Thermal Relaxation Rate in Viscous Multi-Temperature Gas Flows

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Abstract. Rate of thermal relaxation in viscous multi-temperature flows is studied on the basis of the modified Chapman-Enskog method. A single-component gas with slow VT (vibration-translation) transitions is considered, and the relaxation rate is studied in the zero- and first-order approximations. The first-order corrections to the relaxation rate depending on the velocity divergence and difference between vibrational and translational-rotational temperatures are derived, and their estimates are carried out for different flow conditions. For the specific case of harmonic oscillators, the cross terms in the first-order relaxation rate and pressure tensor are obtained in the symmetric form allowing one to prove the reciprocal Onsager relations for the given case.

INTRODUCTION

Modeling of thermal non-equilibrium viscous flows is a challenging task in computational fluid dynamics (CFD) due to their importance in re-entry problems and other modern applications. In multi-temperature flows, the set of governing equations includes conservation equations for mass, momentum and total energy coupled to relaxation equations for the internal energy modes and equations of chemical kinetics (for chemically reacting mixtures). An important point for the accurate prediction of flow parameters is a reliable model for the internal energy production term in the relaxation equations. In CFD, the commonly used expression for the internal energy production rate $\dot{u}_{int}$ (in the absence of chemical reactions, when coupling between reactions and internal energy is not applicable) is

$$\dot{u}_{int} = \rho \frac{u_{int}(T) - u_{int}(T_{int})}{\tau_{int}} \qquad (1)$$

where ρ is the density, T is the gas temperature, T_{int} is the internal temperature, τ_{int} is the relaxation time for the internal energy basically calculated using empirical data. This expression is exact only for the specific case of vibrational relaxation of harmonic oscillators; its derivation requires the Landau-Teller representation of the rate coefficients for vibrational energy transitions and prohibits multi-quantum jumps [1]. For all other kinds of internal energy relaxation, Eq. (1) is approximate and requires additional justification. Moreover, for any kind of internal energy relaxation, the expression (1) is valid only in inviscid gas flows and cannot be used for the Navier-Stokes equations. The main objective of this study is to find the limits of validity of Eq. (1).

Actually, the problem is not new. The rates of strongly non-equilibrium processes in viscous flows were derived theoretically by many authors. Thus non-equilibrium chemical reactions in gases without internal degrees of freedom were considered in Refs. [2, 3, 4], vibrationally excited gases in the state-to-state approach were studied in Refs. [5, 6, 7], the case of coupled vibrational relaxation and dissociation was discussed in Refs. [8, 9, 10]. Recently, the most general theoretical model for any chemical reaction taking into account internal degrees of freedom and non-linear effects was developed in [11], and the symmetric cross coupling terms between the normal mean stress and rates of chemical reactions were derived. In the above references, it was shown that in viscous flows, the rates of non-equilibrium processes include the first-order corrections which, depending on the flow conditions, may be specified by various parameters: mixture composition, velocity divergence, affinities of chemical reactions.

Although the problem received much attention in the literature, it is not completely resolved until the present time. Indeed, the most studies are focused on the rates of chemical reactions, whereas the rate of multi-temperature

vibrational relaxation was not discussed up to now (to the best of our knowledge). Moreover, no numerical estimates of the first-order corrections to the relaxation rate were performed.

The objective of this study is to derive the rate of vibrational relaxation in the zero- and first-order approximations of the modified Chapman-Enskog method [7]. For the sake of simplicity, the case of vibrational relaxation in a single-component gas flow is considered, with the only relaxation channel through vibration-translation VT transitions. The first-order correction to the relaxation rate is obtained as a function of the velocity divergence and of the difference between translational-rotational and vibrational temperatures. For the particular case of harmonic oscillators, the first-order correction is derived in the symmetric form, which makes it possible to determine properly the cross coupling terms between the relaxation rate and pressure tensor and to prove the reciprocal Onsager–Casimir relations for the kinetic coefficients. Numerical estimates of the first-order correction to the relaxation rate are carried out for different flow conditions.

