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# Hysteresis curves and loops for harmonic and impulse perturbations in some non-equilibrium gases

**Research Article**

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**Abstract:** Evolution of sound in a relaxing gas whose properties vary in the course of wave propagation, is studied. A relaxing medium may reveal normal acoustic properties or be acoustically active. In the first case, losses in acoustic energy lead to an increase in internal energy of a gas similarly as it happens in Newtonian fluids. In the second case, acoustic energy increases in the course of sound propagation, and the internal energy of a medium decreases. Variations in the internal energy of a gas are proportional to some generic parameter, the sign of which is responsible for acoustical activity, and depends on intensity and shape of the sound waveform. Hysteresis curves in the plane of thermodynamic states are plotted. Curves for harmonic and several aperiodic sound impulses are plotted, discussed and compared.

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**Keywords:** hysteresis • non-equilibrium gases • thermodynamic relaxation



## 1. Introduction

Thermal conductivity, molecular absorption, scattering and relaxation processes of different origin, lead to attenuation of sound and an irreversible nonlinear increase in the internal energy of a medium [1–3]. A fluid may reveal anomalous absorption and dispersion of sound [4–7]. Some special conditions of external pumping of energy [8–10] or heat release in a chemical reaction [11] make a fluid non-equilibrium, negative dispersive and acoustically active. Anomalous dispersion of sound and negative total viscosity in these two examples of gases are caused by different physical reasons but described by similar equations with a certain generic coefficient which is responsible for dispersion in these fluids, ordinary or anomalous. In the both cases, the relation between acoustic pressure $p_a$ and excess acoustic density $\rho_a$ takes the typical leading-order form for the relaxing media:

$$p_a = c^2\rho_a + \frac{\gamma-1}{2\rho_0}c^2\rho_a^2 + 2Bc^3\int^t \rho_a e^{-(t-t')/\tau}dt', \quad (1)$$

with some parameter $B$ which may take positive or negative values, $\tau$ denotes the relaxation time, $c = \sqrt{\gamma p_0/\rho_0}$ is the propagation speed of infinitely-small magnitude sound, $p_0$, $\rho_0$ are unperturbed pressure and density of a medium, and $\gamma$ is the specific heat ratio in an ideal gas. Three terms in the right-hand side of this equality are

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well-established. In fact, two first specify the isentropic Riemann wave [1–3]. The last therm describes the relaxing properties of a medium. For sound to be a wave process, attenuation (or amplification) and dispersion which follow propagation of sound, are considered to be small over the wavelength, $|B| << \omega/c$, where $\omega$ denotes the characteristic sound frequency. In the low-frequency domain, where $\omega\tau << 1$ and $B < 0$, a fluid behaves as Newtonian with attenuation proportional to $B$ and $\tau$. In view of small $|B|$ as compared to the characteristic wavenumber of sound, attenuation (or enhancement) of low-frequency sound is very low, as well as enlargement of the thermal mode in the field of sound [12, 13]. Eq. (1) reveals that a connection between acoustic pressure and excess acoustic density is not longer algebraic, but depends generally on the prehistory of perturbations in a fluid, thereby causing a medium hysteresis. In the high-frequency regime, $\omega\tau >> 1$, the dynamic equation for acoustic pressure in one-dimensional flow takes the leading-order form:

$$\frac{\partial^2 p_a}{\partial t^2} - c^2\frac{\partial^2 p_a}{\partial x^2} - \frac{\gamma+1}{2c^2\rho_0}\frac{\partial^2 p_a^2}{\partial t^2} + 2Bc\frac{\partial p_a}{\partial t} = 0. \quad (2)$$

