# Instantaneous Heating and Cooling Caused by Periodic or Aperiodic Sound of Any Characteristic Duration in a Gas with Vibrational Relaxation

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#### Summary

Thermodynamic relaxation of internal degrees of a molecule's freedom in a gas occurs with some characteristic time. This makes wave processes in a gas behave differently depending on the ratio of characteristic duration of perturbations and the relaxation time. In particular, generation of the secondary non-wave modes by intense sound in a nonlinear flow dependens on frequency. These kinds of interaction are considered in this study. The exact links between perturbations inside every type of a fluid's motion (modes) and resulting weakly nonlinear equations for interacting modes are derived. These equations are instantaneous and hence are valid for pulsed excitation. Some kind of energy inflow makes a gas with excited degrees of oscillatory molecule's freedom acoustically active. That leads to anomalous acoustic cooling of a gas. The impact of standard viscosity, thermal conductivity, and dependence of the power of energy inflow on temperature is briefly discussed.

PACS no. 43.35.Fj, 43.25.Ed

# 1. Introduction

All thermodynamic processes in fluids occur with some typical times of relaxation. Some processes like Maxwell exchange of translational degrees of molecules freedom possess such small times of relaxation that they may be in fact considered as instantaneous, as compared to the characteristic duration of any macroscopic external perturbation. On the other hand, hydrodynamic perturbations in fluids with internal processes with relatively large time of relaxation, depend strongly on the frequency of external perturbations. Among these relaxation processes, the retarded energy exchange between vibrational, rotational and translational degrees of molecules freedom may be listed. This exchange of energy behaves differently at positive and negative half-periods of perturbations of velocity in sound wave [1, 2, 3]. Chemical reactions in gases are also of importance [4, 5]. The relaxation processes in liquids are much more various. Relaxation is always associated with dispersion and frequency-dependent attenuation.

Relaxation processes in the open systems may be followed by unusual events. This makes them of especial interest in many physical applications. That concerns pumping of energy into vibrational degrees of a molecule's freedom and formation of a non-equilibrium two-temperature

discharge of the excess vibrational energy into translation degrees of a freedom leads to a new high-temperature regime [1, 4, 5]. Actually, the fluid flow in the non-equilibrium media is one of the new and still insufficiently studied fields of

is one of the new and still insufficiently studied fields of modern hydrodynamics, whereas the optical phenomena in the non-equilibrium gases are well-studied due to laser applications.

gas. Such two-temperature state may be unstable, when

In the non-equilibrium fluids, the distribution of energy alters the adiabatic compressibility which leads to anomalous dispersion of sound waves. That was established at first experimentally, and explained theoretically by Herzfeld and Rice [6]. The mathematical content of the nonlinear description of the wave and non-wave processes in a relaxing medium is fairly complex. The nonlinear distortions of sound itself in dispersive fluids are wellunderstood, including formation of the stationary waveforms [7, 8, 9]. Enhancement and nonlinear distortions of sound in acoustically active fluids have been also described in some later references [1, 3, 10]. The nonlinear loss of acoustic energy in the standard thermoviscous fluid increases its background temperature. That occurs due to nonlinear interaction of acoustic and entropy modes in a damping fluid [11]. This transfer of energy from sound to chaotic motion of molecules is called acoustic heating [12, 13]. A rate of temperature increase is proportional to the sound intensity and overall attenuation in a new-

Received 12 January 2017, accepted 23 May 2017.

tonian fluid. As was pointed out by Molevich [3], the nonlinear exchange of energy between sound and the thermal mode may lead to cooling instead of heating in the nonequilibrium gas, if the standard attenuation of a medium is small. The possibility of an anomaly in acoustic streaming, was also pointed out by Molevich [14, 15]. That imposes that the induced vortex motion may occur in an unusual direction. Molevich has considered the harmonic sound of arbitrary frequency in the problems of excitation of the entropy mode. The nonlinear analysis in the papers by Molevich makes use of the method of successive approximations. This means that the solutions to equations take the form of series and result from the set of coupling equations which contain terms of the same order of magnitude [16]. The acoustic force of streaming which was considered in [15], is in fact the averaged quantity over the period of harmonic sound. It is valid for strictly periodic sound and small Reynolds numbers of a flow.

To the best of the author's knowledge, this study is the first which considers exact dispersive properties of nonlinear phenomena in a gas with excited vibrational degrees of molecules freedom caused by sound of any kind not just single frequency excitation.

The method of projecting proposed by the author allows derivation of the dynamic equations for sound and non-wave modes accounting for their interaction in many cases of fluid's flow [17, 18]. That concerns flows of newtonian and non-newtonian fluids perhaps affected by external forces. Equations are formulated in time domain and so do not imply neither periodic sound nor any kind of averaging.

This is the advantage of the method of projecting. The equations governing nonlinear phenomena accompanied sound propagation in a gas with vibrational relaxation have been derived and analyzed by the author in the highand low-frequency limits in [19, 20]. The high- and low-frequency limits are readily traceable. We start from determination of the linear modes as specific types of gas motion in which background parameters are maintained by pumping energy into the vibrational degrees of freedom by power I, and a heat withdrawal from the translational degrees of freedom of power Q (both I and Q refer to unit mass). The relaxation equation for the vibrational energy per unit mass  $\epsilon$  takes the form

$$\frac{\mathrm{d}\varepsilon}{\mathrm{d}t} = -\frac{\varepsilon - \varepsilon_{eq}(T)}{\tau} + I. \tag{1}$$

The equilibrium value of the vibrational energy at a given temperature T is denoted as  $\varepsilon_{eq}(T)$ , and  $\tau$  denotes the vibrational relaxation time. The relaxation time in the most important cases may be thought as a function of temperature [1, 5]. The results may be easily generalized in the case of relaxation time which depends on temperature and density (Sections 3.3-3.5). The quantity  $\varepsilon_{eq}(T)$  relates to a simple model of a molecule as a harmonic oscillator [1, 8],

$$\varepsilon_{eq}(T) = \frac{\hbar\Omega}{m\left(exp(\hbar\Omega/k_BT) - 1\right)},\tag{2}$$

where *m* is the mass of a molecule,  $\hbar\Omega$  is the magnitude of the vibrational quantum,  $k_B$  is the Boltzmann constant. Equation (2) is valid over the temperatures, where anharmonic effects are weak, i.e., below the characteristic temperatures, which are fairly high for the majority of molecules. In particular, the characteristic temperatures for  $H_2$ , CO,  $O_2$  and  $N_2$  equal, respectively, 6300*K*, 3100*K*, 2300*K* and 3400*K* [1, 4, 5].

