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The nonlinear effects of sound in a liquid with relaxation losses

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Abstract: The nonlinear effects of sound in electrolyte with a chemical reaction are examined. The dynamic equations that govern non-wave modes in the field of intense sound are derived, and acoustic forces of vortex, entropy, and relaxation modes are determined in the cases of low-frequency sound and high-frequency sound. The difference in the nonlinear effects of sound in electrolyte and in a gas with excited vibrational degrees of molecules, are specified and discussed.

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Résumé : Nous examinons les effets non linéaires du son dans un électrolyte avec réaction chimique. Nous dérivons les équations dynamiques qui gouvernent les modes non ondulatoires du champ de son intense et nous déterminons, aux fréquences hautes et basses, les forces acoustiques de vortex, l'entropie et les modes de relaxation. Nous soulignons et analysons la différence entre les effets du son dans un électrolyte et dans un gaz avec des modes vibrationnels d'excitation. [Traduit par la Rédaction]

1. Introduction

Attenuation of fluids is of importance in many applications, notably in medical imaging and tissue characterization. The frequency dependence of attenuation in the most popular pattern, in a Newtonian fluid, does not match the observed frequency dependence of attenuation in a tissue [1, 2]. The model of a Maxwell fluid with a thermodynamic relaxation, is one of the samples with the frequency-dependent linear sound speed and attenuation of sound, and the Newtonian model may be considered as a limit of the Maxwell one at low frequencies. A comprehensive summary concerning various absorption in fluids may be found in ref. 3.

Study of absorption and relaxation in water started when the nature of the relaxation mechanism was not known for certain [4]. From the very beginning, the difference in relaxation of fresh water and sea water was well understood. As a rule, several kinds of relaxation are typical for a liquid. Sound absorption in sea water is dominated by chemical relaxations involving magnesium sulfate at high frequencies (relaxation frequency 100 kHz) and boric acid at low frequencies (relaxation frequency 1 kHz). A third intermediate-frequency relaxation involving magnesium and carbonic acid has been also established [5, 6]. Ultrasound has long been utilized in ocean environments for naval, fishery, and biological purposes, among others. The experimental and theoretical study of sound propagation in water, aqueous solutions, and other liquids, including electrolytes, makes it possible to analyze the physical origin of the relaxation processes. That is why they are of key importance. The dispersion that follows relaxation in fluids results in dependence of sound speed and attenuation on its frequency. Various kinds of relaxation yield different frequency dependence. Both the relaxation frequency and the maximum absorption per wavelength can be determined from absorption data. Shear viscosity and thermal conduction also participate in the total attenuation.

In spite of the study of relaxation attenuation in acoustics, which has attracted a number of researchers for a long time, there is a lack of a rigorously derived wave equations for sound propagation with an absorption dominated by relaxation, which is known to be the most important cause of attenuation in tissue. As far as the author knows, the first study considering the linear equation of sound propagation in a liquid with several relaxation mechanisms, was published in 1990 [7]. Until this, the available studies described the properties of attenuation caused by relaxation, but did not develop a wave equation for acoustic propagation in the presence of relaxation loss. The starting point of ref. 7 is a set of classical dynamic equations together with the appropriate thermodynamic and constitutive relations describing a flow in electrolytes [8, 9]. The sound absorption in electrolytes contributes an excess quantity due to a chemical relaxation. The derivation accommodates spatial inhomogeneities in material compressibility, density, and parameters of relaxation. The authors of ref. 7 gave a parallel derivation for gases to show the generality of the method.

While the solution of the linear equation for sound in a fluid with relaxation is readily expressed in terms of the corresponding Green's function [7], a solution of the nonlinear equation in the whole domain of sound frequencies is still unresolved [10]. In a thermoviscous flow, secondary nonlinear effects induced by sound are also of great importance in medical and technical applications. These effects include acoustic streaming and heating, that is, nonlinear generation of vorticity and entropy modes because of losses in momentum and energy of intense sound. Acoustic heating and streaming in tissues depend strongly on thermodynamic processes in a tissue and frequency of sound. As for the nonlinear effects in the field of sound in a relaxing fluid, they are poorly studied. There are a few studies devoted to the nonlinear effects of sound in gases with relaxation and chemical reaction, including those in acoustically active gases. These publications deal with nonlinear effects of periodic, aperiodic, and impulse sound [11–13]. This theoretical study considers the nonlinear effects of sound in electrolytes with relaxation originating from chemical reactions. The nonlinear phenomena in the field of intense sound affect the thermodynamic parameters of a fluid and its bulk velocity [14, 15]. That in turn affects features of sound propagation. The nonlinear phenomena may indicate the kind of relaxation in a fluid; they are frequency-dependent, as is attenuation of sound in a fluid.

