

# Excitation of non-wave modes by sound of arbitrary frequency in a chemically reacting gas

Anna Perelomova  
Gdansk University of Technology,  
Faculty of Applied Physics and Mathematics,  
11/12 Gabriela Narutowicza Street, 80-233 Gdansk, Poland,  
anpe@mif.pg.gda.pl

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## Abstract

The nonlinear phenomena in the field of high intensity sound propagating in a gas with a chemical reaction, are considered. An exotheric chemical reaction of  $A \rightarrow B$  type is followed by dispersion and attenuation of sound which may be atypical during irreversible thermodynamic processes under some conditions. The first and second order derivatives of heat produced in the chemical reaction evaluated at the equilibrium temperature, density and mass fraction of reagent  $A$ , are taken into account. The instantaneous equations are derived which govern dynamics of perturbations in non-acoustic modes, and conclusions of the efficiency of their nonlinear excitation by sound are drawn out. The advantage of this study is in accurate description of dispersion. Acoustic perturbations of any characteristic duration as compared to the duration of chemical reaction are considered, along with periodic, aperiodic perturbations and impulses. The conclusions concern also acoustically active gases.

*Keywords:* Nonlinear acoustics, acoustic activity, acoustic dispersion, chemical reaction, acoustic heating.

## 1 Introduction

The nonlinear losses in acoustic momentum and energy in the thermoviscous flow are the reason for excitation of non-wave modes of a fluid motion. They are known as acoustic heating and streaming relating to the thermal (or, optionally, the entropy) and the vorticity modes. The latter one may exist exclusively in multi-dimensional flows. Regarding to periodic sound which propagates in a Newtonian fluid, the theory of acoustic streaming and heating is well developed. It describes excitation of the non-wave modes in good agreement with experimental data [1, 2, 3]. As for the flows of the non-Newtonian fluids including these with dispersive properties, the nonlinear distortion of sound and its nonlinear effects are still fairly poorly

studied analytically. Thermodynamic relaxation of internal degrees of molecules freedom is one of the reasons for dispersion. Analytical results concerning nonlinear distortions of sound itself in a relaxing medium may be found in Refs [4, 5, 6, 7, 8]. The study [9] incorporates analytical methods and computation in description of nonlinear propagation of sound in bounded volumes of a relaxing medium. The fluids where thermodynamic relaxation occurs, are paid to great attention in connection to their medical and technical applications. Absorption in complex molecular liquids is often connected with excitation of a number of internal molecular degrees of freedom. In gases, the molecules may translate, vibrate and rotate. Molecules in strongly heated gases may ionize. Relaxation times in molecular liquids and biological fluids and tissues are usually densely disturbed [10].

When non-equilibrium effects are considered, the conventional conservation equations should be supplemented by the terms and equations which are responsible for relaxation. Besides acoustic, entropy and vorticity modes, there appear once more non-wave mode (or a number of modes) which reflects relaxation of some thermodynamic parameter (parameters). That concerns flows in chemically reacting fluids and in gases with excited degrees of oscillatory freedom of molecules [11, 12]. The important issue is interaction of different modes which are nonlinearly coupling. Among other problems, we may list controlling of mass concentration of reagents, oscillatory energy of gas molecules, variations in the background temperature and in the velocity of bulk flows by means of sound. The related issue is the self-action of a sound beam with account of thermal lens and the main stream which are induced by it [13]. Processes in open systems often are followed by amplification instead of attenuation of sound. That concerns magnetoacoustic waves that have much in common with sound over gases with thermodynamic relaxation and are described by the similar equations [14]. Excitation of the secondary non-wave modes also occurs unusually in acoustically active plasma [15]. Attention to unusual nonlinear phenomena of sound in chemically reacting gases has been probably first attracted by Molevich [16]. In this study, we consider excitation of the entropy and relaxation modes by sound with precise account of dispersive properties of a flow of a chemically reacting gas. We incorporate analytical methods and numerical simulations in evaluations of nonlinear effects of some kinds of impulsive sound.

The main idea of the analytical method is to determine relations of specific perturbations in a flow of infinitely-small magnitudes of disturbances and to use them in studies of weakly nonlinear flow. That makes possible to derive a set of coupling equations which consider weak nonlinear interaction of all modes. No restrictions concerning the type of sound (periodic or not) are imposed. The links specifying every mode and relative projectors are valid for any spectrum of perturbations. They are exactly evaluated. The method has been successfully applied by the author in studies of acoustic heating and streaming in the standard absorbing fluids and some flows of non-Newtonian fluids [17, 18, 19]. Two limiting cases of high or low-frequency sound as compared with the inverse time of a chemical reaction, have been discussed by the author in Refs [20, 21]. We do not consider the effects connected with boundaries restricting by unbounded volumes of a gas in this study. Boundary conditions in close volumes determine the discrete spectrum of perturbations, and the equilibrium non-uniformity of the background caused by external forces or energy release or/and inclusion of spacial inhomogeneities introduce dispersion which may be of importance and essentially affects the definition of modes in a flow and the subsequent analysis [22, 23].

## 2 Dispersion relations in one-dimensional flow

The one-dimensional gas flow along axis  $OX$  is considered. The momentum, energy, and continuity equations in a gas where an exotheric chemical reaction of  $A \rightarrow B$  type occurs, take the form [12, 16]:

$$\begin{aligned} \rho \frac{d\vec{v}}{dt} &= -\vec{\nabla}P, \\ \frac{C_{V,\infty}}{C_{P,\infty} - C_{V,\infty}} \frac{dT}{dt} - \frac{T}{\rho} \frac{d\rho}{dt} &= Q, \\ \frac{d\rho}{dt} + \rho \vec{\nabla} \cdot \vec{v} &= 0, \end{aligned} \quad (1)$$

where  $\vec{v}$ ,  $\rho$ ,  $P$  denote velocity, density and pressure of a gas, respectively. In the system of equations (1),  $T$  is temperature (it is measured in Joules per molecule),  $C_{V,\infty}$  and  $C_{P,\infty}$  are the frozen heat capacities at constant volume and constant pressure, respectively,  $Q = HmW/\rho$  denotes the heat produced in the medium per one molecule due to a chemical reaction with  $H$  being the reaction enthalpy per unit mass of reagent  $A$ , and  $m$  marks the averaged molecular mass of a gas. The relaxation equation for the mass fraction  $Y$  of reagent  $A$ , and the Clapeyron equation of state for an ideal gas complement the system (1):

