

# Features of Nonlinear Sound Propagation in Vibrationally Excited Gases

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Weakly nonlinear sound propagation in a gas where molecular vibrational relaxation takes place is studied. New equations which govern the sound in media where the irreversible relaxation may take place are derived and discussed. Their form depends on the regime of excitation of oscillatory degrees of freedom, equilibrium (reversible) or non-equilibrium (irreversible), and on the comparative frequency of the sound in relation to the inverse time of relaxation. Additional nonlinear terms increase standard nonlinearity of the high-frequency sound in the equilibrium regime of vibrational excitation and decrease otherwise. As for the nonlinearity of the low-frequency sound, the conclusions are opposite. Appearance of a non-oscillating additional part which is a linear function of the distance from the transducer is an unusual property of nonlinear distortions of harmonic at the transducer high-frequency sound.

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## 1. Introduction. Basic equations and starting points

The non-equilibrium physics was born and started to rapidly develop in the sixties of the XX-th century in connection with the need of a deeper study of unusual hydrodynamics of media where irreversible processes take place (ZELDOVICH, RAIZER, 1966; GORDITS *et al.*, 1973; OSIPOV, UVAROV, 1992). The most important of them are gases with excited degrees of oscillatory freedom of molecules. Non-equilibrium processes are established as well in discharge plasma, the rarified levels of the upper atmosphere, interstellar media, and so on. There was reported relaxation of rotational, translational, and electronic degrees of freedom of a molecule. Difference in relaxation times follows from difference of probabilities of various elementary events. Chemically reacting media are also relaxing; the duration of reaction is the characteristic time of relaxation. If a chemical reaction is irreversible, the sound propagates over the reacting medium unusually. The irreversible relaxation results in anomalous dispersion and absorption of ultrasonics waves in such media

(ZELDOVICH, RAIZER, 1966; OSIPOV, UVAROV, 1992; KOGAN, MOLEVICH, 1986; MOLEVICH *et al.*, 2005). Interest in the non-equilibrium phenomena in the physics of gases was originally connected with the study of these anomalies.

This paper is devoted to the nonlinear features of sound propagation in the low-frequency (when the characteristic frequency of the sound,  $\omega$ , is much smaller than the inverse time of relaxation,  $1/\tau$ ) and high-frequency regimes in the vibrationally relaxing gas. It is well-known, that dispersion, due to reversible relaxation, leads to an increase in the phase speed with enlargement in the sound frequency. Vice versa, in the non-equilibrium regime, the phase speed decreases. The nonlinear features of sound propagation depend on the frequency of the sound and on the type of vibrational excitation, equilibrium or not. The nonlinear distortion is not longer determined by the standard parameter of nonlinearity; and the type of distortion differs from the standard one.

We consider a gas whose steady state is maintained by pumping energy into the vibrational degrees of freedom by power  $I$  and heat withdrawal from the trans-

lational degrees of freedom of power  $Q$ , while both  $I$  and  $Q$  refer to the unit mass (Sec. 2). The relaxation equation for the vibrational energy per unit mass complements the system of conservation equations in the differential form. It takes the form:

$$\frac{d\varepsilon}{dt} = -\frac{\varepsilon - \varepsilon_{eq}(T)}{\tau(\rho, T)} + I. \quad (1)$$

The equilibrium value for the vibrational energy at the given temperature  $T$  is denoted by  $\varepsilon_{eq}(T)$ , and  $\tau(\rho, T)$  is the vibrational relaxation time. The mass, momentum, and energy conservation equations governing thermoviscous flow in a vibrationally relaxing gas read (OSIPOV, UVAROV, 1992):

$$\begin{aligned} \frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{v}) &= 0, \\ \rho \left[ \frac{\partial \mathbf{v}}{\partial t} + (\mathbf{v} \cdot \nabla) \mathbf{v} \right] &= -\nabla p, \end{aligned} \quad (2)$$

$$\rho \left[ \frac{\partial(e+\varepsilon)}{\partial t} + (\mathbf{v} \cdot \nabla)(e+\varepsilon) \right] + p(\nabla \cdot \mathbf{v}) = \rho(I - Q),$$