To conclude about the equation of state, that is, about connection of total perturbations of pressure and density in a medium, one requires knowledge not only of acoustic perturbations, but also of irreversible transfer of sound energy into the energy of the thermal mode. This transfer in Newtonian fluids is followed by an isobaric increase of temperature of a medium, and, as a consequence, in decrease of its density [1, 3, 14]. In non-equilibrium media, under some conditions, sound amplifies [7, 10, 11], and internal energy may decrease, that is followed by enlargement in density of a medium. Along with normal or anomalous attenuation, nonlinearity is the necessary condition for generation of the thermal mode in the field of ultrasound: in linear flow, acoustic and non-wave motions of a fluid do not interact. Variations of internal energy depend on intensity of the wave, on $B$, but also on gradients of acoustic perturbations. The thermodynamic cycles in a strain-stress diagrams for solid materials with hysteresis nonlinearity are usually represented by loops [15, 16]. Less pronounced, but similar loops are typical in Newtonian thermoconducting fluids [17]. Hedberg, Rudenko were the first who have attracted attention to hysteresis in Newtonian and relaxing fluids. Similar hysteresis curves, normal or anomalous, exist in fluids with normal or anomalous attenuation of sound. decrease in the excess density which specifies the entropy mode. These curves are represented by the hysteresis curves in the plane of thermodynamic states (total excess density $\Leftrightarrow$ total excess pressure). Two examples of relaxing gases with inherent coefficients $B$ are described in the subsections below. The following simplifying conditions are made in the both examples:

1) there is no ambient motion of a gas;
2) the necessary transversal variations of ambient parameters (in $OYZ$ plane) do not significantly affect the wave propagation;
3) the shear, bulk viscosity of a gas and its thermal conductivity are not considered;
4) the frequency of sound is much larger than the inverse characteristic relaxation time: $\omega\tau \gg 1$;
5) the magnitude of sound slowly varies over its period: $|B|c/\omega \ll 1$.

## 1.1. Gases in which an exothermic chemical reaction occurs

For these kind of processes in a gas,

$$B = \frac{Q_0(\gamma-1)(Q_\rho + (\gamma-1)Q_T)}{2c^2 m} \quad (3)$$

is the quantity evaluated at unperturbed $p_0$, $T_0$, $Y_0$, where $Y$ denotes the mass fraction of reagent $A^*$ in $A^* \to B^*$ exothermic reaction, $m$ is the averaged molecular mass of a gas, and $Q$ is the heat produced in a medium per one molecule due to a chemical reaction, $Q_0 = Q(T_0, \rho_0, Y_0)$ [11]. The dimensionless quantities $Q_T$, $Q_\rho$ are conditioned by dependence of $Q_0$ on temperature and density of the reacting mixture:

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

The characteristic duration of chemical reaction is

$$\tau = \frac{HmY_0}{Q_0 Q_Y}, \quad (5)$$

where $H$ is the reaction enthalpy per unit mass of reagent $A^*$, $Q_Y = \frac{Y_0}{Q_0}\left(\frac{\partial Q}{\partial Y}\right)_{T_0,\rho_0,Y_0}$.

## 1.2. Gases with excited vibrational degrees of molecule's freedom

The second example of fluid which may be acoustically active, relates to a gas whose steady state is maintained by pumping energy into the vibrational degrees of molecule's freedom by power $I$ ($I$ refers to a unit mass). The relaxation equation for the vibrational energy $\varepsilon$ per unit mass is given by the formula [7]:

$$\frac{d\varepsilon}{dt} = -\frac{\varepsilon - \varepsilon_{eq}(T)}{\tau} + I. \quad (6)$$

The equilibrium value of the vibrational energy at given temperature $T$ is denoted by $\varepsilon_{eq}(T)$, and $\tau(\rho, T)$ marks the vibrational relaxation time. The quantity $\varepsilon_{eq}(T)$ in the case of a system of harmonic oscillators, equals:

$$\varepsilon_{eq}(T) = \frac{\hbar\Omega}{m\,(exp(\hbar\Omega/k_B T) - 1)}, \tag{7}$$

where $m$ denotes a molecule's mass, $\hbar\Omega$ is the magnitude of the vibrational quantum, $k_B$ is the Boltzmann constant. The above equation is valid over the domain of temperatures, where one can neglect anharmonic effects, i.e., below the characteristic temperatures, which are fairly high for most molecules [6, 7]. The quantity

$$B = -\frac{(\gamma-1)^2 T_0 \tau}{2c^3} \left( C_v + \frac{\varepsilon - \varepsilon_{eq}}{\tau} \left( \frac{\partial \tau}{\partial T} + \frac{\rho}{(\gamma-1)T} \frac{\partial \tau}{\partial \rho} \right) \right)_0 \tag{8}$$

may be positive (under some conditions) [8, 9] in a non-equilibrium gas, and negative in an equilibrium one [7, 8, 10]. It is the quantity evaluated at unperturbed $p_0$, $T_0$, and $C_v = d\varepsilon_{eq}/dT$. The relaxation time in the most important cases may be thought as a function of temperature according to Landau and Teller with some positive constants $\tilde{A}$ and $\tilde{B}$, $\tau(T) = \tilde{A}\exp(\tilde{B}T^{-1/3})$ [4, 7]. Acoustical activity of a medium is in principle possible due to negative $d\tau/dT$. There exists the threshold quantity of pumping magnitude $I$ starting from which the excitation is non-equilibrium, since $\varepsilon - \varepsilon_{eq} \approx I\tau$.

