INTERACTION BETWEEN WAVES IN STIMULATED RAMAN SCATTERING

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Submitted to JETP editor December 16, 1963

J. Exptl. Theoret. Phys. (U.S.S.R.) 46, 2126-2131 (June, 1964)

An analysis is given of the amplifying properties of a medium based on the phenomenon of stimulated Raman scattering. In contrast to reports available in the literature, we consider cases when the waves are propagated along various directions. The interaction efficiency of Stokes and Rayleigh wave components of the field is determined in this case by the angle between the electric field of the Rayleigh component and the wave vector of the Stokes component. Interaction of the three wave components (Rayleigh, Stokes and anti-Stokes) is possible only for certain mutual orientations of their wave vectors. As a rule, however, this interaction is much less efficient than the interaction of two components, Rayleigh and Stokes.

1. The phenomenon of stimulated Raman scattering may be described as follows: when certain organic liquids are irradiated by intense light of frequency ω_p, a very intense coherent Stokes component of the light is generated. The frequency of this component ω_c is related to the frequency of the incident light by the relation $\omega_p - \omega_c = \omega_0$, where ω_0 is a molecular vibration frequency present in the ordinary Raman spectrum. The coherent anti-Stokes component of the light, of frequency $\omega_a = \omega_p + \omega_c$, has not been observed in the first experiments [1,2], but it has been observed later [3] that it also occurs under definite conditions.

The quantum mechanical theory of stimulated Raman scattering has been developed in a number of papers [4-6].

The mechanism of this phenomenon has also been studied from the point of view of classical physics [7,8]. We have previously [7] investigated the process of energy transfer due to the interaction of the Rayleigh component of the field (the pumping wave) with its Stokes component in the case where the waves are propagating in the same direction. In this case the anti-Stokes component does not play any significant role in the interaction between the waves.

These conditions (i.e., both waves propagating in one direction) indeed obtained in the first experiments [1,2] on stimulated Raman scattering. However in constructing amplifiers or oscillators using the principle of stimulated Raman scattering it is preferable to have conditions such that the waves are propagating in different directions. When the propagation directions of the waves are different, the equations describing this process contain additional terms which cannot be neglected and which greatly complicate the solution of the equations. As was shown by Garmire, Pandarese, and Townes [8], under the condition

$$2\mathbf{k}_r - \mathbf{k}_s - \mathbf{k}_a = 0, \tag{1}$$

(where $\mathbf{k}_s$, $\mathbf{k}_a$ and $\mathbf{k}_r$ are the wave vectors of the Stokes, anti-Stokes and Rayleigh components of the light respectively) the anti-Stokes component does play an important part in the interaction between the waves. Condition (1) may be satisfied in media exhibiting normal dispersion, but it is then necessary that the wave vectors $\mathbf{k}_s$, $\mathbf{k}_a$ and $\mathbf{k}_r$ be in different directions.

The present paper treats the interaction between the Rayleigh and Stokes components of the light when their propagation directions are different, and also treats the interaction of these components with the anti-Stokes components when this interaction is efficient.

2. We consider the case in which the anti-Stokes component of the field is not efficiently generated and in which the frequency ω_r is close to the frequency difference $\omega_r - \omega_0$ but is not necessarily equal to it. The behavior of the electric field $\mathbf{E}$ in the medium is described by Maxwell's equations:

$$\frac{1}{c^2}\frac{\partial^2}{\partial t^2}(\mathbf{E} + 4\pi\mathbf{P}) = -\operatorname{rot}\operatorname{rot}\mathbf{E}, \quad \operatorname{div}(\mathbf{E} + 4\pi\mathbf{P}) = 0, \tag{2*}$$

*rot = curl.

where $\mathbf{P}$ is the polarization per unit volume. We consider a semi-infinite medium whose boundary is the plane $z = 0$. Light waves of frequency ω_s and ω_r are propagating in this medium and the electric field strength is given by

$$\mathbf{E} = \mathbf{E}_s e^{i(\omega_s t - \mathbf{k}_s \mathbf{r})} + \mathbf{E}_r e^{i(\omega_r t - \mathbf{k}_r \mathbf{r})} + \text{c.c.} \tag{3}$$

Here $\mathbf{E}_s$ and $\mathbf{E}_r$ are slowly varying complex amplitudes which may depend both on time and on the coordinate z. Corresponding to this electric field there is a polarization $\mathbf{P}$ given by

$$\mathbf{P} = \mathbf{P}_s e^{i(\omega_s t - \mathbf{k}_s \mathbf{r})} + \mathbf{P}_r e^{i(\omega_r t - \mathbf{k}_r \mathbf{r})} + \text{c.c.}, \tag{4}$$

where $\mathbf{P}_s$ and $\mathbf{P}_r$ may depend on both $\mathbf{E}_s$ and $\mathbf{E}_r$ simultaneously, since the polarization of the medium depends on the total field strength.

