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Generation of vorticity motion by sound in a chemically reacting gas and inversion of acoustic streaming in the non-equilibrium regime

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Abstract: Nonlinear stimulation of the vorticity mode caused by losses in the momentum of sound in a chemically reacting gas is considered. The instantaneous dynamic equation for the vorticity mode is derived. It includes a quadratic nonlinear acoustic source, which reflects the fact that the reason for the interaction between sound and the vorticity mode is nonlinear. Both periodic and aperiodic sound may be considered as the origin of the vorticity flow. The equation governing the mean flow (the acoustic streaming) in the field of periodic sound is also derived. In the non-equilibrium regime of a chemical reaction, there may exist streaming vortices whose direction of rotation is opposite to that of the vortices in the standard thermoviscous flows. For periodic sound, this is illustrated by an example. The theory and the example describe both equilibrium and non-equilibrium chemical reactions.

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1. Introduction, basic equations, and starting points

The reason for vorticity flow in the field of sound is the nonlinear loss in the momentum of the sound wave. An acoustic source periodic in time can generate mean motion, a phenomenon known as acoustic streaming. There are extensive reviews on this subject [1–3]. Although acoustic streaming has been studied in detail both theoretically and experimentally for over a century, the spatial and temporal distribution of the radiation force as a function of diffraction, absorption, and the geometry of the flow is still poorly understood. The main difficulty is the nonlinear origin of the phenomenon. The second origin of acoustic and non-acoustic mode interactions is absorption. It is evident that any relaxation process contributes to the vortices caused by sound and in particular to the streaming. Relaxation processes of different types manifest themselves, among other attributes, through absorption.

The interest in relaxation phenomena, and especially in

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non–equilibrium ones in the physics of gases, primarily originated from observations of anomalous dispersion and absorption of ultrasonics waves in a gas with excited non–equilibrium internal degrees of freedom. The reason for these anomalies is the mechanism of retarded energy exchange between the internal and translational degrees of freedom of the molecules [4, 5]. Intensive investigations of the flow of thermodynamically non–equilibrium gases began in connection with advances in laser engineering and plasma aerodynamics in the 1960s. The hydrodynamics of the non–equilibrium fluids remains one of the new and quickly developing fields of modern hydrodynamics. Studies in this field are passing through a stage of revealing new physical effects and the formulation of fundamental conclusions. The anomalous dispersion and amplification of sound becomes a physical reality in gases out of thermal equilibrium and in gases where irreversible reactions occur [5, 6]. The peculiarities of sound propagation in gases where a chemical reaction takes place, look similar to those in gases with excited internal degrees of freedom. The nonlinear interaction of sound with non–acoustic types of motion in relaxing fluids is presently poorly analyzed. It was first pointed out by Molevich that acoustic heating (i.e., the generation of a thermal mode) or streaming may be reversed in a vibrationally excited molecular gas with negative second viscosity [7]. The standard attenuation itself always leads to a positive excess temperature associated with the thermal mode and to streaming whose velocity in an unbounded volume is directed accordingly along the direction of sound [2, 8].

We start from the linear determination of modes as specific types of gas motion in a gas where a chemical reaction takes place (Sec. 2). The definition of any mode fixes the relations for the dynamic perturbations belonging to this mode. This is necessary for the correct decomposition of equations governing sound and non–acoustic modes accounting for the interaction of all modes in a weakly nonlinear flow (Sec. 3). This also resolves an existing inconsistency in the traditional theory of streaming. While compressibility is a necessary condition for sound propagation, the traditional analysis is limited to incompressible fluids (for a discussion of this topic, see [9, 10]). This allows the elimination of the equation for energy balance and the derivation of results by averaging the equations of continuity and momentum with respect to the period of sound. The traditional analysis is valid only for periodic sound because it requires the averaged partial derivative of every perturbation with respect to time to be zero [2, 8]. It is then impossible to consider the thermal mode, and it requires verification in the case of gases that are strongly compressible. The decomposition of modes based on instantaneous relations of perturbations specific for every

mode gives the possibility to decompose also all equations governing different modes in a weakly nonlinear flow. Where necessary, we will use the asymptotic methods of nonlinear acoustics, which are based on the presence of parameters of relatively small magnitude. Consequently, under some assumptions the equations can be simplified. For example, the diminutive parameter M (the Mach number) and the weak diffraction of the sound beam μ will be employed along with some other parameters connected with chemical reactions. They will be introduced in the following sections.

The present study considers the simplest model of gas dynamics in which a chemical reaction of the type $A \rightarrow B$ takes place. This model can also be used for the description of acoustical properties of reacting media with complex branching reactions [11]. The system is governed by the two equations for momentum and energy as well as by the continuity equation:

$$\rho \frac{d\vec{v}}{dt} = -\vec{\nabla}P,$$
$$\frac{C_{V,\infty}}{R} \frac{dT}{dt} - \frac{T}{\rho} \frac{d\rho}{dt} = Q, \qquad (1)$$
$$\frac{d\rho}{dt} + \rho(\vec{\nabla} \cdot \vec{v}) = 0.$$

In the equations above, $\vec{v}$ denotes the gas velocity, ρ and P are the density and the pressure of the gas, T is the temperature measured in Joules per molecule (actually the ordinary temperature multiplied by the Boltsmann constant k_B), $C_{V,\infty}$ and $C_{P,\infty}$ are the "frozen" heat capacities at constant volume and constant pressure, respectively (i.e., the corresponding processes take place at infinitely high frequencies), $R = C_{P,\infty} - C_{V,\infty}$ is the universal gas constant, $Q = \frac{HmW}{\rho}$ represents the heat produced in a medium per molecule due to a chemical reaction (W is the volume rate of formation of the reaction product B, H denotes the reaction enthalpy per unit mass of the reagent A, and m denotes the averaged molecular mass). The relaxation equation for the mass fraction Y of reagent A and the equation of state complement the system (1):

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

Equations (1) do not account for the standard attenuation due to shear viscosity and thermal conductivity. A brief discussion of how to include standard attenuation in the initial equations is given in the concluding remarks below.

