for both dense and rarefied polyatomic gases. For hydrogen and deuterium gases in a wide temperature range where the rotational and vibrational modes in a molecule play a role, we compare the dispersion relations with those obtained in experiments and by the classical Navier–Stokes Fourier theory. From the comparison with experiments, we estimate the bulk viscosity and evaluate its temperature dependence. We study the characteristics of attenuation in a gas which has a larger relaxation time related to the dynamic pressure than the other relaxation times related to the shear stress and the heat flux by adopting the ET theory with 6 fields.

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1 Introduction

Extended thermodynamics (ET) for both dense and rarefied polyatomic gases has been established [1–3] based on the general methods in the ET theory [4], recently. This is the theory of 14 fields (ET14) of the mass density, velocity, temperature, shear stress, dynamic pressure, and heat flux with two parallel hierarchical series of field equations of balance type. The constitutive equations are determined explicitly by the thermal and caloric equations of state. Also, as a simplified theory of ET14, the ET theory with 6 fields of the mass density, velocity, temperature and dynamic pressure has been proposed (ET6) [5, 6].

After the establishment of the newly developed ET theory, it has been applied to rarefied polyatomic gases. The sound wave [7], light scattering [8], shock wave [9–11] and heat conduction [12, 13] have been studied, and the features of the characteristic velocities have been discussed [14]. From the kinetic theory, it was proved that ET14 is perfectly consistent with molecular extended thermodynamics (MET) basing on the maximum entropy principle [15]. Recently, MET with many fields has been developed, and the complete equivalence between ET and MET was proved [14] as is the case in rarefied monatomic gases [16].

In the previous study of sound waves [7], the dispersion relations described by the ET14 theory were compared with experimental data for hydrogen, deuterium and hydrogen deuteride gases in a temperature range where the only translational and rotational modes are excited, and the validity of the ET theory was demonstrated. Moreover, it is clarified that hydrogen and deuterium gases have large bulk viscosity.

This paper presents the new results on the study of sound waves. Based on the same procedure in [7], the dispersion relation is studied and the following points are reported: (i) the comparison of the phase velocities and attenuations, in hydrogen and deuterium gases in a wide temperature range where the rotational and vibrational modes in a molecule play a role, with those obtained in experiments and by the classical Navier–Stokes Fourier (NSF) theory based on the local equilibrium assumption [17, 18], (ii) the temperature dependence of the bulk viscosity in hydrogen and deuterium gases, and (iii) the effect of the large bulk viscosity on the waves.

The paper is organized as follows. In Sect. 2, we summarize the basic equations necessary for the present analysis and the dispersion relation. In Sect. 3, the dispersion relations in hydrogen and deuterium gases are compared with both experimental data and those derived from the NSF theory. We also evaluate the relaxation times and the bulk viscosity. In Sect. 4, we study the characteristics of the attenuation by using ET6. The last section is devoted to the summary and concluding remarks.

2 Basic Equations and Dispersion Relation

In this section, we summarize the basic equations for the present analysis and deduce the dispersion relation.

2.1 Linearized Field Equations of the ET Theory

In the present study, the independent variables are the mass density ρ, velocity v_i, temperature T, shear stress $S_{\langle ij \rangle}$ (symmetric traceless part of the viscous stress S_{ij}), dynamic pressure $\Pi(= -S_{ii}/3)$ and heat flux q_i, where $i, j = 1, 2, 3$. The stress is expressed by $t_{ij} = -p\delta_{ij} + S_{ij}$ with p being the pressure. The basic equations in the present study are the linearized field equations of ET14 [1] around an equilibrium state. We adopt the equations

of state for a classical (i.e., non-degenerate) ideal gas, when the temperature is not extremely low, expressed by

$$p = \frac{k_B}{m}\rho T, \qquad \varepsilon = \frac{3}{2}\frac{k_B}{m}T + \varepsilon_{\text{int}}(T), \tag{1}$$

where ε, ε_{int}, k_B and m are, respectively, the specific internal energy, specific internal energy due to the internal modes of a molecular such as rotational and vibrational modes, Boltzmann constant and mass of a molecule. Note that gases are, in general, non-polytropic, that is, the specific heat at constant volume $c_v(= \mathrm{d}\varepsilon/\mathrm{d}T)$ is, in general, not constant but depends on the temperature. Then, the basic equations are given by [7]