In the steady state regime, which is the only one considered here, we have $\partial E_n/\partial t = 0$. The index n takes the values s and r. Then putting (3) and (4) in (2) and taking account of the slowness of the variation of the complex amplitudes $\mathbf{E}_n$, we obtain

$$\frac{\partial \mathbf{E}_n}{\partial z} = \frac{i}{2k_n \cos\gamma_n} \times \left[k_n^2 \mathbf{E}_n - \frac{\omega_n^2}{s^2}(\mathbf{E}_n + 4\pi \mathbf{P}_n) - 4\pi \,\mathrm{grad}\,\mathrm{div}\,\mathbf{P}_n \right], \tag{5}$$

where γ_n is the angle between the z axis and the wave vector $\mathbf{k}_n$. In order to obtain a closed system of equations for the $\mathbf{E}_n$ we must express the amplitudes of the polarizations $\mathbf{P}_n$ in terms of the $\mathbf{E}_n$. We write $\mathbf{P}_n$ in the form

$$\mathbf{P}_n = \mathbf{P}_n^0 + \mathbf{P}_n', \tag{6}$$

where $\mathbf{P}_n^0$ is linear in $\mathbf{E}_n$, and $\mathbf{P}_n'$ is a small non-linear term.

For simplicity we consider the molecules in the irradiated liquid to be isotropic. One may then show [7] that

$$\mathbf{P}_s' = \frac{\Delta - i}{1 + \Delta^2} \varkappa (\mathbf{E}_s \mathbf{E}_r^*) \mathbf{E}_r, \quad \mathbf{P}_r' = \frac{\Delta + i}{1 + \Delta^2} \varkappa (\mathbf{E}_s^* \mathbf{E}_r) \mathbf{E}_s, \tag{7}$$

where $\Delta = (\delta\omega_0)^{-1}[\omega_0^2 - (\omega_r - \omega_s)^2]$ and δ and κ are coefficients depending on the properties of the molecules of the irradiated liquid. Putting (6) in (5) and also putting $k_n^2 = \omega_n^2 c^{-2}(1 + 4\pi P_n/E_n)$, and using (7) we obtain

$$\frac{\partial \mathbf{E}_s}{\partial z} = \alpha_s (\mathbf{E}_s \mathbf{E}_r^*) \left[\mathbf{E}_r - \frac{\mathbf{k}_s}{k_s^2} (\mathbf{k}_s \mathbf{E}_r) \right],$$

$$\frac{\partial \mathbf{E}_r}{\partial z} = -\alpha_r (\mathbf{E}_s^* \mathbf{E}_r) \left[\mathbf{E}_s - \frac{\mathbf{k}_r}{k_r^2} (\mathbf{k}_r \mathbf{E}_s) \right];$$

$$\alpha_s = \frac{1 - i\Delta}{1 + \Delta^2} \varkappa \frac{2\pi k_s}{\cos\gamma_s}, \qquad \alpha_r = \frac{1 + i\Delta}{1 + \Delta^2} \varkappa \frac{2\pi k_r}{\cos\gamma_r}. \tag{8}$$

If the wave vectors $\mathbf{k}_s$ and $\mathbf{k}_r$ are parallel, the scalar products $\mathbf{k}_s \cdot \mathbf{E}_r$ and $\mathbf{k}_r \cdot \mathbf{E}_s$ are zero and Eqs. (8) are very much simplified. This case has been considered by us previously [7]. In the case of non-parallel wave vectors, the terms containing the scalar products are non-zero and because of their presence the differential $d\mathbf{E}_n$ is perpendicular to the wave vector $\mathbf{k}_n$, i.e., no longitudinal component of the field is generated. In general the simultaneous solution of the system (8) is extremely complicated. Hence in what follows we will analyze the case of linear amplification of the signal wave, for which $|\mathbf{E}_r| \gg |\mathbf{E}_c|$ and the reaction of the amplified signal on the pump wave may be neglected; we will also treat several of the simplest nonlinear situations for which the latter approximation is not valid.

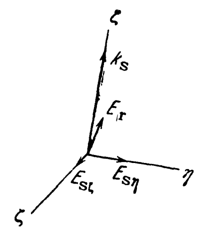

FIG. 1. Orientation of the vectors $\mathbf{k}_s$, $\mathbf{E}_r$ and $\mathbf{E}_s$ with respect to the coordinate system ξ, η, and ζ.