$$\frac{dY}{dt} = -\frac{W}{\rho}, \quad P = \frac{\rho T}{m}. \quad (2)$$

Every quantity in Eq.(1) represents a sum of an unperturbed value and its excess quantity, for example:  $p = p_0 + p'$  (in a weakly nonlinear flow absolute value of any variation is much less than the unperturbed quantity,  $|p'| \ll p_0$ , and so on). The background is homogeneous in the longitudinal direction of wave propagation which is pointed by axis  $OX$ . Following [12, 16], we assume that the stationary quantities  $Y_0, T_0, P_0, \rho_0$  may depend on the coordinate perpendicular to the direction of wave propagation only weakly, and a medium is initially motionless with  $v_0 = 0$ . In this study, we consider nonlinear terms of order not higher than second with respect to powers of perturbations. This imposes weak nonlinearity of a flow. Eqs (1), with account for Eq.(2) within accuracy of quadratic nonlinear terms, has been derived by the author in Ref.[20]:

$$\begin{aligned} \frac{\partial v'}{\partial t} + \frac{T_0}{m\rho_0} \frac{\partial \rho'}{\partial x} + \frac{1}{m} \frac{\partial T'}{\partial x} &= -v' \frac{\partial v'}{\partial x} + \frac{T_0 \rho'}{m\rho_0^2} \frac{\partial \rho'}{\partial x} - \frac{T'}{m\rho_0} \frac{\partial \rho'}{\partial x}, \\ \frac{\partial T'}{\partial t} + (\gamma_\infty - 1) \left( T_0 \frac{\partial v'}{\partial x} - Q_T \frac{Q_0}{T_0} T' - Q_\rho \frac{Q_0}{\rho_0} \rho' - Q_Y \frac{Q_0}{Y_0} Y' \right) &= -v' \frac{\partial T'}{\partial x} - (\gamma_\infty - 1) T' \frac{\partial v'}{\partial x}, \\ \frac{\partial Y'}{\partial t} + \frac{1}{Hm} \left( Q_T \frac{Q_0}{T_0} T' + Q_\rho \frac{Q_0}{\rho_0} \rho' + Q_Y \frac{Q_0}{Y_0} Y' \right) &= -v' \frac{\partial Y'}{\partial x}, \\ \frac{\partial \rho'}{\partial t} + \rho_0 \frac{\partial v'}{\partial x} &= -v' \frac{\partial \rho'}{\partial x} - \rho' \frac{\partial v'}{\partial x}, \end{aligned} \quad (3)$$

where  $\gamma_\infty = \frac{C_{P,\infty}}{C_{V,\infty}}$  denotes the frozen adiabatic exponent, and dimensionless quantities  $Q_T, Q_\rho, Q_Y$  are expressed in terms of the partial derivatives of the heat produced in a chemical reaction,  $Q$ :

$$Q_T = \frac{T_0}{Q_0} \left( \frac{\partial Q}{\partial T} \right)_{T_0, \rho_0, Y_0}, \quad Q_\rho = \frac{\rho_0}{Q_0} \left( \frac{\partial Q}{\partial \rho} \right)_{T_0, \rho_0, Y_0}, \quad Q_Y = \frac{Y_0}{Q_0} \left( \frac{\partial Q}{\partial Y} \right)_{T_0, \rho_0, Y_0}. \quad (4)$$

The second-order derivatives of  $Q$  will be considered in Sec.4.2. They are factors by quadratic nonlinear terms and do not affect nor the definition of modes nor relative projectors. The linearized version of Eqs (3) (referred below as (3)**lin**) describes a flow of infinitely-small perturbations. It differs from Eqs (3) by its right-hand side, which is zero. We start studies of linear motion by usual representing of any perturbation as a sum of planar waves proportional to  $\exp(i\omega t - ikx)$ . Four dispersion relations  $\omega_i(k)$  with ordering numbers from  $i = 1$  till 4, specify different types of fluid motion, two wave modes ( $i = 1, 2$ ), the relaxation mode ( $i = 3$ ) and the entropy mode ( $i = 4$ ). They take the leading-order forms:

$$\omega_1 = u_\infty k + \frac{u_\infty \tau_c Q_0 (\gamma_\infty - 1) (Q_\rho + Q_T (\gamma_\infty - 1))}{2\gamma_\infty T_0 (1 + ik u_\infty \tau_c)} k, \quad \omega_2 = -u_\infty k - \frac{u_\infty \tau_c Q_0 (\gamma_\infty - 1) (Q_\rho + Q_T (\gamma_\infty - 1))}{2\gamma_\infty T_0 (1 - ik u_\infty \tau_c)} k,$$

$$\omega_3 = i \left( \frac{u_\infty^2 Q_0 (\gamma_\infty - 1) (k^2 u_\infty^2 \tau_c^2 (Q_\rho - Q_T) - \gamma_\infty Q_T)}{\gamma_\infty T_0 (1 + k^2 u_\infty^2 \tau_c^2)} + \frac{1}{\tau_c} \right), \quad \omega_4 = 0, \quad (5)$$

where

$$u_\infty = \sqrt{\frac{\gamma_\infty T_0}{m}}$$

denotes the frozen sound speed, and

$$\tau_c = \frac{HmY_0}{Q_0 Q_Y} \quad (6)$$

is the characteristic duration of a chemical reaction. The approximate roots of dispersion equation for both acoustic branches in the case of weak attenuation (amplification) of small-scale sound with  $|ku_\infty \tau_c| \gg 1$  (or, equivalently, high-frequency sound with  $|\omega \tau_c| \gg 1$ ), were firstly derived in Ref.[12]. The form of Eqs(5) is appropriate for the Cauchy problem. The boundary regime problem requires the dispersion relations in the form  $k(\omega)$ . The only limitation of this study, along with weak nonlinearity, is the weak variation of heat release in a chemical reaction with temperature and density, which guarantees weak attenuation/amplification and dispersion of sound over its period, that is

$$\max|Q_\rho, Q_T| \ll \frac{\gamma_\infty T_0}{Q_0 \tau_c}.$$

The acoustic perturbations enhance if

$$Q_\rho + (\gamma_\infty - 1)Q_T > 0. \quad (7)$$

In this case, a gas is acoustically active as it has been established in Ref.[12].

