CERTAIN PROBLEMS IN THE THEORY OF THE STIMULATED MOLECULAR SCATTERING OF LIGHT

V. S. STARUNOV

P. N. Lebedev Physics Institute, USSR Academy of Sciences

Submitted April 15, 1969

Zh. Eksp. Teor. Fiz. 57, 1012–1023 (September, 1969)

A theory is developed for stationary and nonstationary stimulated thermal and concentration scattering of light. The influence of stimulated thermal light scattering on the position of the components of stimulated Mandel'shtam-Brillouin scattering is investigated. The obtained results enable one to explain certain previously incomprehensible experimental data concerning stimulated light scattering.

INTRODUCTION

THE application of giant laser pulses to physical investigations has permitted the observation of a whole series of new nonlinear optical effects, including the stimulated scattering of light.[1-5] Satisfactory theories already exist for such stimulated scattering processes as Raman scattering (see, for example, [6,7]), Mandel'shtam-Brillouin scattering,[8-9] and Raleigh-wing scattering.[10] Considerably less has been done in regard to the theoretical interpretation of various aspects of the recently observed stimulated thermal (entropy) scattering of light[4] and stimulated concentration scattering of light.[5] The present article is devoted to a theoretical investigation of stimulated thermal scattering of light (STS) and its influence on stimulated Mandel'shtam-Brillouin scattering (SMBS), and also a theory of stimulated concentration scattering of light (SConS).

The STS phenomenon consists in the fact that, as a result of the interaction between a wave of intense perturbing light and the weak light waves of the initial thermal scattering by the entropy fluctuations in a nonlinear medium, temperature "waves" appear, and a transfer of energy takes place from the stimulating radiation to the scattered light wave.

In contrast to other kinds of stimulated light scattering, STS may arise as a consequence of two different mechanisms involving the nonlinear interaction of light waves with matter: as a consequence of the electrocaloric effect (STS I)[11] and as a consequence of the direct absorption of light (STS II).[12] The simultaneous influence of these effects on the STS phenomenon has not been considered previously; in the same way the nonstationary processes associated with STS and a whole series of other questions related to this phenomenon have not been analyzed.

In the steady-state regime the change of a medium's dielectric permittivity occurs primarily as a consequence of its thermal expansion. However, during a time interval which is appreciably shorter than the time for establishment of a temperature wave ($\sim 10^{-8}$ sec for liquids), the STS I and STS II phenomena are not able to develop. In this case a third kind of STS (STS III) may be essential; just like STS II this third form arises as a consequence of the absorption of light, but if differs substantially from STS I and STS II due to the kinetic evolution of the scattering process. The physical reason for the appearance of STS III is the change of the medium's dielectric constant (at constant density) as a consequence of the excitation of the molecules' internal degrees of freedom in connection with the absorption of light. The time required to establish this process is much shorter than the time necessary for establishment of a temperature wave, which arises in connection with a change of the temperature of the molecules' external degrees of freedom. The important role of STS III for ultrashort (picosecond) pulses of perturbing light is determined by this, whereas for nanosecond time intervals one can neglect the STS III phenomenon in comparison with STS II since usually $|(\partial\epsilon/\partial T)_\rho| \ll |(\partial\epsilon/\partial T)_p|$.

The large interest in STS which has recently developed, and the appearance of a whole series of articles devoted to experimental investigation of this phenomenon[4,13-17] and to its practical application,[18] indicate the timeliness of this problem.

The theory of steady-state and nonstationary STS can easily be utilized for an analysis of SConS. A theory of SConS is expounded in the last Section of this article.

1. THEORY OF STIMULATED STEADY-STATE THERMAL SCATTERING OF LIGHT AND STIMULATED MANDEL'SHTAM-BRILLOUIN SCATTERING

In connection with STS the effect of the electromagnetic waves on a substance becomes apparent in a heating of the nonlinear medium. The amount of heat Q discharged into the medium is proportional to the square of the intensity of the total electric field of the stimulating and scattered light waves.

One can obtain the quantitative characteristics of the STS phenomenon from a simultaneous solution of the system of nonlinear Maxwell equations and the equation of thermal conductivity:

$$\frac{1}{c^2}\frac{\partial^2 \mathbf{E}}{\partial t^2} - \nabla^2 \mathbf{E} = -\frac{1}{n^2 c^2}\left(\frac{\partial \varepsilon}{\partial T}\right)_p \frac{\partial^2 (\mathbf{E} T_1)}{\partial t^2} \tag{1}$$

$$\frac{\partial T_1}{\partial t} - \chi \Delta T_1 = \frac{1}{4\pi \rho c_p}\left\{2k_\omega n^2 c \mathbf{E}^2 - \left(\frac{\partial \varepsilon}{\partial T}\right)_\rho T_0 \mathbf{E}\dot{\mathbf{E}}\right\}; \tag{2}$$

here $\mathbf{E}$ is the total field of the stimulating and scattered light waves, c is the velocity of light in a medium with index of refraction n, $\epsilon = n^2$ is the dielectric permittivity of the medium, $2k_\omega$ is the absorption coefficient of the light, c is the specific heat at constant pressure

(p = const, since in Eqs. (1) and (2) the SMBS phenomenon is not considered), T_0 is the equilibrium absolute temperature of the medium, T_1 is the deviation of the medium's temperature from its equilibrium value, χ and ρ are, respectively, the coefficient of thermal conductivity and the density of the medium.