GOVERNING EQUATIONS AND ZERO-ORDER SOLUTION

Consider the case of vibrational relaxation in a single-component diatomic gas A_2 of harmonic oscillators under the condition

$$\tau_{tr} < \tau_{rot} < \tau_{VV} \ll \tau_{VT} \sim \theta \tag{2}$$

where τ_{tr}, τ_{rot}, τ_{VV}, and τ_{VT} are, respectively, the characteristic times of translational, rotational, vibration-vibration, and vibration-translation relaxation, θ is the characteristic gas-dynamic time. This case was widely studied in the literature for spatially homogeneous gases and inviscid flows [1]. In the recent paper [12] it is shown that rotational transitions may contribute to the vibrational kinetics, however in the present work we do not take into account rovibrational exchanges.

Under the condition (2), the set of collision invariants of rapid processes includes: mass m, momentum $m\mathbf{u}$ ($\mathbf{u}$ is the particle velocity), total energy $mu^2/2 + \varepsilon_i + \varepsilon_j$ (ε_i and ε_j are, respectively, vibrational and rotational energies of the levels i and j), and vibrational energy ε_i. The corresponding set of macroscopic flow variables consists of density ρ, macroscopic gas velocity $\mathbf{v}$, total specific energy u, and specific vibrational energy u_v, or, which is essentially the same, ρ, $\mathbf{v}$, gas temperature T, and vibrational temperature T_v.

Starting from the Boltzmann equation, the set of governing equations is derived in the form

$$\frac{d\rho}{dt} + \rho \nabla \cdot \mathbf{v} = 0, \tag{3}$$

$$\rho \frac{d\mathbf{v}}{dt} + \nabla \cdot \boldsymbol{P} = 0, \tag{4}$$

$$\rho \frac{du}{dt} + \nabla \cdot \mathbf{q} + \boldsymbol{P} : \nabla \mathbf{v} = 0, \tag{5}$$

$$\rho \frac{du_v}{dt} + \nabla \cdot \mathbf{q}_v = \dot{u}_v, \tag{6}$$

where $\mathbf{q}$, $\mathbf{q}_v$ are the total internal energy and vibrational energy diffusive fluxes, $\boldsymbol{P}$ is the pressure tensor, $\dot{u}_v$ is the rate of vibrational energy production due to VT transitions:

$$\dot{u}_v = \sum_{ij} \varepsilon_i \int J_{ij}^{VT} d\mathbf{u} = \frac{1}{2} \sum_{ii'} \Delta\varepsilon_{ii'} \sum_j \int J_{ij}^{VT} d\mathbf{u} = \frac{1}{2} \sum_{ii'} \Delta\varepsilon_{ii'} \sum_{kk'} \sum_{jlj'l'} \int \left(f_{i'j'} f_{k'l'} \frac{s_{ij}s_{kl}}{s_{i'j'}s_{k'l'}} - f_{ij}f_{kl} \right) g\sigma_{ijkl}^{i'j'k'l'} d^2\Omega d\mathbf{u}_1. \tag{7}$$

Here, $\Delta\varepsilon_{ii'} = \varepsilon_{i'} - \varepsilon_i$ is the vibrational energy variation in a VT transition $A_2(i) + M = A_2(i') + M$, J_{ij}^{VT} is the collision operator for VT exchange (see, for instance [7]), f_{ij} is the distribution function, s_{ij} is the statistical weight. The differential cross section $\sigma_{ijkl}^{i'j'k'l'}$ corresponds to a collision between particles on the internal levels i, j, k, l moving with relative velocity $\mathbf{g}$, which results in the transition to internal levels i', j', k', and l' whereas the relative velocity vector after collision appears within the solid angle $d^2\Omega$; $\mathbf{u}_1$ is the partner velocity. We will assume hereafter that the vibrational state of a partner M does not vary during the VT transition, i.e. $k = k'$.