2. Dispersion relations and motions of infinitely small amplitude and their decomposition

2.1. Dispersion relations

Let us consider a two-dimensional gas flow of infinitely small amplitude in the plane OXY. Every quantity ε represents the sum of an unperturbed value ε_0 and its variation ε', where $|\varepsilon'| \ll |\varepsilon_0|$. Following Molevich [7, 12], we assume that the stationary quantities Y_0, T_0, P_0, and ρ_0 are maintained by transverse pumping, so that the background is homogeneous in the longitudinal direction perpendicular to the plane OXY. The equations for momentum, energy, mass-fraction balance, and continuity read:

$$\frac{\partial v_x'}{\partial t} + \frac{1}{\rho_0}\frac{\partial P'}{\partial x} \equiv \frac{\partial v_x'}{\partial t} + \frac{T_0}{m\rho_0}\frac{\partial \rho'}{\partial x} + \frac{1}{m}\frac{\partial T'}{\partial x} = 0,$$

$$\frac{\partial v_y'}{\partial t} + \frac{1}{\rho_0}\frac{\partial P'}{\partial y} \equiv \frac{\partial v_y'}{\partial t} + \frac{T_0}{m\rho_0}\frac{\partial \rho'}{\partial y} + \frac{1}{m}\frac{\partial T'}{\partial y} = 0,$$

$$\frac{\partial T'}{\partial t} + (\gamma_\infty - 1)\left(T_0 \frac{\partial v_x'}{\partial x} + T_0 \frac{\partial v_y'}{\partial y} - Q_T \frac{Q_0}{T_0} T' \right.$$
$$\left. - Q_\rho \frac{Q_0}{\rho_0}\rho' - Q_Y \frac{Q_0}{Y_0} Y' \right) = 0,$$

$$\frac{\partial Y'}{\partial t} + \frac{1}{Hm}\left(Q_T \frac{Q_0}{T_0} T' + Q_\rho \frac{Q_0}{\rho_0}\rho' + Q_Y \frac{Q_0}{Y_0} Y' \right) = 0,$$

$$\frac{\partial \rho'}{\partial t} + \rho_0 \left(\frac{\partial v_x'}{\partial x} + \frac{\partial v_y'}{\partial y} \right) = 0,$$

$$(3)$$

where

$$\gamma_\infty = \frac{C_{P,\infty}}{C_{V,\infty}}$$

denotes the frozen adiabatic exponent, and the quantities Q_T, Q_ρ, Q_Y are evaluated in the equilibrium state:

$$Q_T = \frac{T_0}{Q_0}\left(\frac{\partial Q}{\partial T} \right)_{T_0,\rho_0,Y_0},$$

$$Q_\rho = \frac{\rho_0}{Q_0}\left(\frac{\partial Q}{\partial \rho} \right)_{T_0,\rho_0,Y_0},$$

$$Q_Y = \frac{Y_0}{Q_0}\left(\frac{\partial Q}{\partial Y} \right)_{T_0,\rho_0,Y_0}.$$

$$(4)$$

In the first two equations in (4), the excess pressure is expressed in terms of the excess density and temperature in accordance with the equation of state (i.e., the second equation in (2)). Studies of motion of infinitely small amplitude usually begin by representing all perturbations as planar waves:

$$\varepsilon'(x, y, t) = \tilde{\varepsilon}(k_x, k_y)\exp\left[i(\omega t - k_x x - k_y y)\right]. \quad (5)$$

Some intermediate steps are necessary to determine the heat capacity under constant pressure, C_P, and under constant volume, C_V. Both of these quantities depend on the frequency ω,

$$C_P = C_{P,\infty} + mHR\left(\frac{\partial Y}{\partial T} \right)_P,$$

$$C_V = C_{V,\infty} + mHR\left(\frac{\partial Y}{\partial T} \right)_V.$$

$$(6)$$

They enter the dispersion equation, whose roots determine all possible types of motion in a reacting gas. From the fourth equation in system (3), the following equalities arise:

$$\left(\frac{\partial Y}{\partial T} \right)_V = -\frac{Q_T}{Q_Y(1 + i\omega\tau_c)}\frac{Y_0}{T_0},$$

$$\left(\frac{\partial Y}{\partial \rho} \right)_T = -\frac{1}{\rho_0^2}\left(\frac{\partial Y}{\partial V} \right)_T = -\frac{Q_\rho}{Q_Y(1 + i\omega\tau_c)}\frac{Y_0}{\rho_0},$$

$$(7)$$

where $V = \frac{1}{\rho}$ is the specific gas volume, and

$$\tau_c = \frac{HmY_0}{Q_0 Q_Y} \quad (8)$$

is the characteristic duration of the chemical reaction. The equation of state (i.e., the second equation in (2)) along with the thermodynamic equality

$$\left(\frac{\partial Y}{\partial T} \right)_P = \left(\frac{\partial Y}{\partial T} \right)_V + \left(\frac{\partial Y}{\partial V} \right)_T \left(\frac{\partial V}{\partial T} \right)_P, \quad (9)$$

result in the following expression

$$\left(\frac{\partial Y}{\partial T} \right)_P = \frac{(Q_\rho - Q_T)}{Q_Y(1 + i\omega\tau_c)}\frac{Y_0}{T_0}. \quad (10)$$

The dispersion equation determining two acoustic (wave) types of motion and three non-wave ones then takes the form:

$$\omega^2 \left(\omega^3 - i\frac{C_{V,0}}{C_{V,\infty}\tau_c}\omega^2 - c_\infty^2 \tilde{\Delta}\omega + i\frac{C_{P,0}}{C_{P,\infty}\tau_c}c_\infty^2 \tilde{\Delta} \right) = 0,$$

$$(11)$$

where $\tilde{\Delta} = k_x^2 + k_y^2$, $c_\infty = \sqrt{\gamma_\infty \frac{T_0}{m}}$ is the frozen linear sound velocity (i.e., the one for sound of infinitely large frequency as compared with the inverse time of the chemical reaction, and infinitely small magnitude), and $C_{P,0}$ and $C_{V,0}$ denote the low-frequency heat capacities

$$C_{P,0} = C_{V,\infty}\left(\gamma_\infty + \frac{(\gamma_\infty - 1)Q_0\tau_c(Q_\rho - Q_T)}{T_0} \right),$$

$$C_{V,0} = C_{V,\infty}\left(1 - \frac{(\gamma_\infty - 1)Q_0\tau_c Q_T}{T_0} \right).$$

$$(12)$$

The approximate roots of the dispersion equation for acoustic branches in one dimension were first derived and adequately studied by Molevich [12]. There are five dispersion relations in two-dimensional flow: two acoustic modes indexed by 1 and 2 and three non-wave ones. The third non-acoustic root describes the relaxation due to the chemical reaction; its approximate value depends on the spatial scale of the perturbation. The last two roots (the fourth denoting the thermal mode and the fifth denoting the vorticity mode) equal zero,

$$\omega_4 = 0, \quad \omega_5 = 0. \tag{13}$$

The vorticity mode appears as one of various possible types of motions in flows exceeding one dimension. As for the two branches of sound, their dispersion relations depend on the ratio between the sound period and the characteristic duration of the chemical reaction τ_c.