## 3 Decomposition of equations in a weakly nonlinear flow

### 3.1 Modes and linear dynamic equations

Four dispersion relations (5) determine four independent modes:  $\psi = \psi_1 + \psi_2 + \psi_3 + \psi_4 = (v', T', Y', \rho')^T$ . In general, every specific perturbation contributes in the total perturbation, for example,  $\rho' = \rho_1 + \rho_2 + \rho_3 + \rho_4$ . That allows to decompose equations governing every mode in their linear parts using specific properties of modes, which are determined by the dispersion relations, and, equivalently, by the links of specific perturbations inside every mode. There is

no coupling between modes in a linear flow. The first acoustic mode (progressive in the positive direction of axis  $OX$ ), the relaxation mode, and the entropy mode are specified by the following leading-order links:

$$\psi_1 = \begin{pmatrix} v_1 \\ T_1 \\ Y_1 \\ \rho_1 \end{pmatrix} = \begin{pmatrix} \frac{u_\infty}{\rho_0} + \frac{Q_0(\gamma_\infty - 1)(Q_\rho + (\gamma_\infty - 1)Q_T)}{2\gamma_\infty T_0 \rho_0} \int_x^\infty dx_1 \exp\left(\frac{x-x_1}{u_\infty \tau_c}\right) \cdot \\ \frac{(\gamma_\infty - 1)T_0}{\rho_0} + \frac{Q_0(\gamma_\infty - 1)(Q_\rho + (\gamma_\infty - 1)Q_T)}{\rho_0 u_\infty} \int_x^\infty dx_1 \exp\left(\frac{x-x_1}{u_\infty \tau_c}\right) \cdot \\ - \frac{Q_0(Q_\rho + (\gamma_\infty - 1)Q_T)}{Hm\rho_0 u_\infty} \int_x^\infty dx_1 \exp\left(\frac{x-x_1}{u_\infty \tau_c}\right) \cdot \\ 1 \end{pmatrix} \rho_1, \quad \psi_3 = \begin{pmatrix} \frac{mu_\infty^2 + Q_0(Q_\rho - Q_T)(\gamma_\infty - 1)\tau_c}{m\rho_0 u_\infty^2 \tau_c} \int^x dx_1 - \frac{Q_0 \tau_c (\gamma_\infty - 1)(Q_\rho + (\gamma_\infty - 1)Q_T)}{2u_\infty^3 \tau_c m \rho_0} \int_{-\infty}^\infty dx_2 \exp\left(-\frac{|x-x_2|}{u_\infty \tau_c}\right) \int^{x_2} dx_1 \cdot \\ - \frac{(\gamma_\infty - 1)T_0}{\rho_0} - \frac{Q_0(\gamma_\infty - 1)(Q_\rho + (\gamma_\infty - 1)Q_T)}{\rho_0 u_\infty} \int_{-\infty}^\infty dx_1 \exp\left(-\frac{|x-x_1|}{u_\infty \tau_c}\right) + \frac{mu_\infty^2 - 2\gamma_\infty \tau_c (\gamma_\infty - 1)Q_0 Q_T}{\rho_0 u_\infty^2 \tau_c^2} \int^x dx_2 \int^{x_2} dx_1 \cdot \\ \frac{u_\infty^2}{H\rho_0(\gamma_\infty - 1)} + \frac{Q_0(Q_\rho + (\gamma_\infty - 1)Q_T)}{Hm\rho_0 u_\infty} \int_{-\infty}^\infty dx_1 \exp\left(-\frac{|x-x_1|}{u_\infty \tau_c}\right) - \frac{mu_\infty^2 - 2\gamma_\infty \tau_c (\gamma_\infty - 1)Q_0 Q_T}{Hm\rho_0 u_\infty^2 \tau_c^2 (\gamma_\infty - 1)} \int^x dx_2 \int^{x_2} dx_1 \cdot \end{pmatrix} \rho_3, \quad \psi_4 = \begin{pmatrix} 0 \\ -\frac{T_0}{\rho_0} \\ -\frac{\tau_c Q_0(Q_\rho - Q_T)}{Hm\rho_0} \\ 1 \end{pmatrix} \rho_4.$$

The lower limit of integration in the double integrals depends on the physical context of a problem. Linear equations for any mode may be immediately decomposed from the system (3) **lin**. The linear dynamic equation governing an excess density in the acoustic wave progressive in the positive direction of axis  $OX$ , and these for the relaxation and the entropy modes are established by the dispersion relations (5):

$$\begin{aligned} \frac{\partial \rho_1}{\partial t} + u_\infty \frac{\partial \rho_1}{\partial x} + \frac{Q_0(\gamma_\infty - 1)(Q_\rho + (\gamma_\infty - 1)Q_T)}{2\gamma_\infty T_0} \int_x^\infty \exp\left(\frac{x-x_1}{u_\infty \tau_c}\right) \frac{\partial \rho_1(x_1, t)}{\partial x_1} dx_1 &= 0, \\ \frac{\partial \rho_3}{\partial t} + \left( \frac{Q_0(\gamma_\infty - 1)(Q_\rho - Q_T)}{T_0 \gamma_\infty} + \frac{1}{\tau_c} \right) \rho_3 - & \\ \frac{Q_0(\gamma_\infty - 1)(Q_\rho + (\gamma_\infty - 1)Q_T)}{2\gamma_\infty \tau_c u_\infty T_0} \int_{-\infty}^\infty \exp\left(-\frac{|x-x_1|}{u_\infty \tau_c}\right) \rho_3(x_1, t) dx_1 &= 0, \quad \frac{\partial \rho_4}{\partial t} = 0. \end{aligned} \quad (9)$$