The quantity $\dot{Q}/\rho c_p$ stands on the right hand side of Eq. (2), where

$$\dot{Q} = \frac{1}{4\pi}\left\{2k_\omega n^2 c\mathbf{E}^2 - \left(\frac{\partial \varepsilon}{\partial T}\right)_p T_0 \mathbf{E}\dot{\mathbf{E}}\right\}. \tag{3}$$

The first term of relation (3) is due to true absorption of light in the medium, and the second is due to the electrocaloric effect.[19] Thus, here both STS as a consequence of the electrocaloric effect previously considered by the author[11] and the scattering caused by light absorption, which has been analyzed by Herman and Gray,[12] [1)] are simultaneously taken into consideration.

The system of Eqs. (1) and (2) clearly indicates the physical nature of STS, and will later be used for an analysis of the nonstationary regime of this phenomenon. Within the limits of the steady-state theory it turns out to be possible to simultaneously consider both the STS phenomenon and its influence on SMBS. For this purpose it is convenient to start from the solution of the following system of the nonlinear equations of Maxwell, hydrodynamics, and thermal conductivity:

$$\frac{\partial^2 u}{\partial t^2} = \nabla^2\left\{\frac{1}{\rho\beta_T}(u + \sigma T_1) + \Gamma\frac{\partial u}{\partial t} - \frac{Y}{8\pi\rho}\mathbf{E}^2\right\}, \tag{4}$$

$$\frac{\partial T_1}{\partial t} - \chi\nabla^2 T_1 - \frac{\gamma - 1}{\sigma}\frac{\partial u}{\partial t} = \frac{1}{4\pi\rho c_v}\left\{2k_\omega n^2 c\mathbf{E}^2 - \left(\frac{\partial \varepsilon}{\partial T}\right)_\rho T_0 \mathbf{E}\dot{\mathbf{E}}\right\} \tag{5}$$

$$\frac{1}{c^2}\frac{\partial^2 \mathbf{E}}{\partial t^2} - \nabla^2\mathbf{E} = -\frac{1}{n^2 c^2}\frac{\partial^2}{\partial t^2}\left\{Y\mathbf{E}u + \left(\frac{\partial \varepsilon}{\partial T}\right)_\rho \mathbf{E}T_1\right\}, \tag{6}$$

where $u = \rho_1/\rho$, ρ_1 is the deviation of the medium's density from its equilibrium value, $\gamma = c_p/c_V$, $Y = (\rho\partial\epsilon/\partial\rho)_T$, $\beta_T = 1/\rho v_T^2$, $\Gamma = [\eta + \frac{4}{3}\eta']/\rho$, β_T and v_T denote the isothermal compressibility and the velocity of sound, σ denotes the temperature coefficient of volume expansion, and η and η' denote the shear and volume coefficients of viscosity.

For the STS gain associated with scattering at a frequency close to the frequency of the stimulating light, the same result is obtained from the system of Eqs. (4)–(6) as from the system of Eqs. (1) and (2). But for SMBS the gain will differ from that obtained without taking the influence of STS into account.

In what follows we neglect the second term on the right hand side of Eq. (6); this term is much smaller than the first term since $(\partial\epsilon/\partial T)_\rho$ is usually a very small quantity.

The steady-state theory is applicable if, for the duration t_0 of the stimulating pulse or for a time t passing after the beginning of the stimulated scattering process, the conditions $t \gg t_{ph}$ and $t \gg \tau_T$ are fulfilled, where τ_{ph} denotes the lifetime of the acoustic phonons and τ_T is the time for establishment of thermal equilibrium in the "temperature wave":

$$\tau_{ph} = 1/2\alpha v, \quad \tau_T = 1/2\chi q^2. \tag{7}$$

In (7) and in what follows $\mathbf{q} = \mathbf{k}_0 - \mathbf{k}_1$, where $\mathbf{k}_0$ and $\mathbf{k}_1$ are the wave vectors, respectively, of the stimulating and scattered light waves, v and α are the velocity and amplitude coefficient for the absorption of hypersound.

Let the total field $\mathbf{E}$ consist of a linearly-polarized stimulating light wave $\mathbf{E}_0$ and a scattered wave $\mathbf{E}_1$. Then one can seek a solution of the system of Eqs. (4)–(6) in the form of Fourier integrals for $\mathbf{E}_0$, $\mathbf{E}_1$, u, and T_1. Representing these quantities in the form of Fourier integrals enables one to take the finite spectral linewidth of the stimulating light into consideration.

Even in the steady-state case it is impossible to obtain a general solution of the system of Eqs. (4)–(6) in analytic form. In order to simplify further, let us linearize the system of Eqs. (4)–(7) assuming that $|\mathbf{E}_0| \gg |\mathbf{E}_1|$ and $|\mathbf{E}_0(\mathbf{r})|^2 \sim$ const. This so-called approximation of a fixed stimulating wave is valid when the intensity of the stimulated scattering is much smaller than the intensity of the stimulating light. In addition, it is assumed that the relative change of the amplitudes is small over distances of the order of a wavelength. This condition allows one to keep only the first spatial derivative of the amplitude in Eq. (6). In Eqs. (4) and (5) in general the derivatives of the amplitudes can be neglected since, in the majority of cases, the absorption of hypersound is large, and the following conditions are satisfied:

$$\left|\frac{1}{u}\frac{\partial u}{\partial x}\right| \ll \alpha, \quad \left|\frac{1}{T_1}\frac{\partial T_1}{\partial x}\right| \ll \alpha_T, \tag{8}$$

which means that the coefficients for the absorption of hypersound α and for absorption of the temperature wave α_T are larger than their gain in a stimulated scattering process.[2)]

