

# The Influence of State-to-State Kinetics on Diffusion and Heat Transfer behind Shock Waves

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**Abstract.** In the paper, the influence of vibrational and chemical kinetics on heat transfer and diffusion in hypersonic flows of  $N_2/N$  mixture in the relaxation zone behind shock waves is studied on the basis of the state-to-state kinetic theory approach. The results of calculations of vibrational level populations  $n_i$ , gas temperature  $T$ , total energy flux  $\mathbf{q}$ , diffusion velocities of molecules at different vibrational states  $\mathbf{V}_i$  and atoms  $\mathbf{V}_a$  in the relaxation zone behind a shock front are presented for the free stream Mach number  $M = 10, 15$ . The contribution of different dissipative processes to the total energy flux is estimated for various flow conditions. The impact of non-equilibrium vibrational distributions in the free stream on molecular level populations and transport properties in the relaxation zone is shown.

**Keywords:** transport coefficients, state-to-state vibrational kinetics, shock waves

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## INTRODUCTION

For prediction of gas dynamic parameters and transport properties in high-temperature gas flows, the non-equilibrium excitation of internal degrees of freedom and chemical reactions should be taken into account. Different approaches are developed in the kinetic theory for description of kinetic and dissipative processes in non-equilibrium flows [1]. The most accurate approach is based on consideration of the detailed state-to-state molecular distributions over internal energy levels and their influence on flow parameters and transport properties [2]. In the present paper this approach is used to study non-equilibrium vibration-dissociation kinetics and its influence on diffusion and heat transfer in reacting flows behind shock waves. In shock heated gases the state-to-state kinetics has been studied by many authors (see [3, 4, 5] and Refs. in [1]), the influence of vibrational level populations on the mass and heat transfer was considered, for the first time, in [5]. The state-to-state transport kinetic theory approach for reacting gases has been proposed in [2] and then applied in some papers for evaluation of transport properties of different flows (see [6] and Refs. in [1]). The simplified algorithms for calculation of state-dependent transport coefficients are proposed in [7]. Specific features of diffusion and energy transfer within the detailed kinetic description are discussed in [8]. However, the influence of strongly non-equilibrium vibrational distributions on the mass and heat transfer as well as the impact of free stream conditions on transport properties behind a shock front are not sufficiently understood up to now. In this paper we present the results of numerical study of state-to-state vibration-dissociation kinetics, heat transfer and diffusion in the flows of  $N_2(i)/N$  mixture in the relaxation zone behind shock waves. We concentrate the attention on the contribution of various processes to the total energy flux including along with heat conduction, thermal and mass diffusion also diffusion of vibrationally excited molecules. This distinctive feature of the state-to-state flow description was mentioned before in [9].

## STATE-TO-STATE DESCRIPTION OF DIFFUSION AND HEAT TRANSFER

Experimental data on relaxation times in shock heated air components [10] show fast equilibration of translational and rotational degrees of freedom compared to vibrational and chemical relaxation. Therefore in numerical simulations distributions of molecules over translational and rotational energies are supposed to be equilibrium or weakly non-equilibrium while vibrational-chemical relaxation is considered in the strongly non-equilibrium regime. In the state-to-state approach the set of gas flow parameters includes not only the macroscopic velocity  $\mathbf{v}(\mathbf{r}, t)$ , gas temperature  $T(\mathbf{r}, t)$  and molar fractions of chemical species, but also the vibrational level populations of molecules. In this case

the governing equations contain the conservation equations of mass, momentum and total energy coupled to the equations for vibrational level populations and atomic number densities. These equations are derived in [1] from the kinetic equations for distribution functions in the zero and first order approximations of the Chapman-Enskog method generalized for the conditions of strongly non-equilibrium vibrational and chemical kinetics.

