DISPERSION AND ABSORPTION OF ULTRASOUND IN A VIBRATIONALLY EXCITED
GAS OF ANHARMONIC MOLECULES

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Measurement of the dispersion and absorption of ultrasound is an important way of studying relaxation processes in molecular gases. With the development of laser and plasma techniques, interest in vibrationally excited molecular gases has increased significantly in recent years. For a high degree of vibrational excitation the theory of Landau and Teller [1], which describes the relaxation of harmonic oscillators, becomes inapplicable. This is because of the significant effect of the anharmonicity of the molecular vibrations on vibrational−translational (V−T) and vibrational−vibrational (V−V) processes of energy exchange. The theoretical study of the dispersion and absorption of ultrasound in a vibrationally excited gas carried out in [2-4] on the basis of the theory [1] did not take into account the anharmonicity of the molecular vibrations and therefore it cannot be applicable under strongly nonequilibrium conditions. The purpose of the present paper is to describe sound propagation in a system of excited anharmonic molecules.

1. Gasdynamical Description. We consider a plane sound wave propagating in an inviscid vibrationally excited gas of diatomic molecules at rest. Let the nonequilibrium state of the system be maintained by an external source q, which pumps energy into the vibrational degree of freedom of the molecules. Since we are considering a significantly nonequilibrium situation, the system is inhomogeneous in principle. However if the wavelength λ is much smaller than the characteristic linear dimension L of the inhomogeneities:

$$\lambda \ll L, \tag{1.1}$$

one can assume that the sound propagates on a homogeneous stationary background.

The system of gasdynamical equations describing the evolution of the density n, velocity u, and temperature T of an inviscid gas has the form

$$\frac{dn}{dt} + n(\nabla u) = 0, \quad \frac{du}{dt} = -\frac{1}{mn}\nabla p, \quad \frac{d}{dt}\left(cT + \frac{mu^2}{2} + \varepsilon\right) = -\frac{1}{n}\nabla(pu) + q. \tag{1.2}$$

Here p = nT is the gas pressure; m is the mass of the component particles; c is the heat capacity at constant volume associated with the translational and rotational degrees of freedom; ε is the vibrational energy per molecule; $d/dt = \partial/\partial t + (u\nabla)$.

The system (1.2) must be supplemented by an equation for the time derivative of the quantity ε:

$$d\varepsilon/dt = -S + q \tag{1.3}$$

(S is the rate of relaxation of vibrational energy). In general S depends on the state of the gas and on the strength of the source. With no loss in generality, we write S in the form

$$S = (\varepsilon - \varepsilon_{eq})/\tau, \tag{1.4}$$

where ε_{eq} is the equilibrium value of ε corresponding to the gas temperature T and $\tau = \tau(n, T, q)$ is an effective vibrational relaxation time.

All variables describing the sound wave are written in the form

$$a = a_0 + a' \exp(-i\omega t + ikx). \tag{1.5}$$

Here ω is the angular frequency; k is the wave number (k = k_0 − iδ); a_0 is a constant corresponding to the gas unperturbed by the sound wave ($a' \ll a_0$). Linearizing (1.2) and (1.3) in the small perturbations (1.5), we obtain the dispersion relation for the propagation of

acoustic oscillations in a nonequilibrium system, assuming the condition (1.1):

$$\frac{k^2 u_T^2}{\omega^2} = \frac{c + \dfrac{i}{\omega}\left(\dfrac{c + c_v}{\tau_0} + \alpha_1\right) + \dfrac{1}{\omega^2}\beta_1}{c + 1 + \dfrac{i}{\omega}\left(\dfrac{c + 1 + c_v}{\tau_0} + \alpha_1 + \alpha_2\right) + \dfrac{1}{\omega^2}(\beta_1 + \beta_2)}, \tag{1.6}$$

where $u_T = (T_0/m)^{1/2}$ is the isothermal speed of sound; $c_v = \partial \varepsilon/\partial T$ is the equilibrium heat capacity of the vibrational degrees of freedom of the molecules; α and β are given by