2.1.1. Dispersion relations for high-frequency sound

The first limiting case pertains to the domain of acoustic frequencies large compared to the inverse duration of the chemical reaction:

$$\frac{1}{|\omega_{1,2}\tau_c|} \approx \frac{1}{|c_\infty\sqrt{\widetilde{\Delta}}\tau_c|} \equiv \delta_\infty \ll 1. \tag{14}$$

The condition below reminds us that sound is a wave process, so that dispersion and attenuation during the sound period are small:

$$|D\delta_\infty| \ll \left|\frac{C_{V,\infty}}{C_{V,0}}\right|, \quad D = \frac{\left(c_\infty^2 - c_0^2\right)}{c_\infty^2}, \tag{15}$$

where D denotes the dispersion, and

$$c_0 = \sqrt{\frac{C_{P,0}T_0}{C_{V,0}m}}$$

is the linear sound velocity at very low frequencies. The inequality (15) is valid if

$$|Q_0[Q_\rho + (\gamma_\infty - 1)Q_T]| \ll \left|\frac{\gamma_\infty T_0}{\delta_\infty(\gamma_\infty - 1)\tau_c}\right|. \tag{16}$$

In view of Eqs. (14) and (15), the leading-order high-frequency acoustic roots of the dispersion equation (11) take the form

$$\begin{aligned} \omega_1 &= c_\infty\sqrt{\widetilde{\Delta}} + i\frac{D}{2}\frac{C_{V,0}}{C_{V,\infty}\tau_c}, \\ \omega_2 &= -c_\infty\sqrt{\widetilde{\Delta}} + i\frac{D}{2}\frac{C_{V,0}}{C_{V,\infty}\tau_c}. \end{aligned} \tag{17}$$

Amplitudes of excess acoustic quantities increase if

$$Q_0[Q_\rho + (\gamma_\infty - 1)Q_T] > 0, \tag{18}$$

and decrease otherwise. The inequality (18) determines the area of irreversibility of the chemical reaction; it also establishes the following inequality [12]:

$$\begin{aligned} c_\infty^2 - c_0^2 &= \frac{T_0}{m}\left(\frac{C_{P,\infty}}{C_{V,\infty}} - \frac{C_{P,0}}{C_{V,0}}\right) \\ &= \frac{(\gamma_\infty - 1)Q_0(Q_\rho + (\gamma_\infty - 1)Q_T)T_0\tau_c}{m(Q_0Q_T\tau_c(\gamma_\infty - 1) - T_0)} < 0, \end{aligned} \tag{19}$$

if $C_{V,0} > 0$. Thus, the sign of D distinguishes between an equilibrium (positive D) and a non-equilibrium irreversible chemical reaction (negative D).

2.1.2. Dispersion relations for low-frequency sound

In the other limiting case,

$$|\omega_{1,2}\tau_c| \approx |c_0\sqrt{\widetilde{\Delta}}\tau_c| \equiv \delta_0 \ll 1, \tag{20}$$

the approximate acoustic roots of Eq. (11) equal

$$\begin{aligned} \omega_1 &= c_0\sqrt{\widetilde{\Delta}} + i\frac{D}{2}\frac{C_{P,\infty}c_0^2\tau_c}{C_{P,0}}\widetilde{\Delta}, \\ \omega_2 &= -c_0\sqrt{\widetilde{\Delta}} + i\frac{D}{2}\frac{C_{P,\infty}c_0^2\tau_c}{C_{P,0}}\widetilde{\Delta}. \end{aligned} \tag{21}$$

The terms responsible for attenuation (amplification) of sound are of second order in the small parameter δ_0. This coincides with the general idea about weak attenuation of low-frequency sound (proportional to its square frequency) in fluids with standard attenuation.

2.1.3. Definition of modes

Substitution of the approximate roots of the dispersion equation in Eqs. (3) readily produces the relationships of the Fourier transforms of the perturbations specific for every mode. The relations for both acoustic branches determined by the roots ω_1 and ω_2 are the following:

$$\tilde{\psi}_i = \begin{pmatrix} \tilde{v}_{x,i} \\ \tilde{v}_{y,i} \\ \tilde{T}'_i \\ \tilde{Y}'_i \\ \tilde{\rho}'_i \end{pmatrix} = \begin{pmatrix} \frac{\omega_i k_x}{\Delta} \\ \frac{\omega_i k_y}{\Delta} \\ T_0\left(\gamma_\infty - 1 - (\gamma_\infty - \gamma_0)\widehat{R}\right) \\ \frac{c_\infty^2 - c_0^2}{H(\gamma_\infty - 1)}\widehat{R} \\ \rho_0 \end{pmatrix} \frac{\tilde{\rho}'_i}{\rho_0}, \tag{22}$$

where $i = 1, 2$, and $\widehat{R}$ denotes an integral operator acting on scalar functions $\phi(x, y, t)$ as follows:

$$\widehat{R}\phi = \frac{C_{V,0}}{C_{V,\infty}\tau_c} \int_{-\infty}^{t} \phi e^{-(t-t')\frac{C_{V,0}}{C_{V,\infty}\tau_c}} dt'. \qquad (23)$$

