Stimulated sound scattering in viscous liquids

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The possibilities of the observation of universal types of stimulated sound scattering (SS) in homogeneous viscous liquids are investigated theoretically. The threshold pumping intensities, the intensity and pulse duration of the scattered wave are calculated for both the stationary and the nonstationary regimes. The conditions are estimated under which the competing processes of harmonic generation, overheating, and forced convection are suppressed.

1. INTRODUCTION

Only the elastic mechanism of nonlinearity has been studied in detail in the acoustics of normal liquids ("the scattering of sound by sound").[1,2] This mechanism guarantees the direct dependence of the sound velocity on the amplitude p of the sound pressure. Much less is known about other mechanisms of nonlinearity, connected with the excitation of modes of a nonacoustical nature ("the inelastic scattering of sound"). Stimulated sound scattering (SS) in a liquid, for example, has not been investigated experimentally up to the present time, although there exists no obstacle to it in principle. At the present time, it is clear that the SS of sound should bear important information on the liquid structure and the dynamics of its internal motions. The prospects in this sense do not differ from those realized in nonlinear optics, where the investigation of Raman light scattering has already become one of the most successful methods of molecular spectroscopy and where practically all forms of SS (Raman, Mandel'shtam-Brillouin, Rayleigh) find application in the optical transformations of frequency and in the elements of optimization of powerful lasers.

The fundamental obstacle in the way of experimental investigation of stimulated sound scattering in a liquid is connected with its large quadratic nonlinearity. This pertains also to the observation of other nonlinear acoustical effects (for example, the self-action of sound beams), the magnitude of which is proportional to the intensity (and not to the amplitude) of the sound wave. Such a nonlinearity, in the absence of the frequency dispersion that is a characteristic of the frequency range up to 1–10 GHz, leads to the effective generation of harmonics that form a shock wave, and which roughly speaking does not leave energy for other nonlinear processes. We note that the situation is directly the opposite in the nonlinear optics of liquids.[4] In the first place, there is the anomalously small nonlinearity, which is quadratic in the electric field intensity **E**, and which in such media can be assured only by a relatively weak spatial dispersion. Second, the strong frequency dispersion in the optical system destroys the phase agreement in the processes of frequency multiplication.

The narrow band nonlinear transformations in acoustics can be realized by introducing artificial dispersion (possible methods have been described in Refs. 1 and 2), i.e., by extending the spatial scale L_h that characterizes the effectiveness of harmonic generation. An alternative to this lies in the assuring of conditions under which the investigated process (in our case, stimulated scattering) develops over scales L_s that are less than L_h. In practice, this reduces to the choice of the medium and to the optimization of the geometry of the experiment. Questions of optimization form the subject of a separate paper. In the present paper, we shall calculate the amplification parameters for different types of stimulated scattering of sound and estimate the conditions of suppressing competing processes—harmonic generation, heating of the liquid, convective mixing.

2. MECHANISM OF STIMULATED SCATTERING OF SOUND IN HOMOGENEOUS LIQUIDS; THRESHOLD PARAMETERS OF THE SCATTERING

1. A closed description of the nonlinear acoustic effects in a homogeneous liquid is provided by a set of equations, including the equation of continuity

$$\partial_t\rho+\partial_b(\rho v_b)=0, \tag{1}$$

the equation of balance of forces

$$\rho(\partial_t+v_b\partial_b)v_a+\partial_a p=\eta\partial_b^2 v_a+(\xi+\eta/3)\partial_a\partial_b v_b, \tag{2}$$

the equation of entropy production

$$\rho T(\partial_t+v_b\partial_b)\mathscr{S}$$
$$=\varkappa\partial_b^2 T+\xi(\partial_b v_b)^2+\eta\partial_a v_b[\partial_a v_b+\partial_b v_a-2\delta_{ab}\partial_c v_c/3], \tag{3}$$

and the equation of state. The latter isolates a pair of independent thermodynamic variables: $p=p(\mathscr{S},\rho)$, $T=T(\rho,\mathscr{S})$. Here ρ, $\mathscr{S}$ and T are the density of the liquid, its specific entropy, and the temperature; v_a is the local velocity of the liquid, η, ξ and $\varkappa$ are the coefficients of shear viscosity, bulk viscosity and thermal conductivity.