3. If $|\mathbf{E}_s| \gg |\mathbf{E}_r|$ we may treat $\mathbf{E}_r$ as a constant. It then follows from (8) that

$$\frac{\partial \mathbf{E}_s \mathbf{E}_r^*}{\partial z} = \alpha_s \mathbf{E}_s \mathbf{E}_r^* |\mathbf{E}_r|^2 \sin^2\beta, \tag{9}$$

where β is the angle between the wave vector $\mathbf{k}_s$ and the vector $\mathbf{E}_r$. Integrating (9) we obtain

$$\mathbf{E}_s \mathbf{E}_r^* = C e^{\chi z};$$

$$C = (\mathbf{E}_s(0) \mathbf{E}_r^*), \qquad \chi = \alpha_s \sin^2\beta |\mathbf{E}_r|^2. \tag{10}$$

We introduce a coordinate system ξ, η, ζ, where the ζ axis is along $\mathbf{k}_s$ and η is coplanar with $\mathbf{k}_s$ and $\mathbf{E}_r$ (Fig. 1). Then putting (10) in (9) and integrating we obtain

$$E_{s\xi} = C_\xi, \qquad E_{s\eta} = E_{r\eta} C e^{\chi z} / |\mathbf{E}_r|^2 \sin\beta + C_\eta;$$
$$C_\xi = E_{s\xi}(0), \qquad C_\eta = E_{s\eta}(0) - C E_{r\eta} / |E_r|^2 \sin\beta. \tag{11}$$

It is clear from (11) that only the component of $\mathbf{E}_s$ parallel to $\mathbf{E}_r$ is amplified. This means that the plane of polarization of the Stokes component varies during propagation and for large z approaches the direction of the projection of the vector $\mathbf{E}_r$ on the plane perpendicular to $\mathbf{k}_s$. The amplification is a maximum when $\mathbf{E}_r$ is perpendicular to $\mathbf{k}_s$, independent of the mutual disposition of the wave vectors. In particular the amplification does not decrease even in the case in which $\mathbf{k}_s$ and $\mathbf{k}_r$ are perpendicular as long as $\mathbf{E}_r \perp \mathbf{k}_s$. The latter case is of significant practical advantage in the construction of amplifiers using stimulated Raman emission. Amplification does not occur in the case of isotropic molecules if $\mathbf{E}_r \parallel \mathbf{k}_s$.

4. For sufficiently large z the condition $|E_s| \ll |E_r|$ is no longer satisfied and Eq. (11), describing the regime of linear amplification, becomes invalid. The nonlinear regime is described by the general system of equations (8), whose solution, as we already have pointed out, is an extremely complex problem. However the nonlinear regime is of interest from the point of view of achieving the maximum gain coefficient for the amplification.

We therefore consider the special case (which has an analytic solution) in which $\mathbf{E}_s$ and $\mathbf{E}_r$ are either polarized in the same plane or else are parallel. In this case the solution of (8) has the form

$$|\mathbf{E}_s|^2 = \frac{UCe^{\lambda z}}{1+Ce^{\lambda z}}, \qquad |\mathbf{E}_r|^2 = \frac{\alpha_r}{\alpha_s}(U - |\mathbf{E}_s|^2),$$

$$\varphi_r = \frac{\Delta}{1+\Delta^2}\cos^{-2}\widehat{\mathbf{k}_s\mathbf{k}_r}\ln(1+Ce^{\lambda z}),$$

$$\varphi_s = \frac{\Delta}{1+\Delta^2}\alpha_r U z - \varphi_r. \tag{12}$$

Here φ_n is the phase of $\mathbf{E}_n$, that is, we have $E_n = |E_n| e^{i\varphi_n}$,

$$U = \frac{\alpha_s}{\alpha_r}|\mathbf{E}_s|^2 + |\mathbf{E}_s|^2, \quad C = \frac{|\mathbf{E}_s(0)|^2}{U - |\mathbf{E}_s(0)|^2}, \quad \lambda = \alpha_r \sin^2\beta\, U.$$

It is clear from (12) that for $z \to \infty$ the value of $|E_s|^2$ does not depend on the mutual orientation of the vectors $\mathbf{k}_s$ and $\mathbf{k}_r$, with the exception of the degenerate case $\beta = 0$. The mutual orientation of the wave vectors is important only in determining the spatial rate of redistribution of energy between the Stokes and Rayleigh components. This rate, however, plays an important role under real conditions in the presence of energy dissipation. For $z \to \infty$ the energy of the Rayleigh component is completely transformed into energy of the Stokes component and energy in the molecular vibrations.