We consider the following kinetic processes in the flows of  $N_2(i)/N$  mixture:

- VV vibrational energy exchanges

$$N_2(i) + N_2(k) = N_2(i \pm 1) + N_2(k \mp 1), \quad (1)$$

- VT (TV) exchanges between vibrational and translational energies

$$N_2(i) + M = N_2(i \pm 1) + M, \quad M = N_2, N, \quad (2)$$

- dissociation and recombination

$$N_2(i) + M = N + N + M, \quad M = N_2, N. \quad (3)$$

The expressions for the diffusion velocities and total energy flux derived in [2, 7] may be written as a sum of contributions of different processes [11, 8]:

$$\mathbf{V}_i = \mathbf{V}_i^{TD} + \mathbf{V}_i^{MD} + \mathbf{V}_i^{DVE}, \quad \mathbf{V}_a = \mathbf{V}_a^{TD} + \mathbf{V}_a^{MD}, \quad (4)$$

$$\mathbf{q} = \mathbf{q}^{HC} + \mathbf{q}^{MD} + \mathbf{q}^{TD} + \mathbf{q}^{DVE}. \quad (5)$$

Here,  $\mathbf{V}_i^{MD}$ ,  $\mathbf{V}_a^{MD}$ ,  $\mathbf{V}_i^{TD}$ ,  $\mathbf{V}_a^{TD}$  and  $\mathbf{V}_i^{DVE}$  are, respectively, the contributions of the mass diffusion, thermal diffusion, and diffusion of vibrational energy:

$$\mathbf{V}_i^{MD} = -D_{mm}\mathbf{d}_m - D_{ma}\mathbf{d}_a, \quad \mathbf{V}_i^{TD} = -D_{Tm}\nabla \ln T, \quad \mathbf{V}_i^{DVE} = -n \left( \frac{n_m}{D_{mm}} + \frac{n_a}{D_{ma}} \right)^{-1} \nabla \ln \frac{n_i}{n_m}, \quad (6)$$

$$\mathbf{V}_a^{MD} = -D_{ma}\mathbf{d}_m - D_{aa}\mathbf{d}_a, \quad \mathbf{V}_a^{TD} = -D_{Ta}\nabla \ln T,$$

$\mathbf{q}^{HC}$ ,  $\mathbf{q}^{MD}$ ,  $\mathbf{q}^{TD}$  and  $\mathbf{q}^{DVE}$  are, respectively, energy fluxes associated with the heat conductivity of translational and rotational degrees of freedom (Fourier flux), mass diffusion, thermal diffusion, and the transfer of vibrational energy:

$$\begin{aligned} \mathbf{q}^{HC} &= -\lambda' \nabla T, \quad \mathbf{q}^{MD} = \rho_m h_m \mathbf{V}_m^{MD} + \rho_a h_a \mathbf{V}_a^{MD}, \quad \mathbf{q}^{TD} = -p(D_{Tm}\mathbf{d}_m + D_{Ta}\mathbf{d}_a) + \rho_m h_m \mathbf{V}_m^{TD} + \rho_a h_a \mathbf{V}_a^{TD}, \\ \mathbf{q}^{DVE} &= \sum_i \left( \frac{5}{2} k_B T + \langle \varepsilon^i \rangle_{rot} + \varepsilon_i \right) n_i \mathbf{V}_i^{DVE}. \end{aligned} \quad (7)$$

In Eqs. (6)-(7)  $D_{mm}$ ,  $D_{ma}$ ,  $D_{aa}$ ,  $D_{Tm}$  and  $D_{Ta}$  are the multi-component diffusion and thermal diffusion coefficients for each molecules and atoms,  $\mathbf{d}_m$  and  $\mathbf{d}_a$  are the diffusive driving forces depending on gradients of the level populations  $n_i$  and the pressure  $p = nk_B T$ ,  $k_B$  is the Boltzmann constant,  $n$ ,  $n_m$  and  $n_a$  are the number densities of a gas mixture, molecules and atoms, respectively,  $\lambda' = \lambda_{tr} + \lambda_{rot}$  is the thermal conductivity coefficient,  $\lambda_{tr}$  and  $\lambda_{rot}$  correspond to energy transfer due to elastic collisions and those with rotational energy transfer,  $h_m$ ,  $h_a$ ,  $\rho_m$  and  $\rho_a$  are the enthalpies per unit mass and the densities of molecular and atom components,  $\langle \varepsilon^i \rangle_{rot}$  is the mean rotational energy,  $\varepsilon_i$  is the vibrational energy of  $N_2(i)$  molecules. In the considered approach the energy flux and diffusion velocities include along with the gradients of temperature  $T$  and atomic number densities  $n_a$ , also the gradients of all vibrational level populations  $n_i$ . Diffusion and heat conductivity coefficients depend on level populations, number densities of atoms, the gas temperature and can be found as a solution of transport linear systems derived for the state-to-state approximation in [2, 7]. Coefficients of these systems are specified by the cross sections of more frequent collisions with transitions of translational and rotational energies.

