# PROPAGATION OF HYDRODYNAMIC PERTURBATIONS IN A VIBRATIONALLY NONEQUILIBRIUM GAS

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The problem of propagation of small and finite perturbations in a continuous medium is associated with the traditional directions of classical hydrodynamics and nonlinear acoustics. Starting with the well-known studies of Riemann [1], Korteveg and De Vries [2], and others, this area has acquired substantial mathematical justification. However, the kinetic solution of specific problems of nonlinear acoustics and the theory of shock waves started substantially later, in the 1950s and 1960s of the present century. This was created, on one hand, by the appearance of sources of intense ultrasound, and, on the other, by the creation of laboratory techniques of investigating shock waves. In this connection one must note the large contribution to the development of nonlinear acoustics by Khokhlov and collaborators [3]. The method developed of slowly varying profiles [4, 5] is still at the present time one of the basic methods of investigating nonlinear interactions.

Recently the theory of nonlinear hydrodynamic perturbations has obtained new momentum. The use of large volumes of an active medium in gas lasers and the clear understanding of the nonequilibrium character of processes occurring in the upper atmospheric layers of earth and other planets generated the necessity of studying the propagation of hydrodynamic perturbations in nonequilibrium media with an excess reserve of internal energy. Along with the well-known dissipative and nonlinear effects in these media there appears a new factor, related to energy transfer from enriched internal degrees of freedom to hydrodynamic modes. Thus, the nonequilibrium medium can emerge as an amplifier of hydrodynamic perturbations. The amplifying properties of the nonequilibrium nonlinear medium change the nature of wave propagation of small and finite amplitudes: sound amplification is possible, stationary disruptions are generated and a detonation regime occurs. The purpose of the present review is also the discussion of recent achievements in this new region of hydrodynamics of a nonequilibrium gas.

1. Sound Propagation in a Vibrationally Nonequilibrium Gas. Sound propagation in a nonequilibrium gas was investigated theoretically in [6-14]. To explain the basic features of sound propagation in this medium consider the simplest case of one-dimensional propagation of a planar sound wave in a gas which is stationary, but whose nonequilibrium state undergoes pumping and heat exchange. The system of equations describing propagation of hydrodynamic perturbations in this gas consists of hydrodynamic and relaxation equations. In the simplest case one can neglect the effect of viscosity and thermal conductivity on sound absorption and consider the following model mechanism of creating a nonequilibrium stationary state [9]. The gas under investigation undergoes action of energy pumping in vibrational degrees of freedom of power I and effective heat transfer from the translational degrees of freedom of power Q (I and Q refer to unit mass). The effect of walls on sound wave propagation is neglected.

The equations of continuity, motion, and energy are in this case

$$\frac{\partial\rho}{\partial t}+\operatorname{div}(\rho\mathbf{v})=0, \tag{1}$$

$$\rho\frac{d\mathbf{v}}{dt}=-\nabla p, \tag{2}$$

$$\frac{dh}{dt}-\frac{1}{\rho}\frac{dp}{dt}=I-Q, \tag{3}$$

where $h=\gamma p/((\gamma-1)\rho)+\varepsilon$; $\gamma=(c+1)/c$.

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The relaxation equation for the vibrational energy is written in the form

$$\frac{d\varepsilon}{dt}=\frac{\varepsilon_{eq}-\varepsilon}{\tau}+I. \tag{4}$$

Further analysis is conducted in a standard manner. The full system of hydrodynamic equations, including the relaxation equation and the equation of state, describes any motion of the medium. Sound oscillations are small perturbations, and can be described by linear equations, obtained if in Eqs. (1)-(4) and the equation of state one substitutes a solution in the form

$$a+a'\exp(ikx-i\omega t), \tag{5}$$

where $a \equiv \rho$, p, T, v, $\varepsilon$; a is a parameter of the unperturbed medium, and a' is a perturbation which is assumed to be small. Substituting (5) in the full system of hydrodynamic equations makes it possible to obtain a dispersion relation, relating the frequency $\omega$ and the wave vector k. The complex wave vector value found from this relation $k(\omega) = \mathrm{Re}\,k + i\,\mathrm{Im}\,k$ corresponds to sound absorption if $\mathrm{Im}\,k > 0$, and sound amplification if $\mathrm{Im}\,k < 0$. These two regions are separated by the limiting frequency value $\omega_{cr}$, obtained from the condition $\mathrm{Im}\,k = 0$. In the simplest case for pumping and heat transfer one can use the expressions $I = I(\varepsilon)$, $Q = k_B\kappa(T - T^*)/(R\rho)$, where T is the gas temperature, T* is the wall temperature, R = V/S, V is the volume of the system, S is the area of the surface, through which heat transfer takes place [9]. In this case the frequency for which the absorption coefficient vanishes is determined by the equality

$$\omega^2_{cr}=\frac{2}{\tau^2}\,\frac{\varkappa}{R\rho I}\,\frac{k_B T}{\left(2c-T\dfrac{\partial\ln\tau}{\partial T}\right)}, \tag{6}$$

if $2c - T(\partial\ln\tau/\partial T) \gg 1$. The result (6), though obtained for a special model of pumping and heat transfer, is quite general. The critical frequency for a strong temperature dependence of the relaxation time can always decrease with increasing pumping intensity and relaxation time, and increases with increasing effectiveness of heat exchange, i.e., is determined by the reverse of vibrational (internal) energy and the rate of its transition into translational degrees of freedom.

The condition of sound amplification, obtained in [9], is

$$\frac{\left(2c-T\dfrac{\partial\ln\tau}{\partial T}\right)\dfrac{I\tau m}{k_B T}-\dfrac{\varkappa m\tau}{R\rho}-c_v}{2c(c+1)}>0, \tag{7}$$

allowing simple physical interpretation. Sound amplification is possible if following a period of the sound wave the energy flow from the outside following pumping and the transition from internal degrees of freedom, proportional to $\left(2c-T\dfrac{\partial\ln\tau}{\partial T}\right)\dfrac{I\tau m}{k_B T}$, exceeds losses due to heat exchange with the walls and relaxation processes, proportional to $\kappa m\tau/(R\rho) + c_v$.