The zero-order distribution function corresponding to the Eq. (2) is well known in the literature [1, 7, 10]

$$f_{ij}^{(0)} = \left(\frac{m}{2\pi kT} \right)^{3/2} \frac{n_i s_j}{Z_{rot}(T)} \exp\left(-\frac{mc^2}{2kT} - \frac{\varepsilon_j}{kT} \right) \tag{8}$$

and represents the Maxwell-Boltzmann distribution over velocities and rotational energy with the gas temperature T and non-equilibrium Boltzmann distribution over vibrational energy with the vibrational temperature T_v

$$n_i = \frac{n}{Z_{vibr}(T_v)} \exp\left(-\frac{\varepsilon_i}{kT_v}\right). \tag{9}$$

Here, k is the Boltzmann constant, $\mathbf{c} = \mathbf{u} - \mathbf{v}$ is the peculiar velocity, Z_{rot} and Z_{vibr} are the rotational and vibrational partition functions, n_i are the vibrational level populations, the vibrational statistical weight $s_i = 1$.

Based on this distribution function, the transport and production terms for an inviscid flow can be calculated. The transport terms take the usual form $\mathbf{P}^{(0)} = p\mathbf{I}$ ($p = nkT$), $\mathbf{q}^{(0)} = 0$. The rate of vibrational relaxation can be calculated substituting Eq. (8) into Eq. (7) and using the microscopic energy conservation. After some transformations, we obtain:

$$\dot{u}_v^{(0)} = -\frac{1}{2}n\sum_{ii'} \omega_{ii'} \Delta\varepsilon_{ii'} n_i k_{ii'}^{(0)}, \tag{10}$$

where $k_{ii'}^{(0)}$ is the zero-order state-specific rate coefficient of the VT transition

$$k_{ii'}^{(0)} = \sum_{jlj'l'} \int \frac{f_{ij}f_{kl}}{n_i n_k} g\sigma_{ijkl}^{i'j'k'l'} d^2\Omega d\mathbf{u}\, d\mathbf{u}_1, \tag{11}$$

and $\omega_{ii'}$ is the parameter characterizing vibrational relaxation

$$\omega_{ii'} = \exp\left(-\frac{\Delta\varepsilon_{ii'}}{kT}\frac{T - T_v}{T_v}\right) - 1. \tag{12}$$

For the case of weak deviations from equilibrium, when $|(T - T_v)/T_v| \ll 1$, this parameter reduces to

$$\omega_{ii'} \approx -\frac{\Delta\varepsilon_{ii'}}{kT}\frac{T - T_v}{T_v} \tag{13}$$

and becomes linear function of the temperature difference. Consequently, for weak disequilibrium, the vibrational energy production depends linearly on $(T - T_v)/T_v$.

Therefore, if the zero-order state-specific rate coefficients are known, the energy production can be easily calculated using Eq. (10). The rate coefficients $k_{ii'}^{(0)}$ can be calculated either by direct integration (see Eq. (11)) for a given cross section or using some analytical or semi-empirical models (SSH, FHO, etc.,..., see [13] for the details).

It can be shown that the expression (10) can be reduced to Eq. (1) if the following assumptions are adopted: 1) the molecular vibrations are modelled by harmonic oscillators with infinite number of levels; 2) only single-quantum transitions are allowed; 3) the state-specific rate coefficients are connected by the relation $k_{i+1,i}^{(0)} = (i+1)k_{10}^{(0)}$. Thus, equation (1) represents a particular case of Eq. (10) for the above restrictions. In the next section we will show that in a viscous flow, Eq. (1) is not valid even for the assumptions 1)–3).