The dispersion equation for the system (1)–(3), linearized about the equilibrium state (we shall denote the equilibrium values by the subscript "0"), has five plane-wave solutions, $k_i=k_i(\omega)$, k_i are the wave numbers, ω is the frequency. For a liquid with moderate viscosity and thermal conductivity, at real values of the parameters, two of these five modes are acoustic, with mutually opposed directions of propagation. The three remaining modes are diffusive in character. The three possible forms of the stimulated scattering of sound are due just to their excitation by sound. This set of stimulated scattering, which is complete for the single-component homogeneous liquid, consists of the stimulated scattering from temperature waves (STW), the stimulated scattering by vortex waves, which has a purely hydrodynam-

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ic origin, and stimulated scattering by waves of longitudinal acoustic flow (WLAF). The first common feature of these types of stimulated scattering is the direct connection with dissipative processes, either through the sound absorption coefficient δ or directly through the viscosity. The second common feature is the phenomenological closeness to the Rayleigh stimulated scattering of light—the small frequency shift, and a linewidth $\Delta\omega$ that is close to it in order of magnitude.

In the first theoretical works only the stationary amplification coefficients were considered and estimated for the stimulated scattering via the vortex[5] and temperature[6,7] and arbitrary flow[8] mechanisms. Only in Refs. 7 and 8 was a procedure indicated for nonstationary calculations. However, as shown in Ref. 9 by the example of another effect that is cubic in the amplitude—the self-action of a sound beam—the characteristic times of the nonlinear processes here are such that with modern sound sources the effect can be observed more readily just in the nonstationary regime. Therefore, along with a consistent account of dissipation, we shall take explicitly into account the finite rates of development of the stimulated scattering.

2. As in the analysis of self-action,[9] we shall use a quadratic expansion of the equation of state in the departures of p and T from their equilibrium values. The relative estimates of the individual terms of this expansion are taken from Ref. 9. The term corresponding to harmonic generation ($\sim p^2$) is omitted, since we have in mind a comparison, at the end of the calculation, of the corresponding scale L_h with the inverse growth rates of the amplification.

We represent the sound field by the superposition of the pump wave (p_p) and the scattered wave (p_s):

$$p=(p_p/2)\exp[i(\omega_p t-\mathbf{k}_p\mathbf{r})]+(p_s/2)\exp[i(\omega_s t-\mathbf{k}_s\mathbf{r})]+\text{c. c.} \quad (4)$$

We also represent the nonacoustic modes in the form of traveling waves with frequency $\Omega=\omega_p-\omega_s$ and wave vector $\mathbf{q}=\mathbf{k}_p-\mathbf{k}_s$:

$$T-T_0=(T'/2)\exp[i(\Omega t-\mathbf{qr})]+\text{c. c.}, \quad (5)$$

and similarly for the velocity of the longitudinal flow $\mathbf{u}$ (curl $\mathbf{u}=0$) and amplitude of the vortex mode $\mathbf{w}=$ curl $\mathbf{u}$.

The standard truncation procedure[4] yields, first, an equation describing the amplification of the scattered wave in the field of the given dissipating pump (for definiteness, propagating along the x axis):

$$i\{(\mathbf{k}_s\nabla)+(\omega_s/c_0^2)\partial/\partial t+\delta k_s\}p_s=-(\omega_p p_p/2c_0^2)(k_s\mathbf{u}^*)$$
$$+(\gamma\omega_s^2 p_p/4c_0^2)T'^*-(i/4c_0)(\mathbf{k}_s[\mathbf{k}_p\times\mathbf{w}^*]);$$
$$p_p=p_0(t-x/c_0)\exp(-\delta x). \quad (6)$$

