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Abstract

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ON THE THEORY OF STIMULATED SCATTERING IN THE WING OF THE RAYLEIGH LINE

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Stimulated light scattering in the wing of the Rayleigh line (SWRL), observed in a number of liquids ([1]), arises as a result of the orientation of anisotropic molecules of the medium in the strong electric fields of the exciting and scattered light waves ([1,2]).

The joint solution of the linearized nonlinear Maxwell equations and of the equation determining the nonlinear polarization of the medium, without taking into account the interaction of the Stokes and anti-Stokes components of the scattered light, gives ([2]) an exponential growth in space of the Stokes component of the scattered light; moreover, the gain coefficient proves to be maximal at the frequency $(\omega_1)_{\max} = \omega_0 - 1/\tau$, where ω_0 is the frequency of the laser radiation, and τ is the anisotropy relaxation time ([2,3]).

In this note a solution is given of the nonlinear problem of stimulated scattering in the Stokes wing of the Rayleigh line, and certain questions are considered concerning the interaction of the laser and Stokes, as well as anti-Stokes, scattered radiation with the anisotropic molecules of the medium.

We shall characterize the deviation of the axes of the anisotropic molecules of the medium from a chaotic distribution by the quantity (see ([2,3])) $s = \overline{\cos^2\theta} - 1/3$, where θ is the angle between the axis of a molecule and the chosen direction z. In the absence of an external force, $s = 0$.

Let the light propagating in the medium consist of three plane waves polarized along the z axis, so that the total electric field is

$$E = E_z = \frac{1}{2}\sum_{l=0}^{2} E_l(\mathbf{r}) e^{i(\omega_l t - \mathbf{k}_l \mathbf{r})} + \text{c.c.}, \tag{1}$$

where the indices $0, 1, 2$ refer respectively to the exciting, Stokes, and anti-Stokes waves. Then ([2])

$$s = \frac{2}{45}\frac{\alpha_1 - \alpha_2}{kT}\left\{\sum_{l=0}^{2}|E_l|^2 + \left[\frac{e^{i\Omega t}}{1 + i\Omega\tau}\left(E_0 E_1^* e^{-i(\mathbf{k}_0 - \mathbf{k}_1)\mathbf{r}} + E_0^* E_2 e^{-i(\mathbf{k}_2 - \mathbf{k}_0)\mathbf{r}}\right) + \text{c.c.}\right]\right\}. \tag{2}$$

Here $\Omega = \omega_0 - \omega_1 = \omega_2 - \omega_0$; α_1 and $\alpha_2 = \alpha_3$ are the principal polarizabilities of the molecule.

1. If the Stokes and anti-Stokes waves do not interact with each other, then the anti-Stokes wave is attenuated, and for E_0 and E_1, from the nonlinear Maxwell equations and (2), neglecting second derivatives of the amplitudes, we obtain the system of equations:

$$2\mathbf{k}_0\nabla E_0 + 2\frac{\omega_0}{c}k_\omega E_0 = -k_0^2 A\frac{\Omega\tau}{1 + \Omega^2\tau^2}|E_1|^2 E_0,$$

$$2\mathbf{k}_1\nabla E_1 + 2\frac{\omega_1}{c}k_\omega E_1 = k_1^2\frac{\Omega\tau}{1 + \Omega^2\tau^2}|E_0|^2 E_1. \tag{3}$$

Above we set

$$|\mathbf{k}_0|^2 = \frac{\omega_0^2}{c^2}\left\{1 + A\left[|E_0|^2 + |E_1|^2\left(1 + \frac{1}{1 + \Omega^2\tau^2}\right)\right]\right\}, \tag{4}$$

$$|\mathbf{k}_1|^2 = \frac{\omega_1^2}{c^2}\left\{1 + A\left[|E_1|^2 + |E_0|^2\left(1 + \frac{1}{1 + \Omega^2\tau^2}\right)\right]\right\}$$

and introduced the notation

$$A = \frac{2}{45}\frac{\alpha_1 - \alpha_2}{\varepsilon_0 kT}\frac{\partial\varepsilon}{\partial s} = \frac{\varepsilon_2}{2\varepsilon_0}, \qquad k_\omega = \frac{\varepsilon''}{2\varepsilon_0}\frac{\omega}{c}, \qquad c = \frac{c_0}{\sqrt{\varepsilon_0}},$$

where $2k_\omega$ and c_0 are, respectively, the absorption coefficient and the speed of light in vacuum.