## 2. Dynamics of total excess density and pressure

The types of a Newtonian fluid motion of infinitely-small magnitude are well-established [18]. The linear classification applies also in a weakly nonlinear flow. In one dimension, there exist two acoustic branches and the entropy (or thermal) mode. The generation of the thermal mode in the field of intense sound alters temperature of the medium of sound propagation. That happens to viscous fluids and yields to scattering of sound at the heated domains [19]. The acoustic heating caused by periodic sound in Newtonian fluids is well-studied [1, 3]. The details of analysis of instantaneous interaction of sound and the thermal mode in a weakly nonlinear flow of Newtonian fluids may be found in [20, 21]. Induced scattering of sound in thermodynamically non-equilibrium media, which considers both domains with altered temperature and vortex flow, was considered in the papers by Molevich [22, 23].

The instantaneous generation of the thermal mode in the field of low-frequency or high-frequency sound in the relaxing fluids where irreversible processes may take place, have been discussed in [13, 24]. In general, the analysis applies not only to periodic sound and describes, among other, instantaneous variations of excess density specifying the thermal mode in the field of intense sound ($\rho_e$ denotes excess quantity associated with the entropy mode, and $Q_a$ is the acoustic source of the entropy mode),

$$\frac{\partial \rho_e}{\partial t} = \rho_0 Q_a, \tag{9}$$

where

$$Q_a = -\frac{2(\gamma-1)Bc}{\rho_0^2} \frac{\partial \rho_a}{\partial t} \int^t \rho_a dt \tag{10}$$

in the case of chemically reacting gas [13], and

$$Q_a = -\frac{2Bc}{\rho_0^2} \left( \rho_a^2 + \gamma \frac{\partial \rho_a}{\partial t} \int^t \rho_a dt \right) \tag{11}$$

in the case of vibrationally excited gas [24]. In this study, we take into account neither thermal conductivity, nor shear and bulk viscosity which make the domain of acoustical activity smaller or may prevent it at all. Inclusion of mechanical viscosity and thermal conduction is discussed in the papers [10, 25]. We concentrate in this study on the thermodynamic relaxation exclusively. The lower limit of integration in the expressions for $Q_a$ should be chosen in accordance to the beginning of the sound transmission. The total excess pressure $p'$ is a sum of acoustic pressure and excess pressure associated with the entropy mode, this last part equals zero: the entropy motion is isobaric. The total excess density consists also of two parts. The leading-order relation takes the form

$$\begin{aligned} \frac{p'}{\rho_0 c^2} &= \frac{p_a}{\rho_0 c^2} = \frac{\rho_a}{\rho_0} + \frac{\gamma-1}{2\rho_0^2}\rho_a^2 + \frac{2Bc}{\rho_0}\int^t \rho_a dt \\ &= \frac{\rho' - \rho_e}{\rho_0} + \frac{\gamma-1}{2\rho_0^2}\rho'^2 + \frac{2Bc}{\rho_0}\int^t \rho' dt \\ &= \frac{\rho'}{\rho_0} + \frac{\gamma-1}{2\rho_0^2}\rho'^2 + \frac{2Bc}{\rho_0}\int^t \rho' dt - \int^t Q_a dt. \end{aligned} \tag{12}$$

The irreversible decrease (or increase, if $B > 0$) in acoustic energy is connected with enlargement (or reduction) of excess temperature which specifies the thermal mode. The analysis below concerns harmonic sound and some impulses. The last term in the right-hand side of Eq. (12), though is smaller in order than other, is of great importance: namely this term is responsible for irreversible losses or enlargement of the internal energy of a medium.