FIRST-ORDER SOLUTION

Following the procedure of the modified Chapman-Enskog method, the first-order distribution function $f_{ij}^{(1)}$ is obtained as a function of the derivatives of the macroscopic parameters:

$$f_{ij}^{(1)} = f_{ij}^{(0)}\left(-\frac{1}{n}\mathbf{A}_{ij}\cdot\nabla\ln T - \frac{1}{n}\mathbf{A}_{ij}^{(v)}\cdot\nabla\ln T_v - \frac{1}{n}\mathbf{B}_{ij}:\nabla\mathbf{v} - \frac{1}{n}F_{ij}\nabla\cdot\mathbf{v} - \frac{1}{n}G_{ij}\right), \tag{14}$$

where functions $\mathbf{A}_{ij}$, $\mathbf{A}_{ij}^{(v)}$, $\mathbf{B}_{ij}$, F_{ij}, and G_{ij} are found from the linear integral equations [7]. The functions $\mathbf{A}_{ij}$, $\mathbf{A}_{ij}^{(v)}$, $\mathbf{B}_{ij}$ determine the energy fluxes and the non-diagonal elements of the pressure tensor. These transport terms as well as associated transport coefficients of thermal conductivity and shear viscosity are widely studied in the literature [7, 10]. The first-order energy production rate $\dot{u}_v$ is specified by the scalar functions F_{ij} and G_{ij} and up to the present time was not thoroughly investigated.

Starting from the integral equations and auxiliary conditions for F_{ij} and G_{ij} given in [7], using the expansion of unknown functions in the series of orthogonal Sonine and Waldmann-Trübenbacher polynomials

$$F_{ij} = \sum_{rp} f_{rp} S_{1/2}^{(r)}\left(\frac{mc^2}{2kT}\right) P_j^{(p)}\left(\frac{\varepsilon_j}{kT}\right), \qquad G_{ij} = \sum_{rp} g_{rp} S_{1/2}^{(r)}\left(\frac{mc^2}{2kT}\right) P_j^{(p)}\left(\frac{\varepsilon_j}{kT}\right), \tag{15}$$

keeping the first non-zero terms of the expansions (15), and assuming that the specific heats of translational and rotational modes are constant ($c_{tr} = 3k/2m$, $c_{rot} = k/m$), one can derive linear algebraic systems for the expansion coefficients f_{rp} and g_{rp}. Thus the system for g_{rp} after simplifications takes the form

$$\beta_{1100} g_{10} + \beta_{1001} g_{01} = \frac{3}{5kT} \dot{u}_v^{(0)} - \sum_{ij} \int J_{ij}^{VT(0)} S_{1/2}^{(1)} P_j^{(0)} d\mathbf{u}$$

$$\frac{3}{2} g_{10} + g_{01} = 0. \tag{16}$$

Here $\beta_{rr'pp'} = \left[S_{1/2}^{(r)} P_j^{(p)}, S_{1/2}^{(r')} P_j^{(p')} \right]$ is the bracket integral over the cross sections of rapid processes which can be expressed in terms of the rotational relaxation time [7, 10]. The system for the coefficients f_{rp} looks similarly to (16), except that the right-hand side of the first equation should be replaced by $-2/5$.

Calculating the integral in the right-hand side of Eq. (16).1 yields

$$\sum_{ij} \int J_{ij}^{VT(0)} S_{1/2}^{(1)} P_j^{(0)} d\mathbf{u} = -\frac{1}{2} n \sum_{ii'} \frac{\Delta\varepsilon_{ii'}}{kT} \omega_{ii'} n_i k_{ii'}^{(0)} - \frac{1}{4} \sum_{ii'kk'} \omega_{ii'} \sum_{jj'll'} \frac{\Delta\varepsilon_{jlj'l'}^{rot}}{kT} \int f_{ij} f_{kl} g \sigma_{ijkl}^{i'j'k'l'} d^2\Omega d\mathbf{u} d\mathbf{u}_1. \tag{17}$$

It is interesting to emphasize that the first-order correction to the rate of vibrational relaxation depends, in the general case, on the variation of rotational energy of colliding particles $\Delta\varepsilon_{jlj'l'}^{rot} = \varepsilon_{j'} + \varepsilon_{l'} - \varepsilon_j - \varepsilon_l$. However, it is reasonable to assume that $\Delta\varepsilon_{jlj'l'}^{rot} \ll \Delta\varepsilon_{ii'}$ and thus to neglect the second term in Eq. (17). Indeed, such an assumption is not justified only for light gases like H_2, D_2 at low temperature conditions. Therefore, taking into account Eq. (10), we obtain

$$\sum_{ij} \int J_{ij}^{VT(0)} S_{1/2}^{(1)} P_j^{(0)} d\mathbf{u} = \frac{\dot{u}_v^{(0)}}{kT} \tag{18}$$

which allows us to reduce the right-hand side of Eq. (16).1 to $-2\dot{u}_v^{(0)}/5kT$.