Following accurate transport algorithms for the considered mixture one should solve  $l + 1$  equations for evaluation of each independent diffusion coefficient and  $3l + 2$  equations for heat conductivity coefficients where  $l$  is the number of excited vibration levels of  $N_2$  molecules. After simplifications proposed in [7], the number of equations for the binary mixture is reduced to 5 and 3 equations, respectively. The self-consistent approach is based on the direct implementation of the state-to-state kinetic theory algorithms to the CFD code and requires the solution of transport

systems at each space and time step of calculations. These procedure occurs very complicated and time consuming. Therefore in this paper we used the simplified way proposed in [2, 7]. First, vibrational level populations, number density of atoms, gas temperature and velocity in the relaxation zone behind a shock wave have been found from the equations of the inviscid flow of the considered mixture in the Euler approximation. Then, the obtained data have been substituted to the accurate transport systems for calculations of the total energy flux and diffusion velocities of atoms and molecules at different vibrational states.

## VIBRATION-DISSOCIATION KINETICS

The flow of  $N_2(i)/N$  mixture in the relaxation zone behind a shock wave is described by the following equations [1]:

$$\frac{d(vn_i)}{dx} = R_i^{vibr} + R_i^{diss-rec}, \quad i = 0, 1, \dots, l, \quad (8)$$

$$\frac{d(vn_a)}{dx} = -2 \sum_i R_i^{diss-rec}, \quad (9)$$

$$\rho_0 v_0^2 + p_0 = \rho v^2 + p, \quad (10)$$

$$h_0 + \frac{v_0^2}{2} = h + \frac{v^2}{2}, \quad (11)$$

where  $x$  is the distance from the shock front, the subscript "0" denotes the parameters in the free stream:

$$h = h_m Y_m + h_a Y_a, \quad (12)$$

$Y_m = \rho_m / \rho$  and  $Y_a = \rho_a / \rho$  are the mass fractions of molecules and atoms,  $\rho$  is the mass density of mixture,

$$h_a = \frac{5}{2} \bar{R}_a T + \frac{\varepsilon}{m_a}, \quad h_m = \frac{7}{2} \bar{R}_m T + \frac{1}{\rho_m} \sum \varepsilon_i n_i,$$

where  $\bar{R}_a$  and  $\bar{R}_m$  are the specific gas constants of molecules  $N_2$  and atoms  $N$ ,  $m_a$  is the atom mass,  $\varepsilon$  is the formation energy of atoms.

The right hand sides of the Eqs. (8)-(11) are given by the relations:

$$R_i^{vibr} = R_i^{VT} + R_i^{VV} = \sum_M n_M \sum_{i'} (n_{i'} k_{i'i}^M - n_i k_{i'i'}^M) + \sum_k \sum_{i'} \sum_{k'} (n_{i'} n_{k'} k_{i'i}^{k'k} - n_i n_k k_{i'i'}^{kk'}), \quad (13)$$