where  $\mathbf{v}$  denotes the velocity of fluid,  $\rho$ ,  $p$  are the density and pressure,  $e$  marks the internal energy per unit mass of translation motion of molecules,  $x_i$  ( $i = 1, 2, 3$ ) are space coordinates. The system (2) may be complemented by the terms which account for viscosity and thermal conductivity but they are insignificant in the studies of nonlinear distortions of the sound. We will consider only effects relating to the oscillatory relaxation. Two thermodynamic functions  $e(p, \rho)$ ,  $T(p, \rho)$  complete the system (2). Thermodynamics of ideal gases provides the equalities:

$$e(p, \rho) = \frac{R}{\mu(\gamma - 1)}, \quad T(p, \rho) = \frac{p}{(\gamma - 1)\rho}, \quad (3)$$

where  $\gamma = C_{P,\infty}/C_{V,\infty}$  is the isentropic exponent without account for vibrational degrees of freedom ( $C_{P,\infty}$  and  $C_{V,\infty}$  denote “frozen” heat capacities correspondent to very quick processes),  $R$  is the universal gas constant, and  $\mu$  is the molar mass of a gas.

## 2. Fundamentals of modes' designation and derivation of dynamic equations

Let start by considering a motion of a gas with an infinitely small magnitude in the case  $I = Q$ . Every quantity  $q$  is represented as a sum of unperturbed value  $q_0$  (in absence of the background flows,  $v_0 = 0$ ) and its variation  $q'$ . The flow is supposed to be one-dimensional along axis  $Ox$ . Following Molevich and Makaryan (MOLEVICH, 2003; 2004; MAKARYAN, MOLEVICH, 2007), we consider weak transversal pumping which may alter the background quantities in the transversal direction of axis  $Ox$ . It is assumed that the

background stationary quantities are constant along axis  $Ox$ .

The system of conservation equations including the quadratic nonlinear terms, which are of major importance in the nonlinear acoustics, with the account of Eqs. (3), takes the form (PERELOMOVA, 2012):

$$\begin{aligned} \frac{\partial v'}{\partial t} + \frac{1}{\rho_0} \frac{\partial p'}{\partial x} &= -v' \frac{\partial v'}{\partial x} + \frac{\rho'}{\rho_0^2} \frac{\partial p'}{\partial x}, \\ \frac{\partial p'}{\partial t} + \gamma p_0 \frac{\partial v'}{\partial x} - (\gamma - 1) \rho_0 \frac{\varepsilon'}{\tau} &+ (\gamma - 1) \rho_0 T_0 \Phi_1 \left( \frac{p'}{p_0} - \frac{\rho'}{\rho_0} \right) = -v' \frac{\partial p'}{\partial x} \\ &- \gamma p' \frac{\partial v'}{\partial x} + (\gamma - 1) \rho' \left( \frac{\varepsilon'}{\tau} - T_0 \Phi_1 \left( \frac{p'}{p_0} - \frac{\rho'}{\rho_0} \right) \right) \\ &- (\gamma - 1) \rho_0 \left( T_0 \left( \frac{1}{\tau^2} \frac{d\tau}{dT} \right)_0 \varepsilon' \left( \frac{p'}{p_0} - \frac{\rho'}{\rho_0} \right) \right. \\ &\left. + T_0 \Phi_1 \left( \frac{\rho'^2}{\rho_0^2} - \frac{p' \rho'}{p_0 \rho_0} \right) + T_0 \Phi_2 \left( \frac{p'}{p_0} - \frac{\rho'}{\rho_0} \right)^2 \right), \end{aligned} \quad (4)$$

$$\frac{\partial \rho'}{\partial t} + \rho_0 \frac{\partial v'}{\partial x} = -v' \frac{\partial \rho'}{\partial x} - \rho' \frac{\partial v'}{\partial x},$$

$$\begin{aligned} \frac{\partial \varepsilon'}{\partial t} + \frac{\varepsilon'}{\tau} - T_0 \Phi_1 \left( \frac{p'}{p_0} - \frac{\rho'}{\rho_0} \right) &= T_0 \left( \frac{1}{\tau^2} \frac{d\tau}{dT} \right)_0 \varepsilon' \left( \frac{p'}{p_0} - \frac{\rho'}{\rho_0} \right) \\ &+ T_0 \Phi_1 \left( \frac{\rho'^2}{\rho_0^2} - \frac{p' \rho'}{p_0 \rho_0} \right) \\ &+ T_0 \Phi_2 \left( \frac{p'}{p_0} - \frac{\rho'}{\rho_0} \right)^2 - v' \frac{\partial \varepsilon'}{\partial x}, \end{aligned}$$