The momentum, energy and mass conservation equations for a flow of vibrationally relaxing gas without thermal and mechanical losses read

$$\rho \left[ \frac{\partial \vec{v}}{\partial t} + (\vec{v} \cdot \vec{\nabla}) \vec{v} \right] = -\vec{\nabla} p,$$

$$\rho \left[ \frac{\partial (e+\varepsilon)}{\partial t} + (\vec{v} \cdot \vec{\nabla}) (e+\varepsilon) \right] + p(\vec{\nabla} \cdot \vec{v}) \qquad (3)$$

$$= \rho (I-Q),$$

$$\frac{\partial \rho}{\partial t} + \vec{\nabla} \cdot (\rho \vec{v}) = 0,$$

where  $\vec{v}$  denotes the velocity of a fluid,  $\rho$ , *p* are its density and pressure, *e* is the internal energy of translation motion of molecules per unit mass.

Apart from Equations (1), (2), two thermodynamic functions  $e(p, \rho)$ ,  $T(p, \rho)$  complete the system (3). We use these ones for an ideal gas,

$$e(p,\rho) = \frac{p}{(\gamma-1)\rho} = \frac{R}{\mu(\gamma-1)}T(p,\rho), \qquad (4)$$

where  $\gamma$  is the isentropic exponent without account for vibrational degrees of freedom, *R* denotes the universal gas constant, and  $\mu$  is the molar mass of a gas.

# 2. Planar motions of infinitely small magnitudes of perturbations and their decomposition

Consider a flow of infinitely small magnitudes of perturbations. A flow is supposed to be planar along axis Ox with Q = const, I = const. We will discuss the impact of thermal conductivity, viscosity, and dependence of Q and I on the temperature in Section 3.5.

Considering every quantity q as a sum of unperturbed value  $q_0$  (the background flows are absent, so that  $v_0 = 0$ ) and its variation q', one readily rearranges the governing equations of momentum, energy balance, and continuity into the linear form which contains only the first powers of perturbations [1],

$$\frac{\partial v'}{\partial t} + \frac{1}{\rho_0} \frac{\partial p'}{\partial x} = 0,$$
  
$$\frac{\partial p'}{\partial t} + \gamma p_0 \frac{\partial v'}{\partial x} - (\gamma - 1)\rho_0 \frac{\varepsilon'}{\tau}$$
  
$$+ (\gamma - 1)\rho_0 T_0 \Phi_1 \left(\frac{p'}{p_0} - \frac{\rho'}{\rho_0}\right) = 0,$$
  
$$\frac{\partial \rho'}{\partial t} + \rho_0 \frac{\partial v'}{\partial x} = 0,$$
  
$$\frac{\partial \varepsilon'}{\partial t} + \frac{\varepsilon'}{\tau} - T_0 \Phi_1 \left(\frac{p'}{p_0} - \frac{\rho'}{\rho_0}\right) = 0,$$
 (5)

where

$$\Phi_1 = \left(\frac{C_v}{\tau} + \frac{\varepsilon - \varepsilon_{eq}}{\tau^2} \frac{\mathrm{d}\tau}{\mathrm{d}T}\right)_0 \tag{6}$$

is the quantity evaluated at equilibrium state at  $p_0$ ,  $T_0$ , and  $C_v = d\varepsilon_{eq}/dT$ . We make use of the expansion in series of Equations (4) to express perturbations of translation temperature and internal translational energy in terms of perturbations of pressure and density,

$$e' = \frac{p_0}{(\gamma - 1)\rho_0} \left(\frac{p'}{p_0} - \frac{\rho'}{\rho_0}\right).$$
 (7)

The last equation in the set (5) follows from Equations (1), (7),

$$\frac{\partial \varepsilon'}{\partial t} + \frac{\varepsilon'}{\tau} = \left(\frac{C_v}{\tau} + \frac{\varepsilon - \varepsilon_{eq}}{\tau^2} \frac{\mathrm{d}\tau}{\mathrm{d}T}\right)_0 T' = T_0 \Phi_1 \left(\frac{p'}{p_0} - \frac{\rho'}{\rho_0}\right).$$
(8)

Without loss of generality,  $\tau$  in the left-hand side of Equation (8) denotes its equilibrium value,  $\tau_0$ .

Studies of fluid motions of infinitely-small amplitudes start usually with representing of all perturbations as a sum of planar waves,

$$q'(x,t) = \int_{-\infty}^{\infty} \tilde{q}(k) \cdot \exp i(\omega t - kx) \, dk + cc, \qquad (9)$$

where  $\tilde{q}(k) \exp(i\omega(k)t)$  is the Fourier-transforms of any perturbation q'.

