

Figure 4 shows one of the frames of the film record which depicts flow about a cylinder in a channel. The interference bands were obtained by the real-time method in accordance with the optical scheme in Fig. 3. Interference lines 2 and the position of the colored marking 1 are visible on the photograph. It can be seen that the marking and the interference lines are poorly defined. This is related to the fact that the interference lines are localized in the region between the object and the hologram. Thus, the objective lens of the film camera must be directed between the sharp image of the colored marking and the lines of the interferogram.

For simultaneous sharp recording of the colored marking and interference bands, it is necessary to significantly stop down the objective lens. This cannot always be done, since it entails the use of highly sensitive film with a large grain size and, thus, low resolution with regard to the information obtained. These problems can be circumvented either by using a light-amplifying apparatus such as the high-speed camera in Fig. 3 or by recording the interferograms together with the image of the colored marking through the use of two exposures with holograms from the focused image.

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INFLUENCE OF GAS FLOW ON THE DISTRIBUTION FUNCTION OF DISSOCIATING MOLECULES IN VIBRATIONAL LEVELS

G. Ya. Dynnikova

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1. The influence of gas motion on the reaction kinetics of strongly excited molecules was examined in [1-4] - where it was established that significant nonequilibrium corrections, in magnitude, that are proportional to the spatial derivative of the macroscopic quantities, their powers, and derivatives, occur in the expressions for the macroscopic reaction rate. It is shown in [5-8] that upon taking more complete account of the properties of the kinetic equation solutions for high molecule energies they depend in a nonlinear manner on the spatial derivatives. In particular, the rate of diatomic molecule dissociation - truncated harmonic oscillators - depends exponentially on \sqrt{u} , where u is the flow velocity [8] in a nonisothermal convective flow.

Nonequilibrium correction to the reaction rate is a result of perturbations of the molecule distribution in the vibrational levels. The solution obtained in [8] for the distribution function possesses the property of locality, which is not fully in agreement with the explanation presented in [8], whence there results that the population of the upper energetic levels do not succeed in following the change in the translational temperature. Another interpretation of the results obtained is given below.

2. As is known [9], the law of conservation of the distribution functions holds in the model of a harmonic oscillator with an infinite number of levels if the initial distribution is a Boltzmann one. The population of the i -th vibrational level x_i at each instant is here determined by the formula

$$x_i = (1 - q)q^i, \quad (2.1)$$

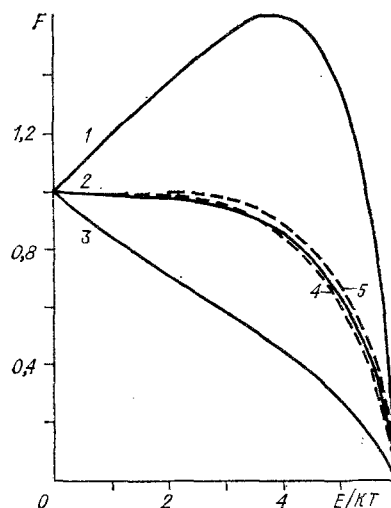


Fig. 1

where q is described by the equation

$$dq/dt = (q - q_*)/\tau. \quad (2.2)$$

Here $\tau^{-1} = P_{10}Z(1 - q)$; $q_* = \exp(-E_1/KT)$; P_{10} is the transition probability from the first level to the ground state; Z is the collision frequency; E_1 is the energy of a vibrational quantum; K is the Boltzmann constant; and T is the translational temperature.

For a slow change in the temperature when

$$\frac{\tau}{q_*} \frac{dq_*}{dt} = \frac{E_1 \tau}{KT} \frac{d(\ln T)}{dt} \ll 1,$$

an approximate solution of (2.2) is $q \approx q_* \left(1 - \frac{\tau}{q_*} \frac{dq_*}{dt}\right)$. In this case the solution (2.1) can be interpreted as a Boltzmann distribution with a lagging temperature T_1

$$q = \exp\left(-\frac{E_1}{KT_1}\right), \quad T_1 \approx T \left(1 - \frac{\tau}{\theta}\right), \quad \theta = \left(\frac{d(\ln T)}{dt}\right)^{-1}. \quad (2.3)$$