The relaxation modes are derived for the two limiting cases, high frequency ($\delta_\infty \ll 1$) and low frequency ($\delta_0 \ll 1$), i.e., we will utilize these limits to simplify calculations of the stimulation of non-acoustic modes in the field of sound. We still use the nomenclature "high frequency" and "low frequency," although they actually refer to the regimes $c_\infty\sqrt{\widetilde{\Delta}}\tau_c \ll 1$ and $c_0\sqrt{\widetilde{\Delta}}\tau_c \ll 1$:

$$\tilde{\psi}_{3,h} = \begin{pmatrix} \tilde{v}_{x,3} \\ \tilde{v}_{y,3} \\ \tilde{T}'_3 \\ \tilde{Y}'_3 \\ \tilde{\rho}'_3 \end{pmatrix}_h = \begin{pmatrix} \frac{\omega_{3,h} k_x}{\Delta} \\ \frac{\omega_{3,h} k_y}{\Delta} \\ -T_0 \\ \frac{c_\infty^2}{H(\gamma_\infty - 1)} \\ \rho_0 \end{pmatrix} \frac{\tilde{\rho}'_3}{\rho_0},$$

$$\tilde{\psi}_{3,l} = \begin{pmatrix} \frac{\omega_{3,l} k_x}{\widetilde{\Delta}} \\ \frac{\omega_{3,l} k_y}{\widetilde{\Delta}} \\ -T_0 - \frac{m}{\widetilde{\Delta}}\left(\frac{C_{V,0}}{C_{V,\infty}\tau_c}\right)^2 \\ \frac{c_\infty^2}{H(\gamma_\infty - 1)} + \frac{1}{H(\gamma_\infty-1)\widetilde{\Delta}}\left(\frac{C_{V,0}}{C_{V,\infty}\tau_c}\right)^2 \\ \rho_0 \end{pmatrix} \frac{\tilde{\rho}'_3}{\rho_0}, \qquad (24)$$

$$\omega_{3,h} = i\frac{C_{P,0}}{C_{P,\infty}\tau_c},$$

$$\omega_{3,l} = i\frac{C_{V,0}}{C_{V,\infty}\tau_c}.$$

The relations for the perturbations in the thermal mode are free of partial derivatives with respect to coordinates and therefore neither refer to the low–frequency nor to the high–frequency regime,

$$\tilde{\psi}_4 = \begin{pmatrix} \tilde{v}_{x,4} \\ \tilde{v}_{y,4} \\ \tilde{T}'_4 \\ \tilde{Y}'_4 \\ \tilde{\rho}'_4 \end{pmatrix} = \begin{pmatrix} 0 \\ 0 \\ -T_0 \\ -\frac{c_\infty^2\left(\frac{C_{P,0}}{C_{P,\infty}}-1\right)}{H(\gamma_\infty-1)} \\ \rho_0 \end{pmatrix} \frac{\tilde{\rho}'_4}{\rho_0}. \qquad (25)$$

The same applies also to the vorticity mode. The latter is determined by the following relationships:

$$\vec{\nabla} \cdot \vec{v}_5 = 0, \quad \tilde{T}'_5 = 0, \quad \tilde{Y}'_5 = 0, \quad \tilde{\rho}'_5 = 0. \qquad (26)$$

The velocity field of the two sound modes and that of the third and fourth modes are potential: $\vec{\nabla} \times \vec{v}_n = \vec{0}$, $n = 1, \ldots, 4$, and the last mode is rotational in accordance with Eqs. (26). The overall linear velocity is a sum of all the individual parts:

$$\vec{v} = \sum_{n=1}^{5} \vec{v}_n. \qquad (27)$$

The linear flow may be uniquely decomposed into its individual modes at any time. This may be achieved by the use of a set of matrix projectors. The matrix projectors were derived and exploited by one of the authors in some problems of nonlinear hydrodynamics in media with standard absorption [13, 14]. For example, in order to extract the vorticity part from the overall velocity-vector field, it is sufficient to apply the operator $\tilde{P}_v$ to the vector of the Fourier transforms of the velocity components:

$$\tilde{P}_v \begin{pmatrix} \tilde{v}_x \\ \tilde{v}_y \end{pmatrix} = \frac{1}{\widetilde{\Delta}} \begin{pmatrix} k_y^2 & -k_x k_y \\ -k_x k_y & k_x^2 \end{pmatrix} \begin{pmatrix} \tilde{v}_x \\ \tilde{v}_y \end{pmatrix}$$

$$= \begin{pmatrix} \tilde{v}_{x,5} \\ \tilde{v}_{y,5} \end{pmatrix}. \qquad (28)$$

P_v, operating in (x, y) space, satisfies the equality

$$P_v\Delta = \begin{pmatrix} \frac{\partial^2}{\partial y^2} & -\frac{\partial^2}{\partial x \partial y} \\ -\frac{\partial^2}{\partial x \partial y} & \frac{\partial^2}{\partial x^2} \end{pmatrix}, \qquad (29)$$

where $\Delta = \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}$ denotes the Laplacian operating in (x, y) space.

2.2. The quasi-planar sound, relations for perturbations and dynamic equations

In order to simplify the mathematical context and to focus on the physically interesting case of quasi–planar sound propagating in the direction of the OX axis, let us assume that all acoustic perturbations vary much faster in the direction of the OX axis than along OY: $k_x \gg k_y$. This allows us to expand the relations for sound perturbations in powers of the small parameter

$$\mu = \frac{k_y^2}{k_x^2}.$$

For propagation in the positive OX direction, the leading-order relationships in the high–frequency and low–frequency limits take the form:

$$
\begin{pmatrix} v'_{x,1}(x,y,t) \\ v'_{y,1}(x,y,t) \\ T'_1(x,t) \\ Y'_1(x,t) \\ \rho'_1(x,t) \end{pmatrix}_h = \begin{pmatrix} c_\infty + \frac{c_\infty}{2}\frac{\partial^2}{\partial y^2}\int dx \int dx + \frac{D}{2}\frac{C_{V,0}}{C_{V,\infty}\tau_c}\int dx \\ c_\infty \frac{\partial}{\partial y}\int dx \\ (\gamma_\infty - 1)T_0 - T_0\gamma_\infty D\frac{C_{V,0}}{C_{V,\infty}\tau_c}\int dt \\ \frac{Dc_\infty^2}{H(\gamma_\infty-1)}\frac{C_{V,0}}{C_{V,\infty}\tau_c}\int dt \\ \rho_0 \end{pmatrix}\frac{\rho'_1}{\rho_0},
$$

$$
\begin{pmatrix} v'_{x,1}(x,y,t) \\ v'_{y,1}(x,y,t) \\ T'_1(x,t) \\ Y'_1(x,t) \\ \rho'_1(x,t) \end{pmatrix}_l = \begin{pmatrix} c_0 + \frac{c_0}{2}\frac{\partial^2}{\partial y^2}\int dx \int dx - \frac{D}{2}\frac{C_{P,\infty}c_0^2\tau_c}{C_{P,0}}\frac{\partial}{\partial x} \\ c_0 \frac{\partial}{\partial y}\int dx \\ (\gamma_0 - 1)T_0 + T_0\gamma_0 D\frac{C_{P,\infty}\tau_c}{C_{P,0}}\frac{\partial}{\partial t} \\ \frac{Dc_\infty^2}{H(\gamma_\infty-1)}\left(1 - \frac{C_{V,\infty}\tau_c}{C_{V,0}}\frac{\partial}{\partial t}\right) \\ \rho_0 \end{pmatrix}\frac{\rho'_1}{\rho_0}.
$$