$$\frac{\partial \rho}{\partial t} + \rho_0 \frac{\partial v_k}{\partial x_k} = 0,$$

$$\rho_0 \frac{\partial v_i}{\partial t} + \frac{k_B}{m}T_0 \frac{\partial \rho}{\partial x_i} + \frac{k_B}{m}\rho_0 \frac{\partial T}{\partial x_i} - \frac{\partial S_{\langle ij \rangle}}{\partial x_j} + \frac{\partial \Pi}{\partial x_i} = 0,$$

$$\frac{k_B}{m}\rho_0 c_v^* \frac{\partial T}{\partial t} + \frac{k_B}{m}\rho_0 T_0 \frac{\partial v_k}{\partial x_k} + \frac{\partial q_k}{\partial x_k} = 0,$$

$$\frac{\partial S_{\langle ij \rangle}}{\partial t} - 2\frac{k_B}{m}\rho_0 T_0 \frac{\partial v_{\langle i}}{\partial x_{j \rangle}} - \frac{2}{1+c_v^*}\frac{\partial q_{\langle i}}{\partial x_{j \rangle}} = -\frac{1}{\tau_S}S_{\langle ij \rangle}, \tag{2}$$

$$\frac{\partial \Pi}{\partial t} + \left(\frac{2}{3} - \frac{1}{c_v^*}\right)\frac{k_B}{m}\rho_0 T_0 \frac{\partial v_k}{\partial x_k} + \frac{2c_v^* - 3}{3c_v^*(1+c_v^*)}\frac{\partial q_k}{\partial x_k} = -\frac{1}{\tau_\Pi}\Pi,$$

$$\frac{\partial q_i}{\partial t} + \left(1 + c_v^*\right)\left(\frac{k_B}{m}\right)^2 \rho_0 T_0 \frac{\partial T}{\partial x_i} - \frac{k_B}{m}T_0 \frac{\partial S_{\langle ik \rangle}}{\partial x_k} + \frac{k_B}{m}T_0 \frac{\partial \Pi}{\partial x_i} = -\frac{1}{\tau_q}q_i,$$

where c_v^* is the dimensionless specific heat at the reference equilibrium state:

$$c_v^* = \frac{c_v(T_0)}{k_B/m}. \tag{3}$$

The quantities with and without the suffix 0 are, respectively, the quantities at the equilibrium state and the deviations from the equilibrium state. By the Maxwellian iteration [1, 4, 19], the relaxation times τ_S, τ_Π and τ_q evaluated at the reference equilibrium state are, respectively, related to the shear viscosity μ, bulk viscosity ν and heat conductivity κ:

$$\mu = \frac{k_B}{m}\rho_0 T_0 \tau_S, \qquad \nu = \left(\frac{2}{3} - \frac{1}{c_v^*}\right)\frac{k_B}{m}\rho_0 T_0 \tau_\Pi, \qquad \kappa = \left(1 + c_v^*\right)\left(\frac{k_B}{m}\right)^2 \rho_0 T_0 \tau_q.$$

2.2 Dispersion Relation, Phase Velocity and Attenuation Factor

We study the one-dimensional problem and consider a plane harmonic wave propagating in the positive x_1-direction with the frequency ω and the complex wave number k such that

$$\mathbf{u} = \mathbf{w}\mathrm{e}^{\mathrm{i}(\omega t - k x_1)},$$

where $\mathbf{u} \equiv (\rho, v_i, T, S_{\langle ij \rangle}, \Pi, q_i)$ and $\mathbf{w}$ is a constant amplitude vector. We assume a longitudinal wave:

$$v_i \equiv \begin{pmatrix} v \\ 0 \\ 0 \end{pmatrix}, \qquad S_{\langle ij \rangle} \equiv \begin{pmatrix} S & 0 & 0 \\ 0 & -\frac{1}{2}S & 0 \\ 0 & 0 & -\frac{1}{2}S \end{pmatrix}, \qquad q_i \equiv \begin{pmatrix} q \\ 0 \\ 0 \end{pmatrix}.$$