The molecule distribution in vibrational levels, obtained in [8] in the model of a truncated harmonic oscillator with dissociation, can also be interpreted as quasistationary that holds in the isothermal case, but corresponding to the lagging temperature T_1 . This is confirmed by comparing the graphs in Fig. 1. Curves 1 and 3 are obtained in [8] and are the molecule distribution functions at the vibrational energy E (in the continuous spectrum approximation), referred to the Boltzmann distribution for $\tau/\theta = -0.17$ and 0.17 , respectively. Line 2 corresponds to the quasistationary distribution in the isothermal case, while lines 4 and 5 are obtained by renormalization of distributions 1 and 3 to a Boltzmann distribution with lagging temperature calculated by means of (2.3). It is seen that lines 4 and 5 practically coincide with 2. Therefore, the distribution functions of [8] are close to a quasistationary distribution with a lagging temperature.

As mentioned above, in the case of a harmonic oscillator with an infinite number of levels the Boltzmann distribution with intrinsic vibrational temperature is conserved independently of the rate of change of the translational temperature. It is interesting to clarify whether the quasistationary form of the distribution is conserved in the truncated harmonic oscillator model for an arbitrary rate of temperature change.

3. To answer this question we first obtain a quasistationary distribution function for the isothermal case in a convenient form. Exactly as in [8], we consider the dissociation process of molecules comprising a small impurity in an inert gas; therefore, we will take account only of the V-T exchange reaction. Moreover, we assume that the atom concentration of the dissociation products is sufficiently small; consequently, we can neglect the influence of recombination. We shall consider that on reaching the level with energy Q ($Q = nE_1$) the molecule instantly dissociates. Under these assumptions, the level-by-level kinetic equations have the form [9]

$$x_i \frac{1}{N} \frac{dN}{dt} = P_{10} Z [(i+1)(x_{i+1} - qx_i) - i(x_i - qx_{i-1})],$$

$$i = 0, 1, \dots, n-1, x_n = 0 \quad (3.1)$$

(N is the number of dissociating molecules).

Applying the double-summation operator to the left and right sides of (3.1)

$$A_{(k)} = q^{k+1} \sum_{j=0}^k \frac{1-q}{(j+1)q^{j+1}} \sum_{i=0}^j$$

and introducing the notation $\bar{x}_i = x_i/x_0$, $\mu = -\frac{\tau}{N} \frac{dN}{dt}$ we convert (3.1) to the form

$$-\mu q^{k+1} \sum_{j=0}^k \frac{1-q}{(j+1)q^{j+1}} \sum_{i=0}^j \bar{x}_i = \bar{x}_{k+1} - q^{k+1}, \quad k = 0, \dots, n-1.$$

After multiple application of the operator $A_{(k)}$ we obtain

$$x_k = q^k \sum_{i=0}^k (-\mu)^i \varphi_k^{(i)}, \quad (3.2)$$

where $\varphi_k^{(0)} = 1$ and the functions $\varphi_k^{(i)}$ are determined by using the recursion relationship

$$\varphi_k^{(i)} = \sum_{v=i}^k \frac{1-q}{vq^v} \sum_{j=i-1}^{v-1} q^j \varphi_j^{(i-1)}. \quad (3.3)$$

For $k = n$ we have a closed equation to determine μ :

$$\sum_{i=0}^n (-\mu)^i \varphi_n^{(i)} = 0. \quad (3.4)$$

The expression (3.2) is an exact solution of the system (3.1). It can be shown that its right side agrees with the Gottlieb polynomial

$$l_k(\mu) = q^n \sum_{r=0}^n (1-q^{-1})^r \binom{n}{r} \binom{\mu}{r}.$$

The connection between the Gottlieb polynomials and the distribution functions for the truncated harmonic oscillator model was established first in [10]. The Gottlieb polynomials are obtained in this paper in another more convenient form for subsequent operations. Let us just note that the double-summation procedure has been used to obtain successive approximations in [11, 12].