## 3. Hysteresis curves for harmonic sound

If excess pressure is a periodic harmonic function of time for any distance from a transducer $x$ (the phase shift is inessential),

$$P = \frac{p'}{Mc_0^2\rho_0} = \sin(\omega t), \tag{13}$$

$$R = \frac{\rho'}{M\rho_0} = \sin(\omega t) + 2b\cos(\omega t) - 2b - \frac{1}{2}M(\gamma-1)\sin^2(\omega t) + Mb\widetilde{Q},$$

where $P$ and $R$ are dimensionless total excess pressure and density of a gas, $b$ is dimensionless acoustic increment or decrement,

$$b = \frac{Bc}{\omega}, \tag{14}$$

$M$ denotes the acoustic Mach number, and

$$\widetilde{Q} = (\gamma-1)\omega t + 0.5(\gamma-1)\sin(2\omega t) \tag{15}$$

in the case of the reacting gas, and

$$\widetilde{Q} = (\gamma-1)\omega t + 0.5(\gamma+1)\sin(2\omega t) \tag{16}$$

in the case of vibrationally excited gas. The lower limit of integration in Eq. (12) is zero. Eqs (12), (13), (15), (16) determine dependence of the total excess pressure $p'$ on the total excess density $\rho'$ in the parametric form. In dependence on sign of $B$, an excess total density unusually enlarges over a period ($b > 0$) or decreases ($b < 0$). Direction of the hysteresis curves is also different in the case of positive and negative $b$. Two cycles of these hysteresis curves in both fluids in acoustically active or normal damping gases are plotted in the Fig. 1. They correspond to parameters $\gamma = 1.4$, $M = 0.5$ and $b = -0.1$ or $b = 0.1$.

All curves start from the point $(0, 0)$ in the plane $RP$. In both thermodynamic processes in gases, the total excess density gets smaller after the whole period in the normal damping medium, and becomes larger in an acoustically active one. That reflects the nonlinear isobaric growth of the entropy mode temperature, that is, the background temperature, in acoustically active gases, and its unusual decrease otherwise. Relative variation in the internal energy $U$ over the period depends on the sign of $B$ and equals

$$\frac{\omega}{2\pi}\frac{\delta U}{U_0} = -\left\langle \frac{1}{\rho_0}\frac{\partial \rho_e}{\partial t}\right\rangle = -\langle Q_a\rangle, \tag{17}$$

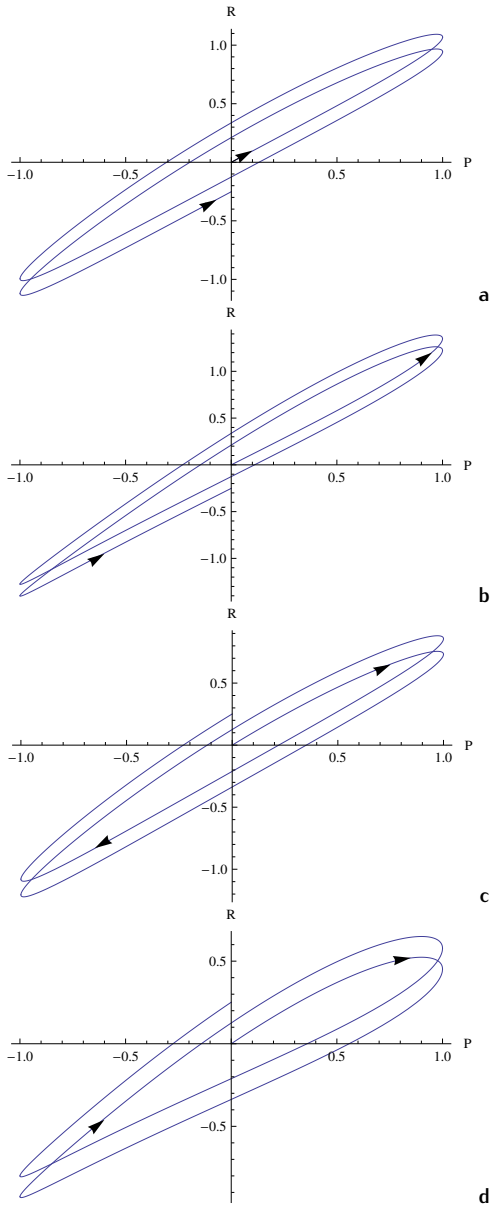


**Figure 1.** Dependence of the total excess dimensionless density, $R$, on the total excess pressure, $P$, in a chemically reacting gas (a,c) and in a gas with excited vibrational degrees of molecule's freedom (b,d). Case of the periodic harmonic sound. Plots (a,b) correspond to the damping medium ($b$ is negative, $b = -0.1$), and plots (c,d) to acoustically active medium ($b$ is positive, $b = 0.1$).