Then, by introducing $z = k/\omega$, the dispersion relation is expressed as follows [7, 20]:

$$\frac{c_v^*(c_0 z)^4}{3\Omega^2(1+c_v^*)^2} \left(-3\frac{1+c_v^*}{\tau_{ps}} - i\Omega\left(5c_v^* + \frac{3+7c_v^*}{\tau_{ps}}\right) + 9\Omega^2 c_v^* \right)$$

$$+ \frac{(c_0 z)^2}{3\Omega^3(1+c_v^*)^2} \left[-3i\frac{(1+c_v^*)^2}{\tau_{qs}\tau_{ps}} + \Omega\left(1+c_v^*\right)\left(\frac{5c_v^*}{\tau_{qs}} + 6\frac{1+c_v^*}{\tau_{ps}} + \frac{3+7c_v^*}{\tau_{qs}\tau_{ps}}\right) \right.$$

$$\left. + i\Omega^2\left(c_v^*\left(13+8c_v^*\right) + 9c_v^*\frac{1+c_v^*}{\tau_{qs}} + \frac{6+20c_v^*+10c_v^{*2}}{\tau_{ps}}\right) - 3\Omega^3 c_v^*\left(7+4c_v^*\right) \right]$$

$$+ \frac{(\Omega - i)(\tau_{ps}\Omega - i)(\tau_{qs}\Omega - i)}{\Omega^3 \tau_{ps}\tau_{qs}} = 0, \tag{4}$$

where the following dimensionless values are introduced

$$\Omega = \tau_S \omega, \qquad \tau_{qs} = \frac{\tau_q}{\tau_S} = \left(1+c_v^*\right)^{-1} \frac{\kappa}{\mu k_B/m}, \qquad \tau_{ps} = \frac{\tau_\Pi}{\tau_S} = \left(\frac{2}{3} - \frac{1}{c_v^*}\right)^{-1} \frac{\nu}{\mu}, \tag{5}$$

with the sound velocity in equilibrium defined by $c_0 = \sqrt{\frac{k_B}{m} T_0 (1 + \frac{1}{c_v^*})}$.

From the dispersion relation, the phase velocity v_{ph} and the attenuation factor α are calculated as the functions of the frequency ω:

$$v_{ph}(\omega) = \frac{\omega}{\Re(k)} = \frac{1}{\Re(z)}, \qquad \alpha(\omega) = -\Im(k) = -\omega\Im(z).$$

In addition, it is useful to introduce the attenuation per wavelength:

$$\alpha_\lambda(\omega) = \alpha\lambda = \frac{2\pi v_{ph}\alpha}{\omega} = -2\pi \frac{\Im(z)}{\Re(z)},$$

where λ is the wavelength. Therefore, for given c_v^*, τ_{qs} and τ_{ps}, the quantity z is calculated from Eq. (4) as the function of Ω. The behavior of the phase velocity and the attenuation factor in high frequency limit has been studied in [7].

Hereafter in the present paper, we will confine our study within the *fastest* sound wave because the experiments discussed in Sect. 3 give us the data on this wave.

3 Comparison with Experimental Data

We compare the dispersion relation obtained above, in particular, the phase velocity v_{ph} and the attenuation per wavelength α_λ as the functions of the frequency ω with the experimental data on normal hydrogen (n-H$_2$) and normal deuterium (n-D$_2$) gases at several temperatures listed in Table 1. The comparison is also made with the predictions by the classical NSF theory.

Table 1 Values of the temperature T_0, dimensionless specific heat c_v^*, sound speed in equilibrium c_0, shear viscosity μ [24–26], heat conductivity κ [24–26] and the ratio of the relaxation times of the heat flux and the shear stress τ_{qs}. The values of the parameter φ, bulk viscosity ν, and the ratio of the relaxation times of the bulk viscosity and the shear viscous stress τ_{ps} evaluated in the reference equilibrium state

Gas	T_0 [K]	c_v^*	c_0 [$\frac{m}{s}$]	μ [μPa s]	κ [$\frac{mW}{m K}$]	τ_{qs}	φ	ν [μPa s]	τ_{ps}
n-H$_2$	273.15	2.42	1260	8.33	173	1.47	41.2	343	162
	295.15	2.45	1310	8.95	187	1.47	32.5	291	126
	873.15	2.54	2240	18.6	403	1.48	38.3	713	141
	1073.15	2.60	2480	21.0	462	1.49	41.4	869	147
n-D$_2$	273.15	2.50	888	11.8	136	1.60	26.0	306	97.4
	295.15	2.50	923	12.6	141	1.55	23.4	296	87.7
	773.15	2.60	1490	24.2	260	1.48	32.0	773	113
	1073.15	2.78	1740	30.4	337	1.42	34.8	1060	114