$$\alpha_1 = \frac{q_0}{\tau_0}\frac{\partial \tau}{\partial T} - \frac{\partial q}{\partial \varepsilon}\left(1 + \frac{q_0}{\tau_0}\frac{\partial \tau}{\partial q}\right), \quad \beta_1 = \frac{1}{\tau_0}\frac{\partial q}{\partial T} + \frac{1}{\tau_0}\frac{\partial q}{\partial \varepsilon}\left(c_v + q_0\frac{\partial \tau}{\partial T}\right),$$

$$\alpha_2 = \frac{q_0}{T_0}\left(1 - \frac{n_0}{\tau_0}\frac{\partial \tau}{\partial n}\right) + \frac{n_0}{T_0}\frac{\partial q}{\partial n} - \frac{\partial q}{\partial \varepsilon}\left(1 + \frac{q_0}{\tau_0}\frac{\partial \tau}{\partial q}\right), \tag{1.7}$$

$$\beta_2 = \frac{q_0}{T_0}\left[\frac{1}{\tau_0}\left(1 + \frac{n_0}{q_0}\frac{\partial q}{\partial n}\right) + \frac{\partial q}{\partial \varepsilon}\left(1 + \frac{q_0}{\tau_0}\frac{\partial \tau}{\partial q}\right) + \frac{n_0}{\tau_0}\frac{\partial q}{\partial \varepsilon}\frac{\partial \tau}{\partial n}\right].$$

In the special case of propagation of ultrasound in a gas in equilibrium ($q = 0$, $\alpha = 0$, $\beta = 0$), (1.6) reduces to the classical equation of relaxation theory [5]. When the deviation from equilibrium is weak we have $\partial \tau/\partial q = 0$ and for the additional conditions $\partial q/\partial T = 0$, $\partial q/\partial n = 0$ assumed in [4], (1.6) reduces to the result obtained in [4]. In order to describe sound propagation in a system of excited anharmonic oscillators, it is necessary to determine τ in (1.6), which can only be done by considering the fundamental processes of vibrational kinetics occurring in the system.

2. Kinetic Model.

As follows from (1.4), τ is defined by the relation

$$\tau = (\varepsilon - \varepsilon_{eq})/S. \tag{2.1}$$

For a system of anharmonic oscillators, S is found from the following system of kinetic equations for the vibrational energy (E_v) distribution function f_v of the molecules

$$\partial f_v/\partial t = -\Pi_{v+1} + \Pi_v + i_v. \tag{2.2}$$

Here Π_v is the flux of population density of the excited molecules in the space of the vibrational quantum number v; i_v is the excitation frequency of the v-th vibrational level by the external source. Explicit expressions for Π_v can be found in [6, 7]. For anharmonic oscillations E_v is usually written in the form $E_v = v[E_1 - \Delta E(v - 1)]$ (ΔE is the energy due to anharmonicity). There is a simple relation between the parameters q and i_v: $q = \sum_v E_v i_v$.

Multiplying (2.2) by E_v and summing over all v, we have

$$\partial \varepsilon/\partial t = -E_1\Phi(v^{**}) + \Delta E\Gamma(v^{**}) + q, \quad \varepsilon = \sum_v E_v f_v. \tag{2.3}$$

Comparing (1.3) and (2.3), we find

$$S = E_1\Phi(v^{**}) - \Delta E\Gamma(v^{**}). \tag{2.4}$$

Here the quantum number v^{**} corresponds to the boundary of the nonequilibrium region of the distribution f_v; $\Phi(v)$ and $\Gamma(v)$ are the flux of quanta and flux of quantum "defect" in the space of the quantum number v. These functions are given by

$$\Phi(v) = v\Pi_v - \sum_{v'=1}^{v}\Pi_{v'}, \quad \Gamma(v) = (v + 1)\Phi(v) - \sum_{v'=2}^{v}\Phi(v'). \tag{2.5}$$

In view of the inequality $\Delta E/E_1 \ll 1$, we will neglect terms proportional to ΔE in order to simplify the expressions.