where angular brackets denote average over the sound period. For the periodic harmonic wave, that yields in the leading order equality valid for both examples of thermodynamic processes in gases:

$$\frac{\omega}{2\pi}\frac{\delta U}{U_0} = -(\gamma-1)\omega b M^2. \tag{18}$$

# 4. Hysteresis curves for some impulses

## 4.1. The Gaussian impulse

If a signal may be approximately considered as Gaussian,

$$P = \exp(-(\omega\eta)^2), \tag{19}$$

where $\omega$ denotes the characteristic inverse duration of an impulse, $\eta = t - x/c$ is the retarded time, Eq. (12) rearranges into

$$R = P - M\frac{\gamma-1}{2}P^2 - 2b\int_{-\infty}^{\eta} Pd(\omega\eta) + \int_{-\infty}^{\eta} Q_a d\eta, \tag{20}$$

where

$$\eta = \begin{cases} -\dfrac{\sqrt{-\ln P}}{\omega}, & \text{if P enlarges,} \\ \dfrac{\sqrt{-\ln P}}{\omega}, & \text{if P decreases.} \end{cases} \tag{21}$$

The total variation in the internal energy is identical in both cases of thermodynamic processes in gases, and equals

$$\frac{\delta U}{U_0} = -\int_{-\infty}^{\infty} Q_a d\eta = -(\gamma-1)\sqrt{2\pi}bM^2. \tag{22}$$

The curves in Fig. 2 are plotted for $M = 0.5$, $\gamma = 1.4$. It is remarkable that the total density increases after passing of a positive pulse both when $b$ is negative and positive. That reflects the integral link between acoustic pressure and density which is larger in order than the acoustic source of the entropy mode. For periodic harmonic sound, it is compensated over the whole period of sound. Nevertheless, the variations in internal energy are still determined exclusively by the acoustic source, $Q_a$. The next section considers a symmetric bipolar impulse for which the integral link becomes compensated similarly as it happens for sinusoidal sound.