Condition (7) for fixed pumping I is better satisfied for molecules with large vibrational quanta and a strong temperature dependence of vibrational relaxation times, since in this case $\partial\ln\tau/\partial T$ is large. For CO with $p = 10^4$ Pa, T = 500 K, T* = 300 K, $T_v$ = 700 K the imaginary part of k is $\mathrm{Im}\,k = 1\ \mathrm{m}^{-1}$. For comparison we mention that the amplification coefficient of IR-radiation in a CO-laser is of the order of $10^{-1}\ \mathrm{m}^{-1}$. In the example considered the sound amplification coefficient exceeds the absorption coefficient (in the absence of pumping) by approximately 10 times.

The existence of a critical frequency (6) leads to filtration of sound oscillations. For sound "running" through a nonequilibrium medium only those oscillations "survive" and are amplified, having a frequency higher than $\omega_{cr}$. We stress that low-frequency sound propagates quasistatically, since for $\omega \to 0$ ($\omega\tau \ll 1$, $\omega \ll \kappa m/(R\rho)$; $Im/(k_BT)$) a quasistationary state can be established.

We note that among the studies [6-14], devoted to sound amplification, the most common case was considered in [9], accounting for the action of pumping and heat exchange.

At present time there exist no direct experiments on sound amplification measurement; there exist, however, experimental results on sound propagation in a gas with the chemical reaction $H_2 + Cl_2 = 2HCl$ [15-17]. A substantial result of these experiments is a noticeable difference between sound absorption in a gas without reactions and in the process of reactions. The decrease in the sound absorption coefficient during the reaction process can be interpreted as appearance of amplification, generated by a source of a heated gas, emerging from an exothermic reaction. At the same time the amplification mechanism is determined by the temperature dependence of the reaction rate. The agreement between theory and experiment in the region of low reaction rates is quite good. For high rates the divergence reaches factors of two. The experiments in [15-17] were conducted in buffered Ar and $SF_6$ gases. To explain the experiment in the latter case it is necessary to account, besides the vibrational—translational energy exchange, for exchange of vibrational energies between HCl and $SF_6$ molecules, which at the present time has not been thoroughly investigated. It must be noted that in interpreting the results of [17], as well as in the numerical simulations [18, 19], the strong vibrational excitation of the HCl molecule has not been included at all [19, 20]. Account of this fact, as well as of the wider frequency spectrum in experiments, allows to account more completely for the acousto-kinetic interaction.

2. Description of Vibrational Nonequilibrium in Terms of Second Viscosity. Dissipative effects in a vibrationally nonequilibrium gas, explained by the retarded energy exchange between the translational and vibrational degrees of freedom, can be described under certain conditions in terms of the second viscosity.

Consider initially the propagation of perturbations in a primarily equilibrium gas. The original system of equations, similarly to (1)-(4), are conveniently written as follows:

$$\frac{\partial\rho}{\partial t} + \operatorname{div}(\rho\mathbf{v}) = 0; \quad \rho\frac{d\mathbf{v}}{dt} = -\operatorname{grad} p;$$
$$\rho\frac{ds}{dt} = -\left(\frac{1}{T} - \frac{1}{T_v}\right)\frac{d\varepsilon}{dt}; \quad \frac{d\varepsilon}{dt} = \frac{\varepsilon_{eq} - \varepsilon}{\tau}. \tag{8}$$

Unlike Eqs. (1)-(4), an entropy equation has been selected in (8) along with the energy equation. Thus, as independent variables in (8) we use ρ, v, ε, and s. The vibrational energy plays the role of the nonequilibrium parameter. Under equilibrium conditions $\varepsilon = \varepsilon_{eq}(\rho, s)$ and the last two relations in (8) are equated to zero.

The second viscosity, as shown in [21], appears only in a special class of solutions of the equations of relaxational hydrodynamics (8). These solutions are characterized by the fact that the relatively fast relaxation process is assumed completed in the zeroth approximation, and local equilibrium at each moment of time is completely determined by the fields of hydrodynamic quantities at the same moment of time. Accordingly, the following expansions for ε and p are utilized:

$$\varepsilon = \varepsilon^{(0)}(\mathbf{r}, \rho, s, \mathbf{v}) + \mu\varepsilon^{(1)}(\mathbf{r}, \rho, s, \mathbf{v}) + \ldots, \tag{9}$$

$$p(\rho, s, \varepsilon) = p^{(0)}(\rho, s) + \mu\left(\frac{\partial p}{\partial\varepsilon}\right)_{\rho, s, \varepsilon^{(0)}}\varepsilon^{(1)} \tag{10}$$

with similar expressions for the derivatives of hydrodynamic quantities. Here $\mu = c_S\tau/L$. Following the determination of $\varepsilon^{(1)}$ from the linearized system (8), we obtain an equation of motion in the required form with the second viscosity:

$$\rho d\mathbf{v}/dt = -\operatorname{grad} p^{(0)} + \operatorname{grad}(\xi \operatorname{div}\mathbf{v}), \tag{11}$$

where

$$\xi = -\frac{\partial p}{\partial\varepsilon}\,\tau\,\frac{\partial\varepsilon^{(0)}}{\partial\rho}\,\rho \tag{12}$$

or $\xi = \tau\rho(c_\infty^2 - c_0^2)$. Here $c_0$ and $c_\infty$ are the propagation rates of low-frequency and high-frequency sound.

The transition in the equations of relaxation gas dynamics to the description in terms of the second viscosity implies contraction in the method of description; instead of the four variables $\rho$, s, v, and $\varepsilon$ there occur three — $\rho$, s, and v. The abbreviated description is reached with the purpose of coarsening the time scale. Processes occurring during times of the order of relaxation times are not considered; only their result, leading to expansion (9), is important.