In studies of weakly nonlinear flows, the author applies an immediate projection of the initial equations onto dynamic equations governing each specific mode [16]. The first step in this process is to determine every branch of acoustic and non-acoustic types of motion in a fluid with attenuation as links of perturbations of all independent variables of a system (perturbations of thermodynamic quantities and compounds of velocity). That allows the derivation of the leading-order individual dynamic equations governing every mode in a weakly nonlinear flow. These equations account for interaction of modes and may be simplified in dependence on the initial intensity of any mode. Acoustic heating and streaming belong to slow flows caused by intense sound, which is considered a dominant mode. The procedure was proposed and applied by the author in analysis of acoustic heating and streaming in fluids with the standard attenuation [16] and some relaxing fluids: Maxwell liquids, gases with excited vibrational degrees of a molecule, gases with chemical reactions. This method is valid for both periodic and aperiodic sound, because it operates on instantaneous quantities and therefore does not require averaging over the sound period at any stage. It applies in a weakly nonlinear flow, yielding the coupling system of dynamic equations for various modes and distributing the nonlinear terms between them correctly. This study investigates weakly nonlinear interaction of sound and non-wave modes in electrolyte with a chemical reaction within an unbounded volume of a liquid. The difference between the nonlinear sound effects in a liquid with this type of relaxation and in a gas with vibrationally excited internal degrees of a molecule, are explained and discussed in Sect. 4.

2. Decomposition of sound, vorticity, and entropy modes

2.1. Basic equations describing motion of a relaxing fluid

In this section, we derive the weakly nonlinear governing equations for a fluid flow with absorption due solely to relaxation mechanisms. The effects of mechanic viscosity and heat conduction are neglected. In gases, these effects are well studied [14, 15], and in liquids and biological tissues, they are small compared to relaxation effects [17–19]. The starting point is

$$\frac{D\rho}{Dt} + \rho(\nabla\cdot\boldsymbol{v}) = 0 \tag{1}$$

for conservation of mass,

$$\rho\frac{D\boldsymbol{v}}{Dt} = -\nabla p \tag{2}$$

for conservation of momentum, and the equation for irreversible thermodynamics detailed below; p, ρ, and $\boldsymbol{v}$ are the total pressure, density, and particle velocity, and D/Dt is the material derivative, defined as

$$\frac{D}{Dt} = \frac{\partial}{\partial t} + (\boldsymbol{v}\cdot\nabla)$$

The Gibbs relation for the rate of variation of entropy s in a fluid

$$T\frac{Ds}{Dt} = \frac{Du}{Dt} - \frac{p}{\rho}\frac{D\rho}{Dt} + \sum_{\nu} A_{\nu}\frac{Dn_{\nu}}{Dt} \tag{3}$$

where A_ν are affinities, and n_ν represent the number of molecules of species ν per unit mass, as in the equations for the chemical reactions in electrolyte [8, 9] (in the equations for gases, n_ν are replaced by the apparent vibrational temperatures $T_{V)}$. The internal energy per unit mass is denoted u. To simplify our analysis and following ref. 7, we assume that all relaxation processes are independent (under conditions listed by Eigen and Tamm [9] or have been expressed in a normal coordinate system that yields the effective independent mechanisms [20].

The entropy is represented by a sum of an equilibrium part (marked by superscript e), and an irreversible part

$$s(p, \rho, \nu_\nu) = s^{e}(p, \rho) + \sum_{\nu}\Delta s_{\nu} \tag{4}$$

where

$$\Delta s_{\nu} = \frac{c_{p}k_{\nu}}{\beta T}\Delta\xi_{\nu} \tag{5}$$

and k_ν is the contribution of the dissolved molecules of species ν to the isothermal compressibility (reciprocal of the bulk modulus). Following refs. 3, 7, and 9, we introduce the quantities $\Delta\xi_\nu$, which are defined as

$$\Delta\xi_{\nu} = \frac{n_{\nu} - n_{\nu}^{e}(p, T)}{\partial n_{\nu}^{e}(p, T)/\partial p} \tag{6}$$

and the appropriate thermodynamic coefficients are given by equalities

$$c_{p} = T\left(\frac{\partial s^{e}}{\partial T}\right)_{p} \qquad \beta = -\frac{1}{\rho}\left(\frac{\partial\rho}{\partial T}\right)_{p} \tag{7}$$

The quantities $\Delta\xi_\nu$ satisfy equations of relaxation, as ~~it has been~~ proved in ref. 7~~:~~

$$\left(\frac{D}{Dt} + \frac{1}{\tau_{\nu}}\right)\Delta\xi_{\nu} = -\frac{Dp}{Dt} \tag{8}$$