Here $\gamma=(\partial\ln c_0^2/\partial T)_p$. Second, all the truncated equations for T', $\mathbf{u}$ and $\mathbf{w}$ are of the diffusion type:

$$(\partial/\partial t-2\chi\mathbf{q}\nabla+\chi q^2+i\Omega)T'=2\delta p_p p_s^*/\rho_0^2 c_0 C_p,$$
$$(\partial/\partial t-2\nu'\mathbf{q}\nabla+\nu' q^2+i\Omega)\mathbf{u}$$
$$=(ip_p p_s^*/4\rho_0^2 c_0^2)\{\mathbf{q}-\mathbf{k}_p(k_s/k_p)-\mathbf{k}_s(k_p/k_s)\}, \quad (7)$$
$$(\partial/\partial t-2\nu\mathbf{q}\nabla+\nu q^2+i\Omega)\mathbf{w}$$
$$=\Omega(\omega_p+\omega_s)p_p p_s^*[\mathbf{k}_p\times\mathbf{k}_s]/2\rho_0^2 c_0^2\omega_p\omega_s.$$

Here

$$\chi=\varkappa(\rho_0 C_p)^{-1},\quad \nu'=(\zeta+4\eta/3)/\rho_0,\quad \nu=\eta/\rho_0.$$

Account of thermal broadening leads to the appearance on the right sides of Eqs. (7) of additional terms that are proportional to $p_p\, p_s^*$. However, they are very small in comparison with those written down in (7). Thus, in the second equation of (7), the source connected with the thermal broadening is $\alpha\delta v c_0/C_p$ times less intense than the fundamental. Here α is the coefficient of volume thermal expansion, C_p is the specific heat at constant pressure, c_0 is the velocity of sound. In liquids with a measured viscosity ($\eta\sim10^{-2}$ P), this relation amounts to 10^{-8}; even in very viscous liquids such as glycerine, it does not exceed 10^{-5}. Here and everywhere we shall give our estimates at the frequency of 1 MHz.

The estimates of Ref. 5 show that the hydrodynamical mechanism of excitation of the vortex mode in normal liquids, taken into account in Eq. (7), is a weak one and can appear only in gases. For this reason, we shall consider below only scattering from temperature waves (STW) and waves of longitudinal acoustic flow (WLAF).

3. For the stationary regime, setting the time derivatives in (6) and (7) equal to zero and neglecting small changes in amplitude in near-boundary layers with thickness of the order of a wavelength [i.e., $\nabla T'\approx0$, $\nabla\times\mathbf{u}\approx0$ in (7)], we find

$$|p_s(\theta,\Omega)|=|p_{s_0}|\exp\{g(\theta,\Omega)J(L)L-\delta L\}. \quad (8)$$

Here θ is the angle between $\mathbf{k}_p$ and $\mathbf{k}_s$, L is the thickness of the liquid layer in the direction of propagation of the scattered wave, p_{s_0} is the input amplitude of the signal at the frequency ω_s (or the mean-square noise amplitude). The factor $J(L)$ is proportional to the intensity $I=p_0^2/2\rho_0 c_0$ of the pump and depends on the geometry of the experiment:

$$J(L)=(I/2\delta L|\cos\theta|)\{1-\exp(-2\delta L|\cos\theta|)\}.$$

In the case of low absorption and in a transverse geometry of the pump, $J(L)\approx I$; with account of finite absorption, $J(L)$ has an upper bound $I/2\delta L$.

The frequency-angle dependence of the amplification is determined by the factor $g(\theta,\Omega)$. For STW, the derivative of the Lorentz contour determines the line shape:

$$g_T=(\gamma\delta k/\varkappa q^2)(\Omega/\chi q^2)[1+(\Omega/\chi q^2)^2]^{-1}, \quad (9)$$

$q\approx2k\sin(\theta/2)$, $k_s\approx k_p\approx k$. Thus, the amplification is a maximum at the shifted frequency $\Omega=\chi q^2\lesssim1$–10 Hz.

The SSAF line has the Lorentzian shape

$$g_u=(4\eta c_0^2)^{-1}[1+(\Omega/\nu' q^2)^2]^{-1} \quad (10)$$

with half-life $\Delta\Omega=\nu' q^2$ up to 100 Hz in liquids with large viscosity.