### 3.2 Non-linear corrections in acoustic mode

The linear links which specify sound should be complemented by the second-order non-linear terms making it isentropic in the leading order in the absence of any kind of attenuation. That concerns attenuation due to relaxation and the Newtonian one. Sound will be associated exclusively with the wave propagating in the positive direction of axis  $OX$ . The following corrected links are inherent to the first acoustic mode:

$$\begin{aligned} v_1(x, t) &= \frac{u_\infty}{\rho_0} \rho_1 + \frac{Q_0(\gamma_\infty - 1)(Q_\rho + (\gamma_\infty - 1)Q_T)}{2\gamma_\infty T_0 \rho_0} \int_x^\infty \exp\left(\frac{x-x_1}{u_\infty \tau_c}\right) \rho_1(x_1, t) dx_1 + \frac{(\gamma_\infty - 3)u_\infty}{4\rho_0^2} \rho_1^2, \\ T_1(x, t) &= \frac{(\gamma_\infty - 1)T_0}{\rho_0} \rho_1 + \frac{Q_0(\gamma_\infty - 1)(Q_\rho + (\gamma_\infty - 1)Q_T)}{\rho_0 u_\infty} \int_x^\infty \exp\left(\frac{x-x_1}{u_\infty \tau_c}\right) \rho_1(x_1, t) dx_1 + \end{aligned}$$

$$\frac{(\gamma_\infty - 1)(\gamma_\infty - 2)T_0}{2\rho_0^2}\rho_1^2. \quad (10)$$

The nonlinear corrections in Eqs (10) are identical to that in the planar Riemann wave [24, 1], propagating over an ideal gas. There is no leading-order correction in the mass fraction of a reagent,  $Y_1$ . One may readily conclude that an excess acoustic density is described by the dynamic equation:

$$\frac{\partial \rho_1}{\partial t} + u_\infty \frac{\partial \rho_1}{\partial x} + \frac{Q_0(\gamma_\infty - 1)(Q_\rho + (\gamma_\infty - 1)Q_T)}{2\gamma_\infty T_0} \int_x^\infty \exp\left(\frac{x - x_1}{u_\infty \tau_c}\right) \frac{\partial \rho_1(x_1, t)}{\partial x_1} dx_1 + \quad (11)$$

$$\frac{(\gamma_\infty + 1)u_\infty}{2\rho_0} \rho_1 \frac{\partial \rho_1}{\partial x} = 0,$$

which recalls the Earnshaw equation [1] but accounts for attenuation or amplification of sound in dependence on the sign of  $Q_\rho + (\gamma_\infty - 1)Q_T$  and sound dispersion.

## 4 Nonlinear effects of intense sound

### 4.1 Variations in the background temperature caused by sound

In this section, we assume magnitudes of perturbations in both non-wave modes small as compared with that of sound,  $|T_3|, |T_4| \ll \max|T_1|$ . Variations in the background temperature associate with an excess temperature in the entropy mode. The row  $d_4$  distinguishes an excess density in the entropy mode from the total vector of perturbations. It is worth noting that it contains constants and does not include derivatives with respect to the spatial coordinate:

$$d_4 \cdot \begin{pmatrix} v'(x, t) \\ T'(x, t) \\ Y'(x, t) \\ \rho'(x, t) \end{pmatrix} = \rho_4, \quad d_4^T = \begin{pmatrix} 0 \\ -\frac{\rho_0}{\gamma_\infty T_0} + \frac{Q_0 \rho_0 (Q_\rho - Q_T)(\gamma_\infty - 1)\tau_c}{\gamma_\infty^2 T_0^2} \\ -\frac{H \rho_0 (\gamma_\infty - 1)}{u_\infty^2} + \frac{H Q_0 \rho_0 (Q_\rho - Q_T)(\gamma_\infty - 1)^2 \tau_c}{\gamma_\infty T_0 u_\infty^2} \\ \frac{\gamma_\infty - 1}{\gamma_\infty} - \frac{Q_0 (Q_\rho - Q_T)(\gamma_\infty - 1)^2 \tau_c}{\gamma_\infty^2 T_0} \end{pmatrix}. \quad (12)$$

The row  $d_4$  reduces all terms belonging to the first, second and third modes in the linear part of the resulting equation when applies at the system (3). As for the nonlinear part of the dynamic equation, only acoustic quadratic terms are considered there. That yields the equation for an excess temperature attributable to the entropy mode:

$$\frac{1}{T_0} \frac{\partial T_4}{\partial t} = -\frac{1}{\rho_0} \frac{\partial \rho_4}{\partial t} = -\frac{Q_0(\gamma_\infty - 1)^2(Q_\rho + (\gamma_\infty - 1)Q_T)}{\gamma_\infty T_0 \rho_0^2} \frac{\partial \rho_1}{\partial x} \int_x^\infty \exp\left(\frac{x - x_1}{u_\infty \tau_c}\right) \rho_1(x_1, t) dx_1. \quad (13)$$

Equation (13) determines heating or cooling excited by an acoustic source which represents its right-hand side. It is instantaneous, valid for any type of sound and describes exactly the dispersive properties of a flow. An excess acoustic density should satisfy the dynamic Eq.(11). In view of the difficulties in establishing of analytical solution to Eq.(11), we make use of the solution to the linear lossless wave equation (that is the case of small nonlinearity as compared to dispersion). For the initially harmonic sound with amplitude  $V_0$ , it takes the form

$$v_1(x, t) = V_0 \exp\left(\frac{k^2 \tau_c^2 u_\infty^2 Q_0(\gamma_\infty - 1)(Q_\rho + Q_T(\gamma_\infty - 1))}{2\gamma_\infty T_0(1 + k^2 u_\infty^2 \tau_c^2)} t\right) \sin(kx - \Omega t), \quad (14)$$

with

$$\Omega = u_\infty k + \frac{\tau_c u_\infty Q_0 (\gamma_\infty - 1) (Q_\rho + Q_T (\gamma_\infty - 1)) k}{2\gamma_\infty T_0 (1 + k^2 u_\infty^2 \tau_c^2)}.$$

Eq.(14) yields the averaged over the sound period leading order expression

$$\frac{1}{T_0} \frac{\partial T_4}{\partial t} = \overline{F_{heat}} = -\frac{V_0^2 Q_0 (\gamma_\infty - 1)^2 (Q_\rho + (\gamma_\infty - 1) Q_T) \Omega^2 \tau_c^2}{2\gamma_\infty T_0 u_\infty^2 (1 + \Omega^2 \tau_c^2)}, \quad (15)$$

where

$$\overline{F_{heat}} = \frac{\Omega}{2\pi} \int_t^{t+2\pi/\Omega} F_{heat} dt.$$

Eq.(15) coincides with Eq.(17) from [21] which was devoted to the effects of high-frequency acoustic perturbations (that is,  $\Omega\tau_c \gg 1$ ). It may be readily concluded that the low-frequency sound with  $\Omega\tau_c \ll 1$  is fairly ineffective in exciting acoustic heating or cooling. If a gas is acoustically active (that is conditioned by inequality (7)), sound enhances in the course of propagation taking energy from the background. It is worth noting that the second derivatives of  $Q(\rho, T, Y)$  with respect to its arguments do not contribute to the acoustic force of heating, since they do not affect corrections due to relaxation in  $\psi_1$ . The terms proportional to the second derivatives may be considered in the second and third equations of Eqs(3). They are completely reduced in view of the form of  $d_4$ .