In the presence of a mismatch in the phases of the waves, the φ_n vary at a rate which is inversely proportional to the energy in the corresponding wave, so that the following relation is fulfilled:

$$\alpha_s |E_r|^2 \frac{\partial\varphi_s}{\partial z} = \alpha_r |E_s|^2 \frac{\partial\varphi_r}{\partial z}. \tag{13}$$

In the absence of mismatch, the phases do not vary along the direction of propagation.

5. As has already been noted above in the general case of the mutual orientation of the wave vectors $\mathbf{k}_s$, $\mathbf{k}_a$ and $\mathbf{k}_r$, the anti-Stokes component of the field does not take part to any appreciable extent in the interaction between the Stokes and the Rayleigh components of the waves. However the anti-Stokes component does take part in the interaction when the relationship (1) between the wave vectors is satisfied. The aim of this section is to discuss this case.

We consider the case in which three waves of frequencies ω_s, ω_a, and ω_r are propagating in the medium. We have $\omega_r - \omega_s = \omega_a - \omega_r$ and as before this difference is close to ω_0 but not necessarily equal to it. Using the same method as led to (8), we may in this case obtain the following system of equations for the $\mathbf{E}_n$:

$$\frac{\partial \mathbf{E}_s}{\partial z} = \alpha_s[\mathbf{E}_s\mathbf{E}_r^* + \mathbf{E}_r\mathbf{E}_a^* e^{i(2\mathbf{k}_r - \mathbf{k}_s - \mathbf{k}_a)\mathbf{r}}]\left[\mathbf{E}_r - \frac{\mathbf{k}_s}{k_s^2}(\mathbf{k}_s\mathbf{E}_r)\right],$$

$$\frac{\partial \mathbf{E}_a^*}{\partial z} = -\alpha_a^*[\mathbf{E}_r\mathbf{E}_a^* + \mathbf{E}_s\mathbf{E}_r^* e^{i(2\mathbf{k}_r - \mathbf{k}_s - \mathbf{k}_a)\mathbf{r}}]\left[\mathbf{E}_r - \frac{\mathbf{k}_a}{k_a^2}(\mathbf{k}_a\mathbf{E}_r)\right],$$

$$\frac{\partial E_r}{\partial z} = -\alpha_r[\mathbf{E}_r\mathbf{E}_s^* + \mathbf{E}_a\mathbf{E}_r^* e^{i(2\mathbf{k}_r - \mathbf{k}_s - \mathbf{k}_a)\mathbf{r}}]\left[\mathbf{E}_s - \frac{\mathbf{k}_r}{k_r^2}(\mathbf{k}_r\mathbf{E}_s)\right]$$

$$+ \alpha_r^*[\mathbf{E}_r\mathbf{E}_a^* + \mathbf{E}_s\mathbf{E}_r^* e^{i(2\mathbf{k}_r - \mathbf{k}_s - \mathbf{k}_a)\mathbf{r}}]\left[\mathbf{E}_a - \frac{\mathbf{k}_r}{k_r^2}(\mathbf{k}_r\mathbf{E}_a)\right]. \tag{14}$$

Here

$$\alpha_a = \frac{1+i\Delta}{1+\Delta^2}\varkappa\frac{2\pi k_a}{\cos\gamma_a}.$$

If condition (1) is not fulfilled one may neglect the oscillating terms in (14). Energy from the wave $\mathbf{E}_a$ will be transferred to the wave $\mathbf{E}_r$ and from the wave $\mathbf{E}_r$ to the wave $\mathbf{E}_s$. We now consider the case in which (1) is satisfied and the exponent in (14) is equal to unity. Then for $|E_s| > |E_a|$ and for $\cos(2\varphi_r - \varphi_s - \varphi_a) < -|E_a|/|E_s|$ both $\mathbf{E}_s$ and $\mathbf{E}_a$ will grow.

Equations (13) have the first integral

$$\frac{|\mathbf{E}_s|^2}{\alpha_s} + \frac{|\mathbf{E}_a|^2}{\alpha_a} + \frac{|\mathbf{E}_r|^2}{\alpha_r} = \text{const}. \tag{15}$$

This equation may be given the following form

$$N_s + N_a + N_r = N, \tag{15'}$$

where N_n is the number of photons at frequency ω_n crossing a unit area perpendicular to z per unit time.