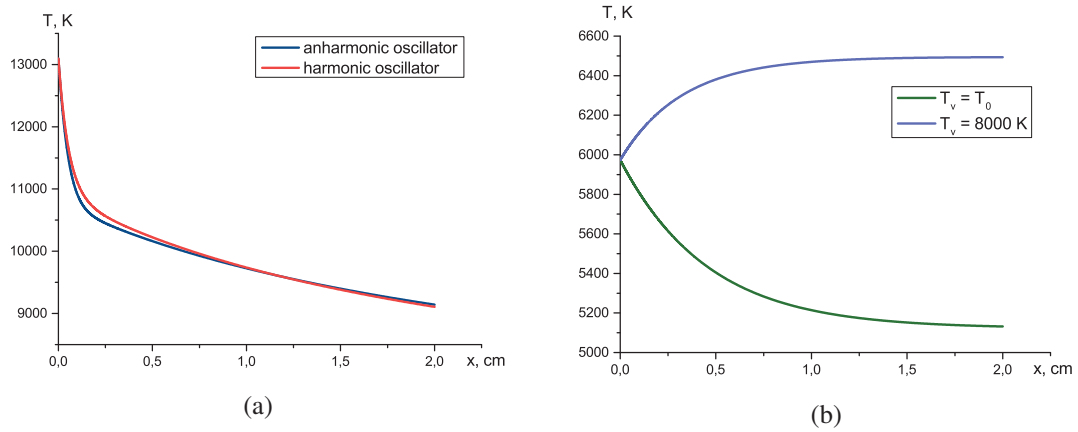
$$R_i^{diss-rec} = \sum_M n_M (n_a^2 k_{rec,i}^M - n_i k_{i,diss}^M), \quad M = N_2, N, \quad i' = i \pm 1, \quad k' = k \mp 1. \quad (14)$$

Here  $k_{i'i}^M$ ,  $k_{i'i'}^{kk'}$ ,  $k_{i,diss}^M$  and  $k_{rec,i}^M$  are state-specific coefficients of VT and VV vibrational energy transitions, dissociation and recombination. The rate coefficients for vibrational energy transitions are calculated using the generalized formulas of SSH-theory [12], dissociation is described by the Treanor-Marrone model [13] modified for the state-to-state approach [1, 4].

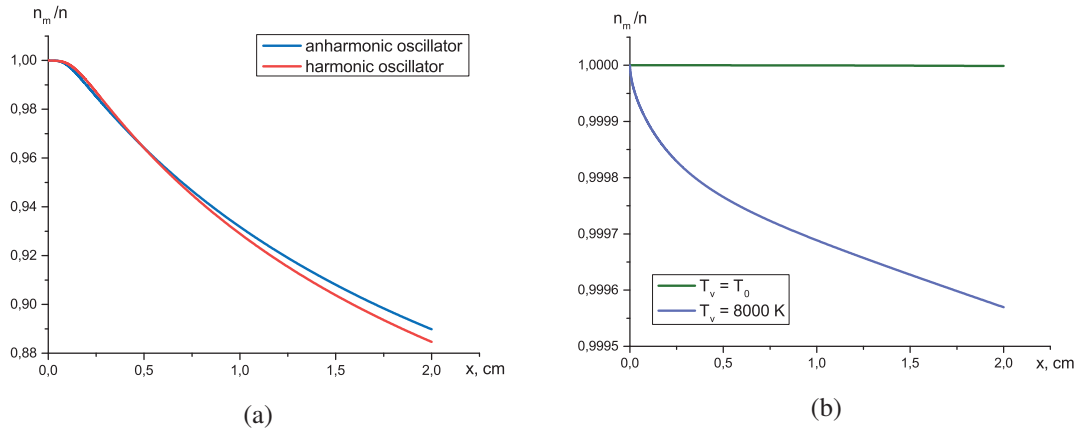
## RESULTS AND DISCUSSION

First, we will consider the vibrational distributions, mixture composition and gas temperature obtained as a solution of Eqs. (8)-(11) with the initial gas parameters just behind the shock found with the use of the Rankine-Hugoniot relations under the assumption of frozen vibrational distributions and mixture composition within the shock front.

Figures. 1-3 show the variation of the gas temperature, vibrational level populations and number densities of  $N_2$  molecules in the relaxation zone obtained for the following conditions in the free stream:  $T_0 = 293$  K,  $p_0 = 100$  Pa,  $n_m = p_0 / k_B T_0$ ,  $n_a = 0$ ,  $M_0 = 15$ , 10. Level populations in the free stream are assumed to be the Boltzmann's with



