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Establishment of the vibrational distribution of diatomic molecules

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An analytic theory is developed of the establishment of the vibrational distribution function of diatomic anharmonic molecules. The solutions obtained are compared with the results of accurate numerical calculations. The rate of heating of a gas is found after the beginning of pumping and in the case of pulsed excitation of the vibrations.

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An interesting example of a system far from the equilibrium is a vibrationally excited gas of diatomic molecules. For many such molecules (for example, N_2, CO, HCl, H_2, etc.) the relaxation time τ_{V-T} of the conversion of the vibrational to the translational energy is long compared with the time taken to establish the vibrational distribution function (τ_{V-V}). This means that it is quite easy to establish a state in which a gas has low translational and rotational temperatures but is highly excited in the vibrational sense. Such a state has a number of special features which affect, for example, the dispersion of sound, the degree of dissociation of molecules, the relaxation of the vibrational energy into heat, and the possibility of a population inversion between neighboring vibrational-rotational levels (for a review see, for example, Ref. 1).

It is shown in Ref. 2 that, in the absence of the $V-T$ relaxation, the vibrational distribution of molecules is affected greatly by their anharmonicity. A further development of the idea of Ref. 2 allowing for the $V-T$ relaxation[1,3–6] has led to the establishment of an analytic theory describing the steady-state distribution of molecules between the vibrational levels.

We shall give an analytic theory of the transient processes of the establishment and relaxation of the vibrational distribution function under conditions far from equilibrium.

The evolution of the vibrational distribution function $f(v)$ can be described by a system of kinetic equations

$$\partial f/\partial t=\Pi_{v+1}-\Pi_v+i_v, \qquad (1)$$

where Π_{v+1} is the flux of vibrational populations in the space of vibrational numbers in a section between the v-th and $(v+1)$-th levels; i_v is the excitation frequency of the v-th level by external sources. The flux Π_{v+1} can be expressed in terms of the frequencies of the $V-V$ exchange $Q_{v+1,v}^{v',v'+1}$ and the frequencies of the collisional and radiative relaxations P_v and A_v:

$$\Pi_{v+1}=\sum_{v'}(Q_{v+1,v}^{v',v'+1}f_{v+1}f_{v'}-Q_{v,v+1}^{v'+1,v'}f_vf_{v'+1})+(P_{v+1}+A_{v+1})f_{v+1}. \qquad (2)$$

Application of a familiar theory[3,5,6] makes it possible to simplify considerably the expression for the flux in the case of a state far from equilibrium. The main assumption of this theory,[3,5,6] which is the smallness of the change in f_v in the $|\Delta v|\leq\Delta v_Q$ case [Δv_Q is the $V-V$ exchange radius representing a reduction in the probability of exchange in processes with an energy defect (as a rule, we have $\Delta v_Q\approx 1-3$)], makes it possible to adopt a continuous variable v, use an analog of the Fokker–Planck approximation, and obtain the following differential form for Π_{v+1}:

$$\Pi(v)=\nu\left\{\frac{\partial}{\partial v}\left[(v+1)^2f^2(v)\left(1-\frac{T}{2\Delta E}\frac{d^2\ln f}{dv^2}\right)\right]\right.$$
$$\left.+2\mu\left(\exp\{\delta_{V-T}v\}+\frac{A_{10}}{P_{10}}\right)f(v)(v+1)\right\}. \qquad (3)$$

Specifically, we can use the following approximation for the $V-V$ exchange rate:[1,5,6]

$$Q_{v+1,v}^{v',v'+1}=Q_{10}(v+1)(v'+1)\exp(-\delta_{V-V}|v-v'|), \qquad (4)$$

where δ_{V-V} is the reciprocal $V-V$ exchange radius[1,5,6] depending on the type of molecule and gas temperature

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T; $\delta_{V-V} \propto T^{-1/2}$. Equation (3) is derived using the following relationships for the $V-T$ relaxation and spontaneous loss rates:

$$P_{v+1}=P_{10}(v+1)\exp\{\delta_{V-T}v\},\; A_{v+1}=A_{10}(v+1),$$

where δ_{V-T} is a number analogous to δ_{V-V}; for a pure gas of diatomic moleucles, we have $\delta_{V-T}=\delta_{V-V}$ (Refs. 1, 5, and 6). Subject to these approximations, we have

$$\nu=\frac{4\Delta E}{T}\delta_{V-V}^{-3}Q_{10}, \tag{5}$$

where ΔE is the anharmonicity energy; $\mu=P_{10}/2\nu \ll 1$.