Hence S depends on Π_v near the boundary of the nonequilibrium region v^{**}. The analysis of [6] for Π_v for the case of a steady-state excitation can be used to find the position of this point. According to [6], the space of the quantum number v can be broken up into three characteristic regions: 1) $0 < v < v^*$ where the dominant contribution to $\Phi(v)$ (or Π_v) is "nonresonant" V—V exchange between the lower and upper vibrational levels; 2) $v^* < v < v^{**}$, where the contribution of "resonant" V—V exchange of upper levels becomes significant; 3) $v > v^{**}$ where V—T exchange processes make the dominant contribution in $\Phi(v)$. Therefore the vibrational quantum number v^{**} is determined from the condition that at $v = v^{**}$ the flux of quanta $\Phi(v)$ transferred upward in v in "resonant" V—V exchange processes is equal to the

total rate of loss of quanta due to V—T relaxation processes. Following [6], we have

$$v^{**} = \delta_{V-T}^{-1} \ln \left(2vc_0 \delta_{V-T} P_{1,0}^{-1} \right),$$ (2.6)

where $c_0 = (q/vE_1)^{1/2}$ is a parameter characterizing the distribution f_v on the "plateau":

$$f_v = c_0/v, \quad v^* < v < v^{**};$$ (2.7)

$v = 4\Delta E Q_{1,0}^{0;1}/T\delta_{V-V}^3$ is the effective frequency of V—V exchange; $P_{1,0}$ and $Q_{1,0}^{0;1}$ are the frequencies of V—T and V—V exchange between the ground and first excited levels; δ_{V-T}, δ_{V-V} are parameters dependent on temperature and the type of the molecules participating in the exchange processes. The number v^* is close to the Treanor number $v_T = E_1 T/(2\Delta E T_v) + 1/2$, which determines the minimum of the Treanor distribution

$$f_v^T = f_0 \exp\left\{ -v \left[E_1/T_v - \Delta E (v-1)/T \right] \right\}$$ (2.8)

in the region $0 < v < v_T^*$ [8] [T_v is the vibrational temperature and is related to c_0 by the equation $c_0 = (v^* + 1)f_{v^*}^T$]. The distribution in region 3) is close to the Boltzmann distribution $f_v \sim \exp(-E_V/T)$.

Obviously the relaxation process can be thought of as V—V-exchange-induced "diffusion" of vibrational excitations from region 1), where the molecules are excited, to the boundary of region 2), at which there is rapid de-activation due to V—T exchange. Since the population density of the lower vibrational levels $v < v^*$ is comparatively large, a quasistationary distribution is established quite rapidly in region 1). But in region 2), where the population density of vibrational levels is not large, "diffusion" of excitations is slow and a quasiresonant distribution cannot be established when the external conditions change sufficiently rapidly, i.e., region 2) ultimately determines the total vibrational energy relaxation rate. We will assume that the characteristic time to establish a quasistationary distribution in region 1) is much shorter than the period of the sound wave. The restrictions on the frequency of the sound imposed by this condition are considered below. We use the diffusion approximation in describing the propagation of excitations into region 2. With this approximation we can transform from the discrete form (2.5) to differential analogs of the expressions for Π_v and Φ_v [6]:

$$\Pi_v = -v \frac{d\left(f_v^2 v^2\right)}{dv}, \quad \Phi(v) = -v \left[v \frac{d\left(f_v^2 v^2\right)}{dv} - f_v^2 v^2 \right].$$ (2.9)

In the derivation of (2.9) we have assumed that the function f_v is smooth [this is known to be satisfied on the "plateau" (2.7)] and that thermal excitation of vibrations can be neglected (which will be correct when the gas temperature is low enough). From (2.2) and (2.9) we obtain an equation describing the evolution of the number density of vibrational quanta $\Psi_v = f_v v$ on the "plateau":

$$\partial \Psi_v/\partial t = vv \partial^2 \Psi_v^2/\partial v^2.$$ (2.10)