Excluding the equilibrium entropy from the entropy-balance equation, (3), and making use of equality

$$\rho A_{\nu}\frac{Dn_{\nu}}{Dt} = k_{\nu}\frac{(\Delta\xi_{\nu})^{2}}{\tau_{\nu}} \tag{9}$$

the entropy-balance equation takes the leading-order form

$$\frac{Dp}{Dt} - c^{2}\frac{D\rho}{Dt} - \frac{B}{2}\frac{D\rho^{2}}{Dt} + \rho c^{2}\sum_{\nu}\frac{D}{Dt}(k_{\nu}\Delta\xi_{\nu}) = \sum_{\nu}\frac{k_{\nu}\beta c^{2}}{\tau_{\nu}c_{p}}(\Delta\xi_{\nu})^{2} \tag{10}$$

where c^2 is squared equilibrium speed of infinitely small magnitude sound, and quantities A and B are determined by equalities [14, 15]

$$c^{2} = \left(\frac{\partial p}{\partial\rho}\right)_{s^{e}} = \frac{A}{\rho_{0}} \qquad B = \left(\frac{\partial^{2} p}{\partial\rho^{2}}\right)_{s^{e}} \tag{11}$$

The nonlinear distortion of a traveling acoustic wave is determined by the parameter of nonlinearity,

$$\varepsilon = \frac{B}{2A} + 1$$

(It is useful to recall that the parameters in an ideal gas: $B = c^2(\gamma - 1)\rho_0$, where γ denotes the specific equilibrium heats ratio under constant pressure, c_p, and constant volume, c_v,

$$c_\mathrm{v} = T\left(\frac{\partial s^\mathrm{e}}{\partial T}\right)_\mathrm{v}$$

and $\varepsilon = (\gamma + 1)/2$ [15, 21].)

Our primary objective is to derive dynamic equations valid at order M^2, where M is the Mach number, that is, up to quadratic nonlinear terms inclusively. The Mach number is a ratio of magnitude of a fluid velocity and the sound speed. It is also of the order of the ratio of the magnitude of perturbation in pressure (density) to the unperturbed pressure (density) of a fluid. The Mach number is a measure of nonlinearity of a flow; a linear flow corresponds to infinitely small Mach number. Flows in liquids are typically weakly nonlinear because of large sound speed as compared with gases. Letting the background values ρ_0 and p_0 be independent of spacial coordinates, one arrives at the system

$$\begin{aligned}
&\frac{\partial \boldsymbol{v}}{\partial t} + \frac{\nabla p'}{\rho_0} = -(\boldsymbol{v}\cdot\nabla)\boldsymbol{v} + \frac{\rho'}{\rho_0^2}\nabla p \\
&\frac{\partial p'}{\partial t} + \rho_0 c^2(\nabla\cdot\boldsymbol{v}) + \rho_0 c^2 \sum_\nu \frac{\partial}{\partial t}(k_\nu \Delta\xi_\nu) = -(\boldsymbol{v}\cdot\nabla)p' - \frac{A+B}{\rho_0}(\nabla\cdot\boldsymbol{v})\rho' \\
&\qquad + \sum_\nu \frac{k_\nu \beta c^2}{\tau_\nu c_p}(\Delta\xi_\nu)^2 - \rho' c^2 \sum_\nu \frac{\partial}{\partial t}(k_\nu \Delta\xi_\nu) - \rho_0 c^2 \sum_\nu (\boldsymbol{v}\cdot\nabla)(k_\nu \Delta\xi_\nu) \qquad (12) \\
&\frac{\partial \rho'}{\partial t} + \rho_0(\nabla\cdot\boldsymbol{v}) = -\rho'(\nabla\cdot\boldsymbol{v}) - (\boldsymbol{v}\cdot\nabla)\rho' \\
&\frac{\partial \Delta\xi_\nu}{\partial t} + \frac{\Delta\xi_\nu}{\tau_\nu} + \frac{\partial p'}{\partial t} = -(\boldsymbol{v}\cdot\nabla)p' - (\boldsymbol{v}\cdot\nabla)\Delta\xi_\nu \qquad (\nu = 1, \ldots N)
\end{aligned}$$

where N is the number of relaxation modes, and the primed quantities denote perturbations. The linear version of this system was derived in ref. 7.