Eq.(13) allows to evaluate instantaneous heating produced by any waveform, for example, by impulses as follows

$$v_1(x, t) = V_0 \exp(-\Omega^2(t - x/u_\infty)^2), \quad (16)$$

$$v_1(x, t) = V_0 \sqrt{2} \exp(0.5 - \Omega^2(t - x/u_\infty)^2) \Omega(t - x/u_\infty), \quad (17)$$

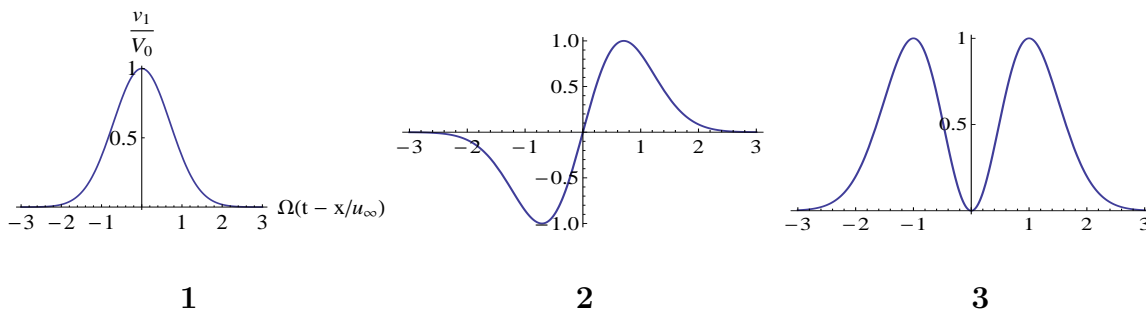
$$v_1(x, t) = V_0 \exp(1 - \Omega^2(t - x/u_\infty)^2) \Omega^3(t - x/u_\infty)^3, \quad (18)$$

$$v_1(x, t) = V_0 (2/3)^{3/2} \exp(3/2 - \Omega^2(t - x/u_\infty)^2) \Omega^2(t - x/u_\infty)^2, \quad (19)$$

$$v_1(x, t) = V_0 \cdot 1.3 \exp(-\Omega^2(t - x/u_\infty)^2) (\Omega^2(t - x/u_\infty)^2 - \Omega(t - x/u_\infty)), \quad (20)$$

$$v_1(x, t) = V_0 \cdot 1.3 \exp(-\Omega^2(t - x/u_\infty)^2) (\Omega^2(t - x/u_\infty)^4 - \Omega(t - x/u_\infty)). \quad (21)$$

In these examples,  $\Omega$  denotes the characteristic inverse sound duration. The impulses are shown in Fig.1. They in fact are solutions to the linear and lossless version of Eq.(11). The production of excess temperature per unit time is shown in Fig.2. The axes are signed only in the first plot in Figs 1,2. Deviation from symmetry of the acoustic force in first four examples of symmetric impulses is one of manifestations of relaxation. An excess temperature of the background after an impulse has gone away is determined by integral of  $F_{heat}$  over time, and it is frequency dependent. Fig.3 shows this quantity divided by  $E = \int_{-\infty}^{\infty} v_1^2 dt$ .



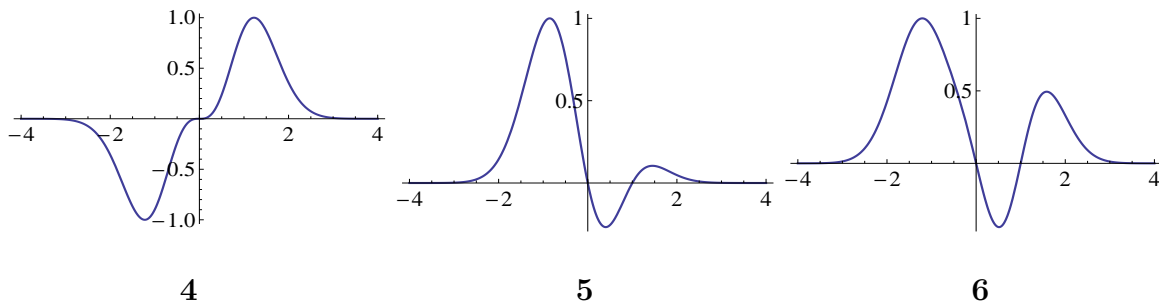


Figure 1. The acoustic impulses, Eq.(16)(1)- Eq.(21)(6).