(30)

Only terms up to linear order in μ have been kept. We consider the small parameters μ, δ_0, δ_∞, and M of the same order. In view of relations (30), the leading–order equations governing the acoustic excess density of sound propagating in the positive OX direction are

$$
\begin{aligned}
\frac{\partial \rho'_1}{\partial t} + c_\infty \frac{\partial \rho'_1}{\partial x} + \frac{c_\infty}{2}\frac{\partial^2}{\partial y^2}\int \rho'_1 dx + \frac{D}{2}\frac{C_{V,0}}{C_{V,\infty}\tau_c}\rho'_1 &= 0 \quad \text{for high frequencies,} \\
\frac{\partial \rho'_1}{\partial t} + c_0 \frac{\partial \rho'_1}{\partial x} + \frac{c_0}{2}\frac{\partial^2}{\partial y^2}\int \rho'_1 dx - \frac{D}{2}\frac{C_{P,\infty}c_0^2\tau_c}{C_{P,0}}\frac{\partial^2 \rho'_1}{\partial x^2} &= 0 \quad \text{for low frequencies.}
\end{aligned}
$$

(31)

The propagation equations (31) can not only be determined from Eqs. (30), but also from the dispersion relations (17) and (21) recalling that

$$
\sqrt{\widetilde{\Delta}} = k_x + \frac{1}{2}\frac{k_y^2}{k_x} + \mathcal{O}(\mu^2) \approx k_x + \frac{1}{2}\frac{k_y^2}{k_x}.
$$

3. Equations governing sound and the vorticity mode in a weakly nonlinear flow

3.1. Second-order nonlinear terms in the hydrodynamic system

Because the quadratic nonlinear terms are of importance when studying weakly nonlinear flows, only these terms will be considered. At quadratic order in the nonlinear terms, the governing system (1) together with (2) is in two dimensions readily rearranged into the following system:

$$
\begin{aligned}
\frac{\partial v_x}{\partial t} + \frac{T_0}{m\rho_0}\frac{\partial \rho'}{\partial x} &+ \frac{1}{m}\frac{\partial T'}{\partial x} + (\vec{v}\cdot\vec{\nabla})v_x \\
&- \frac{T_0\rho'}{m\rho_0^2}\frac{\partial \rho'}{\partial x} + \frac{T'}{m\rho_0}\frac{\partial \rho'}{\partial x} = 0, \\
\frac{\partial v_y}{\partial t} + \frac{T_0}{m\rho_0}\frac{\partial \rho'}{\partial y} &+ \frac{1}{m}\frac{\partial T'}{\partial y} + (\vec{v}\cdot\vec{\nabla})v_y \\
&- \frac{T_0\rho'}{m\rho_0^2}\frac{\partial \rho'}{\partial y} + \frac{T'}{m\rho_0}\frac{\partial \rho'}{\partial y} = 0, \\
\frac{\partial T'}{\partial t} + (\gamma_\infty - 1)&\left[(T_0 + T')(\vec{\nabla}\cdot\vec{v}) - Q_T\frac{Q_0}{T_0}T'\right. \\
&\left. - Q_\rho\frac{Q_0}{\rho_0}\rho' - Q_Y\frac{Q_0}{Y_0}Y'\right] + (\vec{v}\cdot\vec{\nabla})T' = 0, \\
\frac{\partial Y'}{\partial t} + \frac{1}{Hm}&\left[Q_T\frac{Q_0}{T_0}T' + Q_\rho\frac{Q_0}{\rho_0}\rho' + Q_Y\frac{Q_0}{Y_0}Y'\right] \\
&+ (\vec{v}\cdot\vec{\nabla})Y' = 0, \\
\frac{\partial \rho'}{\partial t} + \rho_0(\vec{\nabla}\cdot\vec{v}) &+ (\vec{v}\cdot\vec{\nabla})\rho' + \rho'(\vec{\nabla}\cdot\vec{v}) = 0,
\end{aligned}
$$

(32)

where $\vec{\nabla}$ in two dimensions denotes

$$\vec{i}\,\frac{\partial}{\partial x} + \vec{j}\,\frac{\partial}{\partial y}$$

($\vec{i}$ and $\vec{j}$ are the corresponding basis vectors of unit length). We remark that in Eqs. (32) we have disregarded terms involving second–order derivatives of Q, such as

$$\frac{\partial^2 Q}{\partial T^2}.$$

This restricts the accuracy of our conclusions.

3.2. Decomposition of the intense-sound and vorticity-mode equations and acoustic streaming

In studies of weakly nonlinear dynamics, we still work with linear relations of perturbations in accordance with Eqs. (22), (24), (25), and (26), and we will consider every field perturbation as a sum of perturbations of different modes. The main idea is to decompose the equations governing different modes by applying the corresponding projector to the system that includes weakly nonlinear terms like (32) [13, 14]. Every equation includes nonlinear terms of order not lower than M^2 pertaining to all modes and reflecting the nonlinear interactions of modes in a weakly nonlinear flow. The solution of the final dynamic equations depends on the contribution of every mode in the overall field perturbation. Let the propagation of sound in the positive direction be intense in comparison to all other modes. This means that the characteristic amplitude of the velocity associated with the first branch of sound in the considered domain is much greater than that of other modes:

$$\max |v_1| \gg \max |v_n|, \quad n = 2, \dots, 5. \tag{33}$$

We will only keep dominant terms corresponding to sound propagating towards the right in the nonlinear terms in all formulae below. In view of the relations specific for sound, the governing equations for an excess acoustic density ($\rho_a \equiv \rho_1'$) are at leading order given by

$$\frac{\partial \rho_a}{\partial t} + c_\infty \sqrt{\Delta}\rho_a - c_\infty B \rho_a + \frac{1}{2}\left[\gamma_\infty \rho_a(\vec{\nabla} \cdot \vec{v}_a) + (\vec{v}_a \cdot \vec{\nabla})\rho_a\right] = 0,$$