## 4.2. An example of bipolar impulse

The impulse in the form

$$P = \sin(\omega\eta), \quad \text{if} \quad -\pi \le \eta \le \pi, \tag{23}$$

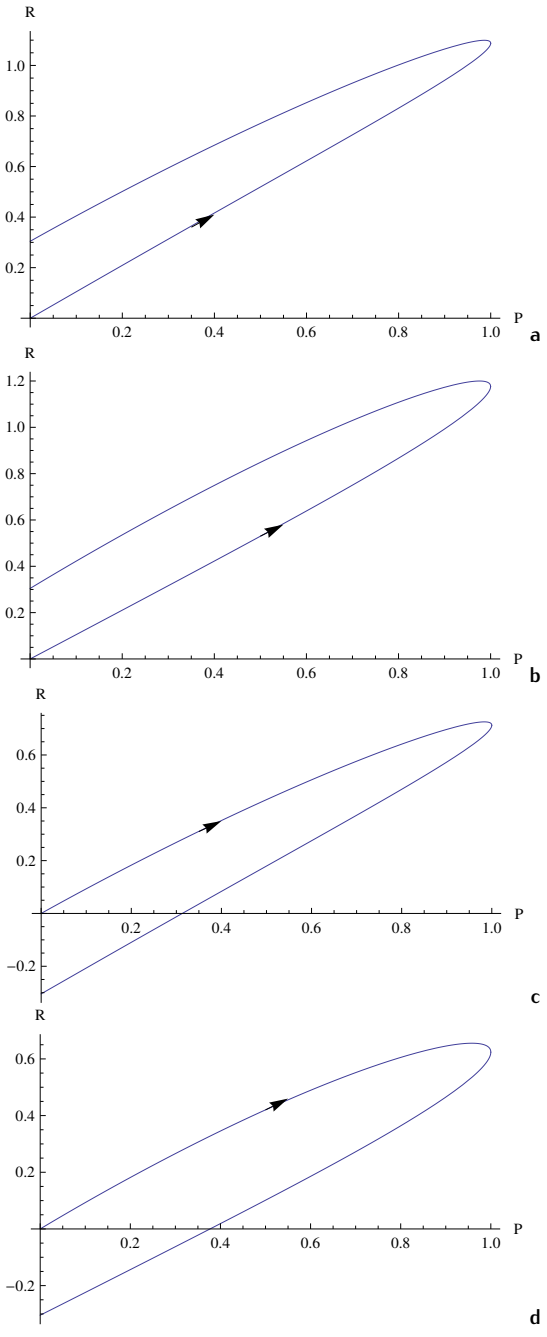


**Figure 2.** Dependence of the total excess density on the total excess pressure in a chemically reacting gas (a,c) and in a gas with excited vibrational degrees of molecule's freedom (b,d). Case of the Gaussian impulse. Plots (a,b) correspond to the damping medium ($b$ is negative, $b = -0.1$), and plots (c,d) to acoustically active medium ($b$ is positive, $b = 0.1$).

and zero otherwise, is asymmetric. Eq. (12) transforms into Eq. (20) with correspondent acoustic source and

$$\eta = \begin{cases} \dfrac{\arcsin P}{\omega}, & \text{if P enlarges,} \\ \dfrac{-\pi - \arcsin P}{\omega}, & \text{if P decreases and negative,} \\ \dfrac{\pi - \arcsin P}{\omega}, & \text{if P decreases and positive.} \end{cases} \tag{24}$$

The hysteresis curves in the Fig. 3 are plotted accordingly to the same set of parameters as in the previous subsections.

The total relative increase in the internal energy which associates with irreversible loss in acoustic energy, in both cases equals

$$\frac{\delta U}{U_0} = -\int_{-\infty}^{\infty} Q_a \, d\tau = -2(\gamma - 1)\pi b M^2. \tag{25}$$

An efficiency of the medium heating due to nonlinear attenuation is more than $2,5$ times larger in the case of the bipolar impulse as compared to the Gaussian one.

## 5. Concluding remarks

In this study, we do not consider thermal conductivity and mechanical viscosity of Newtonian fluids in order to conclude about hysteresis due to pure relaxation. In fact, accounting for thermal conductivity would lead to the additional term in the link of acoustic pressure (that is, total excess pressure) and total excess density, which takes the leading-order form as follows,

$$\begin{aligned} \frac{p_a}{\rho_0 c^2} &= \frac{p'}{\rho_0 c^2} = \frac{\rho'}{\rho_0} + \frac{\gamma - 1}{2\rho_0^2}\rho'^2 + \frac{2Bc}{\rho_0}\int^t \rho' dt \\ &- \int^t Q_a dt + \frac{(\gamma - 1)\chi}{2c^2\rho_0^2 C_p}\frac{\partial \rho'}{\partial t}, \end{aligned} \tag{26}$$

where $\chi$ is thermal conduction of a gas, and $C_p$ is its heat capacity (per unit mass) under constant pressure. The acoustic source, $Q_a$ in this equality should be corrected in view of Newtonian total attenuation [21]. In a thermoconducting gas, loops of the curve $\rho'(p')$ may form over the domains, where pressure decreases with time. Generally, hysteresis curves do not longer start from the point $(0, 0)$ [17]. A difference in the hysteresis curves between two considered examples of gases with relaxation is caused by a difference in acoustic sources of the thermal mode. That makes curves for vibrationally excited gas more asymmetric with thicker part in the positive quadrants in the plane $(R, P)$.