Taking the assumptions which have been made into account and assuming that, as a consequence of the large amplification the spectral line width of the scattered radiation is much smaller than the width of the stimulating radiation, we obtain an exponential growth in the intensity of the scattered radiation in the nonlinear interaction region:

$$|E_1(\xi)|^2 = |E_1(\xi = 0)|^2 \exp\{g(\Omega)\xi\}, \tag{9}$$

where ξ denotes the coordinate in the direction of $\mathbf{k}_1$, and $g(\Omega)$ is the steady-state gain, given by

$$g(\Omega) = \frac{|\mathbf{k}_1|\mathbf{q}^2 Y}{4\pi n^2\rho}\,\mathrm{Re}\int_{-\infty}^{\infty}\frac{A_1}{A_2}|E_0(\omega_0 - \Omega_0)|^2\, d\Omega_0, \tag{10}$$

$$A_1 = \sigma\frac{ik_\omega n^2 c + \frac{1}{4}T_0(\partial\varepsilon/\partial T)_p(\Omega - \Omega_0)}{\rho c_p\beta_s[i(\Omega - \Omega_0) + \chi q^2]} - \frac{1}{4}iY,$$

$$A_2 = (\Omega - \Omega_0)^2 - \frac{\mathbf{q}^2}{\rho\beta_T} - i(\Omega - \Omega_0)\mathbf{q}^2\Gamma - \frac{i\mathbf{q}^2(\gamma - 1)(\Omega - \Omega_0)}{\rho\beta_T[i(\Omega - \Omega_0) + \chi q^2]}.$$

In Eq. (10) $\Omega = \omega_0 - \omega$, $\mathbf{q} = \mathbf{k}_0 - \mathbf{k}_1$, and ω_0 and ω_1 are the frequencies, respectively, of the stimulating and scattered light waves.

The obtained expression (10) for the steady-state gain is valid in the frequency range from $\Omega \sim 0$ to $\Omega \sim \Omega_{MB}$ and, consequently, takes SMBS and STS phenomena into consideration. In order to analyze expres-

[1)] Later the STS phenomenon was also considered by Bespalov et al. (Report at the IVth Symposium on Nonlinear Optics, Kiev, 1968).

[2)] Commonly for liquids and solids $\alpha = 10^2$ to 10^4 cm^{-1} and $\alpha_T \gg \alpha$, but the gain associated with stimulated scattering $\sim 10^{-1}$. Therefore condition (8) is satisfied under actual experimental conditions.

sion (10) it is convenient to represent the frequency distribution of the intensity in the stimulating radiation line in the form of a dispersion distribution with half-width $2\delta\omega_0$:

$$|E_0(\omega_0-\Omega_0)|^2=\frac{|E_0^m|^2}{\pi}\frac{\delta\omega_0}{\Omega_0^2+\delta\omega_0^2}. \quad (11)$$

The gain $g(\Omega)$ has a maximum near the frequency of the stimulating radiation line ($\Omega \sim 0$) and near the frequency of the Mandel'shtam-Brillouin component ($\Omega \sim \Omega_{MB}$). Therefore, without citing the general, cumbersome expression which is obtained after integration of (10) with Eq. (11) taken into account, let us consider the two limiting cases of greatest interest: 1) the scattering spectrum in the vicinity of the frequency of the stimulating radiation; 2) the scattering spectrum in the vicinity of the SMBS frequency $\Omega_{MB} \sim q^2/\rho\beta_S$.

A. Scattering Spectrum in the Vicinity of the Frequency of the Stimulating Radiation

In this case one can assume $q^2/\rho\beta_S \gg \Omega^2$ and then only $-q^2/\rho\beta_S$ is left in the denominator of expression (10). Then, for the gain not far from the frequency of the stimulating radiation we obtain

$$g_T=B_T\frac{\Omega}{\Omega^2+(\chi q^2+\delta\omega_0)^2}|E_0^m|^2, \quad (12)$$

where the constant B_T for a given medium is determined by the expression

$$B_T=\frac{|k_1|}{8\pi n^2\rho c_p}\left(\frac{\partial\varepsilon}{\partial T}\right)_p\left\{2k_\omega n^2c+\frac{1}{2}T_0\left(\frac{\partial\varepsilon}{\partial T}\right)_p\chi q^2\right\}. \quad (13)$$

For STS the gain (12) has a maximum ($g_T > 0$) at the frequency $\omega_1 = \omega_0 - (\chi q^2 + \delta\omega_0)$ sign B_T. Therefore, depending on the sign of B_T either the Stokes or the anti-Stokes part of the scattered light will be amplified.

Usually for solids $(\partial\epsilon/\partial T)_p > 0$, $B_T > 0$, and consequently the maximum of the STS will occur on the Stokes side relative to the frequency of the stimulating radiation.

If $(\partial\epsilon/\partial T)_p < 0$ (for example, in a liquid) then both Stokes as well as anti-Stokes STS are possible depending on the value of the quantity h, where

$$h=\left|\frac{T_0\chi q^2}{4k_\omega n^2c}\left(\frac{\partial\varepsilon}{\partial T}\right)_p\right|. \quad (14)$$

If $h < 1$ (and $(\partial\epsilon/\partial T)_p < 0$) then the anti-Stokes component of the scattered light will be amplified ($g_T > 0$ for $\Omega < 0$), but for $h > 1$ the Stokes component of the scattered light will be amplified ($g_T > 0$ for $\Omega > 0$). For example, for the liquid benzine the condition $h > 1$ means $2k_\omega < 5 \times 10^{-4}$ cm^{-1}.