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 conditions  $\omega \tau \gg 1$  (or  $\omega \tau \ll 1$ ), which restricts consideration by the high-frequency sound (low-frequency sound) [3, 20]. The dispersion relations that are valid for any frequency of sound (alternatively, any wave number k), take the forms

$$\omega_{1} = ck - \frac{(\gamma - 1)^{2}T_{0}k\tau}{2c(1 + ick\tau)}\Phi_{1},$$
  

$$\omega_{2} = -ck + \frac{(\gamma - 1)^{2}T_{0}k\tau}{2c(1 - ick\tau)}\Phi_{1},$$
(10)

where  $c = \sqrt{\gamma RT_0/\mu} = \sqrt{\gamma p_0/\rho_0}$  denotes the infinitely small-signal sound speed in the ideal uniform gas. All evaluations in this study are valid in the case of weakly dispersive and weakly damped perturbations, that is, when

$$\left| \Phi_1 \right| \ll \frac{2c^2}{(\gamma - 1)^2 T_0 \tau}.$$
 (11)

This is the only condition which gives possibility to expand formulas in series and to keep only leading order terms proportional to the zero and first powers of  $\Phi_1$ ,  $\Phi_1^0$  and  $\Phi_1^1$ . The powerful inflow of energy makes the background inhomogeneous and hence may change the conditions of acoustical activity of a gas [21, 22]. The inequality

(11) provides weak distortion of the sound wave caused by dispersion and attenuation, if  $\Phi_1 > 0$  (or amplification, if  $\Phi_1 < 0$ ) over its period. It does not impose restrictions on the spectrum of perturbation in dependence on relaxation time,  $\tau$ . of sound in a gas having internal degrees of freedom. Two last roots of dispersive equation represent the non-wave motions,

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

$$\omega_4 = 0.$$
(12)

The third non-wave mode, which is determined by  $\omega_3$ , originates from the vibrational relaxation, and the fourth root represents the thermal, or entropy, mode.

According to Equations (11),(12), the characteristic time of thermodynamic relaxation weakly differs from  $\tau$ . The perturbations specifying the first acoustic mode, and, in particular, excess density, satisfy the dynamic equation

$$\frac{\partial \rho_1}{\partial t} + c \frac{\partial \rho_1}{\partial x} + \frac{(\gamma - 1)^2 T_0}{2c^2} \Phi_1 \rho_1$$

$$- \frac{(\gamma - 1)^2 T_0}{2c^3 \tau} \Phi_1 \int_x^\infty \exp\left(\frac{x - x'}{c\tau}\right) \rho_1(x', t) \, \mathrm{d}x' = 0.$$
(13)

We will omit prime by the specific perturbations which belong to the different modes. The overall velocity, pressure, density and internal energy are a sum of specific parts:  $v'(x,t) = \sum_{n=1}^{4} v_n(x,t)$ , and so on. In accordance to the roots (10), (12), the Fourier transforms of perturbations may be expressed in terms of four specific Fourier transforms of specific excess densities  $\tilde{\rho}_1$ ,  $\tilde{\rho}_2$ ,  $\tilde{\rho}_3$ ,  $\tilde{\rho}_4$  in the following manner:

$$\tilde{\rho} = \sum_{n=1}^{4} \tilde{\rho}_{n}, \quad \tilde{v} = \sum_{n=1}^{4} \tilde{v}_{n} = \sum_{n=1}^{4} \omega_{n} \tilde{\rho}_{n} / k / \rho_{0},$$

$$\tilde{p} = \sum_{n=1}^{4} \tilde{p}_{n} = \sum_{n=1}^{4} \omega_{n}^{2} \tilde{\rho}_{n} / k^{2},$$

$$\tilde{\varepsilon} = \sum_{n=1}^{4} \tilde{\varepsilon}_{n} = \frac{T_{0} \Phi_{1} \tau}{\rho_{0} c^{2} k^{2}} \sum_{n=1}^{2} \tilde{\rho}_{n} \frac{\gamma \omega_{n}^{2} - c^{2} k^{2}}{1 + i \omega_{n} \tau}.$$
(14)

The links in the (x, t) space follow from Equations (14) and dispersion relations, Equations (10),(12). We may readily establish the operator rows which project the overall vector of the Fourier transforms of perturbations into the Fourier transforms of specific densities. The rows as follow (ordering as third and fourth) distinguish  $\tilde{\rho}_3$  and  $\tilde{\rho}_4$ :

$$\begin{split} \tilde{d}_1^3 \tilde{v} + \tilde{d}_2^3 \tilde{p} + \tilde{d}_3^3 \tilde{\rho} + \tilde{d}_4^3 \tilde{\varepsilon} &= \tilde{\rho}_3, \\ \tilde{d}_1^4 \tilde{v} + \tilde{d}_2^4 \tilde{p} + \tilde{d}_3^4 \tilde{\rho} + \tilde{d}_4^4 \tilde{\varepsilon} &= \tilde{\rho}_4. \end{split}$$

They are

$$\begin{pmatrix} \tilde{d}_{1}^{3} \\ \tilde{d}_{2}^{2} \\ \tilde{d}_{3}^{3} \\ \tilde{d}_{4}^{3} \end{pmatrix} =$$
(15)



Thereby, projecting distinguishes the specific disturbance from the total vector of perturbations at any time. Projecting is in fact a certain way of linear combination of equations in order to keep one specific quantity in the linear part of equations (in this study, the specific excess density; the choice of reference specific variable may be different). It is based on the linear links between perturbations which determine every mode as well as the correspondent dispersion relations. The projectors in (x, t) space readily follow from Equations (15).

## 3. Nonlinear effects of sound

#### 3.1. Weakly nonlinear system of conservation equations

The governing dynamic system with account for quadratic nonlinear terms differs from (5) by the quadratic right-hand side. It has been derived by the author in [20]:

$$\begin{aligned} \frac{\partial v'}{\partial t} &+ \frac{1}{\rho_0} \frac{\partial p'}{\partial x} = -v' \frac{\partial v'}{\partial x} + \frac{\rho'}{\rho_0^2} \frac{\partial p'}{\partial x}, \\ \frac{\partial p'}{\partial t} &+ \gamma p_0 \frac{\partial v'}{\partial x} - (\gamma - 1) \rho_0 \frac{\varepsilon'}{\tau} + (\gamma - 1) \rho_0 T_0 \Phi_1 \left(\frac{p'}{p_0} - \frac{\rho'}{\rho_0}\right) \\ &= -v' \frac{\partial p'}{\partial x} - \gamma p' \frac{\partial v'}{\partial x} + (\gamma - 1) \rho' \left[\frac{\varepsilon'}{\tau} - T_0 \Phi_1 \left(\frac{p'}{p_0} - \frac{\rho'}{\rho_0}\right)\right] \\ &- (\gamma - 1) \rho_0 \left[T_0 \left(\frac{1}{\tau^2} \frac{d\tau}{dT}\right)_0 \varepsilon' \left(\frac{p'}{p_0} - \frac{\rho'}{\rho_0}\right) \\ &+ T_0 \Phi_1 \left(\frac{\rho'^2}{\rho_0^2} - \frac{p' \rho'}{p_0 \rho_0}\right) + T_0 \Phi_2 \left(\frac{p'}{p_0} - \frac{\rho'}{\rho_0}\right)^2\right], \\ &\frac{\partial \rho'}{\partial t} + \rho_0 \frac{\partial v'}{\partial x} = -v' \frac{\partial \rho'}{\partial x} - \rho' \frac{\partial v'}{\partial x}, \end{aligned}$$
(16)  
$$&\frac{\partial \varepsilon'}{\partial t} + \frac{\varepsilon'}{\tau} - T_0 \Phi_1 \left(\frac{p'}{p_0} - \frac{\rho'}{\rho_0}\right) \\ &= T_0 \left(\frac{1}{\tau^2} \frac{d\tau}{dT}\right)_0 \varepsilon' \left(\frac{p'}{p_0} - \frac{\rho'}{\rho_0}\right) + T_0 \Phi_1 \left(\frac{\rho'^2}{\rho_0^2} - \frac{p' \rho'}{p_0 \rho_0}\right) \\ &+ T_0 \Phi_2 \left(\frac{p'}{p_0} - \frac{\rho'}{\rho_0}\right)^2 - v' \frac{\partial \varepsilon'}{\partial x}, \end{aligned}$$

where we made use of the leading order series with respect to powers of perturbations in Equations (1), (4):

$$T' = T_{0} \left( \frac{p'}{p_{0}} - \frac{\rho'}{\rho_{0}} + \frac{\rho'^{2}}{\rho_{0}^{2}} - \frac{p'\rho'}{p_{0}\rho_{0}} \right),$$

$$\frac{d\epsilon'}{dt} = -\frac{\epsilon'}{\tau} + T_{0} \left( \frac{1}{\tau^{2}} \frac{d\tau}{dT} \right)_{0} \epsilon' \left( \frac{p'}{p_{0}} - \frac{\rho'}{\rho_{0}} \right)$$

$$+ T_{0} \Phi_{1} \left( \frac{p'}{p_{0}} - \frac{\rho'}{\rho_{0}} + \frac{\rho'^{2}}{\rho_{0}^{2}} - \frac{p'\rho'}{p_{0}\rho_{0}} \right)$$

$$+ T_{0} \Phi_{2} \left( \frac{p'}{p_{0}} - \frac{\rho'}{\rho_{0}} \right)^{2},$$

$$\Phi_{2} = T_{0} \left[ -\frac{1}{\tau^{2}} C_{v} \frac{d\tau}{dT} - \frac{(\epsilon - \epsilon_{eq})}{\tau^{3}} \left( \frac{d\tau}{dT} \right)^{2} + \frac{1}{2\tau} \frac{dC_{v}}{dT} + \frac{(\epsilon - \epsilon_{eq})}{2\tau^{2}} \frac{d^{2}\tau}{dT^{2}} \right]_{0}.$$
(17)

We consider nonlinear effects in the leading order, that is, these ones connected with the quadratic nonlinear terms which are of the most importance in the nonlinear acoustics.

#### 3.2. Links of perturbations in the intense sound

The problems of generation of the non-acoustic types of motion by the intense sound are of the major importance among all problems which refer to the nonlinear interaction of modes. In turn, the non-wave modes affect the propagation of sound. In the setting of these problems, sound is intense as compared with other types of motion, that is, magnitudes of its perturbations are much larger than that of the non-wave modes. Without loss of generality, we will consider exclusively rightwards propagating disturbances, assuming that all other modes, though may enlarge their magnitudes in time due to nonlinear interaction, are relatively small. Since the order of magnitude of secondary modes is no higher, than the squared Mach number  $M^2$ , the accurate account for the quadratic corrections in the dominative sound, which are of the same order, are necessary [17, 18]. The dynamic equation governing the dominative sound which includes nonlinear term of order  $M^2$ readily follows from the weakly nonlinear corrections to the links between specific acoustic perturbations. Vectors  $\psi_{1,l}$  and  $\psi_{1,nl}$  below represent the linear links and nonlinear corrections for the perturbations in the rightwards progressive sound in the leading order, as it follows from Equations (16).

$$\begin{split} \psi_{1,l} &= \begin{pmatrix} v_1 \\ p_1 \\ \rho_1 \\ \varepsilon_1 \end{pmatrix} (18) \\ &= \begin{pmatrix} 1 \\ \rho_0 c - \frac{(\gamma - 1)^2 T_{0\rho07}}{2c^2 \tau} \Phi_1 \int_x^{\infty} \exp\left(\frac{x - x'}{c\tau}\right) dx' \cdot \\ \frac{\rho_0}{c} + \frac{(\gamma - 1)^2 T_{0\rho07}}{2c^2 \tau} \Phi_1 \int_x^{\infty} \exp\left(\frac{x - x'}{c\tau}\right) dx' \cdot \\ \frac{(\gamma - 1)T_0}{c^2} \Phi_1 \int_x^{\infty} \exp\left(\frac{x - x'}{c\tau}\right) dx' \cdot \end{pmatrix} v_1, \end{split}$$

$$\psi_{1,nl} = \begin{pmatrix} 0 \\ \frac{(3-\gamma)\rho_0}{4c^2} v_1 \\ \frac{(\gamma+1)\rho_0}{4} v_1 \\ 0 \end{pmatrix} v_1$$

 $\psi_{1,nl}$  reflects the nonlinear corrections which agree with that in the Riemann wave [12]. They in fact support adiabaticity of sound in the lossless flow.