We consider the boundary conditions to (2.10). For a strongly nonequilibrium distribution the flux Π_v is considered to be the flux of molecules due to resonant V—V exchange by quanta. Therefore, by definition of the number v^{**}, the flux Π_v at the point v^{**} must vanish with the vanishing of vibrational quanta in V—T exchange processes:

$$-vd\Psi_v^2/dv|_{v=v^{**}} = 0.$$ (2.11)

This is the first boundary condition to (2.10). We note that with the help of (2.9) and (2.11), Eq. (2.4) for S reduces to the form

$$S = E_1 v \Psi_{v^{**}}^2.$$ (2.12)

The second boundary condition is found by writing (2.3) in the form

$$\partial\varepsilon/\partial t = \partial\varepsilon_1/\partial t + \partial\varepsilon_2/\partial t = -E_1 v \Psi_{v^{**}}^2 + q$$

[ε_1 and $\varepsilon_2 = \int_{v^*}^{v^{**}} \Psi_v dv$ are the energies stored in regions 1) and 2)]. Using (2.10) in transforming the derivative $\partial\varepsilon_2/\partial t$ we obtain after integration $\Phi(v^*) = -(1/E_1)(q - \partial\varepsilon_1/\partial t)$. Then, using the assumption that the distribution in region 1) is quasistationary, we finally obtain

$$-v \left[v \frac{d\Psi_v^2}{dv} - \Psi_v^2 \right]\Bigg|_{v=v^*} = \frac{q}{E_1}.$$ (2.13)

For stationary conditions we obtain from (2.1) and (2.12), neglecting $\varepsilon_{eq} \ll \varepsilon$

$$\tau_0 = [\varepsilon_1/E_1 + c_0(v^{**} - v^*)]/vc_0^2,$$

where $\varepsilon_1 = E_1[\exp(E/T_v) - 1]^{-1}$ for moderate excitation (see, e.g., [9]), and $\varepsilon_1 \ll E_1 c_0(v^{**} - v^*)$ for strong excitation as considered here. In a gas perturbed by sound waves the time τ is a function of the frequency of the acoustic perturbation [see (2.10)].

3. Sound Propagation in a Gas of Excited Anharmonic Molecules.
We linearize (2.10) through (2.13), assuming that the perturbation of the number density of vibrational quanta $\Psi_v' = f_v v$ varies periodically according to (1.5), like the other gasdynamical parameters n', T', p', and so on. Then (2.10), with the boundary conditions (2.11) and (2.13), can be written in the form

$$\frac{d\Psi_v'}{dv} = -\frac{i\omega}{2v_0 c_0 v}\Psi_v',$$

$$\left(v\frac{d\Psi_v'}{dv} - \Psi_v'\right)\Big|_{v=v_0^*} = \frac{c_0}{2v_0}v', \quad \frac{d\Psi_v'}{dv}\Big|_{v=v_0^{**}} = 0. \tag{3.1}$$

For simplicity we assume that the acoustic oscillations affect the perturbing source only weakly: q' = 0. Then we do not have to specify the method of maintaining the nonequilibrium state in the system. We note only that when the vibrations are excited by electron collisions in a gas discharge the quantity $\partial q/\partial\xi(\xi \in \{n, T, q\})$ is given in [10], while for excitation of a system of anharmonic molecules by laser radiation it is given in [11].

In (3.1) the boundary conditions are imposed at the points v_0^* and v_0^{**} characterizing the unperturbed distribution function f_v. In the linear theory this assumption is valid since the inclusion of the variation of the numbers v^* and v^{**} leads to terms of higher order than the first in the above equations.