If the spectral width $\delta\omega_0$ of the stimulating radiation is much greater than the linewidth $\delta\Omega_C = \chi q^2$ of the thermal scattering by entropy fluctuations,[20] i.e., if $\delta\omega_0 \gg \chi q^2$, then the STS gain is $\chi q^2/\delta\omega_0$ times smaller than in the case $\delta\omega_0 \ll \chi q^2$.

It follows from Eqs. (12) and (13) that if $h < 1$ then the gain associated with STS is larger the larger the light absorption coefficient $2k_\omega$ is. Taking into consideration that, in addition to the amplification process, a process of light absorption also exists, one can write the condition for growth of the scattered wave in the form $-2k_\omega + g_T > 0$, or

$$\frac{|E_0^m|^2}{8\pi}>\left(\frac{|E_0^m|^2}{8\pi}\right)_{thr}=\left|\frac{2\rho c_p(\chi q^2+\delta\omega_0)}{\omega_0(\partial\varepsilon/\partial T)_p}\right|, \quad h\ll 1. \quad (15)$$

The threshold condition (15) is obtained for such large values of $2k_\omega$ when one can neglect the electrocaloric effect ($h \ll 1$). For benzine condition (15) gives the following threshold value for the intensity of the stimulating light (for the linewidth of a rubidium laser $\sim 10^{-3}$ cm^{-1}): $J_0 > (J_0)_{thr} = 0.2$ MV/cm^2.

We note that the STS effect may be successfully used to Q-switch a laser,[18] including infrared resonators since the effective coefficient of reflection is larger the larger the absorption coefficient of light is. The small frequency shift associated with STS may make such a method for Q switching more effective in certain cases than a utilization of SMBS for this purpose.

B. Scattering Spectrum in the Vicinity of the Frequency of the SMBS Component

Near the frequency of the SMBS component one can assume $\Omega^2 \sim \Omega_{MB}^2 = q^2/\rho\beta_S \gg (\chi q^2)^2$. Neglecting the small terms in (10), we find the following result for the gain:

$$g_{MB}(\Omega)=\left\{\frac{|k_1|\gamma^2\Omega_{MB}\beta_s(2-\gamma)}{32\pi n^2}\frac{\delta\omega_0+\delta\Omega_{MB}}{(\Omega-\Omega_{MB})^2+(\delta\omega_0+\delta\Omega_{MB})^2}\right.$$
$$\left.\times\frac{1}{2}R\frac{\Omega_{MB}-\Omega}{(\Omega-\Omega_{MB})^2+(\delta\omega_0+\delta\Omega_{MB})^2}\right\}|E_0^m|^2, \quad (16)$$

where $2\delta\Omega_{MB} = \Gamma q^2$ is the half-width of the thermal Mandel'shtam-Brillouin scattering, $\Omega_{MB} = q^2/\rho\beta_S$, and the coefficient B_T is determined by expression (13).

The first term in expression (16) has a maximum at the frequency $\Omega = \Omega_{MB}$, and this term differs from previously obtained expressions for the gain associated with SMBS by the factor $2 - \gamma$. This factor appears as a consequence of the influence of STS I (the electrocaloric effect) on SMBS.

The maximum of the second term in (16) does not coincide with the frequency Ω_{MB} and is found on the Stokes side ($\Omega_{max} > \Omega_{MB}$) relative to it for $B_T < 0$, and on the anti-Stokes side ($\Omega_{max} < \Omega_{MB}$) for $B_T > 0$. This maximum is shifted relative to the frequency Ω_{MB} by an amount $\Delta\Omega = \delta\omega_0 + \delta\Omega_{MB}$.

Thus, an asymmetry exists in the gain associated with the Stokes component of the SMBS. Usually $B_T > 0$ for solids, and the maximum of the gain for SMBS will shift toward the anti-Stokes side relative to the frequency Ω_{MB}. If $(\partial\epsilon/\partial T)_p < 0$, then for $h < 1$ the maximum of the second term in (16) will occur at the frequency $\Omega = \Omega_{MB} + (\delta\omega_0 + \delta\Omega_{MB})$, and for $h > 1$ it will occur at the frequency $\Omega = \Omega_{MB} - (\delta\omega_0 + \delta\Omega_{MB})$.

This will happen only if the intensity of the SMBS component is small, and it itself cannot produce the STS phenomenon. However, if the intensity of the SMBS component is large enough, then it itself may excite the STS effect, and then everything indicated in this section about the shift of the SMBS component relative to the frequency Ω_{MB} may reverse its sign, as follows from formula (12). From a comparison of formula (12) and the second term of formula (16) it follows that one can expect such a change in the sign of the frequency shift

of the SMBS component for $J_{MB} > \frac{1}{2} J_0$.[3)]

In fact, let us consider for example the case $B_T < 0$. If J_{MB} is large and begins to affect the saturation process affiliated with SMBS, an intense STS arises both near the frequency of the stimulating radiation and at the frequency of the SMBS component. This leads to a shift of both lines toward the anti-Stokes side. Thus, for a large intensity of the SMBS as a consequence of the STS effect the shift of the SMBS component relative to the frequency of the stimulating radiation turns out to be smaller by roughly the amount $\delta\omega_0 + \delta\Omega_{MB}$. The effectiveness of this process is determined by the gain g_T, in which one should regard the intensity of the SMBS as the intensity of the stimulating radiation. This gain (see Eq. (12)) must be larger than that part of the SMBS gain (the second term in formula (16)), which leads to an opposite shift of the frequency of the SMBS component. From Eqs. (12) and (16) it follows that one can expect a decrease of the shift of the SMBS component if $J_{MB} > \frac{1}{2} J_0$.