The total vector of acoustic perturbations is a sum of  $\psi_{1,l}$  and  $\psi_{1,nl}$ , and weakly nonlinear dynamic equation for the acoustic excess density takes the form

$$\frac{\partial \rho_1}{\partial t} + c \frac{\partial \rho_1}{\partial x} + \frac{(\gamma - 1)^2 T_0}{2c^2} \Phi_1 \rho_1$$

$$- \frac{(\gamma - 1)^2 T_0}{2c^3 \tau} \Phi_1 \int_x^\infty \exp\left(\frac{x - x'}{c\tau}\right) \rho_1(x', t) \, \mathrm{d}x'$$

$$+ \frac{(\gamma + 1)c}{2\rho_0} \rho_1 \frac{\partial \rho_1}{\partial x} = 0.$$
(19)

The nonlinear term in Equation (19) manifests the wellcelebrated nonlinearity originating from nonlinearity in equation of state and hydrodynamic nonlinearity [12, 7] which is of order  $M^2$ .

#### 3.3. Acoustic heating or cooling

The linear projecting is fruitful in investigations of nonlinear interactions of different modes [17, 18, 19]. After application of some projecting row at the system of equations, contributions of all other modes in the linear part of the final dynamic equation are reduced, but the nonlinear terms become distributed between equations. At this point, we make routine manipulations to decompose the dynamic equation for the specific excess density of the entropy mode by means of applying the row  $(d_1^4 \quad d_2^4 \quad d_3^4 \quad d_4^4)$  at the system (16) and collecting together terms of the leading order. Among all coupling nonlinear terms, only pure acoustic ones are considered which are the biggest. They form acoustic force of heating. The links of acoustic perturbations are represented by  $\psi_1 = \psi_{1,l} + \psi_{1,nl}$ . The resulting equation governing the acoustic heating is

$$\frac{1}{T_0} \frac{\partial T_4}{\partial t} = -\frac{1}{\rho_0} \frac{\partial \rho_4}{\partial t} = F_{\text{heat}}$$

$$= -\frac{(\gamma - 1)^2 T_0 \Phi_1}{2c^5 \tau} \left[ \left( c\tau(\gamma + 1) \frac{\partial v_1}{\partial x} + (3 - \gamma) v_1 \right) \int_x^\infty \exp\left(\frac{x - x'}{c\tau}\right) v_1(x', t) \, \mathrm{d}x' - c\tau(3 - \gamma) v_1^2 \right], \quad (20)$$

where  $T_4$  is an excess temperature specific for the entropy mode during an isobaric transfer of acoustic energy into the energy of the entropy mode. On the other hand, velocity in the right-hand side of Equation (20) should satisfy the dynamic equation for sound. Equation (20) is valid at any characteristic frequency of sound. The limits of lowand high- frequency sound may be readily traced. In the low-frequency limit,  $ck\tau \ll 1$ , and the term  $\int_x^{\infty} \exp[(x - x')/(c\tau)]v_1(x', t) dx'$  may be replaced in the leading order with  $c\tau v_1(x, t)$  in Equation (20). In the high-frequency limit,  $ck\tau \gg 1$ , this term may be replaced in the leading order with  $\int_x^{\infty} v_1(x', t) dx'$ . In view of difficulty in establishing the exact solution to Equation (19), the simple estimations of acoustic source  $F_{\text{heat}}$  in the case of periodic sound of any frequency may be implemented on the base of solution of the linear version of equation, into which the governing equation (19) transforms when  $\Phi_1$  tends to zero, that is,

$$\frac{c}{\rho_0}\rho_1 = v_1 = V_0 \sin(\Omega(t - x/c)),$$
(21)

where  $V_0$  denotes the amplitude of velocity. Acoustic force of heating, averaged over the sound period, takes the form

$$\overline{F_{\text{heat}}} = \frac{\Omega}{2\pi} \int_{t}^{t+2\pi/\Omega} F_{\text{heat}} dt$$
$$= \frac{V_0^2 (\gamma - 1)^3 T_0 \Phi_1 \Omega^2 \tau^2}{2c^4 (1 + \Omega^2 \tau^2)}, \qquad (22)$$

which rearranges in the low-frequency and high-frequency limits, respectively, into

$$\overline{F_{\text{heat,low}}} = \frac{V_0^2 (\gamma - 1)^3 T_0 \Phi_1 \Omega^2 \tau^2}{2c^4},$$
  
$$\overline{F_{\text{heat,high}}} = \frac{V_0^2 (\gamma - 1)^3 T_0 \Phi_1}{2c^4}.$$
 (23)

It follows from the above equations, that the low-frequency sound ( $\Omega \tau \ll 1$ ) is fairly ineffective in generation of acoustic heating since the acoustic force is proportional to ( $\Omega \tau$ )<sup>2</sup> [19]. In turn, the high-frequency sound with  $\Omega \tau \gg 1$  produces heat per unit time proportional to  $1 - (\Omega \tau)^{-1}$ . The expression for  $\overline{F_{\text{heat,high}}}$  coincides with Equation (25) from [20].

The sign of the right-hand acoustic source is determined by the sign of  $\Phi_1$ . In the equilibrium regime,  $\Phi_1$  is positive and corresponds to decrease in density and relative increase in temperature of the entropy mode with time. Hence, acoustic heating of the background takes place. When  $\Phi_1 < 0$ , a gas is acoustically active. The anomalous cooling takes place, that is, the background density increases, and its temperature constantly gets smaller. Some simple estimations may be carried out for the case of typical laser mixture  $CO_2 : N_2 : He = 1 : 2 : 3$  at pressure  $p_0 = 1atm$  and temperature  $T_0 = 300K$ . We make use of thermodynamical data from [5, 23, 10]. By using  $\gamma = 1.33$ and  $c = 422m \cdot s^{-1}$ , Equation (22) takes the form

$$\overline{F_{\text{heat}}} = M^2 \frac{\Omega^2 \tau^2}{1 + \Omega^2 \tau^2} 6.65 \cdot 10^{-7} (I_{th} - I),$$

where the acoustic force is measured in 1/s, I is measured in W/kg, and  $I_{th}$  is the threshold quantity,  $I_{th} = 1.99 \cdot 10^6$  W/kg. Hence, the absolute value and sign of the acoustic force depend on the squared Mach number of sound,