2.2. Modes in a flow of infinitely small magnitude and extracting them from the total perturbations

The dispersion relations describing all independent modes follow from the linearized version of (12). They are: acoustic (two branches), thermal (or entropy), and vorticity (two branches) modes in three-dimensional flows; these modes are well-studied in flows of Newtonian fluids [22]. From the dispersion relations, we are able to suggest a linear model propagation equation for every mode in an unbounded fluid. In general, each of the field variables contains contributions from each of three modes, for example, $\boldsymbol{v} = \boldsymbol{v}_\mathrm{a} + \boldsymbol{v}_\mathrm{ent} + \boldsymbol{v}_\mathrm{vor}$. The linearized version of the system (12) takes the form

$$\frac{\partial \Psi}{\partial t} + L\Psi = 0 \qquad (13)$$

where $\Psi = (v_x, v_y, v_z, p', \rho', \Delta\xi_1, \ldots, \Delta\xi_N)^\mathrm{T}$, v_x, v_y, v_z are the Cartesian coordinates of $\boldsymbol{v}$, and L is a linear matrix operator including spatial derivatives. Equation (13) describes the flow with infinitely small magnitudes of perturbations. Studies of motions of infinitely small magnitudes begin usually by representing all perturbations as a sum of planar waves

$$f'(\mathbf{r}, t) = \int\limits_{R^3} \tilde{f}(\boldsymbol{k}) \exp[i\omega(\boldsymbol{k})t - i\boldsymbol{k}\mathbf{r}]d\boldsymbol{k}$$

where $\tilde{f}(\boldsymbol{k})\exp[i\omega(\boldsymbol{k})t]$ denotes the Fourier transform of $f'(\mathbf{r}, t)$, $\tilde{f}(\boldsymbol{k}, t) = (2\pi)^{-3}\int_{R^3} f(\mathbf{r},t)\mathrm{e}^{i\boldsymbol{k}\mathbf{r}}d\mathbf{r}$. In general, the number of dispersion relations, that is, the number of modes, coincides with the number of dynamic and conservation equations. It equals five plus the number of relaxation processes, N, in three dimensions. For simplicity, only one relaxation process with constant k_ν will be considered, ordered as the νth with corresponding time of relaxation. There are two branches of sound ($i = 1$ and 2), and four non-wave modes (the entropy mode, $i = 3$, the relaxation mode, $i = 4$, and two vorticity branches, $i = 5$ and 6) or, in other words, types of motion that may exist in a fluid. The dispersion relations ω_i ($i = 1, \ldots, 6$) are as follows:

$$\begin{aligned}
&\omega_{1,2} = \pm c\mathbf{k} + ik_\nu \frac{c^4\mathbf{k}^2\rho_0\tau_\nu}{2(1 \pm ic\mathbf{k}\tau_\nu)} \qquad \omega_3 = 0 \\
&\omega_4 = \frac{i}{\tau_\nu}\left(1 + \frac{c^2\rho_0 k_\nu}{1 + c^2\mathbf{k}^2\tau_\nu^2}\right) \qquad \omega_{5,6} = 0
\end{aligned} \qquad (14)$$

where $\mathbf{k}$ denotes module of the vector $\boldsymbol{k}$. We consider weakly dispersive flows that are characterized by a small parameter responsible for relaxation,

$$\alpha = c^2\rho_0 k_\nu \ll 1$$

Equations (14) are evaluated with accuracy up to terms proportional to α^0 and α^1 but without any restrictions concerning spatial spectrum of perturbations. In particular, both vorticity modes are solenoidal

$$\nabla\cdot\boldsymbol{v}_{5,6} = 0 \qquad p'_{5,6} = 0 \qquad \rho'_{5,6} = 0 \qquad (15)$$

The solenoidal velocity may be decomposed from the total one by applying the operator $P_{\mathrm{vor},\mathbf{v}}$ at the vector of the overall velocity

$$P_{\mathrm{vor},\boldsymbol{v}}\boldsymbol{v} = \Delta^{-1}\begin{pmatrix} \frac{\partial^2}{\partial y^2} + \frac{\partial^2}{\partial z^2} & -\frac{\partial^2}{\partial x\partial y} & -\frac{\partial^2}{\partial x\partial z} \\ -\frac{\partial^2}{\partial x\partial y} & \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial z^2} & -\frac{\partial^2}{\partial y\partial z} \\ -\frac{\partial^2}{\partial x\partial z} & -\frac{\partial^2}{\partial y\partial z} & \frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2} \end{pmatrix}\begin{pmatrix} \sum_{i=1}^{6} v_{x,i} \\ \sum_{i=1}^{6} v_{y,i} \\ \sum_{i=1}^{6} v_{z,i} \end{pmatrix} = \begin{pmatrix} \sum_{i=5}^{6} v_{x,i} \\ \sum_{i=5}^{6} v_{y,i} \\ \sum_{i=5}^{6} v_{z,i} \end{pmatrix} \qquad (16)$$

where Δ is the Laplacian. Equation (16) manifests in fact a certain way of application of the Helmholtz vector decomposition theorem, which enables us to decompose irrotational and solenoidal vector fields. It may be readily proved that $P_{\mathrm{vor},\mathbf{v}}$ satisfies

$$-\nabla\times(\nabla\times\boldsymbol{\varphi}) = \Delta P_{\mathrm{vor},\boldsymbol{v}}\boldsymbol{\varphi} \tag{17}$$

where $\boldsymbol{\varphi}$ is any smooth three-component vector. The vortex flow may exist in two- and tree-dimensional flows.