**FIGURE 1.** The gas temperature  $T$  behind the shock as a function of  $x$ . (a) -  $M_0 = 15$ , (b) -  $M_0 = 10$ .

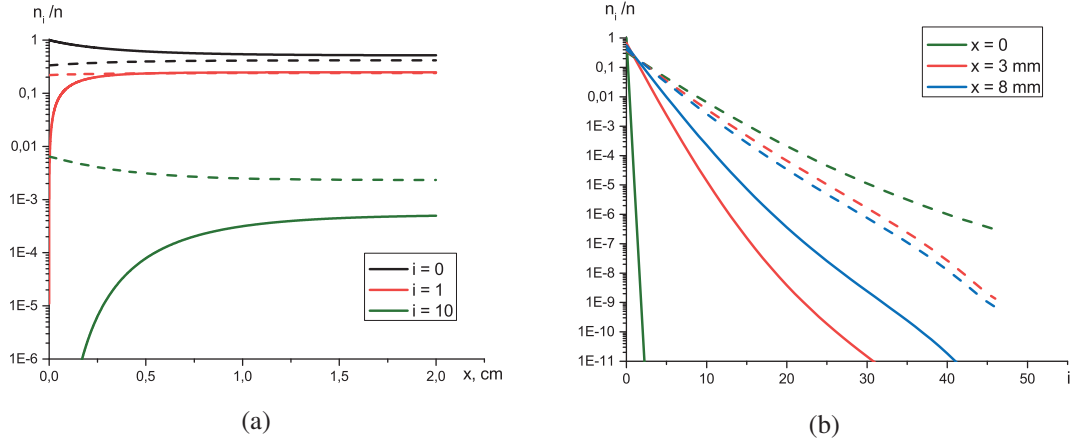


**FIGURE 2.** The molar fraction  $n_m/n$  for  $M_0 = 15$  (a) and for  $M_0 = 10$  (b) behind the shock front as a function of  $x$ .

the vibrational temperature  $T_v$ . The vibrational energy of  $N_2$  molecules is simulated by anharmonic and harmonic oscillator models with the number of excited levels  $l = 46$  and  $l = 32$ , respectively. The influence of anharmonic effects on vibrational distributions, macroscopic parameters and transport properties in the relaxation zone is estimated. We considered both the equilibrium distributions in the undisturbed flow with  $T_v = T_0$  and non-equilibrium distributions when  $T_v = 8000$  K ( $T_v > T_0$ ). Within the shock front the gas temperature rises immediately and its value just behind the shock  $T_1$  is specified by the Mach number in the free stream. On the contrary, the value of vibrational temperature  $T_v$  remains the same before and just behind the shock because the vibrational distributions are supposed frozen within the shock front. Fig. 1 (a) shows that in the first case  $T_v = T_0$  and  $M_0 = 15$ , the gas temperature  $T$  decreases considerably with  $x$  rising from the beginning of the relaxation zone due to  $TV$  translational energy transfer, vibrational excitation and dissociation. We can notice that neglecting of anharmonicity leads to overestimated  $T$  values up to 1.8%.

In the second case (Fig. 1 (b))  $T_v$  occurs higher than  $T_1$  and the character of  $T$  variation changes completely: first, it increases due to  $VT$  transfer of the vibrational energy to the translational one and then, for  $x > 1$  cm, the temperature variation becomes negligible. It means that molecules dissociate mainly due to their high vibrational energy. For  $M_0 = 15$  this effect is not so strong because  $T_1$  becomes higher and closer to  $T_v$ . Vibrationally excited free stream molecules appear in result of some kinds of vibrational energy pumping, for example in shock tubes after a nozzle exit [14]. A similar situation occurs in the problem of conducting by chemical reactions behind shock wave due to vibrational excitation of free stream molecules.

Figures 2a and 2b depict number densities of  $N_2$  molecules for the same conditions as in Figs. 1. Figure 2a



**FIGURE 3.** The vibrational level populations  $n_i/n$ . **(a)** - as functions of  $x$ , **(b)** - for different values of  $x$ .  $M_0 = 10$ . Solid and dash curves correspond to  $T_v = T_0$  and  $T_v = 8000$  K, respectively.