Sound amplification in nonequilibrium media (a negative absorption coefficient) made it possible to suggest the existence of a negative second viscosity coefficient in a vibrationally nonequilibrium gas [12]. On a qualitative level the consideration is the following. The absorption coefficient in the case of a plane wave $\exp(i\omega t \mp ikx)$ is determined by the quantity Im k. The same plane wave in a medium with bulk viscosity decays by the law $\exp(-\tilde{\gamma}x)$, where $\tilde{\gamma} = \omega^2\xi/(2\rho c_S^3)$. Equating the last two expressions, we obtain $\xi = -2\rho c_S^3 \operatorname{Im} k/\omega^2$ or $\xi = -\rho \operatorname{Im}(\omega^2/k^2)/\omega$.

A rigorous derivation of the expression for the second viscosity coefficient in a primarily nonequilibrium medium, not restricted by the acoustic approximation, and based on the general idea of abbreviating the method of description, as discussed earlier, is given in [22]. We note the basic features of the derivation of the second viscosity coefficient in a primarily nonequilibrium gas. The equation for the entropy variation in a nonequilibrium gas with pumping and heat transfer is

$$\frac{ds}{dt} = \left(\frac{1}{T_v} - \frac{1}{T}\right)\frac{\varepsilon_{eq} - \varepsilon}{\tau} + \frac{I}{T_1} - \frac{Q}{T}, \tag{13}$$

whence it is seen that the entropy variation may be of zeroth order of smallness in $\mu$ due to the term $I/T_1 - Q/T$. Thus, the advantage of entropy as one of the independent variables vanishes, since the entropy is no longer constant with an accuracy within second order terms. Along with $\rho$, s another possible choice of independent variables can be $\rho$, E, where E is the total energy of a unit mass. In a primarily equilibrium gas the replacement of $\rho$, s by $\rho$, E does not change anything, since for E = const the entropy is constant within second order terms. In a nonequilibrium gas the constancy of E does not guarantee s = const. Therefore, the $\varepsilon^{(0)}(\rho, s)$ and $\varepsilon^{(0)}(\rho, E)$ values, being the vanishing approximation in expansion (9), are equal, and consequently, the expressions for the second viscosity coefficient are also different. Thus, the choices of variables $\rho$, s and $\rho$, E in a nonequilibrium gas are nonequivalent, since they correspond to different partitions of $\varepsilon$.

The derivation of expressions for the second viscosity coefficient, carried out in [12] on the basis of harmonic analysis, uses as independent variables $\rho$ and E. The nonequilibrium system with pumping and heat transfer is characterized, along with the relaxation time $\tau$, by the effective pumping and heat transfer times, the shorter of which we denote by $\tau_{sou}$. In satisfying the conditions $\omega\tau \ll 1$ and $\omega\tau_{sou} \gg 1$ the intensity of sources (heat transfer and pumping) can be assumed constant. In this case the scheme suggested for calculating the second viscosity coefficient is conserved. Instead of the entropy equation it is only necessary to use the equation for the total energy. The following expression is obtained for the second viscosity coefficient:

$$\xi = \frac{k_B\rho\tau}{m}\,\frac{p^{(0)}(c_v + cI\tau_E)/\rho^2 - cI\tau_\rho}{(c_v + c - c\tau_E I)^2}, \tag{14}$$

where $\tau_E$ and $\tau_\rho$ are the derivatives of $\tau$ with respect to E and $\rho$.

The situation changes for $\tau \sim \tau_{sou}$. In this case, following time $\tau$ not only the fast relaxation process is concluded in the zeroth approximation, but also the fast process of source synchronization with instantaneous hydrodynamic fields. This leads, in turn, to a change in the pressure expression, acquiring the form

$$p = p^{(0)}(\rho) + \mu\frac{\partial p}{\partial \varepsilon}\varepsilon^{(1)} + \mu\frac{\partial p}{\partial E}E^{(1)} + \ldots, \tag{15}$$

where, besides $\varepsilon = \varepsilon^{(0)} + \mu\varepsilon^{(1)} + \ldots$, the expansion $E = E^{(0)} + \mu E^{(1)} + \ldots$ is satisfied. The corresponding second viscosity coefficient has a quite awkward form.

We note two substantial differences in the second viscosity properties of primarily equilibrium and nonequilibrium gases.

Firstly, in deriving the second viscosity coefficient we have further abbreviation of the method of description. Along with the relaxation process, the source synchronization process with hydrodynamic fields is not considered either. In the first case ($\omega\tau_{sou} \gg 1$) this process is assumed frozen, and in the second case ($\omega\tau_{sou} \ll 1$) it is very fast, and only its result is accounted for.

Secondly, the second viscosity coefficient, as well as the absorption coefficient, loses its primary meaning as a characteristic of the medium, since it starts depending on the characteristics of heat exchange and pumping.

The second viscosity coefficient, as well as the absorption coefficient, can acquire negative values [see Eq. (14)]. This fact does not contrdict the second law of thermodynamics, according to which the entropy production $\sigma$ is always positive or equal to zero. Indeed, in the case considered

$$\sigma = (1/T_v - 1/T)(\varepsilon_{eq} - \varepsilon)/\tau, \tag{16}$$

and it is larger than or equal to zero independently of the sign of $\xi$. In Eq. (16) one can isolate the term related to $\xi$:

$$\sigma = \sigma_0 + \tilde{A}\xi \operatorname{div} \mathbf{v}. \tag{17}$$

Unlike ordinary thermodynamics, where $\sigma = \xi(\operatorname{div} v)^2$, the negative $\xi$ value does not change the sign of $\sigma$ [in Eq. (17)], since the additional term in the entropy production, proportional to $\xi \operatorname{div} v$, is small in comparison with the entropy production under unperturbed stationary conditions $\sigma_0$.

3. Nonlinear Hydrodynamic Waves in Vibrationally Nonequilibrium Gas. The propagation of sound oscillations in a vibrationally nonequilibrium gas, can, as shown in Sec. 1, be accompanied by amplification. With the flow of time the amplitude of these waves reaches a finite value, and the further evolution of oscillations must already be described by a system of nonlinear equations.