Fig. 1 Dependence of the dimensionless specific heat c_v^* for n-H$_2$ and n-D$_2$ gases on the temperature T. The *circles* and *triangles* are, respectively, adopted values in the present study and the previous study [7]
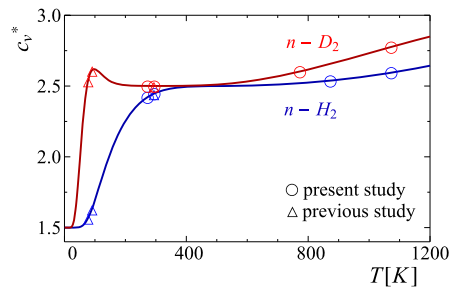
Before discussing the subject, we need to make preliminary calculations for determining the values of c_v^*, τ_{qs} and τ_{ps} defined in (3) and (5).

3.1 Preliminary Calculations

We calculate the specific heat c_v^* of hydrogen and deuterium gases on the basis of statistical mechanics [21, 22]. In the preceding paper [7], we took only the translational and rotational modes into account. In the present paper, to study the dispersion relation at high temperature, we also take into account the vibrational mode described by the harmonic oscillator model. We adopt the rotational constant as 12.09×10^{-22} [J] and 6.047×10^{-22} [J] and the harmonic vibrational frequency as $6332 k_B/\hbar$ [Hz] and $4483 k_B/\hbar$ [Hz] with $\hbar$ being the Planck constant divided by 2π, respectively, for n-H$_2$ and n-D$_2$ [23]. Numerically calculated values of c_v^* are shown in Table 1 and in Fig. 1.

From (5) and with the help of the experimental data on μ, κ and ν, we can in principle, estimate the ratios of relaxation times τ_{qs} and τ_{ps}. However, at present, as we have the suitable data only on μ and κ [24–26], we adopt

$$\varphi = \frac{\nu}{\mu},$$

as an adjustable parameter. We summarize the adopted values of c_v^*, c_0, μ, κ, τ_{qs} and the evaluated values of φ, ν and τ_{ps} in Table 1, details of which will be discussed in the next subsection.
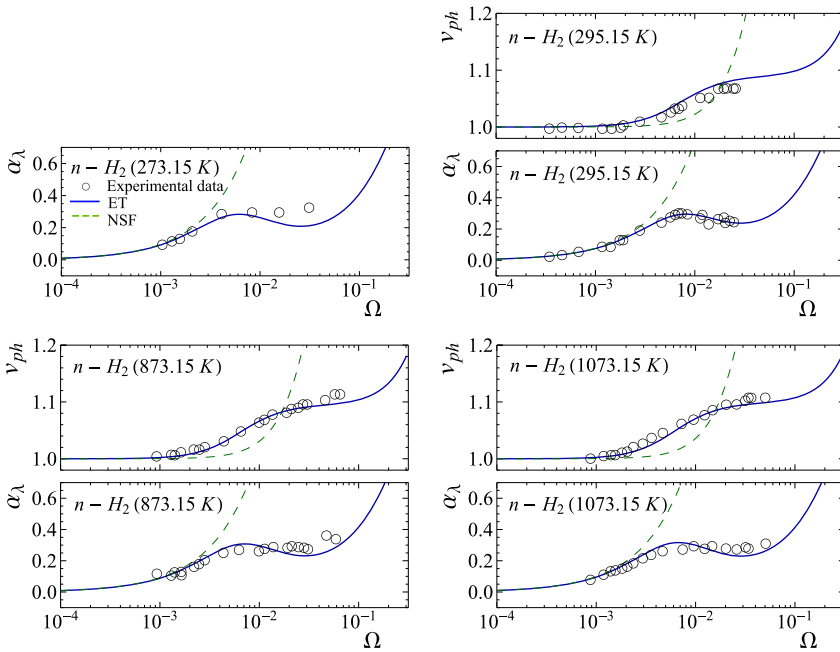
Fig. 2 Dependence of the dimensionless phase velocity v_{ph}/c_0 and the attenuation per wavelength α_λ on the dimensionless frequency Ω for n-H$_2$. The *circles* are the experimental data at $T_0 = 273.15$ K [27], 295.15, 873.15 and 1073.15 K [28]. The *solid* and *dashed lines* are predictions by the ET and NSF theories, respectively