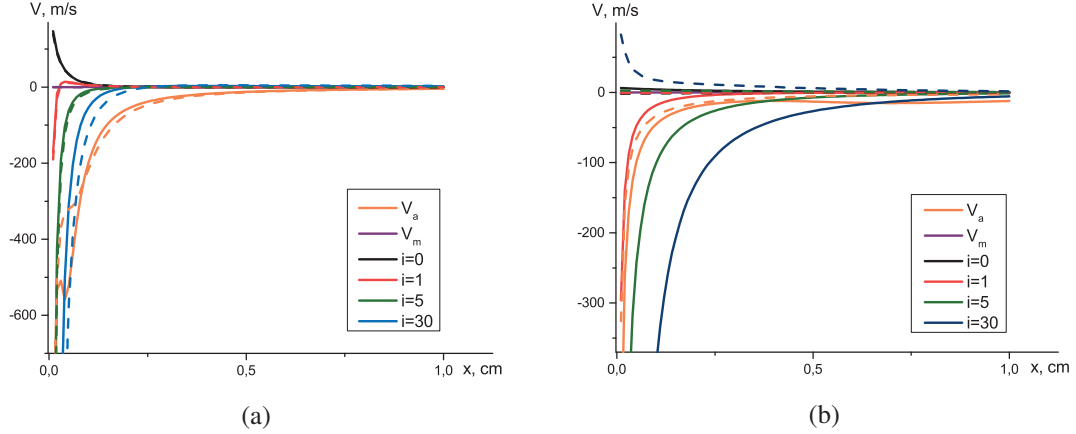
illustrates the influence of anharmonic effects on  $N_2$  molar fraction: we obtain underestimated  $n_m$  values using the harmonic oscillator model up to 0.8% whereas the overestimation of the number densities of atoms reaches 3.7%. Figure 2b shows that vibrational excitation of the free stream molecules ( $T_v > T_1 > T_0$ ,  $M_0 = 10$ ) leads to more intensive dissociation and, consequently, lower values of the molar fractions of molecules compared to the case of the equilibrium free stream conditions ( $T_v = T_0$ ); in the latter case dissociation is practically frozen.

The influence of the initial vibrational excitation in the free stream on level populations in the relaxation zone is demonstrated in Figs. 3. Figure. 3a displays populations  $n_0$ ,  $n_1$  and  $n_{10}$  as functions of  $x$  whereas Fig. 3b presents vibrational distributions calculated for three positions in the relaxation zone. For the equilibrium free stream, populations of all levels, except those for  $i = 0$ , rise markedly in the beginning of relaxation zone as a consequence of  $TV$  excitation and then slightly decrease. On the contrary, in the case of initially excited free stream ( $T_v > T_1 > T_0$ ,  $M_0 = 10$ ) populations of all levels change weakly: for  $i > 1$   $n_i$  decrease from the beginning of the relaxation zone due to  $VT$  deactivation and dissociation whereas  $n_i$  for  $i = 0, 1$  increase weakly in result of  $VT$  and  $VV$  energy transitions.

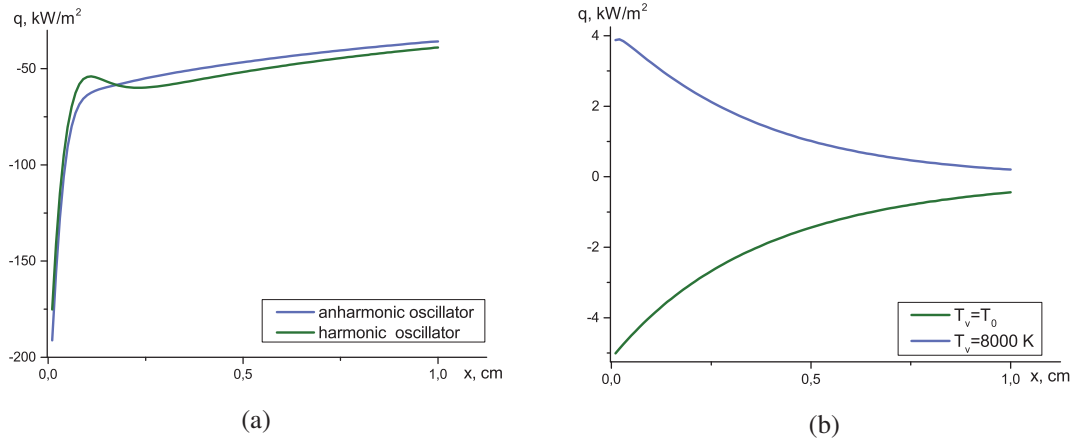
Calculations show quite different limit values of the gas temperature and molar fractions of atoms obtained for the equilibrium and non-equilibrium free stream conditions in the end of the relaxation zone, where the mixture approaches to the thermal equilibrium. It may be explained by the contribution of the initial vibrational energy of  $N_2$  molecules to the total enthalpy in the second case.