To decompose the entropy and relaxation modes from the total vector of perturbations, let us consider the one-dimensional perturbations depending on one spatial coordinate, x. In one dimension, there exist four first modal fields. The total small-magnitude perturbations represent a sum of all modes

$$\begin{aligned}&\tilde{v} = \sum_{i=1}^{4}\tilde{v}_i = \sum_{i=1}^{4}\frac{\omega_i\tilde{\rho}_i}{k\rho_0} \qquad \tilde{p} = \sum_{i=1}^{4}\tilde{p}_i = \sum_{i=1}^{4}\frac{\omega_i^2\tilde{\rho}_i}{k^2}\\ &\tilde{\rho} = \sum_{i=1}^{4}\tilde{\rho}_i \qquad \Delta\tilde{\xi}_\nu = \sum_{i=1}^{4}\Delta\tilde{\xi}_{\nu,i} = -i\sum_{i=1}^{4}\frac{\omega_i^4\tilde{\rho}_i}{k^2[i\omega_i + (1/\tau_\nu)]}\end{aligned} \tag{18}$$

where $\tilde{v} = \tilde{v}_x$, $k = k_{\mathrm{x}}$. Solving algebraic equations

$$P_3(\tilde{v}, \tilde{p}, \tilde{\rho}, \widetilde{\Delta\xi_\nu})^{\mathrm{T}} = \tilde{\rho}_3 \qquad P_4(\tilde{v}, \tilde{p}, \tilde{\rho}, \Delta\tilde{\xi}_\nu)^{\mathrm{T}} = \tilde{\rho}_4$$

one arrives at operator rows that distinguish every specific Fourier transform of excess density for these two modes

$$\begin{aligned}&P_3 = \begin{pmatrix}0 & -\dfrac{1}{c^2} & 1 & -\dfrac{\alpha}{c^2}\end{pmatrix}\\ &P_4 = \begin{bmatrix}\dfrac{i\alpha c^2k^3\rho_0\tau_\nu^3}{(1 + c^2k^2\tau_\nu^2)^2} & \dfrac{\alpha\rho_0c^2k^4\tau_\nu^4}{(1 + c^2k^2\tau_\nu^2)^2} & 0 & \dfrac{\alpha k^2\tau_\nu^2}{1 + c^2k^2\tau_\nu^2}\end{bmatrix}\end{aligned} \tag{19}$$

They are evaluated with accuracy up to terms proportional to α^0 and α^1 but without any restrictions concerning spatial spectrum of perturbations, as well as dispersion relations. The elements of P_3 and P_4 determine some integro-differential spatial operators in the space ($\boldsymbol{r}$, t). When $P_{\mathrm{vor},v}$ applies at the momentum three-dimensional equation, it distinguishes the linear dynamic equation that governs the vortex velocity, and when P_3 and P_4 apply at the system of conservation equations in one dimension, they distinguish equations for the excess specific densities of the entropy and relaxation modes, respectively.

3. Intense sound and dynamics of the non-wave modes in its field

The linearized theory does not indicate any interaction among the modes as long as the domain of interest is far from the boundaries; it may be valid as long as the fluctuations are weak. The special linear combining that uses the properties of individual linearly independent modes, decomposes any specific mode, reducing all other modes in the linear part of the equations. It readily applies in the studies of weakly nonlinear flows and distributes the nonlinear terms between dynamic equations in the correct manner [16]. The nonlinear terms represent the contributions of all modes and may be simplified in the leading order in the context of the physical meaning of a problem. Acoustic streaming and heating impose dominative acoustic mode, so that only pure acoustic terms should be considered among nonlinear ones.

An equation governing excess density in an acoustic planar wave that propagates along axis OX, may be reconstructed using (14)

$$\left(1 - c\tau_\nu\frac{\partial}{\partial x}\right)\left(\frac{\partial\rho_1'}{\partial t} + c\frac{\partial\rho_1'}{\partial x} + \frac{c\varepsilon}{\rho_0}\rho_1'\frac{\partial\rho_1'}{\partial x}\right) = \frac{\alpha c^2\tau_\nu}{2}\frac{\partial^2\rho_1'}{\partial x^2} \tag{20}$$

Its approximate and exact solutions are discussed in ref. 10and references therein, which relate to a stationary waveform that propagates with the equilibrium speed c. Note, that there is an infinite variety of stationary waveforms that take the form of a shock wave and propagate with the speed $\tilde{c}$ greater than c_∞, where c_∞ denotes the frozen sound speed, that is, the sound speed at infinitely large frequency

$$\tilde{c} > c_\infty \qquad c_\infty = (1 + 0.5\alpha)c \tag{21}$$

The density jump in a shock wave equals

$$\Delta\rho_{\mathrm{sh}} = \frac{2\rho_0(\tilde{c} - c)}{c\varepsilon} \tag{22}$$

It is always larger than $\alpha\rho_0/\varepsilon$.