Equation (3) is derived ignoring also the thermal excitation of the vibrations, which may be important in the case of sufficiently high levels.

When an external source excites only the lower vibrational levels, Eq. (1) has the solution $\Pi(v)=\text{const}$ under steady-state conditions for the majority of the vibrational numbers. An analysis of the solution employing $\Pi(v)$ given by Eq. (3) shows that there are two characteristic vibrational numbers v^* and v^{**} (Refs. 3 and 4–6).

The first of them, v^*, is usually called the Treanor number and it is given by

$$v^*=\frac{E_1}{2\Delta E}\frac{T}{T_1}+\frac{1}{2}, \tag{6}$$

where E_1 is the energy of the first vibrational quantum and T_1 is the effective vibrational temperature of the first excited level. The Treanor number characterizes a change from a near-Boltzmann distribution with a temperature T_1 to a plateau $f(v)=C/(v+1)$. The second vibrational number, v^{**}, is found from the condition that the $V-T$ loss rate is negligible in the region of the plateau but becomes comparable with the $V-V$ exchange rate and alters significantly the distribution function:

$$v^{**}=\delta_{V-T}^{-1}\ln C\delta_{V-T}\mu^{-1}. \tag{7}$$

The constant C, which determines the height of the plateau, can be related to the rate of vibrational excitation (pumping):[4,5,7]

$$C=(W\eta_V/E_1\nu)^{1/2}, \tag{8}$$

where W is the pump power per molecule and η_V is the fraction of the power used to excite the vibrations. It should be noted that in the region of the plateau (in the range $v^*<v<v^{**}$) the expression for a particle flux simplifies considerably to

$$\Pi(v)\approx\nu\frac{\partial}{\partial v}[(v+1)^2f^2].$$

We shall be interested mainly in the evolution of the distribution function in the region of the plateau and we shall use a simplified expression for the particle flux. The problem then reduces to the solution of the equation

$$\frac{\partial\psi}{\partial t}=\nu(v+1)\frac{\partial^2\psi^2}{\partial v^2}, \tag{9}$$

where $\psi=(v+1)f(v)$. Equation (9) resembles the nonlinear equation describing conduction of heat in an inhomogeneous medium.[8]

The quantity ν which has the dimensions of frequency may vary with time if, for example, the gas temperature is varied. In this case we can introduce a dimensionless variable

$$\tau=\int_0^t\nu\,dt \tag{10}$$

to reduce Eq. (9) to

$$\frac{\partial\psi}{\partial\tau}=(v+1)\frac{\partial^2\psi^2}{\partial v^2}. \tag{11}$$

The special features of the problems under discussion are that the boundary conditions are not known for Eq. (11). This equation is applicable in a certain range of vibrational numbers (under steady-state conditions from v^* to v^{**}), whose position and dimensions can be found at the end of the solution. Equation (11) has a class of self-similar solutions of the type

$$\psi=\tau^{\alpha-1}\varphi\left(\frac{v+1}{\tau^\alpha}\right), \tag{12}$$

where φ satisfies the ordinary equation

$$\xi(\varphi^2)''+\alpha\xi\varphi'+(1-\alpha)\varphi=0, \tag{13}$$

$\xi=(v+1)/\tau^\alpha$, and a prime denotes differentiation.

It is convenient to classify these solutions by means of the quantity

$$n=\int\psi\,dv$$

representing the average number of vibrational quanta per molecule. Using Eq. (12), we easily find that

$$n=\int\psi\,dv=\tau^{2\alpha-1}\int\varphi(\xi)\,d\xi. \tag{14}$$

If the self-similar function $\varphi(\xi)$ is integrable, then Eq. (14) describes the evolution in time of the average number of quanta per molecule. In particular, $\alpha=1$ corresponds to a sudden change in the pump power as a result of which the average number of quanta begins to vary linearly with time. The case $\alpha=1/2$ corresponds to pulsed excitation of the system for a short time. In this case $n=n_0$ represents the energy contributed per pulse. The case $\alpha=3/2$ corresponds to linear variation of the pump power with time, and so on.