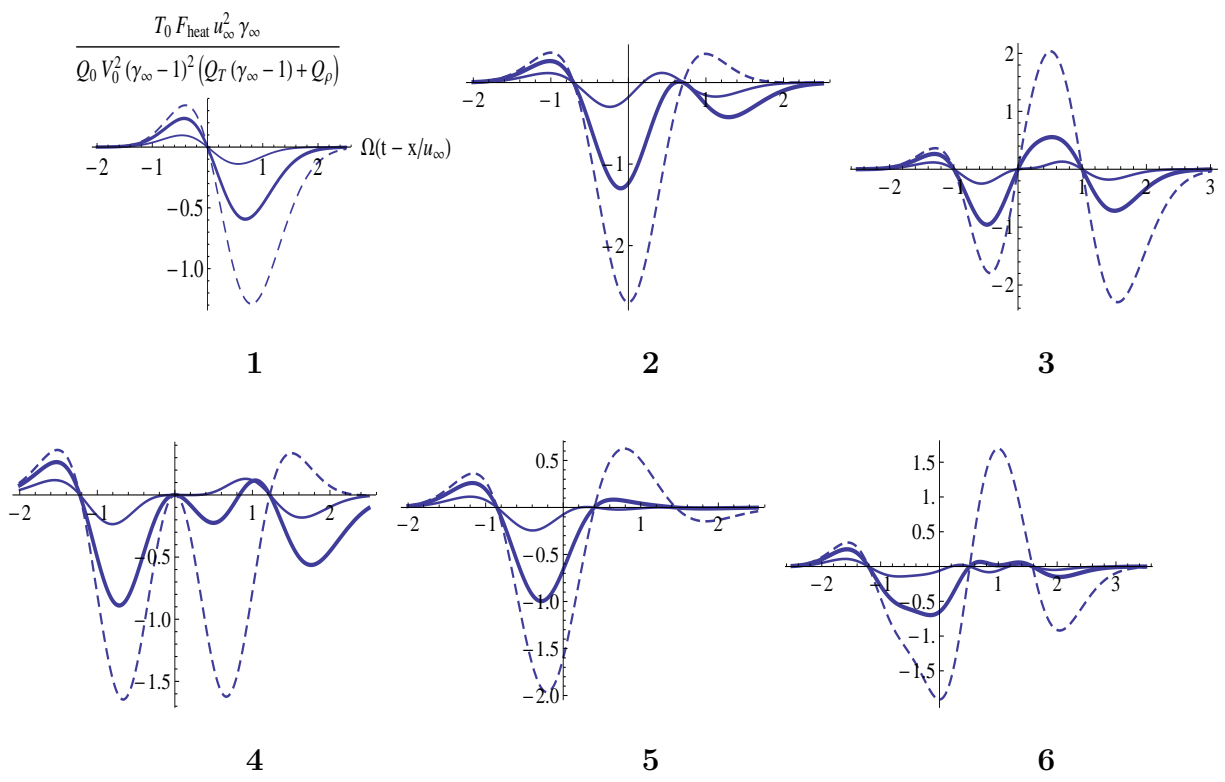


Figure 2. The dimensionless acoustic force of heating (cooling) caused by impulses Eq.(16)(1)- Eq.(21)(6). The normal lines correspond to  $\Omega\tau_c = 0.2$ , the bold lines correspond to  $\Omega\tau_c = 1$ , and the dashed lines correspond to  $\Omega\tau_c = 100$ .

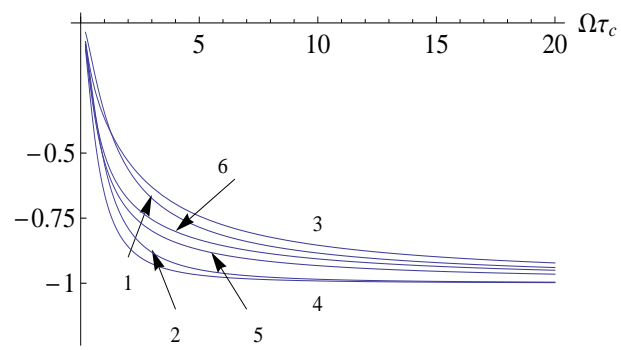




Figure 3. Variations in the background dimensionless temperature  $\frac{T_4\gamma_\infty u_\infty^2}{EQ_0(\gamma_\infty-1)^2(Q_\rho+(\gamma_\infty-1)Q_T)}$  as a function of  $\Omega\tau_c$ , after an impulse has gone away.  $E = \int_{-\infty}^{\infty} v_1^2 dt$  measures the energy of an impulse per unit mass of a medium.

The conclusion is that the fourth impulse is more effective as exciter of heating/cooling among all other impulses, and the third impulse is less effective, especially at low frequencies. Efficiency of heating constantly grows with the inverse characteristic duration of any impulse.

## 4.2 Coupling of sound with the relaxation mode. Account of second-order derivatives of $Q$

In accordance to  $\omega_3$  from Eq.(5), the characteristic time of chemical reaction depends on the spectrum of perturbation and, in general, weakly differs from  $\tau_c$ . It depends also on  $Q_\rho$  and  $Q_T$ . The row  $d_3$  may be evaluated, which distinguishes  $\rho_3$  from the total vector of perturbations,

$$d_3 \cdot \begin{pmatrix} v'(x, t) \\ T'(x, t) \\ Y'(x, t) \\ \rho'(x, t) \end{pmatrix} = \rho_3.$$

Its practical value represents only the part of the third projector applying at  $Y'$ . It is independent on  $Q_\rho$  and  $Q_T$ :

$$d_3^3 = \frac{H\rho_0(\gamma_\infty - 1)}{u_\infty^2} - \frac{H\rho_0(\gamma_\infty - 1)}{2u_\infty^3\tau_c} \left( \int_{-\infty}^{\infty} dx_1 \exp\left(-\frac{|x-x_1|}{u_\infty\tau_c}\right) \right).$$

In contrast to acoustic heating, dynamic equation for the relaxation mode contains terms proportional to the second derivatives of  $Q(\rho, T, Y)$  with respect to its variables. Nonlinear terms including the second order partial derivatives, may be readily considered in the right-hand sides of the second and third equations in the system (3). They do not affect nor definition of modes nor linear projectors but correct the nonlinear dynamic equation for sound perturbations. In particular, the equation which describes perturbation in density of the first acoustic mode progressive in the positive direction of axis  $OX$ , is:

$$\begin{aligned} \frac{\partial \rho_1}{\partial t} + u_\infty \frac{\partial \rho_1}{\partial x} + \frac{Q_0(\gamma_\infty - 1)(Q_\rho + (\gamma_\infty - 1)Q_T)}{2\gamma_\infty T_0} \int_x^\infty \exp\left(\frac{x-x_1}{u_\infty\tau_c}\right) \frac{\partial \rho_1(x_1, t)}{\partial x_1} dx_1 + \\ + \frac{Q_0(\gamma_\infty - 1)}{4\gamma_\infty \rho_0 T_0} (Q_{\rho\rho} + Q_{YY}(\gamma_\infty - 1)^2 + 2(\gamma_\infty - 1)Q_{T\rho}) \int_x^\infty \exp\left(\frac{x-x_1}{u_\infty\tau_c}\right) \frac{\partial \rho_1^2(x_1, t)}{\partial x_1} dx_1 + \\ \frac{(\gamma_\infty + 1)u_\infty}{2\rho_0} \rho_1 \frac{\partial \rho_1}{\partial x} = 0, \end{aligned} \quad (22)$$

where

$$Q_{T\rho} = \frac{T_0\rho_0}{Q_0} \left( \frac{\partial^2 Q}{\partial T \partial \rho} \right)_{T_0, \rho_0, Y_0}, \quad Q_{\rho\rho} = \frac{\rho_0^2}{Q_0} \left( \frac{\partial^2 Q}{\partial \rho^2} \right)_{T_0, \rho_0, Y_0}, \quad Q_{YY} = \frac{Y_0^2}{Q_0} \left( \frac{\partial^2 Q}{\partial Y^2} \right)_{T_0, \rho_0, Y_0}. \quad (23)$$