It is shown in [10] that the general nonstationary solution of the system of level-by-level equations for a truncated harmonic oscillator is expressed in the form

$$Nx_k = \sum_{i=1}^n \alpha_i l_k(\mu_i) \exp\left(-\mu_i \frac{t}{\tau}\right).$$

Here μ_i are roots of the Gottlieb polynomial determined by (3.4) while the coefficients α_i depend on the initial molecule distribution over the levels. All the roots of the Gottlieb polynomials are positive [10], where for each polynomial $l_n(\mu)$ with $n(1-q) \gg 1$ the minimal root is many times less than the remaining roots; consequently, in the quasistationary case the macroscopic dissociation rate equals $\frac{1}{N} \frac{dN}{dt} = -\frac{\mu_0}{\tau}$, where μ_0 is the minimal root of (3.3).

The corresponding distribution function is described by (3.2) for $\mu = \mu_0$.

Analysis of the functions $\varphi_k^{(i)}$ shows that the main contribution to the sum $\sum_{i=1}^k \mu^i \varphi_k^{(i)}$ is given by the first term for $\mu = \mu_0$. This permits obtaining an approximate expression for μ and x_k for $n(1-q) \gg 1$

$$\mu_\theta = (\varphi_n^{(1)})^{-1} = \left(\sum_{v=1}^n \frac{1-q}{vq^v} \sum_{m=0}^{v-1} q^m \right)^{-1} = \left(\sum_{v=1}^n \frac{1-q^v}{vq^v} \right)^{-1} \approx n(1-q)q^n,$$

$$\bar{x}_k \approx q^k \left(1 - \frac{\varphi_k^{(1)}}{\varphi_n^{(1)}} \right) \approx \begin{cases} q^k [1 - q^{n-k} n(1-q)(1-q^k)] \\ \text{for } k(1-q) \leq 1, \\ q^k \left(1 - \frac{n}{k} q^{n-k} \right) \\ \text{for } k(1-q) \gg 1, \end{cases}$$

$$x_0 \approx \left[1 + \sum_{k=1}^{n-1} q^k \left(1 - \frac{\varphi_k^{(1)}}{\varphi_n^{(1)}} \right) \right]^{-1} \approx \frac{1-q}{1 - q^n n(1-q) \ln[n(1-q)]},$$

which are in good agreement with the results of preceding researches.

4. Let us turn to nonisothermal flow. The level-by-level kinetic equations have the form

$$\frac{dx_i}{dt} + x_i \frac{1}{N} \frac{dN}{dt} = P_{10} Z [(i+1)(x_{i+1} - qx_i) - i(x_i - qx_{i-1})], \quad (4.1)$$

$$i = 0, \dots, n-1, \quad x_n = 0.$$

Let us substitute (3.2) in these equations for $q = q_1$ by considering q_1 a quantity not known in advance, and let us use the following properties of the functions $\varphi_k^{(i)}$:

$$\frac{d\varphi_k^{(i)}}{dq} = -\frac{k}{(1-q)q} (\varphi_k^{(i)} - \varphi_{k-1}^{(i)}) \quad \text{for } i \geq k-1,$$

$$\frac{d\varphi_k^{(k)}}{dq} = -\frac{k}{(1-q)q} \varphi_k^{(k)}$$

(see the Appendix for the proof of these properties). We obtain

$$\frac{1}{1-q_1} \frac{dq_1}{dt} \left[x_k - x_{k-1} + \frac{1-q_1}{k} q_1^k \frac{d\mu_0}{dq_1} \sum_{j=1}^k j (-\mu)^{j-1} \varphi_k^{(j)} \right] = P_{10} Z (q - q_1) (\bar{x}_k - \bar{x}_{k-1}). \quad (4.2)$$

There results from these equations that no q_1 exists independent of k and identically satisfying all the equations for $k = 1, \dots, n-1$. Thus, for $k = 1$ there follows $dq_1/dt \approx (q - q_1)/\tau$ from (4.2) while $dq_1/dt \approx q - q_1/[\tau(1 - q_1)]$ for $k = n-1$. If $q_1 \ll 1$ (the low-temperature case $KT \ll E_1$), these equations can be considered equivalent. This means that in this case (3.2) with a lag determined by (2.2) will be a solution of the system (4.1).