Normalization of the distribution function

$$\int f\,dv=1,$$

which should naturally be retained in the full solution, does not appear in the self-similar solutions with the exception of the $\alpha=1$ case. This is a consequence of the approximate nature of Eq. (11). Failure to retain the normalization reflects the accumulation or loss of particles at the levels not described by Eq. (11) (these are the low levels and fairly high levels).

We shall now consider in greater detail the cases of a sudden change in the pumping ($\alpha = 1$) and of pulsed excitation ($\alpha = 1/2$) when the solutions of Eq. (13) are relatively simple. In particular, if $\alpha = 1$, the function $\varphi(\xi)$ is found from

$$\varphi - C_1 + C_0 \ln\frac{\varphi - C_0}{C_1 - C_0} = -\frac{\xi}{2}, \tag{15}$$

where

$$\xi = \frac{v+1}{\tau}, \quad C_1 = \lim_{\xi\to 0}\varphi(\xi), \quad C_0 = \lim_{\xi\to\infty}\varphi(\xi).$$

Thus, the constants C_1 and C_0 represent, in accordance with the meaning of the solution, the "new" and "old" pump powers; C_1 and C_0 can be found using the relationship (8). In particular, if an excitation source is switched on at $t = 0$ ($C_0 = 0$), then

$$\varphi(\xi) = C_1 - \xi/2.$$

In this case, we find that

$$f(v) = C_1/(v+1) - \left(2\int \nu\, dt\right)^{-1}. \tag{16}$$

A comparison of the solution of Eq. (16) with the results of an exact numerical calculation based on the complete system (1), carried out by Kochetov and Pevgov (for the calculation method see Ref. 9), shows that Eq. (16) does indeed describe well the exact solution in a wide range of parameters (Fig. 1) and the constant C_1 is related to the pump power by Eq. (8). Thus, establishment of the vibrational distribution function after switching on the pump power is described by Eq. (16), which is not a trivial expression: the solution (16) differs from that which can be obtained from the steady-state distribution functions (the constant C_1 varies with temperature T_1—see Refs. 4-6). Clearly, the distribution function (16) is applicable when a plateau of significant width appears, i.e., when $2C_1\nu t > v^* + 1$. On the other hand, the validity of Eq. (16) is limited by the condition $v < v^{**}$, known from the steady-state theory.

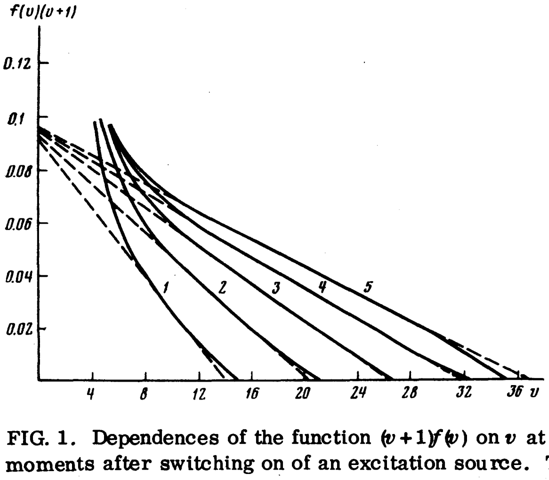

FIG. 1. Dependences of the function $(v+1)f(v)$ on v at various moments after switching on of an excitation source. The continuous curves represent exact numerical calculations and the dashed curves are based on Eq. (16). The results are plotted for a CO–He mixture with $N_{CO} = 10^{16}$ cm^{-3} and $N_{He} = 10^{18}$ cm^{-3} at $T = 175°$ K subjected to excitation at a rate of $W = 0.13$ W/cm^3. Curves 1–5 correspond to $t = 2$, 3, 4, 5, and 6 msec, respectively.