Eq.(22) determines the new kind of nonlinearity dependent on frequency. In the low-frequency limit, Eq.(22) coincides with the Earnshaw equation but refers to the corrected sound speed

$$u_\infty + \frac{\tau_c u_\infty Q_0(\gamma_\infty - 1)(Q_\rho + Q_T(\gamma_\infty - 1))}{2\gamma_\infty T_0}$$

and parameter of nonlinearity

$$\frac{(\gamma_\infty + 1)u_\infty}{2\rho_0} + \frac{Q_0(\gamma_\infty - 1)u_\infty\tau_c}{2\gamma_\infty\rho_0T_0}(Q_{\rho\rho} + Q_{YY}(\gamma_\infty - 1)^2 + 2(\gamma_\infty - 1)Q_{T\rho}).$$

The dynamic equation for an excess density  $\rho_3$  in the field of intense sound, takes the form

$$\begin{aligned} \frac{\partial\rho_3}{\partial t} + \frac{1}{\tau_c}\rho_3 - \frac{Q_0(\gamma_\infty - 1)(Q_\rho + (\gamma_\infty - 1)Q_T)}{2\gamma_\infty\tau_c u_\infty T_0} \int_{-\infty}^{\infty} \exp\left(-\frac{|x - x_1|}{u_\infty\tau_c}\right) \rho_3(x_1, t) dx_1 + \\ + \frac{Q_0(Q_\rho - Q_T)(\gamma_\infty - 1)}{\gamma_\infty T_0} \rho_3 = \\ \frac{H\rho_0(\gamma_\infty - 1)}{u_\infty^2} L(x, t) - \frac{H\rho_0(\gamma_\infty - 1)}{2u_\infty^3\tau_c} \left( \int_{-\infty}^{\infty} \exp\left(-\frac{|x - x_1|}{u_\infty\tau_c}\right) L(x_1, t) dx_1 \right). \\ L(x, t) = \frac{Q_0}{2Hm\rho_0^2} \left( -(\gamma_\infty + 1)(Q_\rho + (\gamma_\infty - 1)Q_T) \int_x^\infty \exp\left(\frac{x - x_1}{u_\infty\tau_c}\right) \rho_1 \frac{\partial\rho_1}{\partial x_1} dx_1 + \right. \\ \left. \rho_1((-2Q_\rho - \gamma_\infty(\gamma_\infty - 1)Q_T)\rho_1 + \frac{2(Q_\rho + (\gamma_\infty - 1)Q_T)}{u_\infty\tau_c} \int_x^\infty \exp\left(\frac{x - x_1}{u_\infty\tau_c}\right) \rho_1 dx_1) - \right. \\ \left. \frac{Q_0\rho_1^2}{2Hm\rho_0^2} (Q_{\rho\rho} + Q_{YY}(\gamma_\infty - 1)^2 + 2(\gamma_\infty - 1)Q_{T\rho}) \right). \end{aligned} \quad (24)$$

The average over the sound period  $\overline{L(x, t)}$  is some constant containing terms proportional to  $Q_\rho$  and  $Q_T$  in the case of acoustic perturbation in the form of Eq.(14)). Hence, the acoustic force of relaxation mode is zero on average in the leading order. This is conditioned by the operator  $d_3^3$  which yield zero when applies at any constant. Aperiodic or impulsive sound may produce weak perturbations in temperature and very weak perturbations in the mass fraction of the reagent,  $Y_3$ . For small-scale perturbations, ( $ku_\infty\tau_c \gg 1$ ), the relaxation mode is isobaric in the leading order with  $T_3 = -\frac{T_0}{\rho_0}\rho_3$ .

The dynamic equation which takes into account the second derivatives of  $Q$  in the high-frequency limit, is

$$\begin{aligned} \frac{\partial\rho_1}{\partial t} + u_\infty \frac{\partial\rho_1}{\partial x} - \frac{Q_0(\gamma_\infty - 1)(Q_\rho + (\gamma_\infty - 1)Q_T)}{2\gamma_\infty T_0} \rho_1 \\ - \frac{Q_0(\gamma_\infty - 1)}{4\gamma_\infty\rho_0 T_0} (Q_{\rho\rho} + Q_{YY}(\gamma_\infty - 1)^2 + 2(\gamma_\infty - 1)Q_{T\rho}) \rho_1^2 + \frac{(\gamma_\infty + 1)u_\infty}{2\rho_0} \rho_1 \frac{\partial\rho_1}{\partial x} = 0. \end{aligned} \quad (25)$$

Eq.(25) takes the similar form as Eq.(20) from Ref.[28] which describes the nonlinear dynamics of magnetoacoustic waves in acoustically active plasma in terms of the longitudinal component of velocity perturbation  $V_z$ . The latter equation includes additionally attenuation due to thermal conduction of a plasma. The sign of term proportional to  $V_z$  in Eq.(15) from Ref.[28] is determined by the first derivatives of the heating-cooling function (it reflects non-specified heating and radiative losses), and the sign of the term proportional to  $V_z^2$  is determined by the its second derivatives. The term proportional to  $V_z^2$  was found to provide a possibility of self-organizing magnetoacoustic waves (so-called autowaves) in a plasma. The authors claim that involving of this term in the evolutionary equation of sound "introduces new physics such as existence of solitary waves". In the more precise analysis, the linear term which follows

from expansion in series of the dispersion relation  $\omega_1(k)$  in Eq.(5) should be accounted in the left-hand side of Eq.(25):

$$-\frac{Q_0(\gamma_\infty - 1)(Q_\rho + (\gamma_\infty - 1)Q_T)}{2\tau_c u_\infty \gamma_\infty T_0} \int^x \rho_1(x_1, t) dx_1.$$

Its absolute value is smaller than that of the third linear term in Eq.(25) but it may be of the same order that the nonlinear term proportional to  $\rho_1^2$ . Taking into account of this term introduces additional dispersion and may alter conclusions of Ref.[28].