where

$$\begin{aligned} \Phi_1 &= \left( \frac{C_{V,eq}}{\tau} + \frac{\varepsilon - \varepsilon_{eq}}{\tau^2} \frac{d\tau}{dT} \right)_0, \\ C_{V,eq} &= \left( \frac{d\varepsilon_{eq}}{dT} \right)_0, \\ \Phi_2 &= T_0 \left( -\frac{1}{\tau^2} C_{V,eq} \frac{d\tau}{dT} - \frac{(\varepsilon_0 - \varepsilon_{eq})}{\tau^3} \left( \frac{d\tau}{dT} \right)^2 \right. \\ &\left. + \frac{1}{2\tau} \frac{dC_{V,eq}}{dT} + \frac{(\varepsilon_0 - \varepsilon_{eq})}{2\tau^2} \frac{d^2\tau}{dT^2} \right)_0. \end{aligned} \quad (5)$$

The relaxation time in the most important cases may be thought as a function of temperature accordingly to Landau and Teller,  $\tau(T) = A \exp(BT^{-1/3})$ , where  $A$  and  $B$  are some positive constants. That gives negative values of  $d\tau/dT$  (ZELDOVICH, RAIZER, 1966; GORDIETS *et al.*, 1973; OSIPOV, UVAROV, 1992) which may

correspond to a negative value of  $\Phi_1$ . The dispersion equation follows from the linear version of Eqs. (4):

$$\omega \left( i\Phi_1(\gamma - 1)T_0\tau(c_\infty^2 k^2 - \gamma\omega^2) + c_\infty^2(c_\infty^2 k^2 - \omega^2)(i - \omega\tau) \right) = 0, \quad (6)$$

where

$$c_\infty = \sqrt{\frac{\gamma RT_0}{\mu}} = \sqrt{\gamma \frac{p_0}{\rho_0}}$$

is the “frozen”, infinitely small-signal sound speed in the ideal uniform gas.

The approximate roots of dispersion equation for both acoustic branches, progressive in the positive and negative directions of axis  $Ox$ , are well known under the simplifying condition  $\omega\tau \gg 1$ , which restricts consideration by the high-frequency sound (OSIPOV, UVAROV, 1992; MOLEVICH, 2003)

$$\omega_{1,\infty} = c_\infty k + \frac{i(\gamma - 1)^2 T_0}{2c_\infty^2} \Phi_1, \quad (7)$$

$$\omega_{2,\infty} = -c_\infty k + \frac{i(\gamma - 1)^2 T_0}{2c_\infty^2} \Phi_1.$$

The last term in both dispersion relations manifests amplification of the sound in the non-equilibrium regime (if  $\Phi_1 < 0$ ) which does not depend on the wave number  $k$ . The approximate roots of dispersion equation for both acoustic branches, progressive in the positive or negative directions of axis  $Ox$  in the low-frequency domain  $\omega\tau \ll 1$ , are as follows (see also (MOLEVICH, 2003; 2004)):

$$\omega_{1,0} = c_0 k, \quad \omega_{2,0} = -c_0 k, \quad (8)$$

where

$$c_0 = c_\infty - \frac{(\gamma - 1)^2 T_0 \tau}{2c_\infty} \Phi_1.$$

The two last roots of the dispersive equation, estimated without the limitation  $\omega\tau \gg 1$  or,  $\omega\tau \ll 1$ , sound:

$$\omega_3 = i \left( \frac{1}{\tau} + \frac{(\gamma - 1)(\gamma + c_\infty^2 k^2 \tau^2) T_0}{c_\infty^2 (1 + c_\infty^2 k^2 \tau^2)} \Phi_1 \right), \quad (9)$$

$$\omega_4 = 0.$$

These last two roots manifest slow varying and stationary, non-wave motions of a gas. Accordingly, perturbation in the velocity, pressure or energy of every dynamic variable may be expressed in terms of specific excess densities. The overall excess velocity, pressure, density, and internal energy are sums of specific parts:

$$\begin{aligned} v'(x, t) &= \sum_{n=1}^4 v'_n(x, t), & p'(x, t) &= \sum_{n=1}^4 p'_n(x, t), \\ \rho'(x, t) &= \sum_{n=1}^4 \rho'_n(x, t), & \varepsilon'(x, t) &= \sum_{n=1}^4 \varepsilon'_n(x, t). \end{aligned} \quad (10)$$