A decrease in the shift of the SMBS component in comparison with the shift measured relative to the thermal light scattering has been repeatedly noted (see, for example, [21]). It was also noted that in liquids in connection with the appearance of STS II scattering, which is shifted toward the anti-Stokes side relative to the frequency of the stimulating radiation (a shift ~200 MHz, half-width of the stimulating radiation line $2\delta\omega_0 \sim 300$ MHz), a decrease in the shift of the SMBS component occurred, also roughly by an amount ~200 MHz,[16] and the width of this shift remained unexplained.

Apparently one of the essential reasons for the change in the position of the Mandel'shtam-Brillouin component affiliated with stimulated scattering is the influence of STS on the position of the SMBS component. It also follows from Eq. (16) that for a sufficiently large and negative value of B_T, amplification of the anti-Stokes component of the SMBS is possible, which was noted previously.[12]

2. NONSTATIONARY THEORY OF STIMULATED THERMAL LIGHT SCATTERING

For an analysis of the nonstationary STS regime, we shall start from the system of Eqs. (1) and (2), assuming that E and T_1 are given in the form

$$E = \tfrac{1}{2}E_0 \exp(i\omega_0 t - i\mathbf{k}_0\mathbf{r}) + \tfrac{1}{2}E_1(t,\mathbf{r}) \exp(i\omega_1 t - i\mathbf{k}_1\mathbf{r}) + \text{c.c.} \quad (17)$$

$$T_1 = \tfrac{1}{2}T(t) \exp(i\Omega t - i\mathbf{q}\mathbf{r}) + \text{c.c.} \quad (17')$$

Let us again assume that $|E_0| \gg |E_1|$ and $|E_0|^2 \sim \text{const}$.

Let the field E_0 be switched on at the moment t = 0 and then it remains constant, but E_1 depends on the coordinates and also on the time, as long as they do not reach the steady-state regime. Substituting (17) into (1) and (2) and, as before, assuming that E_1 and T(t) vary slowly, we obtain a truncated system of equations

$$\frac{\partial E_1^*}{\partial \xi} = -\frac{i|\mathbf{k}_1|}{4n^2}\left(\frac{\partial \varepsilon}{\partial T}\right)_p E_0^* T, \quad (18)$$

$$T(t) = \frac{1}{4\pi\rho c_p} E_0 \exp[-(i\Omega + \chi q^2)t] \int_0^t \left\{ \left[2k_\omega n^2 c - \frac{i}{2}\left(\frac{\partial \varepsilon}{\partial T}\right)_p \Omega T_0 \right] E_1^*(t') - \frac{1}{2}\left(\frac{\partial \varepsilon}{\partial T}\right)_p T_0 \frac{\partial E_1^*(t')}{\partial t'} \right\} \exp[(i\Omega + \chi q^2)t'] dt'. \quad (19)$$

In Eq. (18) the first derivative of E_1^* with respect to the time is omitted ($|c^{-1}\partial E_1^*/\partial t| \ll |qE_1^*/\partial \xi|$, c is large), and in order to obtain Eq. (19) the derivative of T_1 with respect to the coordinate is omitted, which is permissible for $\xi \gg 1/|q|$.

The general solution of Eqs. (18) and (19) has the following form[4)]

$$E_1^*(\xi, t) = E_1^*(0) \exp\{-i\varphi_T(\xi)\} \left\{ \exp[-(i\Omega + \chi q^2)t] I_0[(2iB_T\xi t|E_0|^2)^{1/2}] + (i\Omega + \chi q^2) \int_0^t \exp[-(i\Omega + \chi q^2)t'] I_0[(2iB_T\xi t'|E_0|^2)^{1/2}] dt' \right\}, \quad (20)$$

where $I_0(z)$ denotes a modified Bessel function, and

$$\varphi_T = \frac{|\mathbf{k}_1| T_0 |E_0|^2}{32\pi n^2 \rho c_p}\left(\frac{\partial \varepsilon}{\partial T}\right)_p^2 \xi.$$

As $t \to \infty$ the first term in (20) tends to zero; the second term yields an exponential dependence of $|E_1|^2$ on the coordinates with a steady-state gain g_T which is determined by expression (12) for $\delta\omega_0 = 0$.

Now let us consider what follows from expression (20) in the nonstationary regime when $t \ll (\frac{1}{2}\chi q^2)$. In this case in Eq. (20) one can neglect the second term in comparison with the first. Representing the Bessel function by its asymptotic value (assuming that the amplification is large), we obtain

$$E_1^*(\xi, t) \sim \frac{2 \exp[-i\varphi_T - (i\Omega + \chi q^2)t]}{(2\pi)^{1/2}(2iB_T\xi t|E_0|^2)^{1/4}} \operatorname{ch}(2iB_T\xi t|E_0|^2)^{1/4}. \quad (21)$$