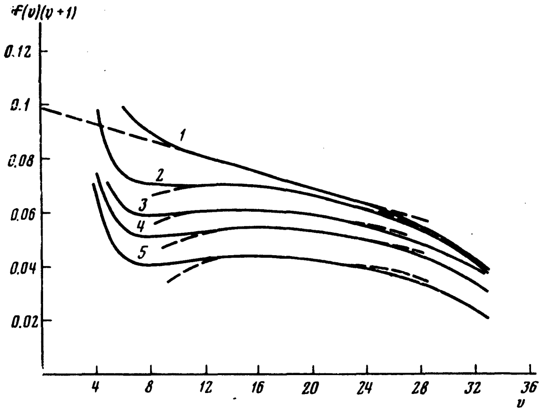

FIG. 2. Dependences of the function $(v+1)f(v)$ on v at various moments after switching off of an excitation source. The mixture and the parameters have the same values as for Fig. 1. The continuous curves represent exact numerical calculations and the dashed curves are based on Eq. (15) with $C_1 = 0$. Curves 1–5 correspond to $t = 0$, 1, 2, 3, and 4 msec, respectively.

The case when $C_1 = 0$ in Eq. (15) corresponds to switching off of a continuously acting excitation source. After a short time $[t \ll (v+1)/2\nu C_0]$, we have

$$f(v) = \frac{C_0}{v+1}\left[1 - \exp\left\{-(v+1)/2C_0\int \nu\, dt\right\}\right],$$

and after a long time $[t \gg (v+1)/2\nu C_0]$, we obtain

$$f(v) = \left[C_0/(v+1)\int \nu\, dt\right]^{1/2}.$$

We can see from these formulas that a perturbation resulting from switching off of a pump source affects first the small values of v and after a considerable time the distribution function deviates significantly from the steady-state form by becoming even flatter.

A comparison of these formulas with the results of the exact numerical calculations of Kochetov and Pevgov also demostrate the validity of the analytic theory (Fig. 2).

In the case of instantaneous (pulsed) excitation the molecules in a gas are usually excited to fairly low vibrational levels ($v \le 8$). The V–V exchange produces two groups of excited moleucles: strongly excited (high values of v) and weakly excited ($v = 1$–2). The strongly excited molecules are transferred to even higher levels because of the energy defect in the V–V exchange, whereas the weakly excited ones drop even lower. This effect is described by the self-similar solution with $\alpha = 1/2$. Integration of Eq. (13) then gives

$$\varphi(\xi) = (^3/_4 n_0)^{1/4}\xi^{1/2} - {}^1/_2\xi,$$

where the integration constant is expressed in terms of n_0 defined by Eq. (14).

The distribution function $f(v,t)$ is

$$f(v,t)=\left(\frac{3}{4}n_0\right)^{1/4}(v+1)^{-1/2}\left(\int\nu\,dt\right)^{-3/4}-\left(2\int\nu\,dt\right)^{-1}$$
$$=\left(2\int\nu\,dt\right)^{-1}\left[\left(12n_0\int\nu\,dt\right)^{1/4}\Big/(v+1)^{1/2}-1\right]. \qquad (17)$$

As pointed out above, the normalization of Eq. (17) is not retained and in fact we have

$$\int f\,dv\sim\frac{1}{t^{1/2}}.$$

The effective reduction in the number of particles now corresponds to their accumulation in the ground state. We can see from Eq. (17) that, for a given v, the function $f(v,t)$ first rises and then falls. For a constant ν the maximum of $f(v,t)$ shifts with time toward higher values of v in accordance with the law $t_{\max}\propto v^2n_0$. The distribution function itself and the parametric dependences which follow from it are in good agreement with the results of exact calculations of Kochetov and Pevgov (Fig. 3).

We shall illustrate the application of the results obtained by calculating the rate of heating of a gas due to conversion into heat of the anharmonic energy defect in the $V-V$ exchange. In the continuum approximation, the expression for the heat evolution rate per molecule can be reduced to

$$q_{V-V}=2\Delta E\nu\int(v+1)^2f^2\left(1-\frac{T}{2\Delta E}\frac{d^2\ln f}{dv^2}\right)dv. \qquad (18)$$