3.2 Experimental Data and Theoretical Predictions for the Dispersion Relation

3.2.1 Hydrogen Gases

For n-H$_2$, the theoretical predictions of the dimensionless phase velocity v_{ph}/c_0 and the attenuation per wavelength α_λ as the functions of the dimensionless frequency Ω by the ET theory are shown in Fig. 2. These are compared with the experimental data and the predictions by the NSF theory for v_{ph}/c_0 at $T_0 = 295.15$, 873.15 and 1073.15 K [28] and for α_λ at $T_0 = 273.15$ K [27], 295.15, 873.15 and 1073.15 K [28].

What is evident from Fig. 2 is the following three points:

(i) In the region with small Ω (under around $\Omega = 10^{-3}$), as is expected, the predictions by the two theories coincide with each other. The values of the parameter φ are determined to be 41.2, 32.5, 38.3 and 41.4 at $T_0 = 273.15$, 295.15, 873.15 and 1073.15 K, respectively, as the best fit of α_λ with the experimental data in this region by using the least-square approach. These adopted values of φ indicate that the order of magnitude of τ_{ps} is 10^2 as shown in Table 1.

(ii) When we go into the ultrasonic frequency region with larger Ω, the ET theory shows its superiority compared with the NSF theory. In particular, this is evidently seen around $\Omega = \omega\tau_s = 10^{-2}$ corresponding to $\omega\tau_\Pi = 1$.

(iii) At all temperatures, the large values of φ are adopted. This means that $\nu \gg \mu$ in a wide temperature range even if the rotational and/or vibrational modes are excited. We will discuss the effect of large bulk viscosity on the attenuation in Sect. 4.
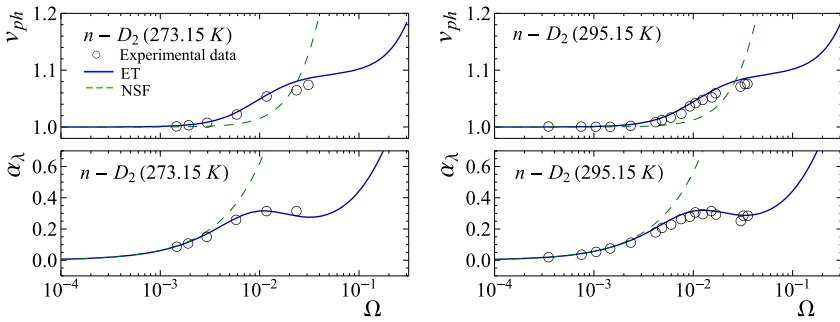
Fig. 3 Dependence of the dimensionless phase velocity v_{ph}/c_0 and the attenuation per wavelength α_λ on the dimensionless frequency Ω for n-D_2. The *circles* are the experimental data at $T_0 = 273.15$ K [27], 295.15, 873.15 and 1073.15 K [28]. The *solid* and *dashed lines* are predictions by the ET and NSF theories, respectively, for each temperature

Fig. 4 The temperature dependence of the bulk viscosity ν for n-H_2 (*solid line*) and n-D_2 (*dashed line*). The *circles* and *squares* are, respectively, the evaluated values of ν for n-H_2 and n-D_2 in the Sect. 3 and in the preceding paper [7]
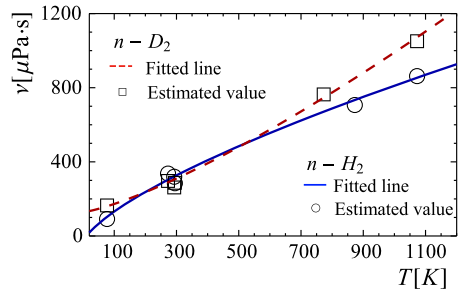
3.2.2 Deuterium Gases

Comparisons are also made for n-D_2 at $T_0 = 273.15$ K [27], 295.15, 773.15 and 1073.15 K with $\varphi = 26.0$, 23.4, 32.0 and 34.8, and shown, respectively, in Fig. 3. From these figures, we have qualitatively the same observations as those in the case of hydrogen gases.