It here possesses local properties only if the inequality $\frac{E_1}{KT} \frac{\tau}{|\theta|} \ll 1$ is satisfied. In the opposite case, the relaxation equation (2.2) must be solved to determine the lag. Let us also note that for $q_1 \ll 1$ the quasistationary distribution function (3.2) is close to a Boltzmann one at all energetic levels.

Let us convert (4.1) by making the substitution $x_i = \bar{x}_i x_0$ and using the equation for x_0 :

$$\frac{dx_0}{dt} + x_0 \frac{1}{N} \frac{dN}{dt} = x_0 P_{10} Z (\bar{x}_1 - q).$$

Consequently,

$$\frac{d\bar{x}_i}{dt} + P_{10} Z (\bar{x}_1 - q) \bar{x}_i = P_{10} Z [(i+1)(\bar{x}_{i+1} - q\bar{x}_i) - i(\bar{x}_i - q\bar{x}_{i-1})], \quad i = 1, \dots, n-1, \quad \bar{x}_n = 0. \quad (4.3)$$

We will seek the solution of (4.3) under the condition $\tau/|\theta| \ll 1$ in the form

$$\bar{x}_k = \left(\prod_{i=1}^k q_i \right) \sum_{j=0}^k (-\mu)^j \varphi_k^{(j)}. \quad (4.4)$$

Here μ is the minimal root of the equation $\sum_{j=0}^n (-\mu)^j \varphi_n^{(j)} = 0$, while the functions $\varphi_n^{(j)}$ are defined by (3.3) for $q = q_1$, where q_1 is as yet unknown. Substituting (4.4) into (4.3), we have

$$\begin{aligned}
& \tau \frac{dq_1}{dt} \left[-k \sum_{i=0}^k (-\mu)^i \varphi_k^{(i)} + \frac{k}{q_1} \sum_{i=0}^{k-1} (-\mu)^i \varphi_{k-1}^{(i)} - \right. \\
& \left. - (1-q_1) \frac{d\mu}{dq} \sum_{i=1}^k i (-\mu)^{i-1} \varphi_k^{(i)} \right] + (q-q_1) \left[k \sum_{i=0}^k (-\mu)^i \varphi_k^{(i)} - \right. \\
& \left. - \frac{k}{q_k} \sum_{i=0}^{k-1} (-\mu)^i \varphi_{k-1}^{(i)} \right] - (q_{k+1}-q_1) (k+1) \sum_{i=0}^{k+1} (-\mu)^i \varphi_{k+1}^{(i)} + \\
& + (q_k-q_1) \frac{k}{q_k} \sum_{i=0}^{k-1} (-\mu)^i \varphi_{k-1}^{(i)} = \tau (1-q_1) \sum_{i=0}^k (-\mu)^i \varphi_k^{(i)} \times \\
& \times \sum_{j=1}^k \left(\frac{1}{q_j} \frac{dq_j}{dt} - \frac{1}{q_1} \frac{dq_1}{dt} \right), \quad q_n = q, \quad k = 1, \dots, n-1.
\end{aligned} \tag{4.5}$$

The quasistationary solution of this system can be found by an iteration method by substituting the value of q_1 from the preceding approximation into the right side of the equations. Thus, if $q_1 = q$ is taken as the zeroth approximation, which corresponds to a distribution for $T = \text{const}$, then the solution of (4.5) with right sides equal to zero will be the first approximation.

The system of equations obtained in such manner contains n equations and n unknowns q_1, \dots, q_n as well as the derivative dq_1/dt . It can be split into one differential equation for q_1 and $n-1$ algebraic equations from which q_2, \dots, q_n can be found as functions of q_1 and dq_1/dt . Operating in such manner, we obtain the first iteration

$$q_k = q_1 + \tau \frac{dq_1}{dt} \frac{1}{k q_1^{k-1} a_k a_{k-1}} \sum_{m=k}^{n-1} q_1^m a_m c_m, \tag{4.6}$$

where $a_m = \sum_{i=0}^m (-\mu)^i \varphi_m^{(i)}$;