For $|E_0| \gg |E_1|$, assuming $|E_0|^2 \sim \text{const}$, from (3) we obtain an exponential growth of $|E_1|^2$ in space (ξ is the coordinate in the direction of $\mathbf{k}_1$),

$$|E_1(\xi)|^2 = |E_1(0)|^2 \exp[g_1(\Omega)\xi] \tag{5}$$

provided that the gain coefficient $g_1(\Omega) > 0$, where

$$g_1(\Omega) = -2k_\omega + A|\mathbf{k}_1|\frac{\Omega\tau}{1+\Omega^2\tau^2}\,|E_0(0)|^2. \tag{6}$$

The solution of (3) in the general case, when $|E_1|$ need not be small, for $\vartheta = 180^\circ$ (backscattering) and $k_\omega = 0$ relates the initial values $|E_0(0)|^2$, $|E_1(L)|^2$ and the final values $|E_0(L)|^2$, $|E_1(0)|^2$ as follows:

$$|E_0(L)|^2 = |E_0(0)|^2 + |E_1(0)|^2 - |E_1(L)|^2, \tag{7}$$

$$|E_1(L)|^2 = \frac{|E_1(0)|^2\left(1 - |E_1(0)|^2/|E_0(0)|^2\right)}{\exp\left\{|\mathbf{k}_0|A\frac{\Omega\tau}{1+\Omega^2\tau^2}\left(|E_0(0)|^2 - |E_1(0)|^2\right)L\right\} - |E_1(0)|^2/|E_0(0)|^2}. \tag{8}$$

Thus, it follows from (8) that at the beginning of the development of the process of stimulated scattering in the Rayleigh-line wing, when $|E_1| \ll |E_0|$, the intensity of the scattered light will grow exponentially. When $|E_1|$ reaches a value comparable with $|E_0|$, the saturation effect will begin to manifest itself and, as is seen from (8), the growth rate of $|E_1|$ will decrease.

2. Relations analogous to (6), (7), and (8) can also be obtained for forward scattering. But, as was pointed out earlier ([2,3]), in stimulated scattering at small angles it is necessary to take into account the interaction of two laser photons, Stokes and anti-Stokes photons. Let us consider such a four-photon interaction in the linear approximation, assuming $|E_0|^2 \sim \text{const}$, $|E_0| \gg |E_1|, |E_2|$. Then from Maxwell' s equations and (2) we obtain the system of equations:

$$i\mathbf{k}_1\nabla E_1 + \gamma_1 E_1 + 2i|\mathbf{k}_1|k_\omega E_1 = |\mathbf{k}_1|^2 A E_0^2 E_2^* \frac{\exp[-i(2\mathbf{k}_0 - \mathbf{k}_1 - \mathbf{k}_2)\mathbf{r}]}{1 - i\Omega\tau}, \tag{9}$$

$$2i\mathbf{k}_2\nabla E_2^* - \gamma_2 E_2^* + 2i|\mathbf{k}_2|k_\omega E_2^* = -|\mathbf{k}_2|^2 A E_0^{*2} E_1 \frac{\exp[i(2\mathbf{k}_0 - \mathbf{k}_1 - \mathbf{k}_2)\mathbf{r}]}{1 - i\Omega\tau},$$

where it must be

$$|\mathbf{k}_0|^2 = \frac{\omega_0^2}{c^2}\left(1 + A|E_0|^2\right). \tag{10}$$

In (9) the notation

$$\gamma_1 = \left\{k_1^2 - \frac{\omega_1^2}{c^2}\left[1 + A|E_0|^2\frac{1}{1 - i\Omega\tau}\right]\right\}$$

has been introduced. For an effective interaction of the laser, Stokes, and anti-Stokes radiations, the condition

$$2\mathbf{k}_0 = \mathbf{k}_1 + \mathbf{k}_2 \tag{11}$$

must be satisfied.