Figure 1. The dimensionless acoustic force of heating (cooling)  $c^4 F_{heat}/(V_0^2 \Phi_1(\gamma-1)^2 T_0)$  for the symmetric pulse (Equation (24), upper plot) and asymmetric pulse (Equation 25, lower plot). The dotted line corresponds to  $\Omega \tau = 0.5$ , the normal line corresponds to  $\Omega \tau = 1$ , and the dashed line relates to  $\Omega \tau = 10$ .

product  $\Omega \tau$ , and difference between *I* and  $I_{th}$ . The relaxation time equals approximately  $5 \cdot 10^{-5}$  s. The exemplary acoustic force at  $\Omega = 1$  MHz,  $M = 10^{-2}$  (this corresponds to the magnitude of velocity 4.2 m/s and to the magnitude of acoustic pressure 1013 Pa) and  $I = 2.5 \cdot 10^6$  W/kg, equals  $-3.4 \cdot 10^{-5}$ /s, that is, the gas tempreature decreases every minute by 0.6 degrees. Equation (20) allows evaluating heating (or cooling) produced by any waveform, for example, by the pulses

$$v_1(x,t) = V_0 \exp\left(-\Omega^2 (t-x/c)^2\right),$$
 (24)

and

$$v_1(x,t) = 2V_0 \exp(0.5 - 2\Omega^2 (t - x/c)^2)$$
  
 
$$\cdot \Omega(t - x/c).$$
(25)

The production of dimensionless excess temperature per unit time in dimensionless quantities and different  $\Omega \tau$  for  $\gamma = 1.33$  is shown in Figure 1.

Deviation from symmetry of the acoustic force in these examples is one of manifestations of relaxation. The residual excess temperature of a gas after pulse passes away is determined by integral of  $F_{\text{heat}}$  over time. Figure 2 shows dependence of the dimensional residual temperature,  $c^4T_4/(E\Phi_1(\gamma-1)^2T_0^2)$ , on  $\Omega\tau$ .  $E = \int_{-\infty}^{\infty} v_1^2 dt$  measures the energy of a pulse. The conclusion is that the symmetric pulse produces larger variations in temperature at large enough characteristic frequency of a pulse,



Figure 2. Residual dimensionless variations in the background temperature  $c^4T_4/(E\Phi_1(\gamma-1)^2T_0^2)$  as a function of  $\Omega\tau$ .  $E = \int_{-\infty}^{\infty} v_1^2 dt$  measures the energy of a pulse per unit mass of a gas. The case of symmetric pulse (Equation 24) is numbered by 1, and the case of asymetric pulse (Equation 25) is numbered by 2.

 $\Omega > 2\tau^{-1}$ . For the laser mixture CO<sub>2</sub> : N<sub>2</sub> : He = 1 : 2 : 3 at normal conditions,  $M = 10^{-2}$  and  $I = 2.5 \cdot 10^6$  W/kg, the residual excess temperature equals  $-2.1 \cdot 10^{-7}$  K in the case of symmetric pulse, and  $-1.9 \cdot 10^{-7}$  K in the case of asymmetric pulse at  $\Omega = 300$  kHz. The acoustic heating (cooling) excited by a single pulse is hardly expected to be significant. It enhances in the case of series of pulses. Equation (20) is suitable for evaluation of instantaneous heating (cooling) for any waveform including series of pulses.

#### 3.4. Generation of the relaxion mode

Applying the row  $(d_1^3 \quad d_2^3 \quad d_3^3 \quad d_3^4)$  at the system (16) and considering the first sound mode as dominative, one readily obtains the leading-order equation which governs an excess density in the relaxation mode

$$\frac{\partial \rho_3}{\partial t} = -\frac{(\gamma - 1)T_0}{c^2} l(x, t)$$

$$+ \frac{(\gamma - 1)T_0}{2c^3\tau} \int_{-\infty}^{\infty} \exp\left(-\frac{|x - x'|}{c\tau}\right) l(x', t) dx',$$
(26)

where

$$l(x,t) = -\frac{(\gamma-1)T_0}{4c^3\tau} \left[ 2c\Phi_1(\gamma+1)\tau \right]$$
$$\int_x^\infty \exp\left(\frac{x-x'}{c\tau}\right) v_1(x',t) \frac{\partial v_1}{\partial x'} dx'$$
$$+ v_1 \left( 4\Phi_1\left((\gamma-1)T_0\left(\frac{1}{\tau}\frac{d\tau}{dT}\right)_0 - 1\right) \right]$$
$$\int_x^\infty \exp\left(\frac{x-x'}{c\tau}\right) v_1(x',t) dx'$$
$$+ c\tau \left( 4\Phi_2(\gamma-1) + \Phi_1(\gamma+3) v_1 \right) \right].$$

The average acoustic source in the case of harmonic (21) or nearly harmonic velocity in the sound wave equals zero at any sound frequency. That is, efficiency of production of

perturbation in the relaxation mode by this kind of sound is insignificant.

The results may be generalized in the case of relaxation time  $\tau$  which depends not just on temperature, but also on density. In the case of  $\tau(T, \rho)$ , dynamic equations for sound and entropy modes (Equations 19, 20) are valid with the rearranged  $\Phi_1$  [10]:

$$\Phi_1 = \left(\frac{C_v}{\tau} + \frac{\varepsilon - \varepsilon_{eq}}{\tau^2} \left(\frac{\mathrm{d}\tau}{\mathrm{d}T} + \frac{\rho}{T(\gamma - 1)}\frac{\mathrm{d}\tau}{\mathrm{d}\rho}\right)\right)_0$$

As for  $\Phi_2$ , it also should be rearranged in view of dependence of  $\tau$  on  $\rho$ . Equations (19),(20) do not contain  $\Phi_2$  at all. Perturbation of density specifying the relaxation mode, is described by Equation (26), which includes  $\Phi_2$  by means of l(x, t). Production of the relaxation mode by periodic or nearly periodic sound is weak independently on the form of l(x, t). Dependence of  $\tau$  on both *T* and  $\rho$  is taken into account in all evaluations in this study which concern CO<sub>2</sub> : N<sub>2</sub> : He = 1 : 2 : 3.