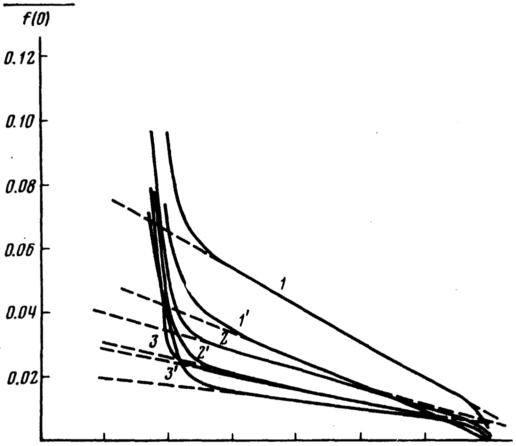

FIG. 3. Dependences of the function $f(v)\sqrt{v+1}/f(0)$ on $\sqrt{v+1}$ for the case of pulsed excitation. The continuous curves represent exact numerical calculations and the dashed curves are based on Eq. (17). The results are given for a CO–Ar = 1 :10 mixture at $P=100$ Torr and $T=83°$ K. For curves 1–3 and 1′–3′ the value of n_0 is 4.9 and 2.45, respectively. Curves 1 and 1′, 2 and 2′, and 3 and 3′ correspond to $t=10$, 20, and 30 msec, respectively.

Since the average number of quanta per molecule is conserved in the $V-V$ exchange processes, it follows that the main contribution to the gas heating is made by the $V-V$ exchange of highly excited particles accompanied by the conversion into heat of a relatively large amount of energy. We can easily show formally that, with a high precision, Eq. (18) can be simplified to

$$q_{V-V}=2\Delta E\nu\int(v+1)^2f^2\,dv=2\Delta E\nu\int\psi^2\,dv. \qquad (19)$$

Application of our self-similar solutions allows us to calculate easily the proportion of the power (or energy) used to heat the gas. In particular, in the case when an excitation source is switched on, we have

$$\frac{q_{V-V}}{E_1\dot{n}}=\frac{4}{3}\frac{\Delta E}{E_1}C_1\int\nu\,dt,\qquad \dot{n}=\nu C_1^2, \qquad (20)$$

whereas after a pump pulse, we obtain

$$\frac{q_{V-V}}{E_1n_0}=\frac{2}{5}\frac{\Delta E}{E_1}\left(3n_0\int\nu\,dt\right)^{1/2}\nu\Big/\int\nu\,dt. \qquad (21)$$

For time intervals such that the heating because of the $V-T$ relaxation is still weak, we obtain the following expression for the temperature rise after pulsed excitation:

$$C_v\frac{dT}{dt}=\frac{2\Delta E}{15}\frac{\nu(T)}{\tau^{1/2}}(3n_0)^{3/2}. \qquad (22)$$

To be specific, we shall consider a gas of a given density (specific heat C_V) and in the range of times of interest to us we shall ignore heat conduction. Bearing in mind that

$$\tau=\int_0^t\nu(T)\,dt,$$

and solving Eq. (22), we obtain

$$\int_{T_0}^{T}\frac{(T-T_0)}{\nu(T)}dT$$
$$=2(3n_0)^3\left(\frac{2\Delta E}{15C_v}\right)^2t, \qquad (23)$$

where T_0 is the initial temperature of the gas. Employing the explicit dependence of quantities Q_{10} and δ_{V-V} on T [see Eq. (5)], we can find from Eqs. (17) and (23) the evolution of the distribution function allowing for the gas heating. Over short time intervals, as long as $\nu(T)$ does not vary and is equal to $\nu(T_0)$, we find from Eq. (23) the dependence of the temperature rise on the contribution of n_0 and on time:

$$\Delta T\propto(3n_0)^{3/2}[\nu(T_0)t]^{1/2}.$$

We can easily calculate also the rate of the $V-T$ relaxation from the levels whose populations are described by our self-similar solutions:

$$q_{V-T}=\hbar\omega(\tilde{v})P_{10}\int(v+1)f(v)\exp\{\delta_{V-T}v\}\,dv. \qquad (24)$$

Here, $\omega(\tilde{v})$ is the frequency of transitions involving

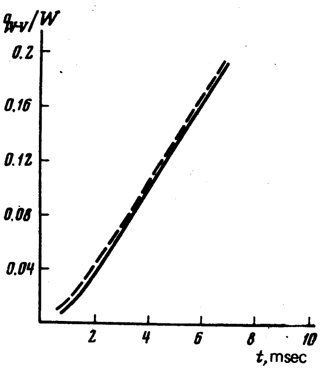

FIG. 4. Proportion of the power converted into heat by the V—V processes in the case of switching on of an excitation source. The mixture and the values of the parameters are the same as for Fig. 1. The continuous curve represents the results of a numerical calculation and the dashed curve is based on Eq. (19).

the levels for which the probability of the $V-T$ relaxation is now low ($\tilde{v} \approx v^{**}$).