It follows from this expression that the change in the intensity of the scattered light in the nonstationary regime is primarily determined by the expression

$$J_1(\xi, t) \sim \exp\{(2|B_T||E_0|^2\xi t)^{1/2}\}. \quad (22)$$

One can determine the time t_{st}, during which the steady-state regime is reached, from the condition that the integrand in Eq. (20) be maximal. This time is given by

$$t_{st} \approx \frac{1}{4}\frac{|B_T|\xi}{\chi q^2}|E_0|^2 \approx \frac{j_T}{2\chi q^2},$$

where $j_T = g_T L$, and L is the nonlinear interaction region. It follows from Eq. (22) that the increase in the intensity of the scattered light does not depend on the frequency. For $t < j_T/2\chi q^2$ one can reach a conclusion about the spectrum of the scattered light from an investigation of expression (21). Taking into account only that term which increases in space and time,[5)] we obtain the result that the broadening of the spectrum of the scattered light is given by the expression

$$\Omega' = \frac{1}{2}\left(\frac{|B_T|\xi}{t}|E_0|^2\right)^{1/2} \operatorname{sign} B_T = \chi q^2 \left(\frac{t_{st}}{t}\right)^{1/2} \operatorname{sign} B_T$$

and consequently it is Stokes broadening for $B_T > 0$ and anti-Stokes broadening for $B_T < 0$. For $t \sim t_{st}$ when the steady-state regime is reached, $|\Omega'|$ reaches its minimal value $\Omega' \sim \chi q^2 \operatorname{sign} B_T$, just as one would expect from a steady-state investigation of STS.

We note that by assuming that $|E_0|^2$ does not depend

[3)] This relation is, of course, approximate since for $J_{MB} \sim (1/2)J_0$ the nonlinear solutions of the system of Eqs. (4) – (6) must be considered.

[4)] One can obtain this solution by applying the Laplace-Carson integral transform to Eqs. (18) and (19).

[5)] This depends on the sign of B_T.

on the time, by the same token we exclude from investigation the broadening as a consequence of an effect analogous to the self-diffusion of light.[22] In fact, as is evident from Eqs. (20) and (21), if $\partial|E_0|^2/\partial t \neq 0$, then a broadening $\Omega'' \sim -\partial\varphi_T/\partial t$ must occur whose sign depends on the sign of $\partial|E_0|^2/\partial t$.

3. STIMULATED THERMAL SCATTERING AS A CONSEQUENCE OF THE EXCITATION OF INTRAMOLECULAR VIBRATIONS

In connection with the propagation of intense light through a medium consisting of complex molecules, a change Δn_i of the index of refraction is possible as a consequence of light absorption and a subsequent change in the "temperature" of the intramolecular degrees of freedom,[23] which was not taken into account above. For nanosecond pulses this effect is small in comparison with electrostriction and heating of the medium (external degrees of freedom of a molecule). But for picosecond time intervals, when electrostriction and thermal expansion of the medium cease to play an important role, in absorbing media this effect may become dominant in the phenomena of self-focusing, self-diffusion, light scattering, and other nonlinear effects.[6)]

The time required to establish this effect should be equal to the average relaxation time associated with the "temperature" of a molecule's internal degrees of freedom. This time is experimentally determined by a measurement of the dispersion of the sound velocity or by hypersonic absorption,[21] and for a liquid it has a value $\sim 10^{-10}$ to 10^{-11} sec, which is less than the lifetime of the acoustic phonons or the time for establishment of a temperature in the medium. In a simple model possessing a single relaxation time, the change of the temperature T_i of the molecules' internal degrees of freedom as a consequence of light absorption[7)] can be determined from the equation

$$\frac{\partial T_i}{\partial t} + \frac{1}{\tau_i} T_i = \frac{k_\omega n^2 c}{2\pi\rho c_i} E^2. \qquad (23)$$

If the medium's thermal expansion is not taken into account ($t \ll \tau_T$), then the change of the medium's dielectric constant occurs at constant density. Therefore, the nonlinear correction Δn_i to the index of refraction for $E = \frac{1}{2} E_0 \exp(i\omega_0 t - i\mathbf{k}_0 \cdot \mathbf{r}) + \text{c.c.}$ will have the form

$$\Delta n_i = \left(\frac{\partial \varepsilon}{\partial T}\right)_\rho \frac{k_\omega n c \tau_i}{8\pi\rho c_i} |E_0|^2 \left[1 - \exp\left(-\frac{t}{\tau_i}\right)\right]. \qquad (24)$$

This quantity is positive if $(\partial\epsilon/\partial T)_\rho > 0$ and therefore such a change in the value of n as a consequence of light absorption will lead to the self-focusing of light,[23] in contrast to the change in n as a consequence of a liquid's thermal expansion ($(\partial\epsilon/\partial T)_\rho < 0$) for long light pulses, when defocusing is observed. The solution of the problem of stimulated scattering (STS III) as a consequence of the effect under consideration can be obtained from the simultaneous solution of Eq. (23) and the nonlinear Maxwell Eq. (1) in which, however, $(\partial\epsilon/\partial T)_\rho$ is replaced by $(\partial\epsilon/\partial T)_\rho$ and T_1 by T_i. If $(\partial\epsilon/\partial T)_\rho > 0$ then in connection with STS III the Stokes part of the scattered light will be amplified, and the steady-state gain g_{i1} of such scattering is given by

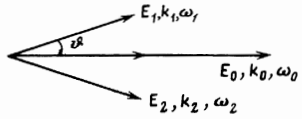
$$g_{i1} = \frac{|k_1| k_\omega c}{4\pi\rho c_i} \left(\frac{\partial \varepsilon}{\partial T}\right)_\rho \frac{\Omega}{\Omega^2 + (\delta\omega_0 + 1/\tau_i)^2} |E_0^m|^2. \qquad (25)$$

The steady-state regime is established during a time $t_{st} \sim j_i \tau_i$, where $j_i = g_{i1} L$. In the nonstationary regime the intensity of the scattered light is determined by formula (22) in which the quantity

$$B_i = \frac{|k_1| k_\omega c}{4\pi\rho c_i} \left(\frac{\partial \varepsilon}{\partial T}\right)_\rho.$$

should be substituted in place of B_T.