3.3 Temperature Dependence of Bulk Viscosity

With the evaluated values of the bulk viscosity listed in Table 1 and in the preceding paper [7], we can estimate its temperature dependence. If we assume that the bulk viscosity depends only on the temperature [29] and that it is expressed as a power law, the temperature dependence of ν for n-H_2 and n-D_2 in a temperature range from 77 to 1073 K may be approximated as

$$\nu_{\text{n-}H_2} = 6.58T^{0.703} - 36,$$

$$\nu_{\text{n-}D_2} = 0.113T^{1.29} + 129,$$

where the unit is [μPa s]. In Fig. 4, these temperature dependences and estimated values are shown.

With this temperature dependence of ν and a rich store of data on μ and κ [24–26, 30], various non-equilibrium phenomena in rarefied polyatomic gases, e.g., shock waves and heat conduction, can be studied.

4 The Characteristics of the Attenuation

In order to study the effect of the large value of the relaxation time τ_Π on the attenuation, it seems to be appropriate to adopt a simpler model, that is, the ET theory with only 6 independent field variables (ρ, v_i, T, Π) developed in [5, 6] (ET6). In the present section, by using this theory, we study the characteristics of attenuation of the sound waves. Other than the hydrogen and deuterium gases, it has been reported that some gases have the large relaxation time of dynamic pressure such as the carbon dioxide gases and nitrous oxide gases [10, 31].

The linearized basic equations of ET6 for rarefied polyatomic gases are expressed as follows:

$$
\begin{aligned}
&\frac{\partial \rho}{\partial t} + \rho_0 \frac{\partial v_k}{\partial x_k} = 0, \\
&\rho_0 \frac{\partial v_i}{\partial t} + \frac{k_B}{m} T_0 \frac{\partial \rho}{\partial x_i} + \frac{k_B}{m} \rho_0 \frac{\partial T}{\partial x_i} + \frac{\partial \Pi}{\partial x_i} = 0, \\
&\frac{k_B}{m} \rho_0 c_v^* \frac{\partial T}{\partial t} + \frac{k_B}{m} \rho_0 T_0 \frac{\partial v_k}{\partial x_k} = 0, \\
&\frac{\partial \Pi}{\partial t} + \left(\frac{2}{3} - \frac{1}{c_v^*} \right) \frac{k_B}{m} \rho_0 T_0 \frac{\partial v_k}{\partial x_k} = -\frac{1}{\tau_\Pi} \Pi.
\end{aligned}
\tag{6}
$$

The dispersion relation of this system is expressed as follows:

$$
(c_0 z)^2 \left(-\frac{5}{3} \frac{c_v^*}{1 + c_v^*} + \frac{i}{\Omega'} \right) + 1 - \frac{i}{\Omega'} = 0,
\tag{7}
$$

where $\Omega' = \tau_\Pi \omega$ is the dimensionless frequency instead of $(5)_1$. It is proved in [5] that this dispersion relation coincides with that of the Meixner's theory based on the local equilibrium assumption [17, 32, 33].

One of the characteristic values for the study of a sound wave is the ratio of the attenuation per wavelength α_λ and the squared phase velocity [34, 35]:

$$
\beta = \frac{\alpha_\lambda}{(v_{ph}/c_0)^2} = -2\pi \left(\frac{c_0}{\omega} \right)^2 \Re(k) \Im(k).
$$

In Fig. 5, the dependences of β derived from the ET6, ET14 and NSF theories on Ω' for different $\tau_{ps}(= \tau_\Pi/\tau_S)$ with fixed $\tau_{qs}(= \tau_q/\tau_S) = 1$ and $c_v^* = 7.5$ are shown. The following features are pointed out:

(i) We may see that, in the case of large τ_Π shown in the top figure of Fig. 5, ET6 coincides with ET14 in the region with small Ω'. This indicates that ET6 is useful to study the attenuation for a gas which has a large bulk viscosity.

(ii) For ET6, the explicit form of β may be easily calculated as follows:

$$
\beta^{\text{ET6}} = \pi \frac{\Omega'(1 + c_v^*)(2c_v^* - 3)}{25(c_v^* \Omega')^2 + 9(1 + c_v^*)^2}.
\tag{8}
$$

This is a similar form with the well-known single relaxation theory [34, 35]. The frequency dependence of β^{ET6} shows a single-peak curve centered around $\Omega' = 1$ due to the relaxation time τ_Π as we can see in Fig. 5. The expression (8) points out that the