Let us now calculate the first-order relaxation rate. If we substitute the first-order distribution function (14) into Eq. (7) then after some transformations we can found

$$\dot{u}_v = \dot{u}_v^{(0)} + \dot{u}_v^{(1)} = \dot{u}_v^{(0)} - \frac{1}{n}\left(f_{10}\nabla\cdot\mathbf{v} + g_{10}\right)\left(3\dot{u}_v^{(0)} - \frac{3}{4} n \sum_{ii'}\left(\frac{\Delta\varepsilon_{ii'}}{kT}\right)^2 n_i k_{ii'}^{(0)} + I\left(\Delta\varepsilon_{jlj'l'}^{rot}\right)\right) \tag{19}$$

where $I\left(\Delta\varepsilon_{jlj'l'}^{rot}\right)$ is the integral depending on the variation of rotational energy similar to that in Eq. (17). Again, if we assume $\Delta\varepsilon_{jlj'l'}^{rot} \ll \Delta\varepsilon_{ii'}$, this integral can be neglected compared to other terms in the parenthesis. Then, taking into account Eq. (10), we obtain the first-order correction $\dot{u}_v^{(1)}$ in the final form

$$\dot{u}_v^{(1)} = \frac{3}{2}\left(f_{10}\nabla\cdot\mathbf{v} + g_{10}\right)\sum_{ii'}\Delta\varepsilon_{ii'} n_i k_{ii'}^{(0)}\left(\omega_{ii'} + \frac{1}{2}\frac{\Delta\varepsilon_{ii'}}{kT}\right). \tag{20}$$

It should be noted that the first-order correction to the relaxation rate depends on the velocity divergence $\nabla\cdot\mathbf{v}$ and on the temperature difference $(T - T_v)/T_v$ through the parameter $\omega_{ii'}$. These corrections appear due to spatial non-homogeneity, compressibility and deviations from the Maxwell-Boltzmann distributions. It is clear that Eqs. (19), (20) are not compatible with Eq. (1).

In the linearized case of weak deviations from thermal equilibrium, the last expression can be simplified

$$\dot{u}_v^{(1)} = \frac{3}{2}\left(f_{10}\nabla\cdot\mathbf{v} + g_{10}\right)\sum_{ii'}\frac{(\Delta\varepsilon_{ii'})^2}{kT} n_i k_{ii'}^{(0)}\left(\frac{1}{2} - \frac{T - T_v}{T_v}\right). \tag{21}$$
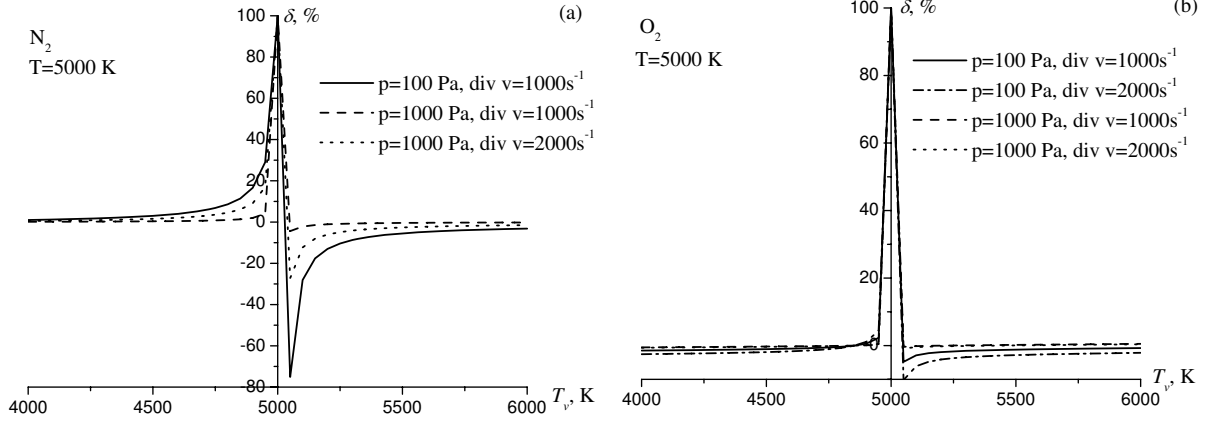
FIGURE 1. Contribution of the first-order correction to the total rate δ as a function of T_v for N_2 (a) and O_2 (b).