Let the laser radiation propagate along the x-axis. Put $k_{1y} = -k_{2y} = k_y$, $k_{1x} = k_{2x} = k_x$. Considering the variation of the amplitudes E_1 and E_2^* along the x-axis, assuming $E_1 \sim \exp(\beta_1 x)$ and $E_2^* \sim \exp(\beta_2 x)$, we find

$$\beta_1 + i(k_0 - k_x) = \beta_2 - i(k_0 - k_x) = -i\frac{k_x}{|\mathbf{k}_1|}k_\omega \pm$$

$$\pm\frac{k_x|\mathbf{k}_0|}{2}\left\{-\left[\frac{1}{k_0} - \frac{1}{k_1}\right]^2\left[\frac{2A|E_0|^2}{1 - i\Omega\tau} + |\mathbf{k}_0|^2\left(\frac{1}{k_0} - \frac{1}{k_1}\right)^2\right]\right\}^{1/2}. \tag{12}$$

For small scattering angles, assuming condition (11) to be satisfied, ($k_0 \simeq k_x$), we obtain ($\beta_1 = \beta_2 = \beta$)

$$\beta = -k_\omega \pm \frac{|k_0|}{2}\,\vartheta\left(\frac{2A|E_0|^2}{1 - i\Omega\tau} - \vartheta^2\right)^{1/2}, \tag{13}$$

where $\vartheta = k_y/k_0$. From (13), at the angle $\vartheta^2_{\mathrm{opt}} = A|E_0|^2$, it is easy to find the maximum gain coefficient, identical for the Stokes and anti-Stokes parts of the wing,

$$g_2 = -2k_\omega + \frac{|\mathbf{k}_0|\,A|E_0|^2}{(1 + \Omega^2\tau^2)^{1/2}}. \tag{14}$$

The coefficient g_2, in contrast to g_1 (see (5)), has its maximum at $\Omega = 0$.

For small scattering angles, formulas (13) and (14) pass, at $k_\omega = 0$, into expressions obtained earlier in [4].

From the general expression (12), for the case when condition (11) is not fulfilled, for example in backscattering, one obtains a result coinciding with (6).

Thus, the four-photon interaction in stimulated Rayleigh-wing scattering can be observed only in a narrow range of angles in a direction close to the direction of propagation of the exciting laser radiation. An analogous phenomenon can also be observed in the lines of stimulated combination scattering of light.

It may be supposed that a phenomenon analogous to stimulated Rayleigh-wing scattering in liquids can also be observed in solid transparent media (for example, with orientation of molecules in molecular crystals). Then this phenomenon, along with others already discussed [2,3], could contribute to the formation of microcracks in transparent media.

3. Consideration of the effect of orientation of anisotropic molecules of the medium in the strong electric fields of light waves on stimulated Mandelstam–Brillouin scattering (stimulated M.B.) shows that for a two-photon process, instead of the light absorption coefficient $2k_\omega$, one should take a certain effective value

$$2(k_\omega)_{\text{eff}} = 2k_\omega - |\mathbf{k}_0| A |E_0|^2 \frac{\Omega\tau}{1+\Omega^2\tau^2}. \tag{15}$$

In this case the shift of the stimulated M.B. components relative to the laser radiation must be greater than in thermal scattering. The usually observed [2,3] decrease of the shift of the stimulated M.B. components means that, apparently, other effects of the opposite sign are more substantial.

If, in successive stimulated M.B., the light scattered backward and amplified in the laser again enters, together with the laser radiation, a medium consisting of anisotropic molecules, then, owing to the four-photon interaction, at an angle ϑ_{opt} to the direction of the laser radiation not only the Stokes but also the anti-Stokes component of stimulated M.B. will be amplified. This mechanism, together with repeated stimulated M.B. in the interaction region [5], may contribute to the appearance, in stimulated M.B., of an anti-Stokes component and substantially facilitate the appearance of a large number of Stokes components.

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Note: Figure translations are in progress. See original paper for figures.