Relations of acoustic rightwards propagating wave in the high-frequency regime ( $\omega_1 k \gg 1$  or, alternatively,  $c_\infty k \tau \gg 1$ ) follow from the dispersion relation  $\omega_1(k)$  (PERELOMOVA, 2012):

$$\begin{aligned} v'_{1,h}(x, t) &= \frac{c_\infty}{\rho_0} \left( 1 - B \int dx \right) \rho'_{1,h}(x, t), \\ p'_{1,h}(x, t) &= c_\infty^2 \left( 1 - 2B \int dx \right) \rho'_{1,h}(x, t), \end{aligned} \quad (11)$$

$$\varepsilon'_{1,h}(x, t) = \frac{2Bc_\infty^2}{(\gamma - 1)\rho_0} \int dx \rho'_{1,h}(x, t),$$

where

$$B = -\frac{(\gamma - 1)^2 T_0}{2c_\infty^3} \Phi_1. \quad (12)$$

The sound is imposed to be a wave process, so that it attenuates (or amplifies, in dependence on the sign of  $B$ ) weakly over the wavelength,  $|B|k^{-1} \ll 1$ . In the low-frequency regime, the relations take the leading-order form (PERELOMOVA, 2010a):

$$\begin{aligned} v'_{1,l}(x, t) &= \frac{c_0}{\rho_0} \rho'_{1,l}(x, t), \\ p'_{1,l}(x, t) &= c_0^2 \rho'_{1,l}(x, t), \end{aligned} \quad (13)$$

$$\varepsilon'_{1,l}(x, t) = \frac{2Bc_0^2}{(\gamma - 1)\rho_0} \int dx \rho'_{1,l}(x, t).$$

Relations (11), (13), along with the linear property of superposition, Eqs. (10), point out a way of combination of four equations from (4) in order to get dynamic equations describing perturbation of only one specific mode. Formally, that may be done by means of the complete set of orthogonal projectors. The remarkable property of projectors is to decompose the dynamic equations governing the correspondent mode by immediate appliance on the linear system. The details of establishing of the projectors in the low- and high-frequency regimes may be found in the papers by one of the authors (PERELOMOVA, 2003; 2006; 2008). Application of the matrix operator on the vector of overall perturbations ( $v'$   $p'$   $\rho'$   $\varepsilon'$ ) actually decomposes the correspondent specific mode:

$$P_n \begin{pmatrix} v'(x, t) \\ p'(x, t) \\ \rho'(x, t) \\ \varepsilon'(x, t) \end{pmatrix} = \begin{pmatrix} v'_n(x, t) \\ p'_n(x, t) \\ \rho'_n(x, t) \\ \varepsilon'_n(x, t) \end{pmatrix}, \quad n = 1, \dots, 4. \quad (14)$$

Within the accuracy up to the terms of the first order in  $(c_\infty k \tau)^{-1}$ ,  $|B|k^{-1}$ , the third row of projector  $P_{1,h}$ , which projects the overall vector of perturbations into an excess density belonging to the first high-frequency acoustic mode, takes the following form:

$$\left( \frac{\rho_0}{2c_\infty} + \frac{B\rho_0}{c_\infty} \int dx \quad \frac{1}{2c_\infty^2} + \frac{B(\gamma-3)}{2(\gamma-1)c_\infty^2} \int dx \quad - \frac{c_\infty(\gamma+1)}{2\rho_0} \rho_{1,h} \frac{\partial \rho_{1,h}}{\partial x} \right. \\ \left. \frac{B}{(\gamma-1)} \int dx \quad \frac{(\gamma-1)\rho_0}{2c_\infty^3 \tau} \int dx \right). \quad (15)$$

The row which projects the overall vector of perturbations into the first branch of the low-frequency sound, calculated with the accuracy up to the terms of the first order in  $(c_0 k \tau)^1$ ,  $|B|k^{-1}$ , is

$$\left( \frac{\rho_0}{2c_0} - \frac{B\rho_0}{c_0} \int dx \quad \frac{1}{2c_0^2} + \frac{B\tau}{(\gamma-1)c_0} \right. \\ \left. - \frac{Bc_0\tau}{(\gamma-1)} \quad \frac{(\gamma-1)\rho_0}{2c_0^2} - \frac{B(\gamma-2)\rho_0\tau}{c_0} \right). \quad (16)$$