In connection with stimulated scattering at small angles, as a consequence of the mechanism under consideration a four-photon interaction is possible in which two photons of the intense perturbing radiation with wave vectors $\mathbf{k}_0$ are transformed into two scattered photons belonging to weak light waves with wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$, which make a small angle with the direction of $\mathbf{k}_0$ (see the figure). This phenomenon may be investigated



according to the same scheme as the four-photon interaction associated with stimulated Rayleigh-wing scattering.[10] By fulfilment of the conservation laws $2\mathbf{k}_0 = \mathbf{k}_1 + \mathbf{k}_2$ and $2\omega_0 = \omega_1 + \omega_2$ the gains g_{i2} are identical for the scattered Stokes (ω_1) and anti-Stokes (ω_2) waves, the gain is maximal at an angle $\theta = \theta_{opt} = (B_i\tau_i/|k_1|)^{1/2}|E_0|$ and for $1/\tau_i \gg \delta\omega_0$ is given by

$$g_{i2} = \frac{B_i \tau_i |E_0|^2}{(1 + \Omega^2\tau_i^2)^{1/2}}. \qquad (26)$$

Thus, the gain associated with such a four-photon interaction is maximal at the unshifted frequency.

Such a four-photon interaction was apparently observed by Mack[24] using picosecond light pulses, although he interprets his own results differently. Stokes STS and the formation in a medium with $\chi = 0$ of a temperature wave as a consequence of a thermal change of ϵ at constant density are considered in [24]. However, such a wave cannot be established during a finite time interval (since $\tau_T \sim \frac{1}{2}\chi q^2$) if one has in mind the temperature of the molecules' external degrees of freedom (as is done in [24]).[8)]

4. STIMULATED CONCENTRATION SCATTERING OF LIGHT

Thermal scattering of light by fluctuations of the concentration in solutions[21] has, just like all forms of

[6)] This effect may also be used to Q-switch lasers in order to obtain ultra-short pulses (10^{-10} to 10^{-11} sec).

[7)] In a strong electromagnetic field the excitation of intramolecular vibrations may also occur as a consequence of other effects, for example, as a consequence of stimulated Raman light scattering. One can take this fact into consideration by understanding $2k_\omega$ as an effective value, which is larger than the absorption coefficient of light.

[8)] In addition, apparently due to a misunderstanding, Mack [24] assumes that the STS associated with a change of ϵ at constant ρ was investigated in [4] whereas that reference deals only with STS, or alternatively the stimulated entropy scattering of light as a consequence of the electrocaloric effect, caused by a thermal change in ϵ at constant p and being the direct stimulated analog of thermal light scattering by entropy fluctuations.

molecular scattering processes, its analog in stimulated scattering. The theory of SConS in a weak solution may be investigated by starting from the system of the nonlinear Maxwell equations and the diffusion equation:

$$\frac{\partial C_1}{\partial t} - D\Delta C_1 = -\frac{C_0}{4\pi kTN}\frac{\partial \varepsilon}{\partial C}\dot{\mathbf{E}}\dot{\mathbf{E}}, \tag{27}$$

$$\frac{1}{c^2}\frac{\partial^2 \mathbf{E}}{\partial t^2} - \nabla^2 \mathbf{E} = -\frac{1}{n^2c^2}\left(\frac{\partial \varepsilon}{\partial C}\right)\frac{\partial^2 (\mathbf{E}C_1)}{\partial t^2}. \tag{28}$$

Here C_1 is the deviation of the concentration of the solvent from its equilibrium value C_0, D is the coefficient of mutual diffusion, and N denotes the number of molecules per 1 cm^3. A steady-state theory of SConS without taking the linewidth of the stimulating radiation into account was considered by Aref'ev.[25] If the finite linewidth of the stimulating radiation (see Eq. (11)) is taken into account, then calculations similar to those carried out in Sec. 1 for STS give the following expression for the gain associated with Stokes SConS:

$$g_C = B_C \frac{\Omega}{\Omega^2 + (Dq^2 + \delta\omega_0)^2}|E_0^n|^2, \tag{29}$$

where B_C, a constant for any given medium, is given by

$$B_C = \frac{|\mathbf{k}_1|C_0}{16\pi n^2 kTN}\left(\frac{\partial \varepsilon}{\partial C}\right)^2 Dq^2. \tag{30}$$

Therefore, as follows from Eq. (29), in the steady-state regime the maximum value of the gain will occur at the frequency $\omega_1 = \omega_0 - (\delta\omega_0 + Dq^2)$. An analysis of Eqs. (27) and (28) indicates that the steady-state regime is reached after a time $t > t_{st} = j_C/2Dq^2$, where $j_C = g_C L$. Since D for a liquid is three orders of magnitude smaller than χ, a time $\sim 10^{-5}$ sec is required to reach the steady-state regime in the case of SConS, instead of a time $\sim 10^{-8}$ sec for STS. In the nonstationary regime ($t < t_{st}$), representing E in the form (17) and making the same assumptions as for the analysis of STS in the nonstationary regime, we obtain

$$E_1^*(\xi, t) = E_1^*(0)\exp[-i\varphi_C(\xi)]\Big\{\exp[-(i\Omega + Dq^2)t]I_0[(2iB_C|E_0|^2\xi t)^{1/2}] + (i\Omega + Dq^2)\int_0^t \exp[-(i\Omega + Dq^2)t']I_0[(2iB_C|E_0|^2\xi t')^{1/2}]dt'\Big\}, \tag{31}$$

where

$$\varphi_C = \frac{|\mathbf{k}_1|C_0\xi}{32\pi n^2 kTN}\left(\frac{\partial \varepsilon}{\partial C}\right)^2 |E_0|^2.$$