3.1. Acoustic streaming

Applying $P_{\mathrm{vor},\mathbf{v}}$ on the first equation from (12) (the momentum equation), results in dynamic equation governing velocity of the vorticity mode with an acoustic source of streaming, $\boldsymbol{F}_{\mathrm{a,vor}}$, in the right-hand side

$$\frac{\partial\boldsymbol{v}_{\mathrm{vor}}}{\partial t} = -\frac{1}{\rho_0}P_{\mathrm{vor},\boldsymbol{v}}\left(\rho_{\mathrm{a}}'\frac{\partial}{\partial t}\boldsymbol{v}_{\mathrm{a}}\right) = -\frac{1}{\rho_0}P_{\mathrm{vor},\boldsymbol{v}}\left(\sum_{n=1}^{2}\rho_n'\frac{\partial}{\partial t}\sum_{n=1}^{2}\boldsymbol{v}_n\right) = \boldsymbol{F}_{\mathrm{a,vor}} \tag{23}$$

or, in terms of vorticity $\boldsymbol{\Omega} = \nabla\times\boldsymbol{v}_{\mathrm{vor}}$, it sounds

$$\frac{\partial\boldsymbol{\Omega}}{\partial t} = -\frac{1}{\rho_0}\nabla\times\left(\sum_{n=1}^{2}\rho_n'\frac{\partial}{\partial t}\sum_{n=1}^{2}\boldsymbol{v}_n\right) \approx \frac{1}{\rho_0^2}\nabla\times\left(\sum_{n=1}^{2}\rho_n'\sum_{n=1}^{2}\nabla p_n'\right) \tag{24}$$

The links between perturbations in density and pressure for both the acoustic modes are

$$\tilde{p}_1 = c^2\tilde{\rho}_1 + \frac{\alpha c^3\mathbf{k}\tau_\nu}{-i + c\mathbf{k}\tau_\nu}\tilde{\rho}_1 \qquad \tilde{p}_2 = c^2\tilde{\rho}_2 + \frac{\alpha c^3\mathbf{k}\tau_\nu}{i + c\mathbf{k}\tau_\nu}\tilde{\rho}_2 \tag{25}$$

Hence, the acoustic force of streaming is proportional to α, because the first terms in both relations (25) do not contribute to the acoustic force. An acoustic source is nonlinear and originates from the nonlinear attenuation in a relaxing medium. An acoustic force may be expressed in terms of specific excess acoustic perturbations ρ_1', ρ_2' with some integro-differential operators that are determined by links of Fourier transforms of acoustic pressure and density, (25). In the limiting cases of the low-frequency ($\omega\tau_\nu \ll 1$) or high-frequency sound ($\omega\tau_\nu \gg 1$), we obtain the relations for the first acoustic mode

$$\begin{aligned}&\text{low-frequency:} \quad \tilde{p}_1 = c^2(1 + i\alpha c\mathbf{k}\tau_\nu)\tilde{\rho}_1\\ &\text{high-frequency:} \quad \tilde{p}_1 = c_\infty^2\left(1 + i\frac{\alpha}{c\mathbf{k}\tau_\nu}\right)\tilde{\rho}_1\end{aligned} \tag{26}$$

To evaluate the quadratic nonlinear terms associated with sound, these links should be complemented by the terms that make sound isentropic in the leading order. That is, the term $(B/2)\rho_1'^2$ should be added in the linear expression for p_1' in terms of ρ_1'.

If we consider only the first acoustic branch, (24) simplifies as

$$\begin{aligned} &\text{low-frequency:} \quad \frac{\partial \boldsymbol{\Omega}}{\partial t} = -\frac{\alpha c^3 \tau_\nu}{\rho_0} \nabla \times \left(\rho_1' \cdot \nabla \Delta^{1/2} \rho_1'\right) \\ &\text{high-frequency:} \quad \frac{\partial \boldsymbol{\Omega}}{\partial t} = \frac{\alpha c}{\rho_0 \tau_\nu} \nabla \times \left(\rho_1' \cdot \nabla \Delta^{-1/2} \rho_1'\right) \end{aligned} \tag{27}$$