The nonlinear equations of motion (the Euler equations) without assuming smallness of oscillations were first integrated by Riemann in 1860 [1]. Riemann started from the equations of hydrodynamics in the one-dimensional case [see the first two equations of system (8)], augmented by the equation of state $p = p(\rho)$. The Riemann method consists of the fact that by interchanging dependent and independent variables the nonlinear hydrodynamic equations become linear. This can be done in the one-dimensional case. The method of integrating the linear differential equation obtained, suggested by Riemann, became the prototype for solving the general problem of integrating second order linear equations in partial derivatives of hyperbolic type.

The Riemann solution for the adiabatic equation of state $p = p_0(\rho/\rho_0)^\gamma$ is (for a wave traveling to the right)

$$v = \Phi\left(t - x\Big/\left(c_S - \frac{\gamma+1}{2} v\right)\right). \tag{18}$$

Here the sound velocity is $c_S = \sqrt{\partial p/\partial \rho} = (c_S)_0 + (\gamma - 1)v/2$, and the shape of the function $\Phi$ is determined by the condition $v = \Phi(t)$ at $x = 0$. Expression (18) shows that relative to a fixed coordinate system the perturbation of the medium, corresponding to a fixed value of the velocity v, moves with velocity

$$\mathbf{u} = (c_S)_0 + \frac{\gamma+1}{2} \mathbf{v}, \tag{19}$$

i.e., the displacement rate of different points of the profile is different. Both positive and negative v values are possible for a harmonic excitation ($\Phi$ is a sinusoidal function). The case $v > 0$ corresponds to the region of compression, and $v < 0$ corresponds to dilatation. It is seen from expression (19) that the compression region moves with velocities $v > c_S$, while the dilatation region moves with velocities $v < c_S$. Thus, the original wave profile gets deformed with the propagation. The wave front becomes steeper until the condensation wave turns into a shock wave. Further treatment stops being valid, because neither viscosity nor thermal conductivity, whose rôle increases with gradients (front steepness), are accounted for.

In the Riemann solution, as we saw, the nonlinearity is not assumed to be small. However, from the physical point of view the statement of the problem according to Riemann is restricted by the validity region of the equation of state $p = p(\rho)$. An adiabatic equation of state describes isoentropic processes, while the real entropy variation for acoustic waves is of third order of smallness (in the Mach number $M_S = v/c_S$, being for nonlinear sound waves $M_S \sim 10^{-2}$-$10^{-4}$). Thus, the Riemann solution describes correctly the behavior of an intense sound wave within the second approximation [23-25].

A simultaneous account of nonlinear and dissipative effects can be carried out by means of the Burgers equation [23-25]. However, it is not accurate, since it cannot be obtained from the equations of hydrodynamics of a viscous fluid without further simplifications. It also describes quite well nonlinear waves in media with dissipation, since it includes all basic terms of second order of smallness.

Denoting by primes deviations of hydrodynamic quantities from their stationary values, the Burger's equation for p' is

$$\frac{\partial p'}{\partial x} - bp' \frac{\partial p'}{\partial \theta} - \tilde{\alpha} \frac{\partial^2 p'}{\partial \theta^2} = 0. \tag{20}$$

Here $\theta = t - x/c_S$, the dissipative coefficient is $\tilde{\alpha} = \left[\frac{4}{3} \eta + \xi + x\left(\frac{1}{c_m} - \frac{1}{c_{pm}}\right)\right]/(2c_S^3 \rho)$, and the nonlinear parameter is $b = (\gamma + 1)/(2c_S{}^3\rho)$. The equation for v has also the same shape, since for simple waves $\rho = \rho(v)$, $p = p(v)$.

The Burgers equation for small nonlinearity transforms to the well-known linear equation for media with dissipation, and in the absence of dissipation – to the Riemann equation for simple waves [23-25].

The quantitative pattern of evolution of a sound wave of finite amplitude, described by the Burgers equation, looks as follows. At the first phase of the wave propagation process there occurs distortion of the profile shape, consisting of formation of a sawtooth signal. At the second phase the front shape is stabilized. An "equilibrium" starts in the competition of nonlinear and dissipative effects. However, the oscillating signal value decreases, since the action of dissipative effects is not compensated. At the third phase the wave amplitude no longer depends on its input value. The wave again becomes harmonic, and decays by the laws of linear acoustics. The solution of the Burgers equation, describing the evolution of an initial harmonic perturbation, was obtained by Khokhlov [25].

During the propagation of nonlinear hydrodynamic gases with excess values of vibrational energies there appears a new factor – energy exchange between vibrational degrees of freedom and hydrodynamic motion. Further energetic "maintenance" of the wave process can lead to two new effects. Firstly, the process of wave reversal is accelerated. Wave amplification in a vibrationally nonequilibrium gas enhances the role of the nonlinear factor, and the nonlinear effects appear earlier. Secondly, energy pumping starts competing with dissipative processes. This can lead to the fact that following wave reversal a stationary regime is formed, for which dissipative losses are totally compensated by energy influx from internal degrees of freedom.

The quantitative description of the evolution of nonlinear hydrodynamic perturbations in a vibrationally nonequilibrium gas was given in [26, 27]. The starting point is the generalized Burgers equation (for $\mu \gg 1$):

$$\frac{\partial p'}{\partial x} - bp' \frac{\partial p'}{\partial \theta} - \alpha \frac{\partial^2 p'}{\partial \theta^2} - \beta p' = 0, \tag{21}$$

which differs from (20) by the term $\beta p'$, taking into account wave amplification due to energy exchange between internal degrees of freedom and hydrodynamic modes. In Eq. (21)

$$p = -\rho c_S \left[\frac{T}{p} \frac{c_v}{c+1} + \frac{I}{\tau} \frac{\partial \tau}{\partial p} - \frac{1}{\tau} \frac{\partial Q}{\partial p}\right]/[2(c+1)\tau T].$$