The macroscopic parameters obtained above as well as their derivatives were used as input data for the calculation of transport coefficients, diffusion velocities and heat flux as functions of the distance from the shock front. The diffusion velocities of atoms, molecules and various vibrational states are given in Figs. 4. Since the diffusion coefficients are almost inversely proportional to the species concentrations and vibrational level populations, the diffusion velocities depend significantly on  $n_i$ ,  $n_a$ ,  $n_m$ . The absolute values of all diffusion velocities decrease with  $x$  because the gradients of number densities of species and vibrational states tend to zero in the final equilibrium state. The sign of the diffusion velocity is specified by the sign of the corresponding species derivative. Thus for equilibrium initial conditions, the diffusion velocities of molecules and of molecules at the vibrational level  $i = 0$  are positive whereas  $V_a$  and other  $V_i$  ( $i > 0$ ) are negative. For non-equilibrium initial conditions, the behaviour of  $V_i$  is opposite because the main process in this case is  $VT$  deactivation and not excitation. Therefore, the diffusion velocities of high levels are positive. The behaviour of  $V_m$ ,  $V_a$  is similar in both cases. It is worth mentioning that for  $M = 10$ , although the rate of dissociation in the case of non-equilibrium initial conditions is much higher compared to that in the case of initial equilibrium (and therefore the gradient of  $n_a$  is higher), the values of  $V_a$  are close to those obtained in the case  $T_v = T_0$ . The reason for this is in low concentration of atoms in the equilibrium case, which causes large self-diffusion coefficients of atoms compensating the small gradient of  $n_a$ . The influence of anharmonicity on diffusion velocities is found to be weak for low levels, however it becomes noticeable for higher levels.

Figure 5 present the variation of the total heat flux in the relaxation zone behind the shock wave for different initial conditions. For  $M_0 = 15$  the absolute value of the total heat flux is rather high near the shock wave front, then (for



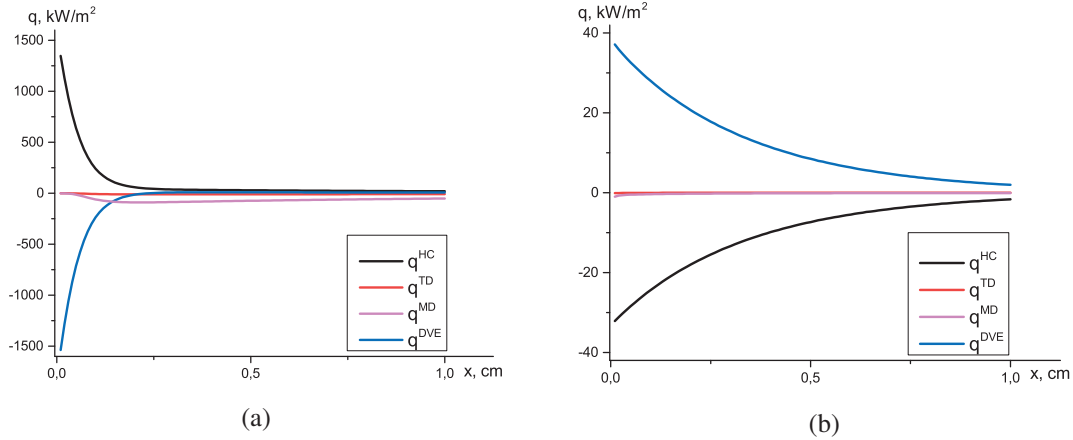
**FIGURE 4.** The diffusion velocity behind the shock as a function of  $x$ . (a) -  $M_0 = 15$ , (b) -  $M_0 = 10$ . Solid and dash curves correspond to (a) - anharmonic and harmonic oscillator, (b) -  $T_v = T_0$  and  $T_v = 8000$  K, respectively.