$$\begin{aligned}
c_m &= (1-q_1) \frac{d\mu}{dq_1} \sum_{i=1}^m i (-\mu)^{i-1} \varphi_m^{(i)} + \delta m \left(a_m - \frac{a_{m-1}}{q_1} \right); \\
\delta &= (1-q_1) \frac{d\mu}{dq_1} \left[\sum_{k=1}^{n-1} q_1 a_k \sum_{i=1}^k i (-\mu)^{i-1} \varphi_k^{(i)} \right] \left[\sum_{k=1}^{n-1} k a_k (q_1^{k-1} a_{k-1} - q_1^k a_k) \right]^{-1},
\end{aligned}$$

or approximately for $n(1-q_1) \gg 1$, $(1-q_1) \ll 1$

$$\begin{aligned}
\delta &\approx n^2 (1-q_1)^2 q_1^n \ln [n(1-q_1)], \\
q_k &= \begin{cases} q_1 + \tau \frac{dq_1}{dt} \delta \frac{1-q_1^{k-1}}{k(1-q_1) q_1^{k-1}}, & k(1-q_1) \ll 1, \\ q_1 + \tau \frac{dq_1}{dt} \frac{n^2 (1-q_1) q_1^n}{k q_1^{k-1}} \ln \frac{n}{k}, & \begin{cases} k(1-q_1) \gg 1, \\ (n-k)(1-q_1) \gg 1, \end{cases} \\ q_1 + \tau \frac{dq_1}{dt} \frac{n q_1^{n-k+1}}{2k} \left[1 - \frac{1}{3} (n-k)(1-q_1) \right], & (n-k)(1-q_1) \ll 1. \end{cases}
\end{aligned} \tag{4.7}$$

The differential equation for q_1 in this approximation has the form

$$dq_1/dt = (q-q_1)/[\tau(1-\delta)], \tag{4.8}$$

for $\frac{\tau dq}{q dt} \ll 1$, $q_1 \approx q \left[1 - \frac{\tau dq}{q dt} (1-\delta) \right]$. The expression (4.7) for δ can be considered for $(1-q) \ll 1$ approximately a function of $Y = \theta/KT = nE_1/KT \approx n(1-q)$; thus: $\delta \approx Y^2 e^{-Y} \ln Y$. The maximal value $\delta \approx 0.49$ is reached for $Y \approx 3$, while as $Y \rightarrow \infty$, $\delta \rightarrow 0$.

We make the next iteration by substituting the solution (4.6) into the right side of (4.5). Estimates show that the second approximation found in this manner differs from the first by corrections of the order of $(\tau/\theta)^2$.

Using (4.7) we find the function $\prod_{i=1}^k q_i$ in the expression (4.4) for the population of the k-th level:

$$\prod_{i=1}^k q_i = q_1^k \prod_{i=1}^k \left(1 + \frac{q_i - q_1}{q_1} \right) \approx q_1^k \exp \left(\sum_{i=1}^k \frac{q_i - q_1}{q_1} \right),$$

$$\sum_{i=1}^k \frac{q_i - q_1}{q_1} \approx \begin{cases} \frac{\tau}{\theta} \delta \frac{1 - q_1^{k-1}}{q_1^{k-1}}, & k(1 - q_1) \ll 1, \\ \frac{\tau}{\theta} \frac{\delta \ln(n/k)}{k(1 - q_1) q_1^{k-1} \ln[n(1 - q_1)]}, & \begin{cases} k(1 - q_1) \gg 1, \\ (n - k)(1 - q_1) \gg 1. \end{cases} \\ \frac{\tau}{\theta}, & k = n - 1, \end{cases}$$

It is hence seen that the distribution obtained differs from the lagging one by a correction of the order of τ/θ . Correspondingly, the macroscopic dissociation rate $(1/N)(dN/dt)$ equals the dissociation rate for a lagging temperature to the accuracy of terms of order τ/θ :

$$\frac{1}{N} \frac{dN}{dt} = -P_{10} Z n q x_{n-1} \approx -P_{10} Z n^2 (1 - q_1)^2 q_1^n \left(1 + \frac{\tau}{\theta} \right).$$

The function q_1 and its corresponding lagging temperature $T_1 = \frac{E_1}{K} \left[\ln \frac{1}{q_1} \right]^{-1}$ are determined from (4.8). The correction δ here exerts substantial influence on the dissociation rate if $(\tau/|\theta|) Y^3 e^{-Y} \ln Y$ is of the order of or greater than unity.