The generalized Burgers equation (21) is obtained from the equations of continuity and motion, supplemented by the relaxation and entropy equations (2) and (13) within the approximation $\omega\tau \gg 1$ accurately up to second order terms (inclusively) in small hydrodynamic perturbations. In the frequency region $\omega\tau \gg 1$, where the generalized Burgers equation (21) is valid, the dissipative (viscous), nonlinear, and relaxation effects are differently manifested. The ratio of the viscous term $\alpha(\partial^2 p'/\partial\theta^2)$ to the relaxation term $\beta p'$ in (21) is of the order of $\omega^2\tau\tau_*$. Thus, at frequencies

$$\frac{1}{\tau} \ll \omega \ll \frac{1}{(\tau\tau_*)^{1/2}} \tag{22}$$

the relaxation effects are predominant over the viscous terms, since in (21) one can neglect the third term, describing Stokes (viscous) absorption. The quasilinear equation obtained in this case is relatively simple to analyze [26]. If a perturbation propagates in the medium, whose profile at $x = 0$ is

$$p'(0, \theta) = f(\theta), \tag{23}$$

then [28]

$$p' = \exp(\beta x)\, f(z), \quad \theta - z = bf(z)\,(1 - \exp(\beta x))/\beta. \tag{24}$$

The solution (24) describes wave amplification (damping) and the distortion of its profile. The latter is easily verified by considering the case $\beta x \ll 1$. In this approximation

$$p'(x, \theta) = f(\theta - x(1/c_S - bp'))$$

and relative to an immobile medium the fixed $p'$ value will propagate with velocity $c_S^* = (1/c_S - bp')^{-1}$, which is larger the larger $p'$ is. The distance at which wave reversal occurs and a discontinuity is formed, determined by the condition $dp'/dx = \infty$, equals

$$l_p = \ln[1 + \beta/(bf')]/\beta. \tag{25}$$

It follows from (25) that a discontinuity is formed only for $\beta/(bf') > -1$. For positive $\beta$, corresponding to an excess vibrational energy in a nonequilibrium gas, this condition is always satisfied. For $\beta < 0$ a discontinuity is formed only when the profile is quite steep $f' < -\beta/b$.

A distortion of the wave profile in the propagation process changes the harmonic content of the perturbation. If $f(z) = p_0' \sin(\omega z)$, the solution of (24) acquires the form

$$p' = p_0' \exp(\beta x) \sum_{n=1}^{\infty} \frac{2J_n(n\Delta)\sin(n\omega\theta)}{n\Delta}, \quad \Delta = \frac{(\gamma+1)\,\omega p_0'}{2\gamma c_S p}\,\frac{\exp(\beta x) - 1}{\beta}. \tag{26}$$

It is seen from (26) that, along with uniformly increasing amplitudes of all harmonics, proportional to $\exp(\beta x)$, we have acceleration of energy pumping from the first harmonic to the higher ones, since $(\exp(\beta x) - 1)/\beta > x$ and the discontinuity is formed faster. With increasing amplitudes of higher harmonics Stokes absorption starts playing a substantial role, therefore in the region of wave reversal it is necessary to include the discarded viscous term. Equation (21) can be solved in general form by using the Hopf–Cole replacement $p' = 2\alpha \dfrac{\partial \ln \varphi}{\partial\theta}/b$. To sum up, we obtain

$$\frac{\partial\varphi}{\partial x} - \beta\varphi \ln\varphi = \alpha\,\frac{\partial^2\varphi}{\partial\theta^2}. \tag{27}$$

In the stationary case $\varphi(\theta, x) = \varphi(\theta)$, and Eq. (27) is easily integrated:

$$\frac{\partial y}{\partial\theta} = \pm\left[\frac{\beta}{\alpha}\,(C - C\exp(-2y) - y)\right]^{1/2}, \tag{28}$$

where $y = \ln\varphi$, and the C value is determined by initial and boundary conditions.

The pressure profile in the stationary regime is determined by (28). For small y, corresponding to small $\theta$:

$$p' = \frac{2\alpha}{b}\,\frac{\partial y}{\partial\theta} = -\beta(1 - 2C)\,\theta/b. \tag{29}$$

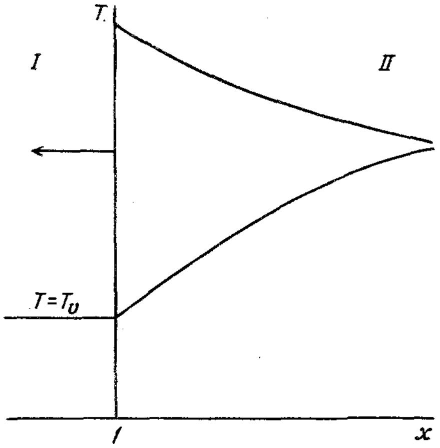

Fig. 1

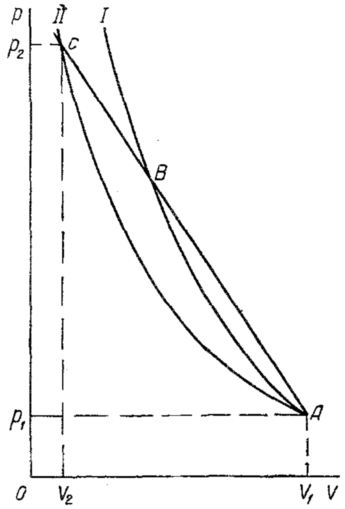

Fig. 2

Fig. 1. Temperature distribution in a shock wave. Region I is the unperturbed gas, region II is the relaxation zone, 1 is a shock wave (viscous condensation discontinuity), and the wave moves from right to left.

Fig. 2. Hugoniot adiabat. Curves I and II differ by the value of $\gamma$ ($\gamma_I > \gamma_{II}$).

Near $y_0 = \ln(2C)/2$, corresponding to the extremum of $\partial y/\partial\theta$:

$$p' = 2\left(C - \frac{1}{2} - \frac{\ln(2C)}{2}\right)(\alpha\beta)^{1/2}\cos[(\beta/\alpha)^{1/2}(\theta - \theta^*)]/b, \tag{30}$$

where $y(\theta^*) = y_0$. Finally, for $y \sim y_2$, where $\theta \sim \tilde{T}/2$ ($\tilde{T}$ is the perturbation period):

$$p' = (1 - 2C\exp(-2y_2))\,\frac{\beta}{b}\left(\frac{\tilde{T}}{2} - \theta\right). \tag{31}$$

The solutions (29)-(31) show that a primary sinusoidal signal, propagating in a vibrationally excited gas, acquires with the flow of time a sawtooth shape, and later evolves without change of shape. We stress that the discussions provided do not take into account the variation in the temperature regime of the medium, and describe the formation of discontinuities moving with a Mach number of the order of unity.