$$\frac{\partial \rho_a}{\partial t} + c_\infty \frac{\partial \rho_a}{\partial x} + \frac{c_\infty}{2}\frac{\partial^2}{\partial y^2}\int \rho_a\, dx - c_\infty B \rho_a + \frac{(\gamma_\infty + 1)c_\infty}{2\rho_0}\frac{\partial \rho_a}{\partial x}\rho_a = 0,$$

$$\frac{\partial \rho_a}{\partial t} + c_0 \sqrt{\Delta}\rho_a - \frac{D}{2}\frac{C_{P,\infty}c_0^2\tau_c}{C_{P,0}}\Delta\rho_a + \frac{1}{2}\left[\gamma_0 \rho_a(\vec{\nabla} \cdot \vec{v}_a) + (\vec{v}_a \cdot \vec{\nabla})\rho_a\right] = 0,$$

$$\frac{\partial \rho_a}{\partial t} + c_0 \frac{\partial \rho_a}{\partial x} + \frac{c_0}{2}\frac{\partial^2}{\partial y^2}\int \rho_a\, dx - \frac{D}{2}\frac{C_{P,\infty}c_0^2\tau_c}{C_{P,0}}\frac{\partial^2 \rho_a}{\partial x^2} + \frac{(\gamma_0 + 1)c_0}{2\rho_0}\frac{\partial \rho_a}{\partial x}\rho_a = 0,$$

$$\tag{34}$$

for the high-frequency, the high–frequency quasi–planar, the low–frequency, and the low–frequency quasi–planar cases, respectively. In these equations, we have abbreviated

$$B = -\frac{D}{2c_\infty}\frac{C_{V,0}}{C_{V,\infty}\tau_c}. \tag{35}$$

In order to decompose the dynamical equation for the velocity of the vorticity mode, it is sufficient to apply the matrix operator P_v, determined by Eq. (29), to the momentum equations (i.e., the first two equations in the system (32)). As a result, all terms corresponding to the potential velocity vector are reduced in the linear part of equations. On the right–hand side, we keep only acoustic terms. Applying of P_v then yields the dynamic equation for the vorticity mode in the field of intense sound in these two equivalent forms:

$$\frac{\partial \vec{\Omega}}{\partial t} = \frac{1}{\rho_0}\vec{\nabla} \times \left(\rho_a \frac{\partial \vec{v}_a}{\partial t}\right), \quad \frac{\partial \vec{v}_v}{\partial t} = \frac{1}{\rho_0}P_v\left(\rho_a \frac{\partial \vec{v}_a}{\partial t}\right). \tag{36}$$

Here, $\vec{\Omega} = \vec{\nabla} \times \vec{v}_v$ is the vorticity of the flow, with $\vec{v}_v$ replacing $\vec{v}_5$. Accounting for Eqs. (22), one finally gets the following equations in terms of $\vec{v}_v$ and $\vec{\Omega}$:

$$\frac{\partial \vec{v}_v}{\partial t} = -\frac{2Bc_\infty^3}{\rho_0^2} P_v \left[(\vec{\nabla}\rho_a) \int \rho_a dt \right], \quad \frac{\partial \vec{\Omega}}{\partial t} = \frac{2Bc_\infty^3}{\rho_0^2} (\vec{\nabla}\rho_a) \times \int \vec{\nabla}\rho_a dt,$$

$$\frac{\partial \vec{v}_v}{\partial t} = \frac{2Bc_\infty^2}{\rho_0^2} P_v \left[(\vec{\nabla}\rho_a) \int \rho_a dx \right], \quad \frac{\partial \vec{\Omega}}{\partial t} = -\frac{2Bc_\infty^2}{\rho_0^2} (\vec{\nabla}\rho_a) \times \int \vec{\nabla}\rho_a dx,$$

$$\frac{\partial \vec{v}_v}{\partial t} = -\frac{Dc_0^2}{\rho_0^2} \frac{C_{P,\infty} \tau_c}{C_{P,0}} P_v \left[(\vec{\nabla}\rho_a) \frac{\partial \rho_a}{\partial t} \right], \quad \frac{\partial \vec{\Omega}}{\partial t} = \frac{Dc_0^2}{\rho_0^2} \frac{C_{P,\infty} \tau_c}{C_{P,0}} (\vec{\nabla}\rho_a) \times \vec{\nabla}\frac{\partial \rho_a}{\partial t},$$

$$\frac{\partial \vec{v}_v}{\partial t} = \frac{Dc_0^3}{\rho_0^2} \frac{C_{P,\infty} \tau_c}{C_{P,0}} P_v \left[(\vec{\nabla}\rho_a) \frac{\partial \rho_a}{\partial x} \right], \quad \frac{\partial \vec{\Omega}}{\partial t} = -\frac{Dc_0^3}{\rho_0^2} \frac{C_{P,\infty} \tau_c}{C_{P,0}} (\vec{\nabla}\rho_a) \times \vec{\nabla}\frac{\partial \rho_a}{\partial x}. \tag{37}$$

Here, the first, second, third, and fourth line refers to the high-frequency, the high-frequency quasi-planar, the low-frequency, and the low-frequency quasi-planar case, respectively. In the evaluation of the equations describing the effects of quasi-planar sound, we have approximated P_v by the leading terms of a power series in μ according to Eq. (28):

$$P_v \approx \begin{pmatrix} \frac{\partial^2}{\partial y^2} \int dx \int dx & -\frac{\partial}{\partial y} \int dx \\ \\ -\frac{\partial}{\partial y} \int dx & 1 \end{pmatrix}. \tag{38}$$