We shall now give the results of a calculation of q_{V-T} for time intervals $t \ll P_{10}^{-1}$ in the case when pumping is switched on:

$$q_{V-T}=\frac{1}{2}\hbar\omega(\tilde{v})P_{10}\delta_{V-T}^{-2}\left(\int\nu\,dt\right)^{-1}\exp\left\{2\delta_{V-T}C_1\int\nu\,dt\right\} \tag{25}$$

and in the case of pulsed excitation:

$$q_{V-T}=\frac{1}{4}\hbar\omega(\tilde{v})P_{10}\delta_{V-T}^{-2}\left(\int\nu\,dt\right)^{-1}\exp\left\{4\delta_{V-T}\left(\frac{3}{4}n_0\int\nu\,dt\right)^{1/2}\right\}. \tag{26}$$

We shall conclude by considering briefly the limits of validity of our solutions. As pointed out above, Eq. (9) describes the change in the distribution function in the intermediate range of vibrational numbers. It is therefore clear that the self-similar solutions are inapplicable to those cases when there are significant changes in $f(v)$ at the lower levels, not described by Eq. (9). This is true, for example, after an abrupt interaction (switching on of a continuously acting or a pulsed pump source) for time intervals shorter than the time needed for the excitation wave to reach the first characteristic vibrational number v^*, known as the Treanor number $[t \lesssim v^*/2\nu C$ or $t \lesssim (v^*)^2/12n_0\nu]$.

On the other hand, the part of the distribution function corresponding to fairly high levels may be affected considerably by the $V-T$ processes. When the excitation wave reaches this region ($v \approx v^{**}$), Eqs. (20), (21), (25), and (26) all become invalid; to calculate q_{V-V} and q_{V-T}, we have to allow for the distortion of the distribution function by the $V-T$ processes. This can be done approximately by regarding the relaxation terms in Eq. (3) as a perturbation. The values of q_{V-V} found in this way are compared with other calculations in Figs. 4 and 5. (The formulas are not given because they are too cumbersome.) We can see that the precision of the analytic theory is quite satisfactory.

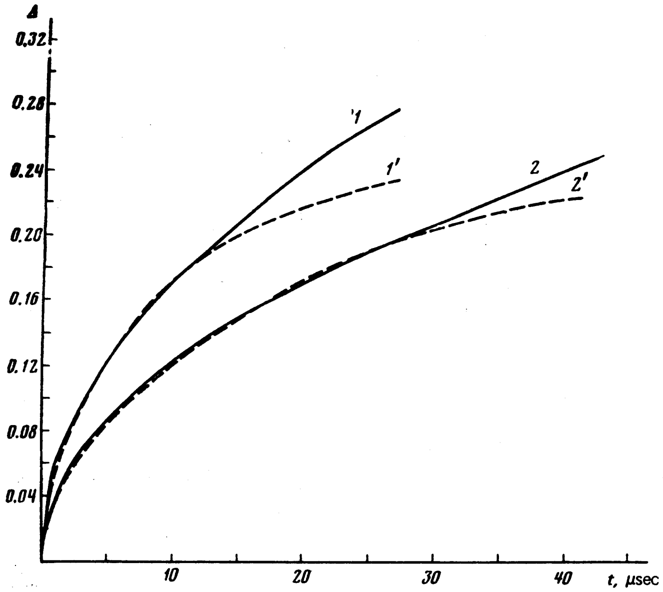

FIG. 5. Proportion of the energy $\Delta=(1/E_1n_0)\int_0^t q_{V-V}dt$, converted into heat by the V—V processes in the case of pulsed excitation. The dashed curves represent exact numerical calculations and the continuous curves are based on Eq. (21) for $T=\text{const}$. The mixture and the parameters are the same as for Fig. 3. For curves 1 and 1′ we have $n_0=4.9$ and for curves 2 and 2′ we have $n_0=2.45$.