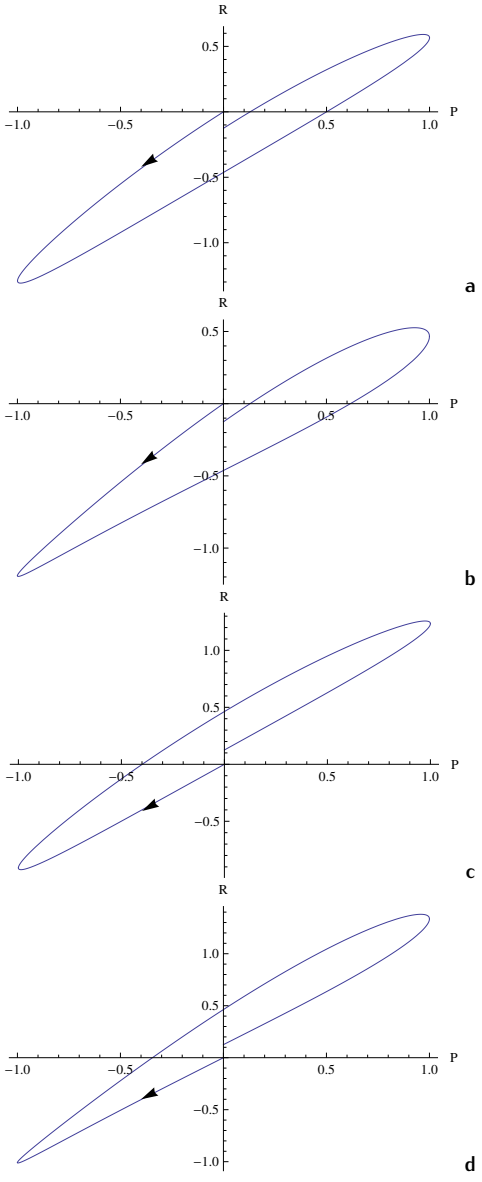


**Figure 3.** The $R \Leftrightarrow P$ diagrams in a chemically reacting gas (a,c) and in vibrationally excited gas (b,c) for the asymmetric sinusoidal impulse. Curves (a,b) corresponds to normal attenuation ($b$ is negative, $b = -0.1$), and curves (c,d) to acoustical enhancement ($b$ is positive, $b = 0.1$).

The nonlinear propagation of sound in Newtonian fluids and these with normal attenuation is always followed by irreversible loss in acoustic energy. The macroscopic wave energy transfers into the thermal energy of chaotic motion of molecules. In the plane of the thermodynamic states ($\rho'$, $p'$), that means that the total density gets smaller by the nature of the case over period of harmonic sound and after passing of an asymmetric acoustic pulse (examples of Sections 3 and 4.2). That is true for any sound pulse,

for which $\int_{-\infty}^{\infty} p_a dt = 0$ at a transducer (this condition makes the third term in the right-hand side of Eq. (12) zero after a pulse passing). In acoustically active media, attenuation and dispersion of sound are anomalous, that leads to anomalous cooling (enlargement of $\rho_e$) of the background of sound propagation instead of heating. That leads also to increase of total density in the course of propagation of harmonic sound or asymmetric sound pulse. In an example of the positive Gaussian impulse, the total density decreases in acoustically active medium. That corresponds to decrease in acoustic part of the total density due to integral "memory" link between acoustic pressure and excess acoustic density (the second term in the right-hand side of Eq. (12)), but the non-wave part of the total density, $\rho_e$, enlarges. Newtonian attenuation may prevent anomalous attenuation of sound during weakly non-equilibrium processes in a medium [7, 8].

An efficiency of heating (or cooling) of a medium depends strongly on the shape of a signal. The bipolar harmonic impulse produces larger variations of temperature of a medium after the pulse has gone, as compared with the Gaussian one. Impulses are widely used in medical and technical applications of ultrasound, where accurate estimates of the thermodynamic state of a fluid are important. The relation which describes hysteresis in a fluid, Eq. (12), is also valid in the quasi-planar geometry of slowly divergent sound beams. The hysteresis curves in the plane of thermodynamic states may be useful in reconstruction of dispersive and viscous properties of a medium [1, 17], including these anomalous and in evaluation of a degree of disequilibrium. equals the inverse time The analogous problems in optics are usually solved by laser spectroscopy [27]. Also, the remote acoustic source may be reconstructed for a medium with known thermodynamic properties. Ultrasonic relaxation makes it possible to heat or to cool a medium remotely.

First authors who have attracted attention to the pressure-volume diagrams, hysteresis curves and loops, and physical distinction between different irreversible processes accompanying nonlinear propagation of intense sound in fluids, were Rudenko, Hedberg [17]. They have outlined an analogy and difference between hysteresis in Newtonian fluids and solids. The study [17] considers loops in the plane of acoustic perturbations for periodic harmonic and saw-tooth sound in a Newtonian fluid and in a fluid with relaxation which is described by some kernel in integral link between acoustic pressure and excess acoustic density. It includes important conclusions about dissipative and hysteresis processes in linear and nonlinear acoustic fields in fluids.

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