The most applications refer to a weakly diffracting acoustic beam, that is, to a quasi-planar wave progressing along a straight line, which may be identified with axis OX. In this case, the small parameter that is responsible for weak diffraction, may be defined as [14]

$$\mu = \frac{k_y^2 + k_z^2}{k_x^2}$$

and evaluations can be readily performed in the leading order up to terms proportional to $\mu^{1/2}$. One arrives at

$$\begin{aligned} &\text{low-frequency:} \quad \frac{\partial \boldsymbol{\Omega}}{\partial t} = \frac{\alpha c^3 \tau_\nu}{\rho_0^2} \left[\boldsymbol{e}_y \left(\frac{\partial \rho_1'}{\partial x} \frac{\partial^2 \rho_1'}{\partial x \partial z} - \frac{\partial \rho_1'}{\partial z} \frac{\partial^2 \rho_1'}{\partial x^2} \right) \right. \\ &\qquad\qquad \left. - \boldsymbol{e}_z \left(\frac{\partial \rho_1'}{\partial x} \frac{\partial^2 \rho_1'}{\partial x \partial y} - \frac{\partial \rho_1'}{\partial y} \frac{\partial^2 \rho_1'}{\partial x^2} \right) \right] \\ &\text{high-frequency:} \quad \frac{\partial \boldsymbol{\Omega}}{\partial t} = -\frac{\alpha c}{\rho_0^2 \tau_\nu} \left[\boldsymbol{e}_y \left(\frac{\partial \rho_1'}{\partial x} \frac{\partial}{\partial z} \int \rho_1' dx \right) \right. \\ &\qquad\qquad \left. - \boldsymbol{e}_z \left(\frac{\partial \rho_1'}{\partial x} \frac{\partial}{\partial y} \int \rho_1' dx \right) \right] \end{aligned} \tag{28}$$

where $\boldsymbol{e}_y$ and $\boldsymbol{e}_z$ are unit vectors along axes OY and OZ. In the leading order, sound of all frequencies induces insignificant longitudinal velocity of the vortex flow.

3.2. Acoustic heating and generation of the relaxation mode

We consider the one-dimensional flow along axis OX in issues concerning acoustic heating. The conclusions are valid in the quasi-planar geometry of sound as well. Links for the perturbations in the first acoustic branch (which will be solely considered for simplicity), take the forms

$$\tilde{v}_1 = \frac{c}{\rho_0}\left(1 + \alpha \frac{ck\tau_\nu}{2}\right)\tilde{\rho}_1 \qquad \Delta\xi_{\nu,1} = \frac{c^3 k \tau_\nu}{i - ck\tau_\nu}\left[1 + \frac{\alpha ck\tau_\nu}{2(-i + ck\tau_\nu)^2}\right]\tilde{\rho}_1 \tag{29}$$

They allow expression of an acoustic source in terms of disturbance of density attributable to the acoustic mode. Perturbations specifying all other modes are considered as negligibly small among a wide variety of nonlinear ones. The leading order low-frequency and high-frequency limits of these relations are

$$\begin{aligned} &\text{low-frequency:} \quad \tilde{v}_1 = \frac{c}{\rho_0}\tilde{\rho}_1 \qquad \Delta\tilde{\xi}_\nu = -ic^3\tau_\nu k\tilde{\rho}_1 \\ &\text{high-frequency:} \quad \tilde{v}_1 = \frac{c_\infty}{\rho_0}\tilde{\rho}_1 \qquad \Delta\tilde{\xi}_\nu = -c_\infty^2\tilde{\rho}_1 - i\frac{c^2}{ck\tau_\nu}\tilde{\rho}_1 \end{aligned} \tag{30}$$

Applying P_3 from (19) on (12) reduces all acoustic and relaxation terms in the left-hand linear side and yields the leading-order equations that govern acoustic heating

$$\begin{aligned} &\text{low-frequency:} \quad \frac{\partial \rho_3'}{\partial t} = -\frac{\alpha\beta c^4 \tau_\nu}{c_p \rho_0}\left(\frac{\partial \rho_1}{\partial x}\right)^2 + \frac{\alpha c}{2\rho_0}\frac{\partial \rho_1'^2}{\partial x} \\ &\text{high-frequency:} \quad \frac{\partial \rho_3'}{\partial t} = -\frac{\alpha\beta c^2}{\tau_\nu c_p \rho_0}\rho_1'^2 \end{aligned} \tag{31}$$

As expected, an excess density that specifies the entropy mode in both regimes decreases with time, because $\partial\rho'^2_1/\partial x < 0$ because of irreversible losses in acoustic energy associated with relaxation. The first term in the low-frequency source is of major importance in the case of almost periodic sound. The density decrease is followed by the isobaric enlargement of temperature.