The general solution of (3.1) can be expressed in terms of Bessel functions of the first $J_m(x_v)$ and second $N_m(x_v)$ kinds:

$$\Psi_v' = v'\sqrt{vc_0}Z(x_v)/2v_0,$$

$$Z(x_v) = \frac{2}{\sqrt{v_0^*}}\frac{J_1(x_v)N_0^{**} - N_1(x_v)J_0^{**}}{x_v^* (J_0^*N_0^{**} - J_0^{**}N_0^*) - 2(J_1^*N_0^{**} - J_0^{**}N_1^*)}, \tag{3.2}$$

where $x_v = (i\omega\tau_v)^{1/2}$; $\tau_v = 2v/v_0 c_0$ is the effective time of propagation of the perturbation along the v axis [7]; the asterisks above J_m and N_m imply that these functions are evaluated at the points v_0^* and v_0^{**}. The fact that (3.2) is complex indicates a phase shift of the parameters T' and n' with respect to Ψ_v' [5].

With the help of (2.12) and (3.2) we can easily find S' (the perturbation of the relaxation rate S): $S' = q_0\left(\frac{v'}{v_0} + \frac{2}{c_0}\Psi_{v_0^{**}}'\right)$. From the definition of the vibrational relaxation time (2.1), knowing S' and using (1.3), we obtain

$$\tau' = [(1 - i\omega\tau_0)/i\omega]S'/S_0. \tag{3.3}$$

Using (3.2) and (3.3), we can find expressions for the derivatives $\partial\tau/\partial n$ and $\partial\tau/\partial T$, which determine, through (1.7), the dispersion and absorption of ultrasound:

$$\frac{\partial\tau}{\partial\xi} = \frac{1 - i\omega\tau_0}{i\omega}(1 + Z(x_v))\frac{1}{\xi}\frac{\partial\ln v}{\partial\ln\xi}, \quad \xi \in \{n, T\}. \tag{3.4}$$

Substituting (3.4) into (1.6), we have the dispersion relation

$$\frac{k^2 u^2}{\omega^2} = \frac{c + \frac{1}{i\omega}\frac{q_0}{T_0}(1 + Z(x_v))\frac{\partial\ln v}{\partial\ln T}}{c + 1 + \frac{1}{i\omega}\frac{q_0}{T_0}(1 + Z(x_v))\left(\frac{\partial\ln v}{\partial\ln T} - \frac{\partial\ln v}{\partial\ln n}\right)}. \tag{3.5}$$

The domain of applicability of (3.5) is bounded from the low-frequency direction by the applicability of the plane-wave approximation (1.1), and from the high-frequency direction by the assumption that the Treanor distribution function varies in a quasistationary way. Estimating [9] the time t_1 to establish the Treanor distribution as $t_1 = \varepsilon_1/q_0$, the latter condition can be written in the form $\omega t_1 \ll 1$. For the problem considered here, this condition is satisfied more exactly the larger the supply of quanta on the "plateau": $c_0(v^{**} - v^*) \gg \varepsilon_1/E_1$.
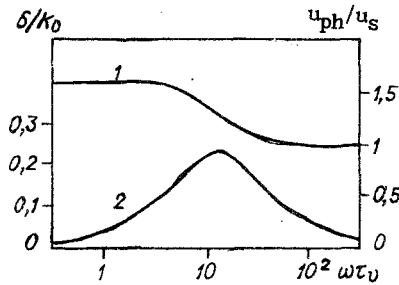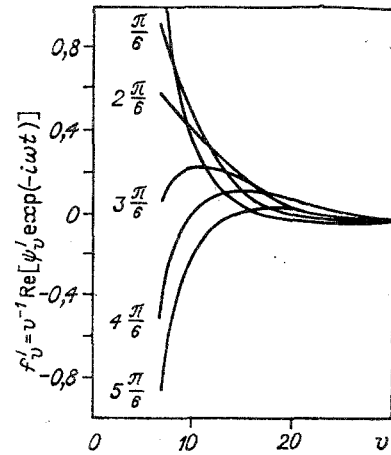
Fig. 1

Fig. 2

In view of the complicated nature of the complex expression $Z(x_v)$ an analysis of (3.5) is possible only in the limiting cases of low ($\omega\tau_v \ll 1$) and high ($\omega\tau_v \gg 1$) frequencies. We note that τ_v is a function of the parameter c_0 (or q_0). The quantity τ_v varies within very wide limits as a function of c_0. For strong vibrational nonequilibrium c_0 typically lies between 10^{-3} and 10^{-2}, and τ_v lies between 10^{-2} and 10^{-3} sec. Hence the frequency region $\omega\tau_v \ll 1$ will correspond to long-wavelength perturbations for which the condition (1.1) may not be applicable. Therefore the low-frequency limit of (3.5) is of interest only from the point of view of studying the asymptotic behavior of the dispersion characteristics.