4. The vorticity mode generated by periodic sound

The difficulties in the description of the vorticity mode caused by sound are obviously nonlinearity, absorption, and diffraction in both equations governing sound and the vorticity mode. The solution of the planar version of the second equation in Eqs. (34) for a periodic transducer,

$$\frac{\partial \rho_a}{\partial t} + c_\infty \frac{\partial \rho_a}{\partial x} - c_\infty B \rho_a + \frac{(\gamma_\infty + 1)c_\infty}{2\rho_0} \frac{\partial \rho_a}{\partial x} \rho_a = 0, \tag{39}$$

takes the form [2, 5]:

$$\rho_a(X, \tau) = \rho_A \exp(B_{sh}X)$$
$$\sum_{n=1}^{\infty} \frac{2J_n(nK\{\exp[B_{sh}X] - 1\}) \sin(n\omega\tau)}{nK\{\exp[B_{sh}X] - 1\}}, \tag{40}$$

where $\tau = \frac{t-x}{c_\infty}$ denotes the retarded time, ω is the sound frequency,

$$K = \frac{(\gamma_\infty + 1)\omega\rho_A}{2\rho_0 c_\infty B},$$

$$x_{sh} = \frac{1}{B} \ln\left(1 + \frac{1}{K}\right)$$

is the distance for forming a shock front of sound,

$$X = \frac{x}{x_{sh}}$$

is a dimensionless coordinate, and $B_{sh} = Bx_{sh}$. J_n denotes the Bessel function of order n. The solution (40) accounts for nonlinearity and absorption; it is valid at distances from the transducer within which a saw-like front has not formed yet: $0 \leqslant X < 1$. In the case of a weakly diffracting beam, it can be represented by the following formula:

$$\rho_a(X, Y, \tau) = \rho_A \exp(B_{sh}X - Y^2)$$
$$\sum_{n=1}^{\infty} \frac{2J_n(nK\{\exp[B_{sh}X] - 1\}) \sin(n\omega\tau)}{nK\{\exp[B_{sh}X] - 1\}}. \tag{41}$$

$Y = \frac{y}{L}$ denotes the dimensionless transverse coordinate, where L marks the characteristic transverse width of the sound beam. Substituting the solution (41) into the "high-frequency" equation for Ω (i.e., Eq. (37)), and averaging the equation over one period of sound $\frac{2\pi}{\omega}$, one arrives at a solution for $\langle \vec{\Omega}_t \rangle$, which equals the driving force of the vorticity mode averaged over the sound period

$$\langle \vec{\Omega}_t \rangle = \frac{\omega}{2\pi} \int_t^{t + \frac{2\pi}{\omega}} \frac{\partial \vec{\Omega}}{\partial t} dt$$
$$= \frac{2Bc_\infty^3}{\rho_0^2} \left\langle \left[(\vec{\nabla}\rho_a) \times \int \vec{\nabla}\rho_a dt \right] \right\rangle. \tag{42}$$

Considering averaged quantities reduces the problem of the generation of the vorticity mode to the problem of acoustic streaming in its classic meaning. The first two components $\langle \Omega_{t,x} \rangle$ and $\langle \Omega_{t,y} \rangle$ are zero, and the third one

$\langle \Omega_{t,z} \rangle$ is nonzero:

$$E\langle \Omega_{t,z} \rangle = \frac{8Y \exp[2(B_{sh}X - Y^2)]}{K^2 \{\exp[B_{sh}X] - 1\}} \tag{43}$$
$$\sum_{n=1}^{\infty} \frac{1}{n^2} J_n(nK\{\exp[B_{sh}X] - 1\})^2,$$

where

$$E = \frac{\rho_0^2 L}{\rho_A^2 Dc_\infty} \frac{C_{V,\infty}\tau_c}{C_{V,0}} = -\frac{L}{2M^2 B c_\infty^2}. \tag{44}$$

Figure 1 shows the distribution of the longitudinal force of acoustic streaming along the OX axis. We have plotted $\langle \Omega_{t,z} \rangle$ versus X for different values of K (and thus different B_{sh}) and different values of Y. In the evaluations, only the first five terms in the series (43) were taken into account.
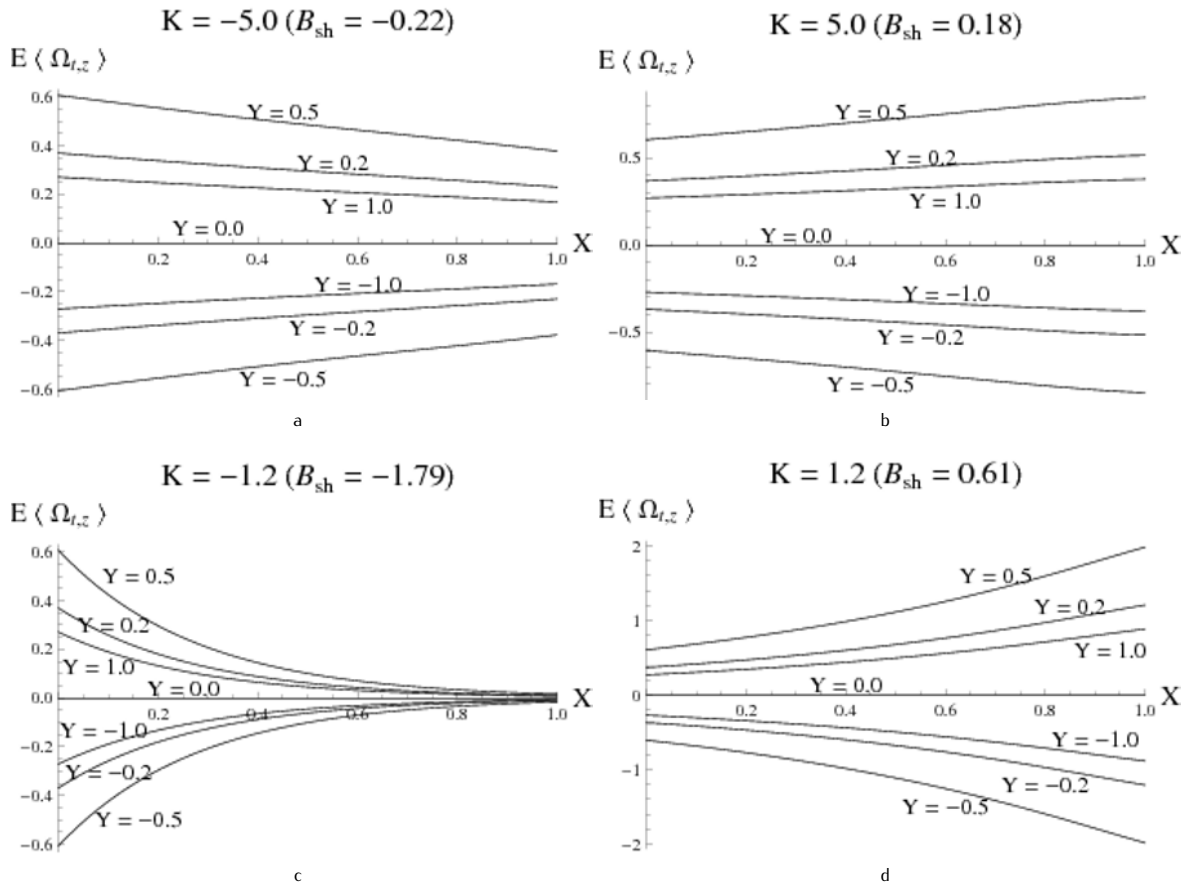

Figure 1. Longitudinal force of acoustic streaming versus X for different transversal distances from the sound beam Y and different K (i.e., different B_{sh}).