In conclusion, we note that the results obtained in this paper refer to the case of convective flow without taking account of diffusion transfer. As follows from [13], the diffusion process in a spatially inhomogeneous flow can exert substantial influence on the molecule distribution function over vibrational levels.

APPENDIX

We shall prove the equalities

$$\frac{d\varphi_k^{(k)}}{dq} = -\frac{k}{q(1-q)} \varphi_k^{(k)}; \quad (\text{A.1})$$

$$\frac{d\varphi_k^{(i)}}{dq} = -\frac{k}{q(1-q)} (\varphi_k^{(i)} - \varphi_{k-1}^{(i)}), \quad k > i. \quad (\text{A.2})$$

The validity of (A.1) is seen easily by differentiating the expression resulting from the definition of $\varphi_k^{(k)}$: $\varphi_k^{(k)} = (1 - q)^k / k! q^k$. The equality (A.2) is evident for $i = 0$. For $i > 0$ we prove it by induction. For all $i \geq m$ let (A.2) be true. For $i = m + 1$ we have

$$\frac{d\varphi_k^{(m+1)}}{dq} = \frac{d}{dq} \sum_{p=m+1}^k \sum_{r=m}^{p-1} \frac{(1-q) q^{r-p}}{p} \varphi_r^{(m)} = \sum_{p=m+1}^k \sum_{r=m}^{p-1} \frac{1}{p} [(r-p) q^{r-p-1} (1-q) \varphi_r^{(m)} - q^{r-p} \varphi_r^{(m)} - r q^{r-p-1} (\varphi_r^{(m)} - \varphi_{r-1}^{(m)})] = -\sum_{p=m}^{k-1} \frac{\varphi_p^{(m)}}{q} - \sum_{r=m}^{k-1} \frac{\varphi_r^{(m)}}{q} \left(\frac{1 - q^{k-r}}{q^{k-r}} \right) = -\sum_{r=m}^{k-1} \frac{\varphi_r^{(m)}}{q^{k-r+1}},$$

while on the other hand

$$-\frac{k}{(1-q)} [\varphi_k^{(m+1)} - \varphi_{k-1}^{(m+1)}] = -\frac{1}{q^{k+1}} \sum_{r=m}^{k-1} q^r \varphi_r^{(m)}$$

results from the definition of the functions $\varphi_k^{(i)}$. Therefore, the equality (A.2) is also true for $i = m + 1$, meaning for all i ($0 < i < k$) also.

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NONISOTHERMAL FLOW OF A POLYATOMIC GAS IN A CHANNEL
AND THE THERMOMOLECULAR PRESSURE DIFFERENCE EFFECT

V. M. Zhdanov, V. A. Zaznoba, and I. V. Safonova

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In [1] the nonisothermal flow of a rarefied polyatomic gas in a plane channel was considered, using the method of [2-4]. One of the results of [1] was an expression for the thermomolecular pressure difference (TPD) arising at the ends of the channel for a fixed temperature difference. It is known [5-13] that measurement of the TPD for polyatomic gases can serve as an independent source of information on the characteristics of the inelastic collisions between molecules, since the effect depends upon the translational part of the thermal conductivity (λ^t), which in turn depends explicitly on the rotational and vibrational collision numbers Z_r and Z_v . This was first discussed in [5, 14], where a theoretical expression for the TPD was obtained using the "dusty-gas" model. For small Knudsen numbers the dependence of the TPD effect on λ^t also follows from a rigorous kinetic analysis of viscous and thermal slipping in a polyatomic gas [15, 16]. The use of the dusty-gas model to interpret the experimental results involves, however, a whole set of parameters resulting from the model itself (empirical constants) and from the choice of an "average" temperature of the gas in the channel [6, 17]. This is evidently the reason for the rather large scatter in the results for Z_r for several polyatomic gases, as obtained by different authors [18]. A second cause of discrepancies between the results is the use in certain papers of nonrigorous expressions for the translational Eucken factor, in which the combined effect of the rotational and vibrational degrees of freedom of the molecules is not taken into account in a sufficiently correct way.

Since measurements of the TPD effect are usually done using packets of circular cylindrical capillaries, a more reliable method of interpreting the results should be based on

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