Finally, for very high frequencies, when $\omega^2\tau\tau_* \gg 1$, relaxation processes are frozen and can be neglected in comparison with Stokes absorption. In this case the propagation process is described with good accuracy by the ordinary Burgers equation (20).

The problem of generation of shock discontinuities in a nonequilibrium gas starts attracting attention in recent years. Along with [26, 27], the studies [29, 30] are devoted to this problem, where the transformation of a weak discontinuity into a shock wave has been investigated. At the same time we noted that at the present time there exist no direct experimental studies of propagation of hydrodynamic perturbations in a nonequilibrium vibrationally excited gas, though the technique of obtaining large volumes of a nonequilibrium gas is well developed.

## 4. Shock Waves in a Nonequilibrium Vibrationally Excited Gas.

The propagation of shock waves in diatomic or polyatomic gases is accompanied by gas heating. The heating process has a two-step character (at least for shock waves, at whose fronts there are no chemical reactions and ionization) [31]. Initially a temperature enhancement of translational and rotational degrees of freedom is observed at the front of the shock wave in the viscous condensation discontinuity. The width of the viscous condensation discontinuity for quite strong shock waves is of the order of the mean free path. Later, in a quite extended relaxation zone we have excitation of vibrational degrees of freedom. The temperature of vibrational degrees of freedom increases, while the translational temperature drops. All temperatures

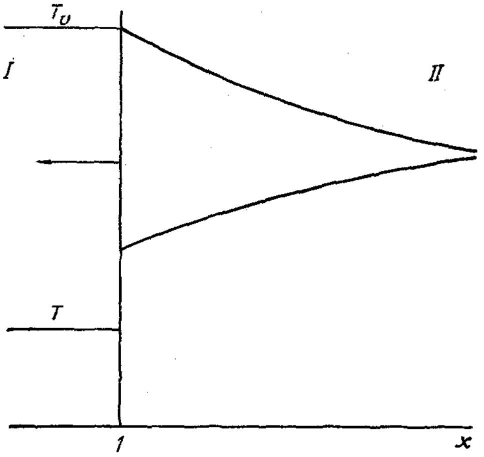

Fig. 3

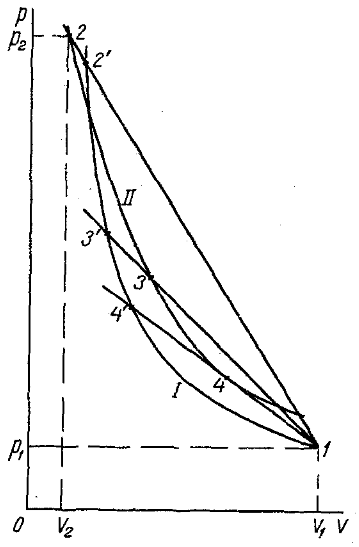

Fig. 4

Fig. 3. Temperature distribution in a shock wave, propagating in a nonequilibrium gas. Region I is an unperturbed nonequilibrium gas, region II is the relaxation zone, and the wave moves from right to left.

Fig. 4. Hugoniot adiabat for a shock wave in a nonequilibrium gas.

are equalized at the edge of the relaxation zone. The length of the relaxation zone is of the order of $v\tau$. This process is shown schematically in Fig. 1. The parameter distribution at the front of the shock wave is conveniently analyzed on a pV-diagram. The thermodyanmic parameters on both sides of the shock discontinuity are related by the conservation laws of fluxes of mass, momentum, and energy. For one-dimensional flow in a coordinate system attached to the front of the shock wave the conservation laws can be written in the form [31]:

$$\rho_1 v_1 = \rho_2 v_2; \quad p_1 + \rho_1 v_1^2 = p_2 + \rho_2 v_2^2, \quad h_1 + \frac{v_1^2}{2} = h_2 + \frac{v_2^2}{2}. \tag{32}$$

By the subscript 1 we denote parameters of the gas in the region up to the shock wave, and by the subscript 2 – following the shock wave. From relations (32) one can establish a relation between $p_1$, $V_1 = 1/\rho_1$ and $p_2$, $V_2 = 1/\rho_2$. It is of the form

$$h_1 - h_2 + (V_1 + V_2)(p_2 - p_1)/2 = 0. \tag{33}$$

Here $h_1 = h(p_1, V_1)$, and $h_2 = h(p_2, V_2)$. Similarly to the Poisson adiabat $p = p(V, s)$ relation (33) is called the Hugoniot shock adiabat. For an ideal gas

$$h = c_{pm} T = \frac{\gamma}{\gamma - 1} pV,$$

therefore (33) acquires the form

$$\frac{p_2}{p_1} = \frac{(\gamma + 1) V_1 - (\gamma - 1) V_2}{(\gamma + 1) V_2 - (\gamma - 1) V_1}. \tag{34}$$

Fixing the initial state of the gas $p_1$, $V_1$, the shock adiabat (34) determines a set of possible finite states of the gas (see Fig. 2). The specific choice of $p_2$, $V_2$ depends on the shock wave velocity $v_1$. It follows from (32) that

$$v_2^2 = v_1^2 (p_2 - p_1)/(V_1 - V_2). \tag{35}$$

Graphically this velocity is determined by the slope of the line AB, drawn from the initial state into the final one (the ratio $(p_2 - p_1)/(V_1 - V_2)$ is equal to the tangent of the inclination angle of the line). Thus, given the initial conditions $p_1$, $V_1$ and the velocity of the shock wave (the slope of the line AB), one can find the final state $p_2$, $V_2$. The position of the Hugoniot adiabat on the pV plane depends on the quantity $\gamma$. The $\gamma$ value is different at the origin of the relaxation zone and at its end. At the origin of the relaxation zone the vibrational degrees of freedom can be assumed to be "frozen," since $\tau \gg \tau_*$. Therefore, at the origin of the zone for diatomic molecules $\gamma_1 = c_{pm}/c_{vm} = 7/5$. At the end of the relaxation zone starts the state of total thermodynamic equilibrium, and for diatomic molecules at sufficiently high temperatures $\gamma_2 = 9/7$. Thus, two Hugoniot adiabats are, generally speaking, possible from the point $p_1$, $V_1$. One of them (I) corresponds to excitation of quickly relaxing degrees of freedom (translational and rotational), and describes the gas state at the origin of the relaxation zone. The second (II) determines the gas parameters at the end of the relaxation zone, i.e., in the region of achieving total statistical equilibrium. The adiabat I appears to be steeper than II, since for identical densities the pressure under conditions of frozen vibrational degrees of freedom is higher than in the state of full equilibrium, since the compression energy is distributed over a smaller number of degrees of freedom.