The vorticity $\langle \Omega_z \rangle$ depends on time and may be calculated by the use of $\langle \Omega_{t,z} \rangle$ by means of the relation

$$\langle \Omega_z \rangle = \left(t + \frac{\pi}{\omega}\right)\langle \Omega_{t,z} \rangle. \tag{45}$$

The plots in the Fig. 1 show that the sign of the acoustic force of streaming depends on the sign of B, which is apparent from dependence of E on B. Therefore, an inversion of the direction of streamlines occurs in the

equilibrium regime of a chemical reaction (when $B < 0$) as opposed to non-equilibrium chemical reactions (when $B > 0$). This is the main conclusion of this section. For Gaussian beams, the absolute value of the vorticity production per unit time, $|\langle \Omega_{t,z} \rangle|$, is maximal for $Y = 0.5$ and $Y = -0.5$, and the streamlines are symmetric with respect to the beam-propagation axis. The absolute value of the vorticity production in the equilibrium regime of a chemical reaction (when $B < 0$, Figs. 1a and 1c) decreases with the distance from the transducer, otherwise (when $B > 0$, Figs. 1b and 1d) it increases. The rate of enhancement or reduction of the absolute value of the vorticity depends on $|K|$: it is larger for smaller $|K|$ (and thus for larger values of $|B|$). The absolute value of the mean vorticity $|\langle \Omega_z \rangle|$ grows linearly with time for any dimensional coordinates X and Y.

5. Concluding remarks

The objective of this study has been to make possible detailed evaluations of the vorticity-mode generation by sound (in particular, acoustic streaming) in a chemically reacting gas. Acoustic streaming in gases is of greater importance than in liquids; its velocity may achieve dozens of meters per second. The mean flow associated with streaming may transport not only heat perturbations, but also solid or fluid particles. It may be governed remotely by sound. Data on the streaming velocity may be useful for investigations of chemical reactions that take place in a gas. The novelty of this study is the analysis of nonlinear phenomena differing from those in a Newtonian fluid. The equations describe sound and are associated with nonlinear phenomena in a gas with excited (reversibly or not) oscillatory degrees of freedom of molecules [5, 7, 15]. These gases are widely used in lasers. It may be concluded that our results are applicable to some classes of relaxation processes; although conceptually somewhat different, these processes are described by similar equations. The equations derived in this study (Eqs. (37)) describe the nonlinear generation of the vorticity mode that is not necessarily associated with acoustic streaming; they are valid for any sound, including aperiodic, and they are not averaged. A chemically reacting gas is a dispersive medium, so that two limits of sound frequencies (as compared to the inverse duration of the chemical reaction) were considered.

The two-dimensional weakly nonlinear flow of a chemically reacting gas was considered in this study. The inclusion of one more dimension would yield one more vorticity mode and essentially complicate the mathematical content of the study. This would, in fact, not give new

results in comparison with those obtained in the present study. Equations (34) and (36), which describe the dynamics of sound and the vorticity mode, respectively, were derived within an approximation taking terms up to order M^2 into account. The accuracy of our conclusions is restricted because we have neglected second partial derivatives of the heat release $Q(\rho, T, Y)$. An analysis undertaken by the authors has revealed that the inclusion of higher-order derivatives in Q would result in the same equation governing streaming, but would yield corrections to the dynamical equation for the dominant sound mode. Our conclusions are valid in temporally and spatially confined domains, where sound remains dominant with respect to other modes (vorticity, entropy, and chemical).

It is of importance to note that the attenuation (or amplification) of sound considered in this study occurs exclusively due to the presence of a chemical reaction as well as the nonlinear generation of the vorticity mode. In particular, the necessary condition of mode interaction differs from Newtonian viscosity. Moreover, there exists a regime in the chemical reaction, in which sound is amplified during its propagation, and the generation of the vorticity mode is different from that in Newtonian flows. This study does not take into account the thermal and viscous (standard, Newtonian) attenuation of a reacting gas. The terms reflecting these phenomena (they originate from the stress tensor and the energy flux associated with thermal conductivity) should complement the momentum and energy equations in the system (1). The attenuation of low-frequency sound is ignorable. The equation describing the high-frequency excess density in a planar sound wave, which accounts for the standard attenuation, takes the form

$$\frac{\partial \rho_a}{\partial t} + c_\infty \frac{\partial \rho_a}{\partial x} + c_\infty \frac{(\gamma_\infty + 1)}{2\rho_0} \rho_a \frac{\partial \rho_a}{\partial x} - c_\infty B \rho_a - \frac{b}{2} \frac{\partial^2 \rho_a}{\partial x^2} = 0. \tag{46}$$

Here, $b = b_V + b_T$ is the diffusivity of sound, where

$$b_V = \frac{4\eta}{3\rho_0}$$

and

$$b_T = \frac{\kappa \left(\frac{1}{C_{V,\infty}} - \frac{1}{C_{P,\infty}} \right)}{\rho_0}.$$

The quantities η and κ denote the shear viscosity and the thermal conductivity, respectively. The standard attenuation always leads to a linear damping of sound during its propagation. The balance of the last two terms in Eq. (46) determines the *linear* amplification or damping of sound: if, for periodic sound,

$$2c_\infty B - b \frac{\omega^2}{c_\infty^2} < 0,$$

the amplitude of the various sound quantities decrease. Otherwise they increase with time during sound propagation. This is of importance in evaluation of the vorticity mode caused by sound.

An important problem to overcome is the transverse spatial inhomogeneity of the ambient quantities of the gas, which grows simultaneously with increasing heat power Q_0. Taking into account the background inhomogeneity essentially complicates the mathematical analysis, but it may lead to new, physically significant conclusions. The study of the spatial inhomogeneity of a gas with excited internal degrees of freedom has proved that the area of non-equilibrium states of such gas becomes much larger [16, 17]. This may also hold true for a chemically reacting gas.

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