We assume further that all three waves are either polarized parallel or are in the same plane. We consider the region of linear amplification for which $|E_r| \gg |E_s|$, $|E_r| \gg |E_a|$ and $\mathbf{E}_r$ may be considered constant. It is convenient to chose the time origin so that $\mathbf{E}_r$ is real. If $\mathbf{E}_r \parallel \mathbf{E}_s \parallel \mathbf{E}_a$ it follows from (14) that

$$\partial\mathbf{E}_s/\partial z = \alpha_s|\mathbf{E}_r|^2(\mathbf{E}_s + \mathbf{E}_a^*),$$

$$\partial\mathbf{E}_a^*/\partial z = -\alpha_a^*|\mathbf{E}_r|^2(\mathbf{E}_s + \mathbf{E}_a^*). \tag{16}$$

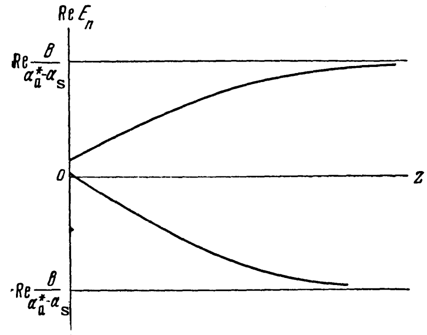

FIG. 2. The behavior of the amplitudes of the Stokes and anti-Stokes waves with increasing coordinate z.

If the vectors $\mathbf{E}_r$, $\mathbf{E}_s$, $\mathbf{E}_a$, $\mathbf{k}_r$, $\mathbf{k}_s$ and $\mathbf{k}_a$ are coplanar, then we obtain along with (16)

$$\partial \mathbf{E}_s/\partial z = \alpha_s \cos \widehat{\mathbf{k}_s\mathbf{k}_r}\, |\mathbf{E}_r|^2 (E_s \cos \widehat{\mathbf{k}_s\mathbf{k}_r} + E_a^* \cos \widehat{\mathbf{k}_a\mathbf{k}_r})\, \mathbf{n}_s,$$

$$\partial \mathbf{E}_a^*/\partial z = -\alpha_a^* \cos \widehat{\mathbf{k}_a\mathbf{k}_r}\, |\mathbf{E}_r|^2 (E_s \cos \widehat{\mathbf{k}_s\mathbf{k}_r} + E_a^* \cos \widehat{\mathbf{k}_a\mathbf{k}_r})\, \mathbf{n}_a. \tag{16'}$$

In the right hand side of (16′) E_n signifies the scalar amplitude, $\mathbf{n}_s$ and $\mathbf{n}_a$ are unit vectors directed along $\mathbf{E}_s$ and $\mathbf{E}_a$, respectively.

Integrating (16), we obtain

$$\alpha_a^* \mathbf{E}_s + \alpha_s \mathbf{E}_a^* = \mathbf{B}, \tag{17}$$

$$\mathbf{E}_s = \mathbf{B} / (\alpha_a^* - \alpha_s) + C_0 \exp[(\alpha_s - \alpha_a^*) E_r^2 z], \tag{18}$$

where

$$C_0 = \mathbf{E}_s(0) - \mathbf{B} / (\alpha_a^* - \alpha_s).$$

The relations (17) and (18) are valid for (16′) if one replaces α_s and α_a^* by $\alpha_s \cos^2(\mathbf{k}_s \cdot \mathbf{k}_r / k_s \parallel k_r)$ and $\alpha_a^* \cos^2(\mathbf{k}_a \cdot \mathbf{k}_r / k_a k_r)$.

It follows from (17) and (18) that in the most usual case, $\mathrm{Re}\,\alpha_a > \mathrm{Re}\,\alpha_s$, even in the linear regime the amplitudes $\mathbf{E}_s$ and $\mathbf{E}_a$ will approach the stationary values $\mathbf{B}/(\alpha_a^* - \alpha_s)$ and $-\mathbf{B}/(\alpha_a^* - \alpha_s)$ respectively (Fig. 2). The case $\mathrm{Re}\,\alpha_a < \mathrm{Re}\,\alpha_s$ is not very probable and will not be treated here. The maximum amplification coefficients for the Stokes and anti-Stokes components are given by the quantities

$$\frac{E_s(\infty)}{E_s(0)} = \frac{\alpha_a^*}{\alpha_a^* - \alpha_s}\left[1 + \frac{\alpha_s E_a^*(0)}{\alpha_a^* E_s(0)}\right],$$

$$\frac{E_a^* \infty}{E_a^*(0)} = \frac{\alpha_s}{\alpha_a^* - \alpha_s}\left[1 + \frac{\alpha_a^*}{\alpha_s}\frac{E_s(0)}{E_a^*(0)}\right]. \tag{19}$$

The first of these coefficients under real conditions does not exceed several times 10, whereas the second coefficient may be somewhat larger. However the nonlinear regime is not realized as a rule in this case.

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Translated by J. A. Armstrong
316