## 5 Remarks and Conclusions

This study incorporates the analytical method in derivation of equations which describes nonlinear dynamics of sound and their nonlinear effects, and numerical simulations. As a medium of sound propagation, we consider gases with exothermic chemical reactions. Thermodynamic relaxation, which follows an exothermic chemical reaction, results not only in a linear amplification (when inequality (7) is valid) or attenuation of sound itself but affects the character of nonlinear interaction of sound and non-acoustic modes. Relaxation makes the distortions of sound and its nonlinear phenomena frequency-dependent. The linear absorption (enhancement) of the low-frequency sound is weak, and efficiency of acoustic heating or cooling is weak also. The high-frequency sound reveals noticeable heating and variations in temperature. That was probably first established and analyzed by Molevich for example of parametric interaction of harmonic waves [16]. The advantage of this study is accurate description of evolution of nonlinear phenomena caused by sound of any characteristic duration, periodic or not. The main results of this study are instantaneous equations (13) and (24) which describe nonlinear excitation of the entropy and relaxation modes. The comparative analysis of efficiency of some impulses as exciters of acoustic heating (cooling) of a gas is done in Sec.4.1. Eq.(13) may be used in evaluations of the thermal self-action of sound beam.

It would be useful to compare the instantaneous equation for acoustic heating (cooling) with that obtained by the author in gases with the Maxwell relaxation. We reproduce its from [25]:

$$\frac{\partial T_{4,Maxwell}}{\partial t} = \frac{\mu}{\rho_0 C_V \kappa} \frac{\partial \rho_1}{\partial x} \int_{-\infty}^t \exp\left(\frac{t_1 - t}{\tau_M}\right) \frac{\partial \rho_1}{\partial x} dt_1, \quad (26)$$

where  $\mu = c_\infty^2/c_0^2 - 1$  denotes the dimensionless coefficient of dispersion,  $c_\infty$  and  $c_0$  are the sound speeds in a medium at very high and low frequencies,  $\tau_M$  is the characteristic time of Maxwell relaxation, and  $\kappa$  is the compressibility of a gas. Eq.(26) represents relaxation differently from (13). At low frequencies, it corresponds to the heating in the Newtonian fluids which is proportional to  $(\frac{\partial \rho_1}{\partial x})^2$ , that is, in the case of harmonic sound, to its square frequency.

The vortex mode which associates with acoustic streaming if caused by losses in acoustic momentum, is described by the general equation for vorticity  $\vec{\Gamma}$  [26]:

$$\frac{\partial \vec{\Gamma}}{\partial t} = -\frac{1}{\rho_0} \vec{\nabla} \times \left( \rho_a \frac{\partial \vec{v}_a}{\partial t} \right), \quad (27)$$

where  $\rho_a$ ,  $\vec{v}_a$  are summary acoustic quantities. By use of links of acoustic perturbations in a quasi-planar beam which propagates in the positive direction of axis  $OX$ , Eq.(27) rearranges

in the leading order as

$$\frac{\partial \vec{\Gamma}}{\partial t} = -\frac{Q_0(\gamma_\infty - 1)(Q_\rho + (\gamma_\infty - 1)Q_T)}{\rho_0\gamma_\infty T_0} \vec{\nabla} \rho_1 \times \left( \int_x^\infty \exp\left(\frac{x-x_1}{u_\infty\tau_c}\right) \frac{\partial \vec{v}_1}{\partial x_1} dx_1 \right), \quad (28)$$

which yields the leading-order equation for the longitudinal stream's velocity,  $V_x$ , in terms of acoustic pressure  $p_1$ :

$$\frac{\partial V_x}{\partial t} = F_s = \frac{Q_0(\gamma_\infty - 1)(Q_\rho + (\gamma_\infty - 1)Q_T)}{\rho_0^2\gamma_\infty u_\infty^3 T_0} p_1 \left( \int_x^\infty \exp\left(\frac{x-x_1}{u_\infty\tau_c}\right) \frac{\partial p_1}{\partial x_1} dx_1 \right). \quad (29)$$

In the case  $ku_\infty\tau_c \ll 1$ ,

$$F_s \approx \frac{Q_0\tau_c(\gamma_\infty - 1)(Q_\rho + (\gamma_\infty - 1)Q_T)}{\rho_0^2\gamma_\infty u_\infty^2 T_0} p_1 \frac{\partial p_1}{\partial x},$$

and in the case  $ku_\infty\tau_c \gg 1$ ,

$$F_s \approx -\frac{Q_0(\gamma_\infty - 1)(Q_\rho + (\gamma_\infty - 1)Q_T)}{\rho_0^2\gamma_\infty u_\infty^3 T_0} p_1^2.$$

Initially harmonic sound with magnitude of acoustic pressure  $P_0$  and frequency  $\Omega$  yields the leading-order average acoustic force of streaming,

$$\overline{F_s} = -\frac{P_0^2 Q_0 (\gamma_\infty - 1)^2 (Q_\rho + (\gamma_\infty - 1)Q_T) \Omega^2 \tau_c^2}{2\rho_0^2 \gamma_\infty u_\infty^3 T_0 (1 + \Omega^2 \tau_c^2)}.$$

The streamlines of vortex flow in acoustically active flows are directed unusually as compared to flows with the standard attenuation. The conclusions concerning nonlinear effects of low- and high-frequency sound coincide with the previous ones. In particular, acoustic force of heating produced by the high-frequency sound, Eq.(13), coincides with Eq.(17) from Ref.[21], and high- and low-frequency limits of acoustic force of streaming coincide with results of Ref.[27]. Low-frequency sound may excite very weak streaming. In this study, we did not consider neither standard attenuation nor thermal conductivity of a gas. These effects may impose on the impact of chemical reaction and prevent strengthening of sound in a gas during non-equilibrium processes. Taken alone, the standard attenuation always leads to damping of sound, which is proportional to its square frequency in the case of periodic sound [1]. Account of heat production in a chemical reaction in zero-order hydrodynamic equations would lead to dependence of the background temperature and density on spacial coordinates. Eqs.(3) are derived imposing small transversal gradients of the background thermodynamic parameters. For large  $Q_0$ , nor definition of modes, nor projectors which follows from (3), are longer valid. That introduces additional dispersion and may lead to the different domain of the wave stability. In spite of similarity of thermodynamic processes in gases with vibrational relaxation and these in which chemical reaction occurs, chemically reacting gases are most important for accurate description of sound dispersion and its nonlinear phenomena in view of comparatively large relaxation times of chemical reactions.

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