Note that for the general case, even for weak disequilibrium, the dependence of $\dot{u}_v^{(1)}$ on $\nabla \cdot \mathbf{v}$ and $(T - T_v)/T_v$ is not linear (there is a cross term containing their production). This is not in line with linear irreversible thermodynamics, where $\nabla \cdot \mathbf{v}$ and $(T - T_v)/T_v$ are considered as independent thermodynamic forces.

For the case of harmonic oscillators when only single-quantum jumps are allowed this contradiction can be eliminated. Moreover, it is possible to obtain symmetric expressions for the relaxation rate and pressure tensor. Indeed, for harmonic oscillators, the vibrational energy variation in a collision is constant and we can write

$$\Delta \varepsilon_{ii'} = \pm (\varepsilon_1 - \varepsilon_0) = \pm \Delta \varepsilon_v, \qquad \omega_{ii'} = \exp\left(\mp \frac{\Delta \varepsilon_v}{kT} \frac{T - T_v}{T_v}\right) - 1, \quad \forall\, i, i'. \tag{22}$$

Let $\Delta \varepsilon_1 = \Delta \varepsilon_v$, $\Delta \varepsilon_2 = -\Delta \varepsilon_v$. Similarly, ω_1 and ω_2 correspond to the "−" and "+" signs in the above expression. Then, after some algebra, we can write the last term in Eq. (14) as $1/n \sum_{r=1,2} G_{ij}^r \omega_r$. This makes it possible to derive the following expressions for the diagonal terms of the pressure tensor π and the first-order correction to the energy production rate:

$$p + \pi = -kT\,[F,F]\,\nabla \cdot \mathbf{v} - kT \sum_{r=1,2} [F,G^r]\,\omega_r, \tag{23}$$

$$\dot{u}_v^{(1)} = \sum_{r=1,2} \frac{\Delta \varepsilon_r}{kT} \dot{u}_v^{r(1)}, \qquad \dot{u}_v^{r(1)} = kT\,[G^r,F]\,\nabla \cdot \mathbf{v} + kT \sum_{s=1,2} [G^r,G^s]\,\omega_s. \tag{24}$$

It is seen that the normal mean stress and the energy production rate depend on the same forces $\nabla \cdot \mathbf{v}$ and ω_r. Moreover, the cross coefficients in these expressions are equal due to the symmetry properties of bracket integrals. Therefore, for the case of harmonic oscillators, the reciprocal Onsager-Casimir relations usually postulated by thermodynamics can be accurately proven. The reciprocal relations are valid for both non-linear and linearized ($|(T - T_v)/T_v| \ll 1$) cases. Note that for the general case, such a proof is still not obvious.

Once again, Eq. (24) cannot be reduced to Eq. (1). Consequently, even for the present particular case of harmonic oscillators with single-quantum jumps, using Eq. (1) in viscous flows is not justified.