Both rows include operators. Employment of the first row on the linearized system (4), i.e., application of the first operator on the first equation from this set, the second operator on the second one, and so on, and calculation of the sum of all four equalities, result in the dynamic equation for an excess density of the first acoustic high-frequency mode:

$$\frac{\partial \rho_{1,h}}{\partial t} + c_\infty \frac{\partial \rho_{1,h}}{\partial x} - c_\infty B \rho_{1,h} = 0, \quad (17)$$

where  $\rho_{1,h}$  is the excess acoustic density,  $\rho'_1$ , in the case of the high-frequency sound. Application of the second row on the system (4) yields the dynamic equation for the first low-frequency acoustic mode:

$$\frac{\partial \rho_{1,l}}{\partial t} + c_0 \frac{\partial \rho_{1,l}}{\partial x} = 0, \quad (18)$$

where  $\rho_{1,l}$  is the excess acoustic density in the case of the low-frequency sound. Equations (17), (18) obviously coincide with the roots of the dispersion relation  $\omega_{1,0}$  and  $\omega_{1,\infty}$  from Eqs. (7), (8). It may be readily established that terms relating to all other modes become reduced. That follows from the properties of projectors.

Applying projectors on the nonlinear vector of system (4) yields quadratic nonlinear corrections in the final dynamic equations originating from the right-hand side of Eqs. (4). We will keep among all of them only those belonging to the progressive in the positive direction of axis  $Ox$  sound. That is valid over spatial and temporal domains, where magnitude of this branch of sound is much larger than that of other modes.

### 3. Nonlinear features of sound propagation

#### 3.1. The high-frequency sound

Application of the row (15) at the column of nonlinear equations (4) and account for the links (11) results, after some ordering, in the equation governing the first sound branch. The term proportional to  $B^0$  in the right-hand side of the equation takes the form:

The term associated with the corrections of order  $B^1$  in the operators, is:

$$- \frac{Bc_\infty(\gamma-1)}{2\rho_0} \int \rho_{1,h} \frac{\partial \rho_{1,h}}{\partial x} dx = - \frac{Bc_\infty(\gamma-1)}{4\rho_0} \rho_{1,h}^2, \quad (20)$$

and the terms originated from the links between acoustic perturbations (15), are:

$$\frac{Bc_\infty(\gamma+1)}{2\rho_0} \left( \rho_{1,h}^2 + 2 \frac{\partial \rho_{1,h}}{\partial x} \int \rho_{1,h} dx \right) \\ - \frac{T_0 \Phi_2 (\gamma-1)^3}{\rho_0} \rho_{1,h}^2. \quad (21)$$

The overall weakly nonlinear equation governing sound contains the sum of all nonlinear terms:

$$\frac{\partial \rho_{1,h}}{\partial t} + c_\infty \frac{\partial \rho_{1,h}}{\partial x} + \frac{c_\infty(\gamma+1)}{2\rho_0} \rho_{1,h} \frac{\partial \rho_{1,h}}{\partial x} - c_\infty B \rho_{1,h} \\ = \frac{Bc_\infty}{2\rho_0} \left( \frac{\gamma+3}{2} \rho_{1,h}^2 + 2(\gamma+1) \frac{\partial \rho_{1,h}}{\partial x} \int \rho_{1,h} dx \right) \\ - \frac{T_0 \Phi_2 (\gamma-1)^3}{\rho_0} \rho_{1,h}^2. \quad (22)$$

The limiting case of Eq. (22) when  $B = 0$  and  $\Phi_2 = 0$  is the famous Earnshaw equation for a simple wave in non-viscous ideal gas (RUDENKO, SOLUYAN, 1977). The analysis of Eq. (22) may be readily proceeded by means of the standard method of successive approximations. In order to establish pure nonlinear distortions, we put the linear term proportional to  $B$  in the left-hand side of Eq. (22) equal to zero. If a transducer placed at  $x = 0$  transmits a harmonic wave,

$$\rho_{1,h}(x = 0, t) = R_A \sin(\omega t), \quad (23)$$

the spectrum is enriched at some distances from the transducer due to nonlinearity of the medium. Simple evaluations yield the approximate solution:

$$\rho_{1,h}(x, t) = R_A \sin(\omega t - kx) \\ + \frac{x(\gamma+1)\omega R_A^2}{4c_\infty \rho_0} \sin(2\omega t - 2kx) \\ + \frac{x R_A^2 (\alpha + \beta)}{2c_\infty \rho_0} \cos(2\omega t - 2kx) \\ + \frac{x R_A^2 (-\alpha + \beta)}{2c_\infty \rho_0}, \quad (24)$$

where

$$\alpha = - \frac{Bc_\infty(\gamma+3)}{4} + T_0 \Phi_2 (\gamma-1)^3, \quad (25) \\ \beta = -Bc_\infty(\gamma+1).$$

The second term in the right-hand side of Eq. (24) reflects the “standard” weakly nonlinear distortions. The third one originates from the vibrational relaxation, it

also varies with time. Assuming that the sign of  $B$  is more important than that of  $\Phi_2$ , one may conclude that the magnitude of nonlinear distortions relating to this oscillating term decrease with  $x$  for the positive  $B$  and increase otherwise. The last monotonic term, analogously, decreases when  $B$  is positive. It causes the non-zero mean value of perturbations in the periodic sound wave. Thus, the conclusion is that the non-equilibrium gases possess unusual nonlinearity which gets smaller with the increase in the degree of disequilibrium. Equilibrium media, *vice versa*, makes the nonlinearity larger. For any periodic in time acoustic wave, the averaged over the sound period term in the right-hand side of Eq. (22) takes the leading-order form:

$$\frac{-\alpha + \beta}{\rho_0} \langle \rho_{1,h}^2 \rangle. \quad (26)$$

It differs from the “standard” nonlinearity,  $\frac{c_\infty(\gamma+1)}{2\rho_0} \rho_{1,h} \frac{\partial \rho_{1,h}}{\partial x}$ , which is zero on average in the leading order. As for the momentum of the acoustic impulse, it is known that it is constant independent on the distance from a transducer before or after formation of a discontinuity in a simple wave. Integrating Eq. (22) from  $x = -\infty$  till  $x = \infty$  and assuming that excess acoustic density and all its spatial derivatives tend to zero at infinities, we finally arrive at the dynamic equation describing the momentum,

$$\frac{\partial P_{1,h}}{\partial t} = c_\infty B P_{1,h} + \frac{(-\alpha + \beta)c_\infty}{\rho_0} \int_{-\infty}^{\infty} \rho_{1,h}^2 dx, \quad (27)$$

where

$$P_{1,h} = c_\infty \int_{-\infty}^{\infty} \rho_{1,h} dx \quad (28)$$

is the density of the acoustic momentum. If we would not consider nonlinearity associated with excitation of vibrational degrees of a molecule freedom, the last term in the right-hand side of Eq. (27) were zero and the acoustic momentum would vary with time proportionally to  $\exp(c_\infty B t)$ . It would increase for a positive  $B$ , in the non-equilibrium case, and decrease otherwise. Dependence of the acoustic momentum on time is connected with exchange of the momentum of internal and external degrees of molecules of a relaxing gas. Account for nonlinearity makes variations of  $P$  with time faster in the equilibrium regime, in view of the fact that the integral in the right-hand side of Eq. (27) is always positive.

If the vibrational relaxation is equilibrium, the discontinuity in the sound wave may not form at all. That happens for enough large  $|B|$ . Without account for nonlinearity connected with vibrational relaxation, for initially sinusoidal wave, the front forms if (PERELOMOVA, WOJDA, 2011)

$$\frac{2\rho_0 c_\infty B \pi}{R_A(\gamma+1)\omega} < -1. \quad (29)$$

One may expect that the threshold of the discontinuity appearance is even lower in view of additional nonlinearity. Vice versa, in the non-equilibrium regime, for the positive  $B$ , discontinuity always forms. Account for the specific nonlinearity originating from relaxation predicts larger distances from a transducer where that happens.