Thus, relaxation processes in a shock wave occur as follows according to the shock adiabat. Initially the system transforms jumpwise from state A to state B. In Fig. 2 this process is localized at the front of the shock wave I. Then the system evolves slowly from state B to state C along the segment BC. This process is localized in region II.

Consider now how the propagation of shock waves occurs in a nonequilibrium vibrationally excited gas. This problem becomes relevant in connection with the study of flow of a nonequlibrium gas in nozzles (gas-dynamic lasers), where the formation of shock waves is possible. Similar problems are also encountered during interactions of shock waves with a nonequilibrium atmosphere and with a gas discharge region.

Let a shock wave propagate through a nonequilibrium gas, with a velocity such that the translational temperature directly behind the front of the shock wave is lower than the vibrational temperature ahead of the front. This is illustrated schematically in Fig. 3. In the relaxation zone II there we have primarily vibrational–translational energy exchange processes, but they now lead to a different result. Under the conditions corresponding to Fig. 1 there occurs heating of the vibrational degrees of freedom and cooling of the translational ones. Under the conditions of Fig. 3 the process occurs in the opposite direction – the vibrational degrees of freedom are cooled, and the translational ones are heated. At first glance this seems a curious but nonessential difference. In reality the inversion in temperatures leads to principal differences.

In the problem corresponding to Fig. 1 the parameter distribution behind the shock wave is completely determined by the shock wave velocity or by the Mach number. For example, for an ideal gas the temperatures in region I ($T_1$) and in region II ($T_2$) are related by [32]

$$\frac{T_2}{T_1} = \frac{[2\gamma M_1^2 - (\gamma - 1)][(\gamma - 1) M_1^2 + 2]}{(\gamma + 1)^2 M_1^2}, \tag{36}$$

where, as usual, the Mach number M is equal to the ratio of the shock wave velocity $v_1$ to the local sound velocity in the unperturbed gas.

Under the conditions corresponding to Fig. 3 the situation changes drastically. The temperature $T_2$ now depends not only on the number $M_1$, but also on the initial conditions. Thus, the shock wave, entering the nonequilibrium gas, varies (under certain conditions) its velocity until all parameters start matching. As shown below, this is possible only for velocities exceeding or being equal to some minimum value, determined by the initial temperature distribution and by the nature of the gas.

Indeed, the conservation equations in the form (32) remain valid even in the case corresponding to Fig. 3. The single difference is that in the specific enthalpy in region I

$$h_1 = 5k_B T_1/(2m) + \varepsilon_1 + p/\rho, \tag{37}$$

The mean vibrational energy per unit mass is no longer equal to the equilibrium value determined by the temperature $T_1$. In the simplest case nonequilibrium recording of vibrational energy ahead of the front of the shock wave can be characterized by the vibrational temperature $T_{v1}$. In this case

$$\varepsilon_1 = \frac{\hbar\omega}{m}\left(\exp\left(\frac{\hbar\tilde{\omega}}{k_B T_{v1}}\right) - 1\right)^{-1}. \tag{38}$$

For $T_{V1} = T_1$ the problem reduces to the preceding one, i.e., to the problem of propagation of shock waves in a primarily equilibrium gas.

The solution of system (32) with account of (37) leads to the relation [33, 34]:

$$\frac{p_2}{p_1} = \frac{7M_1^2 - 1}{6} + \frac{7(M_1^2 - 1)}{12}\left\{\left(1 - \frac{9{,}6M_1^2 m(\varepsilon_1 - \varepsilon_2)}{7(M_1^2 - 1) k_B T_1}\right)^{1/2} - 1\right\}. \tag{39}$$

Here $p_2$ and $\varepsilon_2$ refer to the edge of the relaxation zone, and the Mach number was calculated for sound velocities with frozen oscillations.

It is seen from relation (39) that stationary shock waves exist for

$$\left(\frac{M_1 - 1}{M_1}\right)^2 \geqslant \frac{9{,}6m(\varepsilon_1 - \varepsilon_2)}{7k_B T_1}. \tag{40}$$

Under usual conditions, i.e., for shock wave propagation in an equilibrium gas, $\varepsilon_2 > \varepsilon_1$ (see Fig. 1), therefore inequality (41) is satisfied for any $M_1$. On the other hand, for shock waves in a vibrationally excited gas under the condition $\varepsilon_2 < \varepsilon_1$ (see Fig. 3) there exists a minimum Mach number, whose value is determined by the initial conditions and by the gas properties. For example, for HF with the initial conditions $T_1$ = 300 K, $T_{V1}$ = 2000 K, $p_1$= 5 the minimum number is $M_1$ = 1.8. At the same initial translational temperature and pressure at $T_{V1}$ = 3000 K the minimum number $M_1$ equals 2.6. The occurrence of a minimum Mach number can be interpreted on the basis of detonation representations.