The vanishing of the total amplification coefficient determines the threshold intensity of the pump. For a frequency Ω corresponding to maximal amplification,

$$I_T=8\varkappa\omega/c_0|\gamma|,\quad I_u=4\eta c_0^2\delta. \quad (11)$$

Typical values of the threshold intensities at $\omega/2\pi=1$ MHz are the following: I_T(W·cm^2) = 75 (acetone); 83 (benzene); 180 (water); 390 (glycerin); $I_u=0.014$; 0.4; 0.17; 5.2×10^3. The thresholds (11) increase with increase in the frequency; however, they do it at different rates: $I_T\sim\omega$, $I_u\sim\omega^2$. Consequently, the WLAF dominates at low frequencies $\omega<\omega^*$.

Assuming a classical absorption mechanism, we obtain $\omega^* = 3\varkappa\rho_0/|\gamma|\eta^2$ for the limiting frequency ω^*. In benzene, for example, the WLAF is the basic form of scattering up to frequencies of the order of 10 GHz.

3. CALCULATION OF THE PARAMETERS OF NONSTATIONARY STIMULATED SCATTERING OF SOUND

1. The characteristic "proper" times of the processes of diffusion of the acoustic flow velocity and temperature are estimated from (7) as $\tau_u \sim (\nu' q^2)^{-1} \sim (10^{-2} - 1)$ s and $\tau_T \sim (\chi q^2)^{-1} \sim (10^{-1} - 1)$ s (all for a frequency of 1 MHz). However, it is not possible to assume that they are the only factors determining the rate of growth of the stimulated scattering, since the external source also participates in it (a strong sound wave). Furthermore, experiments on the observation of the phenomenologically close STW of light show[10] that the pulse of scattered radiation was frequently shorter than the pump pulse, and, as a rule, a decrease of the intensity of the scattering already occurs at the maximum of the pump.

For definiteness, we turn to the nonstationary WLAF in backscattering geometry, $\theta = 180°$ (generalization to an arbitrary geometry follows directly). We assume that the pulse length of the pump, τ, is greater than the time of flight of the sound through the liquid layer: $\tau > L/c_0 \sim 10^{-4}$ s at $L \sim 10$ cm. Upon satisfaction of this condition, which is natural for the proposed acoustic experiment, the only spatial scale of amplification in the subthreshold regime is the reciprocal growth rate $G^{-1} = (gJ)^{-1}$. The growth rate of the stimulated scattering in time is determined by a still unknown scale $\tilde{\tau}$. It is intuitively clear that the coordinate dependence of the amplification is basically established by the fast acoustic mode, and the spatial dependence by the slow barotropic mode. Relative estimates of the space and time derivatives in (6) and (7) show that this takes place at

$$1 < c_0\tilde{\tau}G < c_0^2/4\nu'\omega. \qquad (12)$$

The coefficient on the right in (12) at $\omega/2\pi = 1$ MHz ranges from 10^5 (acetone) to 10^2 (glycerin). Thus, both these inequalities must be satisfied; the left differs little from the stimulated scattering threshold condition, the right is violated only at intensities exceeding the threshold of cavitation.

2. Upon satisfaction of (12), making the substitution

$$p_s = p\exp[\delta(L-x) - (\nu' q^2 - i\Omega)t]$$

we get the following equation from (6) and (7):

$$[\partial^2/\partial x\partial t + R\exp(-2\delta x)]p = 0, \quad R = \omega^2 I/\rho_0 c_0^4. \qquad (13)$$

In the approximation (12), any of the forms of stimulated scattering enumerated above is described by a similar equation (in general, with a complex coefficient R). Satisfaction of the inequality $\tau > L/c_0$ guarantees the possibility of substitution of the arguments:

$$\xi = \{1 - \exp[-2\delta(L-x)]\}/2\delta, \quad \bar{\theta} = \int_0^t dt' R, \qquad (14)$$

which reduces (13) to an equation of the Riemann type with constant coefficients. The Riemann problem for this equation is solved exactly in quadratures. In the case of a small excess over threshold ($I = \tilde{K}I_u$, where $\tilde{K}_u$ is not more than several units), the self-similar substitution $r = 2(\xi\theta)^{1/2}$ immediately gives sufficient accuracy. It reduces (13) to the Infeld equation of zero order:

$$\frac{d}{dr}\left(r\frac{dp}{dr}\right) = rp.$$

Choosing its solution to be bounded at zero, we obtain the result that the total amplification coefficient $K(t)$ over the length L amounts to

$$K(t) = |p_s(0,t)/p_{s0}| \approx I_0[r(0,t)]\exp(-\delta L - \nu' q^2 t), \qquad (15)$$