Repeating the procedure by applying P_4 and making use of (30), one arrives at the leading-order dynamic equations for the relaxation excess density in the low-frequency and high-frequency regimes

$$\begin{aligned} &\text{low-frequency:} \quad \frac{\partial \rho_4'}{\partial t} + \frac{1 + \alpha}{\tau_\nu}\rho_4' = 0 \\ &\text{high-frequency:} \quad \frac{\partial \rho_4'}{\partial t} + \frac{\rho_4'}{\tau_\nu} = -\frac{2\alpha c}{\tau_\nu}\left(1 + \frac{A + B}{\rho_0 c^2}\right)\rho_1'^2 \end{aligned} \tag{32}$$

4. Concluding remarks

There is a difference of the considered case of relaxation in liquids and relaxation due to vibrationally excited internal degrees of a molecule's freedom in gases (or relaxation in gases due to exothermic chemical reactions which parallels that in the vibrationally excited gases). The relaxation equation in liquids, (8), includes in its right-hand side the material derivative of the thermodynamic parameter, pressure, while the corresponding equation describing a disturbance of apparent vibrational temperatures, T_V', takes the form

$$\left(\frac{D}{Dt} + \frac{1}{\tau_V}\right)T_V' = \frac{T'}{\tau_V} = \frac{T_0}{\tau_V}\left(\frac{p'}{p_0} - \frac{\rho'}{\rho_0}\right) \tag{33}$$

with the right-hand side proportional to excess temperature, T', but not to its derivative (τ_V denotes the vibrational relaxation time).

The difference in the types of relaxation in liquids and gases affects both linear and nonlinear characteristics of a flow, first of all, the dispersion relations of sound and the entropy mode, and hence, linear dynamic equations describing these modes. The modes as the links of specific perturbations also are different. As for the high-frequency sound, the dispersion relations of sound in a liquid readily follow from (14)

$$\omega_{1,2} = \pm\left(c\mathbf{k} + 0.5k_\nu c^3 \mathbf{k}\rho_0\right) \tag{34}$$

while the dispersion relations of sound in a gas with vibrational relaxation determine the frequency-independent attenuation of the high-frequency sound [23, 24]

$$\omega_{1,2} = \pm c\mathbf{k} + \frac{0.5i(\gamma - 1)^2 c_v T_0}{\tau_V c^2}$$

The relaxation dispersion relation in the gases with vibrational relaxation also looks different as compared with ω_4 from (14) [25]

$$\omega_4 = \frac{i}{\tau_V} + \frac{i(\gamma - 1)\left(\gamma + c^2\mathbf{k}^2\tau_V^2\right)T_0 c_v}{c^2\tau_V\left(1 + c^2\mathbf{k}^2\tau_V^2\right)} \tag{35}$$

It determines different behavior especially at intermediate wavenumbers of perturbation. The relaxation mode is isobaric in the case of a gas with excited vibrational degrees of a molecule's freedom, but in the case of relaxation in a liquid, it is not. The Fourier transform of perturbation in pressure, associated with the relaxation mode, takes the leading-order form in terms of the Fourier transform of its excess density

$$\tilde{p}_4 = -\left(1 + \frac{2c^2\rho_0 k_\nu}{1 + c^2\mathbf{k}^2\tau_\nu^2}\right)\frac{\tilde{\rho}_4}{\mathbf{k}^2\tau_\nu^2} \tag{36}$$

The nonlinear effects of sound also look different in gases and liquids. Acoustic streaming, acoustic heating, and generation of the relaxation mode due to both types of relaxation are fairly weak effects for the low frequencies of sound [25]. As for the high-frequency sound in a vibrationally excited gas, an acoustic force of heating is proportional to $-\gamma(\partial\rho_1'/\partial x)\int\rho_1' dx - \rho_1'^2$ [26], not to $-\rho_1'^2$, as in elecrolytes. Hence, heating in the media of these different types of relaxation is fairly similar in the case of periodic sound, but is different for aperiodic sound, including impulses. The acoustic force of the relaxation mode in gases is proportional to $\rho_1'(\partial\rho_1'/\partial x)$ in the high-frequency regime [27], not to $\rho_1'^2$, (32), hence, the nearly periodic sound in a gas is hardly expected to produce a noticeable perturbation in vibrational energy.

In this study, we consider weakly nonlinear effects of sound in an electrolyte. Nonlinearity and attenuation of sound, which follows relaxation, are the necessary conditions for modes to interact and for transfer of acoustic energy and momentum into that of the non-wave modes. Parameters of the nonlinear phenomena, such as excess temperature and bulk velocity of a fluid, slowly vary with time and may be readily measured. They may be useful in establishing details of relaxation processes in a liquid. The results also may be employed to model nonlinear scattering in applications such as image reconstruction in a liquid.

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