Using the well-known expansions of the Bessel functions for small values of the argument, we obtain

$$\frac{k^2 u_T^2}{\omega^2} = \frac{c - \frac{\tau_v}{\tau_q}\frac{\partial \ln v}{\partial \ln T}}{c + 1 - \frac{\tau_v}{\tau_q}\left(\frac{\partial \ln v}{\partial \ln T} - \frac{\partial \ln v}{\partial \ln n}\right)}, \tag{3.6}$$

where $\tau_q = T_0/q_0$ and hence we usually have $\tau_v/\tau_q > 1$. The reciprocal of the right-hand side of (3.6) can be considered as an effective adiabatic index γ^*. Using the dependence $\nu_{CO} \sim nT^{-1/2}$ given in [12] for the frequency of V—V exchange between CO molecules in a CO—He mixture, we find that $\gamma^* > \gamma = (c + 1)/c$. This means that the phase velocity $u_{ph} = \omega/k_0$ of low-frequency sound exceeds the isoentropic speed of sound $u_{ph} > u_s = (\gamma T_0/m)^{1/2}$ and there is no absorption in this case ($\operatorname{Im} k = 0$). For comparison we note that the propagation velocity of low-frequency perturbations in a gas relaxing according to the Landau and Teller theory is smaller than u_s [2-4]. This feature ($u_{ph} > u_s$) of the propagation velocity of long-wavelength perturbations must be considered a general property of a strongly excited system of anharmonic molecules. Such a system has a negative vibrational heat capacity (with increasing temperature the rate of V—T processes grows more rapidly ($\sim\exp(-\beta T^{-1/3})$) than the rate of V—V processes ($\sim T^n$), and this leads to a decrease in the population density of the upper energy levels].

Analysis of the high-frequency ($\omega\tau_v \gg 1$) limit of (3.5) shows, as expected, that the vibrational degrees of freedom do not participate in the periodic change of state of the gas. They are "frozen out" and do not affect the adiabatic relation between the change of pressure and change of density. Hence $u_{ph} = u_s$ and there is no absorption (or amplification) of sound.

In the intermediate frequency region there is a gradual variation of the speed of sound from the value $(\gamma^* T_0/m)^{1/2}$ to u_s, as is shown in Fig. 1 (curve 1) and amplification of sound is possible in this region (curve 2). The dispersion u_{ph}/u_s and amplification δ/k_0 parameters are shown as functions of $\omega\tau_v$ for a CO—He mixture with $n_{CO} = 10^{16}$ cm^{-3}, $n_{He} = 10^{18}$ cm^{-3}, T = 175 K, and q = 0.75 W·cm^{-3}.

Returning to the solution (3.2) of (3.1), we note that the perturbation of the distribution function $f_v' = v^{-1} \operatorname{Re} \Psi_v'$ is a function of the frequency ω and the quantum number v and varies nonmonotonically for times after a half-period of the sound wave (Fig. 2, $\omega t = 0$ to π, $\omega\tau_v = 10$). The formation of such structures on the distribution f_v would be of interest in the study of emission and absorption spectra of anharmonic dipole molecules under nonequilibrium conditions. It is also of importance in the physics of intense CO lasers, where

the effects of generation are due to the existence of a "plateau" in the distribution function. Because of the build-up of acoustic vibrations there should be lines corresponding to the Q and R branches in the spectra of such lasers, whereas generation occurs only in the P branch when the system is unperturbed by sound [13].

Finally we note that these effects can already be tested experimentally with the help of the methods of ultrasonic acoustics [14] or the techniques of measuring the coefficient of amplification of weak signals with the help of an IR-laser [15].

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