I_0 is the Infeld function. In the nonstationary regime of amplification, a pulse of scattered wave is thus formed. Its maximum is reached at $t = \tilde{\tau}$, when $dK/dt = 0$. It is just this scale $\tilde{\tau}$ that determines the nonstationary processes: its approximate value is

$$\tilde{\tau} \approx \delta L(I/I_u)(c_0^2/8\nu'\omega^2). \qquad (16)$$

At the frequency 1 MHz and at $L = 10$ cm, the threshold (11) is exceeded by a factor of 1.5–2 and $\tilde{\tau}$ changes from 10^{-4}–10^{-3} s in low-viscosity liquids (acetone, benzene, water) to 10^{-5}–10^{-6} s in liquids of the glycerin type. Substitution of (16) in (12) reduces the latter, according to these estimates, to the requirement $10^{-1} < GL < 10^4$; therefore, the approximation assumed in our calculation can be regarded as sufficiently confirmed.

We note that Eq. (16) allows us to calculate with less difficulty the energy of the measured scattered wave,

$$E \sim S\tilde{\tau}p_{s0}^2|K(\tilde{\tau})|/2\rho_0 c_0$$

(S is the area of the transverse cross section of the scattered beam).

3. For the description of a pair of SS mechanisms that are close in effectiveness, we must generalize the results obtained above. Such a situation (two forms of SS with similar thresholds), in addition to WLAF and STW at $\omega \approx \omega^*$, is characteristic for SS near phase transitions of the liquid. As the transition point is approached, the frequency shift tends to zero as also the threshold of SS by oscillations of the corresponding order parameter (the density in a gas-liquid mixture,[11] the concentration in a stratifying solution[12]); thus these forms of SS are "compared" here with the effective universal mechanisms (WLAF and STW).

Analysis of Eq. (6), jointly with two equations of the system (7) (simultaneous closeness to one another of three SS thresholds in a homogeneous liquid is not realistic) can be carried out in the approximations of Sec 3, par. 2, by a method that directly generalizes the method applied there, precisely for the important case $|I_u - I_T|/(I_u + I_T) \ll 1$. The basic qualitatively new result here is connected with the beats of two modes that have close quality factors. The total amplification coefficient of the scattered wave, i.e., the one corresponding to the simultaneous action of two SS mechanisms, is equal to

$$K(t) \approx M_0[2H(t)\xi^{1/2}(0)]\exp[-2\omega^2(\chi+\nu')t/c_0^2 - \delta L] \times\cos[2\omega^2(\nu'-\chi)t/c_0^2]. \qquad (17)$$

Here $M_0(z)$ is the modulus of the modified Bessel function of zero order. The coordinate dependence of $\xi(x)$ in (17) is the same as in (14). The time dependence in (17) is different than under the action of a single SS mechanism (15). The time argument of $H(t)$ is determined by the differential equation

$$Hd^2H/d\tilde{\theta}^2 =2\omega^2\delta\left[(2\Omega I/I_T+\chi q^2I/I_u)^2+(\Omega I/I_u+2\nu' q^2I/I_T)^2\right]^{1/2}/c_0^2e^{2\delta L}. \quad (18)$$

Its solution is expressed in terms of the reciprocal error function of imaginary argument and increases with increase in t more rapidly than $\theta^{1/2}(t)$, i.e., the envelope of the pulse in (17) has a steeper leading edge than in (15).