Consider the process of variation of gas parameters in a shock wave, propagating in a primarily nonequilibrium gas on a pV-diagram (Fig. 4). Curve I in the figure corresponds to the Hugoniot adiabat for a gas with frozen vibrational degrees of freedom. The point $p_1V_1$ corresponds to the initial conditions. The adiabat I describes the state of the gas directly behind the front of the shock wave at the origin of the relaxation zone. One may construct the shock adiabat for the equilibrium state of the gas at the end of the relaxation zone. It is primarily described by Eq. (33) with the only difference that $h_1$ is determined by relation (37) with the nonequilibrium value $\varepsilon_1$, and $h_2$ – by the equilibrium value $\varepsilon_2$. The last fact is significant, since $h_2$ does not transform to $h_1$ at $p_2 = p_1$, $V_2 = V_1$; therefore the equilibrium adiabat (curve II) occurs above the frozen one. The point of adiabat intersection corresponds to the $p_2$ and $V_2$ values, for which the $\varepsilon_2$ equilibrium value equals $\varepsilon_1$. The transition from the state $p_1V_1$ to the state $p_2V_2$, located above the adiabat intersection point, occurs as in the equilibrium case. Initially, at the shock front the gas transforms with a jump from state 1 to state 2' on the frozen adiabat, then in the relaxation zone the gas is further preheated, and its pressure is enhanced. Another situation arises if the final state 3 is located below the adiabat intersection point. As in the preceding case, the gas transforms jumpwise into state 3' on the frozen adibat. Then, in the relaxation zone, the gas reaches state 3, while the parameter variation on the path 3'-3 is opposite to that observed during the transition 2'-2; more precisely, in the relaxation zone on the path 3'-3 the translational temperature increases, while the pressure and density drop. The minimum Mach number for which stationary propagation of shock waves is possible is determined by the slope of the tangent to the equilibrium adiabat, traced from point 1.

The detonation regime of shock wave propagation in a nonequilibrium gas is generated by the same physical reasons as for detonation in a reacting gas. The similarity between nonequilibrium chemically reacting and vibrationally excited gas was noted in [34]. The structural problem of stationary shock waves, propagating in a gas with vibrational and dissipative equilibrium, was first solved in [31, 32, 35, 36], and the conditions of generating the detonation regime were formulated. Numerical estimates were also carried out in [32, 33] for the structure of shock waves in the regime of recompressed detonation. The numerical calculation of the relaxation zone in this regime was carried out in [37]. It must be noted, however, that at the present time there exists no rigorous solution of the problem of shock wave evolution in a nonequilibrium gas. There also exist no direct experiments on studying the behavior of shock waves in a vibrationally excited gas. The latter fact is, obviously, of timely nature, since this region is reached by experimental studies.

The nonstationary problem of shock wave propagation in a nonequilibrium gas was investigated in [38, 39]. Unlike usual detonation in a chemically reacting gas with an Arrhenius rate constant the vibrational relaxation time depends on temperature according to a different law. For anharmonic molecules, in particular, this dependence can be opposite that of Arrhenius or Landau–Teller. An increase of the vibrational relaxation time with temperature leads, as shown in [38, 39], to the appearance of a quasistationary regime of shock wave propagation, preceding the transition to spontaneous detonation. Numerical estimates for nitrogen were carried out in [38, 39]. In this connection we note that due to the slow dependence of vibrational relaxation process in nitrogen the emergence time at the stationary regime at not too high temperatures is of the order of 1 [38]. Therefore, this is not very convenient for experimental studies of the detonation region of shock wave propagation.

Conclusion. Nonlinear acoustics as a self-contained branch of physical hydrodynamics was formulated fairly recently. This happened approximately 25-30 years ago. A still younger area of nonlinear acoustics deals with wave propagation problems in nonequilibrium media. It has increased substantially in recent years. As seen from this review, only first steps have been taken so far. The main result in this region at the present stage of development consists of clarifying the role of the nonequilibrium factor, playing as important a role as nonlinear and dissipative effects.

Hydrodynamics of nonequilibrium media as a generalization of nonlinear acoustics has made only first steps. At the present time it is difficult to estimate not only possible results, but even the field of activity. Nonlinear acoustics, as well as all of physical gas dynamics, was generated at the junction of various sciences. Therefore, its successes and range of interests are determined by successes and interests of adjacent sciences. Today the development of laser physics has initiated the problem of studying acoustic perturbations in actual media. Other problems will be generated tomorrow. In this connection it is difficult to enumerate even the basic problems requiring solution. Nevertheless, several of them already stand out. Among these problems are the study of physicochemical processes in intense acoustic waves, including also laser generation, the interaction of acoustic and shock waves with shock waves with the inclusion of nonequilibrium regions both behind and ahead shock waves, self-focusing of acoustic waves in nonequilibrium media, etc.

At the present time it is difficult and sometimes simply impossible to estimate the scientific and practical importance of some effect or another in nonlinear acoustics. At the same time it is already quite clear now that nonlinear acoustics of nonequilibrium state, as well as all of physical gas dynamics, is of explicit practical importance. From the very start they were formulated on the basis of solving practical problems, and this intimate connection with practice and technology has been conserved so far.

## NOTATION

I, pumping intensity in vibrational degrees of freedom per unit mass; Q, intensity of heat removal from translational degrees of freedom per unit mass; $\rho$, gas density; p, pressure; $\varepsilon$, mean vibrational energy per unit mass; c, heat capacity of translational–rotational degrees of freedom in molecular calculations at constant volume in units of the Boltzmann constant; $\gamma$, adiabatic index; $\varepsilon_{eq}$, equilibrium value of $\varepsilon$; $\tau$, vibrational relaxation time; T, translational temperature; $k_B$, Boltzmann constant; k, wave vector; $\omega$, frequency; $\kappa$, heat-transfer coefficient; m, molecular mass; $c_V$, heat capacity of vibrational degrees of freedom at temperature T in molecular calculations in units of the Boltzmann constant; $T_V$, vibrational temperature; $\xi$, second viscosity coefficient; $c_S$, sound velocity; E, total energy per unit mass; s, entropy per unit mass; $\tau_{sou}$, characteristic relaxation time of sources; $\sigma$, entropy production; $\lambda$, thermal conductivity; $c_{pm}$ and $c_{vm}$, heat capacities at constant pressure and volume per unit mass; $\eta$, shear viscosity coefficient; $\tau_*$, mean free path time; and L, characteristic perturbation length.

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