In the problem of switching off an excitation source the initial state is such that the influence of the $V-T$ processes cannot be ignored even during the first few moments. The $V-T$ processes at the upper levels ($v \approx v^{**}$) are then governed by the gas heating.

The approximate solution for $\Pi(v)$ is valid when the distribution function $f(v)$ has a region where there is little change in $f(v)$. Clearly, this happens in the case of strongly inhomogeneous vibrational excitation. A quantitative criterion for strong inhomogeneity (lower limit of the pump power of energy) governing the validity of our self-similar solutions is of interest on its own account and will be considered in a separate communication.

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Radiation emitted by a shock-compressed high-pressure argon plasma

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Results are presented of measurements of the coefficient of optical absorption of a non-ideal argon plasma produced by powerful ionizing shock waves. The experiments were performed with explosive generators of rectangular shock waves, using condensed explosives. The obtained radiation of the shock-compressed plasma turned out to be much less than theoretical. The "transparentization" of the plasma is explained by using a bounded-atom model that takes into account the deformation of the electron energy spectrum.

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1. INTRODUCTION

The solution of many present-day problems of radiation gasdynamics and plasma physics, such as laser irradiation,[1] hypersonic aerodynamics,[2] explosive light sources,[3] gas-phase nuclear reactors,[4] and others call for detailed information on the optical properties of a high-density plasma in which the effects of interaction between particles play an essential role. An investigation of the radiation of such a plasma is also of definite fundamental interest, since it makes it possible to trace the influence of deviation from ideal conditions on the dynamics and on the energy spectrum of a dense disordered plasma.

The theoretical premises developed to date[5] pertain to a tenuous plasma, where the elementary processes can be easily separated, and the influence of deviations from ideal conditions reduces to a broadening of the spectral lines and to shifts of the photorecombination thresholds. The region of increased densities has become the object of investigations only most recently (see Refs. 6–8), and the degree to which it has so far been investigated is patently insufficient. The data available at present correspond to non-ideality parameters $\Gamma = e^3(8\pi m_e)^{1/2}/(kT)^{3/2} \lesssim 0.2-0.3$, $n_e \leqslant 10^{18}$ cm^{-3} and are to a great degree contradictory, but indicate nevertheless that the continuum radiation exceeds the calculated value when the plasma is increased. At the same time, the plasms screening[9] and the strongly fluctuating microfields[10] can noticably distort the near-threshold region of the spectrum, by decreasing the intensity of the recombination radiation of the dense plasma.[7,8,11]

In this paper we measure the optical properties of a strongly non-ideal ($\Gamma \sim 1.6$) plasma obtained by compression and irreversible heating of a high-pressure gas in the front of a high-power ionizing shock wave (SW). The use of dynamic methods has made it possible to obtain a plasma with extremely high parameters in volumes that are homogeneous and of controllable dimensions. In addition, in view of the small dimensions of the zone of relaxation processes and of the small thickness of the viscous shock discontinuity, the optical radiation is capable of emerging unobstructed from the plasma volume and carries therefore information on the equilibrium properties of the shock-compressed plasma. The absence of complex molecular and ion-molecular formations, the fact that the electron energy spectrum in the elementary radiative processes has been investigated in detail, as well as the high molecular weight, have all dictated the choice of argon as the object of the investigation. In addition, extensive information on the gas dynamic, thermodynamic,[12,13] and electrophysical[14] properties of a dense argon plasma has by now been obtained, so that the state of the plasma can be assessed with great certainty and the interpretation of the optical measurements is greatly facilitated.

The use of explosive dynamic methods has made it possible to trace the influence of the non-ideality on the absorption coefficient of the plasma in a broad and continuously varying range from the previously investigated states with $n_e \lesssim 10^{18}$ cm^{-3} and $\Gamma \lesssim 0.3$ to the extremely high $n_e \sim 2 \times 10^{20}$ cm^{-3} and $\Gamma \sim 1.6$ ($P \sim 5000$ atm), where the effect of "transperentization" of the plasma manifests itself. This effect is interpreted in the present paper from the point of view of distortion of the energy spectrum of the strongly compressed atoms by the strong interparticle interaction.

For a qualitative description of this effect, we use the