Fig. 5 The dependence of β on Ω' for $\tau_{ps} = 10^4$ (*top*), 10^2 (*medium*) and 1 (*bottom*) with $\tau_{qS} = 1$ and $c_v^* = 7.5$
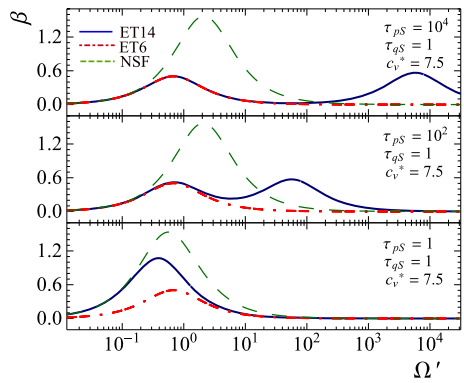
character of the relaxation process is determined only through the specific heat. This is the manifestation of the fact that τ_Π is in the same order of magnitude as the relaxation time of the energy exchange between the molecular translational mode and the internal modes as shown in [5]. In particular, the maximum of β^{ET6} may be calculated as

$$\beta_{\max}^{\mathrm{ET6}} = \frac{\pi(2c_v^* - 3)}{10c_v^*} \quad \text{at } \Omega'_{\max} = \frac{3}{5}\left(\frac{1}{c_v^*} + 1\right). \tag{9}$$

When c_v^* increases, this value monotonically increases. It should be noted that in the limit $c_v^* \to 3/2$, that is, the limit from polyatomic to monatomic rarefied gases, β^{ET6} disappears [36]. On the other hand, in the limit $c_v^* \to \infty$, $\beta_{\max}^{\mathrm{ET6}}$ reaches $\pi/5$ with $\Omega'_{\max} = 3/5$. The behavior of $\beta_{\max}$ is shown in Fig. 6.

(iii) For ET14, β shows a triple-peak curve consisted of three kinds of a single-peak curve corresponding to the effect of the relaxation by the dynamic pressure, shear stress and heat flux, respectively. The positions of the peaks are related to the differences of the order of magnitude of the ratio of relaxation times.

Further studies on this analysis will be reported elsewhere.

5 Summary and Concluding Remarks

Based on the ET theory for rarefied polyatomic gases, we have studied the dispersion relations for hydrogen and deuterium gases in a wide temperature range where the rotational and
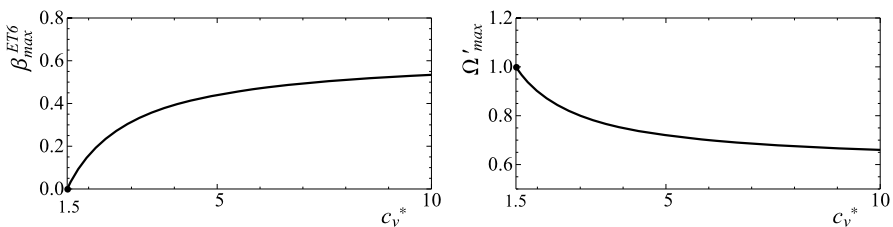
Fig. 6 The dependence of the maximum value of β^{ET6} and the dimensionless frequency of $\Omega'_{\max}$ on the dimensionless specific heat c_v^*

vibrational modes in a molecule play a role. We have properly taken the temperature dependence of the specific heats of these gases into the dispersion relations. The comparison of the theoretical predictions with experimental data on the phase velocity and the attenuation per wavelength has revealed that the ET theory is valid even in the case that the vibrational mode of a molecule is excited. We have also evaluated the temperature dependence of the bulk viscosity and the relaxation times. We have clarified the effect of the large bulk viscosity on the attenuation, and have shown the usefulness of the ET6 theory.

Finally some concluding remarks are made:

(i) The study of the sound wave has usually been done in the framework of thermodynamics of irreversible processes [17, 34, 35] based on the local equilibrium assumption. In the present paper, we have compared the ET theory with the NSF theory as a representative one, and have demonstrated the superiority of the ET theory. We have also shown that the Meixner's theory is included as a special case of the ET theory. These results indicate that the ET theory is valid beyond the local equilibrium assumption.

(ii) We have analyzed the experimental data only on rarefied hydrogen and deuterium gases. The ET theory can be applied to many other rarefied polyatomic gases. Comprehensive study of this must be a promising future work.

(iii) As a future study, it is interesting to study the dispersion relation for sound in dense gases.

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