The first-order correction to the rate of vibrational relaxation was calculated in N_2 and O_2 for different flow conditions. In Fig. 1, the percent contribution of the first-order correction to the total rate $\delta = \dot{u}_v^{(1)}/\dot{u}_v \cdot 100\%$ is given as a function of vibrational temperature for nitrogen (Fig. 1a) and oxygen (Fig. 1b). The temperature is fixed at $T = 5000$ K, the pressure and $\nabla \cdot \mathbf{v}$ are varied. It is seen that for N_2, the contribution of $\dot{u}_v^{(1)}$ is not negligible. For low pressure and large velocity divergence $\dot{u}_v^{(1)}$ may be of the same order as $\dot{u}_v^{(0)}$. For O_2 the effect is weaker, the mean contribution of $\dot{u}_v^{(1)}$ is within 1-2%. High values of δ at $T \approx T_v$ are explained by the fact that while approaching to thermal equilibrium, $\dot{u}_v^{(0)}$ tends to zero and, therefore, the only contribution to the relaxation rate is given by $\dot{u}_v^{(1)}$. However, one should be careful applying multi-temperature models under near-equilibrium conditions, since the limits of their validity are restricted in this case.
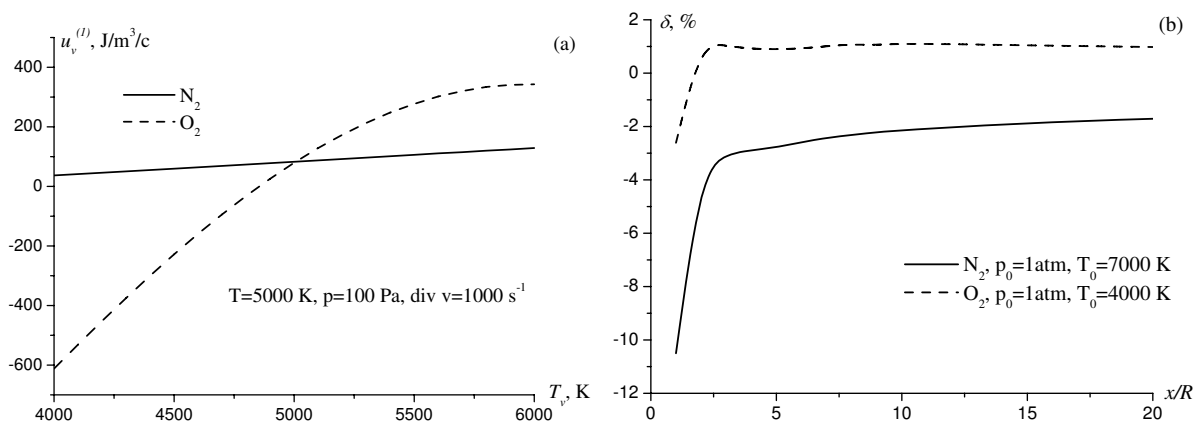
FIGURE 2. First-order correction $\dot{u}_v^{(1)}$ as a function of T_v (a) and contribution of the first-order correction to the total rate δ as a function of x/R in a nozzle.

Fig. 2a shows typical behaviour of the first-order correction $\dot{u}_v^{(1)}$ for nitrogen and oxygen. In Fig. 2b, the contribution of the first-order correction δ to the total rate is presented for a supersonic expanding flow. A flow in a conic nozzle with an angle $21°$ is considered. The throat conditions are (1) $T_0 = 7000$ K, $p_0 = 1$ atm for N_2 and (2) $T_0 = 4000$ K, $p_0 = 1$ atm for O_2. It is seen that close to the throat (particularly for nitrogen), the first-order effects can influence noticeably the rate of vibrational relaxation, whereas with rising x/R (R is the throat radius), the contribution of the first-order correction decreases. Similar calculations have been performed in shock heated flows. The first-order effects are found to be weak in compressive flows which is not surprising since δ becomes small at high pressures.

CONCLUSIONS

The first-order thermal relaxation rate depending on the velocity divergence and temperature difference is derived for the case of VT relaxation. For the particular case of harmonic oscillators, the cross coupling terms between the relaxation rate and pressure tensor are obtained in the symmetric form. The first-order effects are found to be non-negligible in expanding flows; for high pressures, these effects become weak. The results can be generalized for chemical-vibrational coupling, relaxation of different types of internal energy, and vibrational relaxation in complex polyatomic molecules.

ACKNOWLEDGMENTS

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