Observation of the temporal oscillations [with period $\pi c_0^2/\omega^2(\nu'-\chi)$] of the amplification coefficient in SS of sound represents an attractive method of fixing the instant of turning-on the additional SS mechanism in the measurement of external parameters. This method can be used, for example, for observation of SS near phase transitions. In binary liquids, the threshold of concentration SS is quite high under ordinary conditions, but tends to zero at the critical point of stratification[12]; it becomes comparable with the thresholds of the universal mechanisms (STW and WLAF) in a pre-transition region of width $(10^{-1}-10^{-2})$ K. Under the conditions of strong linear scattering, the procedures necessary for the investigation of SS in this region are not connected with the absolute measurements of the pressure or the intensity of the sound. One of such methods can be based exactly on the noted effects of the beating of the scattering modes.

4. COMPARATIVE ESTIMATE OF THE EFFECTIVENESS OF PROCESSES COMPETING WITH THE STIMULATED SCATTERING

The results of Sec. 3 show that the threshold parameters of STW and WLAF are such that these effects can be observed using existing sound sources in liquids, whose choice is not so difficult. Competing sound-induced processes impose more stringent requirements on the parameters of the source and of the investigated medium, either by diverting the energy of the pump or by distorting its frequency-angle spectrum. The cavitation thresholds in normal liquids are much higher than the SS thresholds, which are estimated by Eq. (11). The effect of the stimulated convection here is the same as in the observation of the self-action of sound.[12] There remains the necessary estimate of the generation of harmonics by the elastic nonlinearity and the superheating of the liquid.

The generation of harmonics does not change the observation of the SS if the amplification coefficient of the test wave $K(\tilde{\tau})$ at $L=L_h$ is larger than a certain value $K_0>1$ determined by the sensitivity of the receiving apparatus (the condition $\delta L_h<1$ is satisfied for the entire set of liquids cited by way of example, in the frequency range up to 10 MHz). The length L_h is inversely proportional to the amplitude of the pressure of the pump wave:

$$L_h=(\rho_0c_0^5/2I\omega^2\varepsilon^2)^{1/2}, \quad (19)$$

ε is the coefficient of elastic nonlinearity. Using the asymptotic representation of the Infeld function in (15), we arrive with the aid of (11) and (16) at the required inequality

$$\delta L_hI/I_u\geqslant K_0, \quad (20)$$

or $I\gtrsim I_h=32(K_0\eta\omega\varepsilon)^2/\rho_0c_0$. For STW, it is necessary to replace I_u by I_T in the inequality (20). The inequality (20) is stronger than the threshold condition (11)—the quantity I_h, for example, for acetone, amounts to $(0.2K_0)^2$ W·cm^{-2} at a frequency of 1 MHz. There is an obviously pronounced weakening of (20) with decrease in the frequency: $I_h\sim\omega^2$.

Since $I_h>I_u$ at the frequencies of interest to us, it is necessary to carry out an estimate of the heating of the liquid by the sound pump, assuming $I=I_h$ in the equation of thermal conduction. We require that the increase in the temperature within the time τ not exceed a certain value ΔT (that does not change the phase state of the liquid). This yields

$$\Delta T<T_h=2\delta I_h\tilde{\tau}(I_h, L_h)/\rho_0C_p. \quad (21)$$

A typical value (for acetone) is $T_h=6K_0^3(\delta\eta c_0\varepsilon^2/\rho_0C_p)\approx 3\cdot10^{-6}K_0^3$ deg.

Thus, all the conditions (11), (20) and (21) that are necessary for the observation of WLAF and STW of sound are actually satisfied in liquids with moderate viscosity with the use of pulsed sources whose intensity is of the order of 1–100 W-cm^2 at a frequency of 10^5–10^{-6} Hz.

A scheme with transverse pumping (scattering at 90°) is more realistic for the observation of universal SS of sound under these stringent conditions, although the growth rate G is less by a factor of $\sqrt{2}$ than in the backscattering scheme. In the first place, we can use cylindrical focusing of the pump with a caustic length $h<L_h$, which makes (20) unnecessary and leads to an easing of the condition (21) in the case of a decrease in $\tilde{\tau}$. Second, the background contribution of the multiply reflected pump is lower. Third, use of a resonator is possible, which greatly lowers the threshold intensity. And, finally, the requirements for coherence of the pump are moderated.

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