### 3.2. The low-frequency sound

As for the low-frequency sound, taking into account relations (13) along with the application of row (16) on the nonlinear right-hand side of Eqs. (4) yields:

$$\begin{aligned} \frac{\partial \rho_{1,l}}{\partial t} + c_0 \frac{\partial \rho_{1,l}}{\partial x} + \frac{c_0(\gamma+1)}{2\rho_0} \rho_{1,l} \frac{\partial \rho_{1,l}}{\partial x} \\ = \frac{B}{\rho_0 \tau} \rho_{1,l} \int \rho_{1,l} dx. \end{aligned} \quad (30)$$

For an excess acoustic density at a transducer being a harmonic function of time, the approximate solution at some distances from the transducer takes the form:

$$\begin{aligned} \rho_{1,l}(x, t) = R_A \sin(\omega t - kx) \\ + \frac{x(\gamma+1)\omega R_A^2}{4c_0 \rho_0} \sin(2\omega t - 2kx) \\ + \frac{x B R_A^2}{2\omega \tau \rho_0} \sin(2\omega t - 2kx). \end{aligned} \quad (31)$$

Also, in this case, there is a part oscillating at a double frequency which associates with the standard nonlinearity and one which reflects the effects of relaxation. In the non-equilibrium regime, if  $B > 0$ , the nonlinearity enhances, and it decreases otherwise. Thus, conclusions are opposite to those of the high-frequency sound. Also, perturbations in the sound remain zero on average. For any periodic in time acoustic wave, the averaged over the sound period term in the right-hand side of Eq. (30) takes the leading-order form:

$$\frac{B}{2\rho_0 \tau} \left\langle \left( \int \rho_{1,l} dx \right)^2 \right\rangle, \quad (32)$$

and the conclusion about enhancement of nonlinearity in the non-equilibrium regime of excitation is valid. This type of non-linearity differs both from that in the simple wave and that in the high-frequency case described by Eq. (26).

As for the acoustic momentum, it varies with time in accordance to the following dynamic equation:

$$\frac{\partial P_{1,l}}{\partial t} = \frac{B c_\infty}{2\rho_0 \tau} \left( \int \rho_{1,l} dx \right)^2. \quad (33)$$

It enlarges in the non-equilibrium regime and decreases otherwise.

The unusual feature is that in contrast to the high-frequency sound where the ratio of nonlinear terms associated with the vibrational relaxation and the standard one is of the order  $|B|k^{-1}$  is much less than unity, the ratio of the similar terms in the low-frequency regime in the evolution equation (30) is of the order of the ratio of two small parameters,  $|B|k^{-1}$  and  $\omega\tau$ . It is not rigorously small and may make the nonlinear term originating from the vibrational relaxation larger than the standard one.

#### 4. Concluding remarks

In this study we consider the nonlinear distortions of the sound associated with the equilibrium or non-equilibrium type of relaxation. The linear effect of relaxation is in increase (or unusual decrease, if  $B > 0$ ) in the phase speed of a signal in equilibrium relaxation, while its frequency enlarges. Accordingly, sound enhances in the non-equilibrium regime. As for nonlinear distortions, they depend also on the domain of the sound frequency emitted by a transducer and on the type of relaxation. In addition to standard non-linearity, there appear terms which enlarge nonlinearity or make it smaller. The main conclusion is that nonlinear distortion of the high-frequency sound decreases and that of the low-frequency regime increases, while sound propagates over a gas in vibrational non-equilibrium. This may be of especial importance in the low-frequency domain because the relative magnitude of the term associated with relaxation is proportional to the ratio of small parameters,  $|B|k^{-1}$  and  $\omega\tau$ . Since the low-frequency sound almost does not attenuate linearly, this may lead to unusual short (if  $B > 0$ ) or large (if  $B < 0$ ) distances from a transducer where the sawtooth wave forms. In the equilibrium gases, vice versa, nonlinear distortion of the high-frequency sound enhances and nonlinear distortion of the low-frequency sound declines.

Gases, where vibrational relaxation of the internal degrees of molecules takes place, are just one example among fluids with different thermodynamic relaxation processes which may occur in an irreversible way. In spite of that, flows over these relaxing fluids are described quantitatively by different parameters, the equations governing sound and relative nonlinear phenomena, are quite similar (MOLEVICH, 1986). The nonlinear effects caused by the sound in a chemically reacting gas are discussed in the papers by one of the authors (PERELOMOVA, 2010b). Thus, the conclusions of this study may be expanded over a wide class of fluids with thermodynamical relaxation processes of different kinds. Theoretical predictions hopefully will allow to conclude about qualitative and quantitative relaxation processes in a gas remotely, basing on data on nonlinear distortions of the sound and on variations in its momentum during propagation over a gas.

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