**FIGURE 5.** The total heat flux behind the shock as a function of  $x$ . (a) -  $M_0 = 15$ , (b) -  $M_0 = 10$ .

$x > 0.2$  cm) it decreases together with the derivatives of macroscopic parameters. The sign of the total flux is opposite to that of the Fourier flux due to heat conductivity (it follows from Eq. (7) and decreasing gas temperature (Fig. 1a). For harmonic oscillators, dissociation proceeds faster, and it leads to the shift of the distance  $x$  where the contribution of the mass diffusion process becomes important (see discussion below). This is the reason for the non-monotonic behaviour of the total heat flux in the case of harmonic vibrations. Quantitatively, the effect of anharmonicity on the heat flux is within 10-15%. For  $M_0 = 10$ , the absolute value of the heat flux is much lower because of small gradients of gas-dynamic parameters in the relaxation zone. Note that in the case of initial non-equilibrium the sign of the total heat flux is positive which again is opposite to the Fourier flux sign (see Fig. 1b for the temperature gradient). It is interesting to understand which process is responsible for such a behaviour of the total heat flux.

Figures 6 show the contribution of the various processes to the heat flux. The Fourier heat flux and heat flux caused by the diffusion of vibrational energy give the contribution of the same order near the shock front; for  $M_0 = 15$  their absolute values are rather high at  $x < 0.2$  cm. Nevertheless the signs of these terms are opposite which causes a strong compensation effect leading to much lower values of the total flux. The impact of the vibrational energy diffusion on the heat flux in shock heated flows occurs stronger, it leads to changing the sign of  $\mathbf{q}$  compared to the Fourier flux. Thermal diffusion has practically no effect on the heat flux behind the shock wave. Concerning the mass diffusion, its contribution becomes significant only at  $x > 0.2$  cm due to the dissociation incubation time preventing strong gradients of  $n_m$ ,  $n_a$  close to the front (see Fig. 2a). However in the region where  $\mathbf{q}^{MD}$  is important, the total



**FIGURE 6.** The contribution of the various processes in the heat flux behind the shock as a function of  $x$ , anharmonic oscillator. (a) -  $M_0 = 15$ ,  $T_0 = T_v$ , (b) -  $M_0 = 10$ ,  $T_v = 8000$  K.

flux is small because vibrational distributions reach quasi-equilibrium and vary weakly; the temperature variation is also weak. For  $M_0 = 10$  in the case  $T_0 = T_v$  the contributions to the heat flux are much smaller than for  $M_0 = 15$ , but they are qualitatively similar. For initially non-equilibrium conditions  $T_v = 8000$  K, the Fourier heat flux and heat flux caused by the vibrational energy diffusion reverse the signs because excitation process is replaced by deactivation; the contribution of  $\mathbf{q}^{DVE}$  is again dominating and determines the sign of  $\mathbf{q}$ . Thus we can conclude that the main role in the heat transfer behind shock waves belongs to heat conduction and diffusion of vibrational energy, and its sign is specified by  $\mathbf{q}^{DVE}$ .

## CONCLUSIONS

The influence of state-to-state vibration-dissociation kinetics on diffusion velocities and total energy flux in shock heated  $N_2/N$  mixture is studied in two cases of equilibrium and vibrationally excited distributions before the shock front. Results showed a dramatic impact of initial vibrational excitation of molecules before the shock on the gas temperature, level populations and energy transfer in the relaxation zone. The evolution of diffusion velocities of atoms and molecules in different vibrational states as well as energy flux variation along the relaxation zone is studied and the new effect of contribution of vibrational energy diffusion to the total energy flux is demonstrated for different conditions. A compensation effect between heat conduction and vibrational energy diffusion leading to the significant decrease of the total heat flux is revealed in all considered test cases, and the dominating role of vibrational energy diffusion in the direction of the heat transfer is shown. While the total heat flux is negative under conditions of vibrational excitation, it becomes positive if deactivation processes prevail. The effect of anharmonicity does not exceed 3-4% for gas-dynamic parameters and 10-15% for the total energy flux.

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