# **3.5.** Inclusion of viscosity, thermal conductivity, and dependence of power *I* and heat withdrawal *Q* on temperature

We consider only temperature-dependent power and heat withdrawal in this section, although they may also depend on density. Newtonian attenuation alone always leads to attenuation of sound and heating of a medium, and the analysis depends on balance of degree of non-equilibrium, newtonian attenuation and that due to thermal conduction. Taking into account thermal conductivity and shear and bulk viscosity results in corrections in the dispersion relations. Two acoustic ones are

$$\omega_{1} = ck - \frac{(\gamma - 1)^{2}T_{0}k\tau}{2c(1 + ick\tau)}\Phi_{1} - \frac{i(\gamma - 1)^{2}T_{0}}{2c^{2}(1 + ick\tau)}I_{T} + \frac{i(\gamma - 1)^{2}T_{0}}{2c^{2}}Q_{T} + \frac{ibk^{2}}{2\rho_{0}}, \omega_{2} = -ck + \frac{(\gamma - 1)^{2}T_{0}k\tau}{2c(1 - ick\tau)}\Phi_{1} - \frac{i(\gamma - 1)^{2}T_{0}}{2c^{2}(1 - ick\tau)}I_{T} + \frac{i(\gamma - 1)^{2}T_{0}}{2c^{2}}Q_{T} + \frac{ibk^{2}}{2\rho_{0}},$$
(27)

where  $I_T = (dI/dT)_0$ ,  $Q_T = (dQ/dT)_0$ , and b denotes the total attenuation, which depends on shear viscosity  $\eta$ , bulk viscosity  $\zeta$  and thermal conduction of a gas,  $\chi$ ,

$$b = \zeta + 4/3\eta + \chi \frac{(\gamma - 1)^2 \mu}{\gamma R}.$$

Acoustic harmonic wave of frequency  $\Omega$  is damped in the non-equilibrium regime if the standard attenuation is large enough. This is expressed by inequality

$$\frac{b\Omega^2}{2\rho_0} + \frac{(\gamma - 1)^2 T_0}{2} \left( \frac{\Omega^2 \tau^2}{1 + \Omega^2 \tau^2} \Phi_1 - \frac{I_T}{1 + \Omega^2 \tau^2} + Q_T \right) > 0.$$
(28)

It is remarkable that while newtonian attenuation of sound depends on frequency, as well as attenuation and dispersion due to degree of non-equilibrium,  $\Phi_1$ , and that due to

non-zero  $I_T$ , the part of attenuation due to non-zero  $Q_T$  is frequency independent. If we neglect  $I_T$  and  $Q_T$ , the condition of acoustical activity takes the form

$$\Phi_1 < -\frac{b(1+\Omega^2\tau^2)}{(\gamma-1)^2\rho_0 T_0\tau^2}$$

instead of  $\Phi_1 < 0$ . One may easily evaluate variation in the threshold pumping intensity  $I_{th}$  for the typical laser mixture CO<sub>2</sub> : N<sub>2</sub> : He = 1 : 2 : 3 at normal conditions  $p_0 = 1$  atm, T = 300 K, which is in a frictionless gas equals  $I_{th} = 1.99 \cdot 10^6$  W/kg.

The dependence of the relaxation time  $\tau$  on temperature and density is as

$$\tau = 10^{-7} \frac{\mu}{\rho} \left[ 0.22 \exp\left( -62.75T^{-1/3} \right) + 0.99 \exp\left( -75.46T^{-1/3} \right) + 0.55 \cdot 10^{-2} \sqrt{T} \exp\left( -58.82T^{-1/3} \right) \right]^{-1} (29)$$

where  $\tau$  is measured in seconds,  $\mu = 0.019$  in  $kg \cdot mol^{-1}$ , *T* in Kelvins,  $\rho$  in  $kg \cdot m^{-3}$  [5]. That gives approximately equal values of terms  $(T/\tau)(\partial \tau/\partial T) = -3.4$  and  $\rho/[(\gamma - 1)\tau](\partial \tau/\partial \rho) = -3$ . Taking into account thermal conductivity results in the threshold intensity which depends on the sound frequency,  $I_{th} = (2.00 + 0.08\Omega^2\tau^2) \cdot 10^6$  W/kg. The impact of viscosity is important in the case of highfrequency sound. The threshold intensity doubles at frequency 287 kHz due to taking viscosity into account.

The dispersion relations for the non-wave modes are

$$\omega_{3} = i \left( \frac{1}{\tau} + \frac{T_{0}(\gamma - 1)(\gamma + c^{2}k^{2}\tau^{2})}{c^{2}(1 + c^{2}k^{2}\tau^{2})} I_{T} \right), \quad (30)$$
  
$$\omega_{4} = i \left( \frac{\chi k \mu(\gamma - 1)}{\gamma R \rho_{0}} + \frac{(\gamma - 1)T_{0}}{c^{2}} (Q_{T} - I_{T}) \right).$$