For $t < t_{st}$ and a sufficiently large gain, it follows from Eq. (31) that the intensity of the SConS varies according to the law

$$J_C \sim \exp\{2(B_C|E_0|^2\xi t)^{1/2}\}. \tag{32}$$

From Eq. (31) one can obtain the result that in the nonstationary regime the scattered radiation is broadened in the Stokes region by an amount

$$\Omega' = \frac{1}{2}\left(\frac{B_C|E_0|^2\xi}{t}\right)^{1/2} = Dq^2\left(\frac{t_{st}}{t}\right)^{1/2},$$

which reaches its limiting value $\Omega' \sim Dq^2$ for $t \sim t_{st}$, i.e., at the transition to the steady-state regime. Here, just as in the analysis of STS, the broadening $\Omega'' \sim -\partial\varphi_C/\partial t$ as a consequence of the self-diffusion of light is not taken into account.

In conclusion the author expresses his gratitude to I. L. Fabelinskiĭ for his interest and for a helpful discussion of the obtained results.

Note added in proof (August 25, 1969). Formula (16) is valid for $t > \tau_{ph}$ and $t > \tau_T$ (see Eq. (7)). For $\tau_{ph} < t < \tau_T$ the influence of STS on SMBS must diminish, and in (16) one can set the second term of the sum inside the curly brackets equal to zero, but in the first term of this sum the factor $2 - \gamma$ should be replaced by unity.

[1] E. J. Woodbury and W. K. Ng, Proc. IRE **50**, 2367 (1962).
[2] R. Y. Chiao, C. H. Townes, and B. P. Stoicheff, Phys. Rev. Letters **12**, 592 (1964).
[3] D. I. Mash, V. V. Morozov, V. S. Starunov, and I. L. Fabelinskiĭ, ZhETF Pis Red. **2**, 41 (1965) [JETP Lett. **2**, 25 (1965)].
[4] G. I. Zaĭtsev, Yu. I. Kyzylasov, V. S. Starunov, and I. L. Fabelinskiĭ, ZhETF Pis. Red. **6**, 802 (1967) [JETP Lett. **6**, 255 (1967)].
[5] I. M. Aref'ev and V. V. Morozov, ZhETF Pis. Red. **9**, 448 (1969) [JETP Lett. **9**, 269 (1969)].
[6] N. Bloembergen, Usp. Fiz. Nauk **97**, 307 (1969).
[7] V. N. Lugovoĭ, Vvedenie v teoriyu vynuzhdennogo kombinatsionnogo rasseyaniya sveta (Introduction to the Theory of Stimulated Raman Scattering of Light), Nauka, M. 1968.
[8] N. M. Kroll, J. Appl. Phys. **36**, 34 (1965).
[9] C. L. Tang, J. Appl. Phys. **37**, 2945 (1966).
[10] V. S. Starunov, Tr. FIAN **39**, 151 (1967); Dokl. Akad. Nauk SSSR **179**, 65 (1968) [Sov. Phys.-Doklady **13**, 217 (1968)].
[11] V. S. Starunov, Phys. Letters **26A**, 428 (1968).
[12] R. M. Herman and M. A. Gray, Phys. Rev. Letters **19**, 824 (1967).
[13] I. L. Fabelinskiĭ, D. I. Mash, V. V. Morozov, and V. S. Starunov, Phys. Letters **27A**, 253 (1968).
[14] D. H. Rank, C. W. Cho, N. D. Foltz, and T. A. Wiggins, Phys. Rev. Letters **19**, 828 (1967).
[15] T. A. Wiggins, C. W. Cho, D. R. Dietz, and N. D. Foltz, Phys. Rev. Letters **20**, 831 (1968).
[16] J. L. Emmet and A. L. Schawlow, Phys. Rev. **170**, 358 (1968).
[17] D. Pohl, I. Reinhold, and W. Kaiser, Phys. Rev. Letters **20**, 1141 (1968).
[18] F. Gires and B. Soep, Proc. IEEE **56**, 1613 (1968).
[19] L. D. Landau and E. M. Lifshitz, Élektrodinamika sploshnykh sred (Electrodynamics of Continuous Media), Gostekhizdat, M. 1959 (English Transl., Addison-Wesley, 1960).
[20] I. L. Fabelinskiĭ, Molekulyarnoe rasseyaniyae sveta (Molecular Scattering of Light), Izd. Nauka, M. 1965.
[21] D. I. Mash, V. V. Morozov, V. S. Starunov, E. V. Tiganov, and I. L. Fabelinskiĭ, ZhETF Pis. Red. **2**, 246 (1965) [JETP Lett. **2**, 157 (1965)].
[22] L. A. Ostrovskiĭ, ZhETF Pis. Red. **6**, 807 (1967) [JETP Lett. **6**, 260 (1967)].
[23] G. A. Askar'yan, ZhETF Pis. Red. **4**, 400 (1966) [JETP Lett. **4**, 770 (1966)].
[24] M. E. Mack, Phys. Rev. Letters **22**, 13 (1969).
[25] I. M. Aref'ev, Éksperimental'noe issledovanie teplovogo i vynuzhdennogo molekulyarnogo rasseyaniya sveta v rastvorakh v shirokom spektral'nom intervale (Experimental Investigation of Thermal and Stimulated Molecular Scattering of Light in Solutions in a Broad Spectral Interval), Dissertation, FIAN, M. 1968.

Translated by H. H. Nickle
117