Equations (27), (30) are valid up to terms proportional to  $Q_T^1$  and  $I_T^1$ . The linear modes and projectors take the new form. Links for the rightwards progressive sound are

$$\begin{split} \psi_{1,l} &= \begin{pmatrix} v_1(x,t) \\ p_1(x,t) \\ \rho_1(x,t) \\ \varepsilon_1(x,t) \end{pmatrix} \tag{31} \\ &= \begin{pmatrix} 1 \\ \left[ \rho_0 c - \frac{(\gamma-1)^2 T_0 \rho_0}{2c^2} (\Phi_1 + I_T) \int_x^\infty \exp\left(\frac{x-x'}{c\tau}\right) dx' \\ - \frac{(\gamma-1)^2 \rho_0 T_0 Q_T}{2c^2} \int_x^\infty dx' \cdot + \frac{\chi(\gamma-1)^2}{\gamma R \mu} \frac{\partial}{\partial x} \right] \\ \left[ \frac{\rho_0}{c} + \frac{(\gamma-1)^2 T_0 \rho_0}{2c^4} (\Phi_1 + I_T) \int_x^\infty \exp\left(\frac{x-x'}{c\tau}\right) dx' \\ + \frac{(\gamma-1)^2 \rho_0 T_0 Q_T}{2c^4} \int_x^\infty dx' \cdot + \frac{b}{2c^2} \frac{\partial}{\partial x} \right] \\ \left[ \frac{(\gamma-1) T_0}{c^2} (\Phi_1 + I_T) \int_x^\infty \exp\left(\frac{x-x'}{c\tau}\right) dx' \cdot \right] \end{split}$$

In accordance to the dispersion relations (27), the linear governing equation for the excess density in the first sound mode, takes the form

$$\frac{\partial \rho_1}{\partial t} + c \frac{\partial \rho_1}{\partial x} + \frac{(\gamma - 1)^2 T_0}{2c^2} (\Phi_1 + Q_T) \rho_1 - \frac{b}{2c^2 \rho_0} \frac{\partial^2 \rho_1}{\partial x^2} - \frac{(\gamma - 1)^2 T_0}{2c^3 \tau} (\Phi_1 + I_T) \int_x^\infty \exp\left(\frac{x - x'}{c\tau}\right) \rho_1(x', t) \, \mathrm{d}x' = 0.$$
(32)

The linear equation for an excess density which specifies the thermal mode, is

$$\frac{\partial \rho_4}{\partial t} - \frac{\chi(\gamma-1)\mu}{\gamma R \rho_0} \frac{\partial^2 \rho_4}{\partial x^2} + \frac{(\gamma-1)(Q_T - I_T)T_0}{c^2} \rho_4 = 0.$$

The nonlinear terms which account for nonlinear interaction of modes, includes expressions proportional to  $Q_T$ and  $I_T$  but also terms proportional to  $Q_{TT} = (d^2Q/dT^2)_0$ and  $I_{TT} = (d^2I/dT^2)_0$ . The leading-order averaged form of the dynamic equation of heating which considers effects due to relaxation exclusively, and referes to the periodic sound (21), is

$$\frac{1}{T_0} \left( \frac{\partial T_4}{\partial t} + \frac{(\gamma - 1)T_0(Q_T - I_T)}{c^2} T_4 \right) = \overline{F_{\text{heat}}} 
= \frac{V_0^2(\gamma - 1)^3 \Omega^2 \tau^2 T_0 \Phi_1}{2c^4 (1 + \Omega^2 \tau^2)} + \frac{V_0^2(2\gamma - 3)(\gamma - 1)^2 T_0 Q_T}{4c^4} 
+ \frac{V_0^2(\gamma - 1)^2 (5 - \gamma + (3\gamma + 1)\Omega^2 \tau^2) T_0 I_T}{8c^4 (1 + \Omega^2 \tau^2)} 
+ \frac{V_0^2(\gamma - 1)^3 T_0^2}{4c^4} (Q_{TT} + I_{TT}).$$
(33)

Equation (33) with constant acoustic force if  $Q_T \neq I_T$ , has a solution

$$T_{4} = \frac{c^{2}}{(\gamma - 1)(Q_{T} - I_{T})}\overline{F_{\text{heat}}}$$

$$\cdot \left[1 - \exp\left(-\frac{(\gamma - 1)T_{0}(Q_{T} - I_{T})t}{c^{2}}\right)\right],$$
(34)

which satisfies zero initial condition, that is, an excess temperature specific for the entropy mode achieves positive maximum with time if  $\overline{F_{heat}} > 0$ , and negative minimum otherwise. Taking into account for temperaturedependent Q and T yields in the diffusion equation for  $T_4$ . That imposes better extension of entropy perturbations in the space. The coefficient of diffusion may be negative. If  $Q_T = I_T$ , Equation (33) is readily integrated. The part of acoustic force which associates with  $\Phi_1$ , is always positive for positive  $\Phi_1$  and dependeds  $\Omega \tau$ , but this one which associates with  $Q_T$ ,  $Q_{TT}$ ,  $I_{TT}$  does not depend on  $\Omega \tau$  at all. We drop the terms proportional to product of the first and second derivatives of Q and I with respect to temperature in Equation (33).

### 4. Concluding remarks

The main result of this study is the equation governing an excess density which specifies the thermal mode, Equation

(20). Perturbations in density and temperature of the thermal mode vary due to nonlinear interaction with sound. This interaction is anomalous in acoustically active gas. Equation (20) is instantaneous and consequently valid at any time for any type of acoustic excitation.

The only limitations are small nonlinearity  $M \ll 1$ , and conditions of weak attenuation or amplification of sound over its period, determined by Equation (11).

This last inequality also corresponds to small gradients of the background parameters of a gas, which we set constants in this study. The non-uniformity of the background may play important role in the evaluations of domain of equilibrium.

Reference [21] studies amplification of sound in a flat layer of the non-equilibrium gas. It has revealed some new properties, as compared to the case of the uniform gas. In particular, the domain of instability in the plane pumping intensity - an inverse time of relaxation becomes smaller. We consider unbounded volumes of a relaxing gas. The boundary conditions in close volumes lead to a discrete spectrum of sound wavenumbers and frequencies.

The standard thermoviscosity always results in sound attenuation and heating in a nonlinear fluid flow. It enlarges the threshold power of pumping  $I_{th}$  making it dependent on sound frequency. The results may be useful in evaluations of variations in the background temperature and density which reflect the nonlinear phenomena of sound. The domains with residual excess temperature form thermal lenses that in turn have impact on the sound propagation. Finally, the results may be useful for evaluations of degree of non-equilibrium, threshold power, sound intensity and viscosity of a gas by means of acoustic observations.

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