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STIMULATED FOUR-PHOTON INTERACTION AND ITS INFLUENCE ON STIMULATED RAYLEIGH-WING SCATTERING

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The series of closely related phenomena known as stimulated Raman, Brillouin, and Rayleigh-wing scattering and beam trapping have given rise to a large number of interesting results. We consider here another related phenomenon of stimulated light scattering which involves a stimulated four-photon or light-by-light process (briefly discussed previously[1]) with zero or near-zero frequency shift. This process will be shown to have some aspects not discussed previously and will be shown further to play a significant role in nonlinear processes in liquids.

The intensity-dependent dielectric constant[2] or third-order susceptibility[3] responsible for the coupling of the waves in this process can be considered to have two parts, one part which can respond to sum frequencies and depends for its size on nearby two-photon states (e.g., electronic) and another part which can respond to difference frequencies and depends for its size on nearby Raman states (e.g., acoustical, rotational, and vibrational). The latter part is usually dominant because of near resonance of difference frequencies with low-lying excitations. We pay particular attention to the molecular-orientation Kerr effect.

Consider the interaction of two plane light waves in a medium with an intensity-dependent refractive index. For these two waves the total electric field is $E=E_0+E_1$, where

$$E_i=\tfrac{1}{2}\{\mathcal{E}_i\exp[i(\vec{k}_i\cdot\vec{r}-\omega_i t)]+\text{c.c.}\}\quad(i=0,1). \tag{1}$$

We assume E_0 is a strong incident wave and E_1 is a weak scattered wave. Let us for the present consider only the degenerate case $\omega_0=\omega_1$. The intensity-dependent part of the dielectric constant is given by

$$\Delta\epsilon=\epsilon_2E^2=\tfrac{1}{2}\epsilon_2[|\mathcal{E}_0|^2+(\mathcal{E}_0\mathcal{E}_1{}^*e^{i\vec{q}\cdot\vec{r}}+\text{c.c.})], \tag{2}$$

where we assume $\epsilon_2>0$ and real, and where $\vec{q}=\vec{k}_0-\vec{k}_1$. Also, we have neglected higher order terms in $\mathcal{E}_1$ as well as terms oscillating at $2\omega_0$. The second term indicates that stationary periodic layers are set up in the medium which can produce reflections in a manner similar to the Raman-Nath effect. The change in ϵ gives rise to an induced nonlinear polarization wave, which to first order in the weak wave is

$$\Delta P=(\epsilon_2/8\pi)\{|\mathcal{E}_0|^2(E_0+2E_1)+\tfrac{1}{2}[\mathcal{E}_0{}^2\mathcal{E}_1{}^*e^{i(\vec{k}_0+\vec{q})\cdot\vec{r}-i\omega_0t}+\text{c.c.}]\}. \tag{3}$$

The terms containing $|\mathcal{E}_0|^2$ correspond to changes in phase velocity of the strong and weak waves, respectively: There is no energy transferred to either wave from these terms since the polarization waves are in phase with their respective fields. Note that the weak wave has an additional increase in refractive index

$$\Delta n=\Delta n_{\text{weak}}-\Delta n_{\text{strong}}=\epsilon_2|\mathcal{E}_0|^2/4n_0, \tag{4}$$

where n_0 is the zero-field index of refraction.

Consequently, there exists an additional lengthening of the weak-wave propagation vector $\Delta k = \Delta n(\omega_0/c)$ (weak-wave retardation).

The last term in (3) represents a polarization wave with a wave vector $\vec{k}_0+\vec{q}$ which should radiate. Hence, if we add a second weak wave E_2 to the total field, where $E_2=\frac{1}{2}\mathcal{E}_2\exp(i\vec{k}_2\cdot\vec{r}-i\omega_0 t)$ + c.c., it will be possible to amplify this wave if $\vec{k}_2=\vec{k}_0+\vec{q}$ and if the phase of this wave is properly chosen. Because of the additional lengthening of the wave vectors of the weak waves, $\vec{k}_1$ and $\vec{k}_2$ are not necessarily collinear with $\vec{k}_0$. The power transferred into E_2 is

$$\mathcal{P}_2=E_2\left(\frac{d\Delta P}{dt}\right)_2=\frac{\omega_0\epsilon_2}{16\pi}\mathrm{Im}(\mathcal{E}_0{}^2\mathcal{E}_1{}^*\mathcal{E}_2{}^*)=\mathcal{P}_1, \qquad (5)$$

where the last equality indicates that power is also transferred symmetrically into E_1. The power gain will be positive if the phases φ_1 and φ_2 of the fields $\mathcal{E}_i=|\mathcal{E}_i|\exp(i\varphi_i)$ are chosen so that $\pi>2\varphi_0-\varphi_1-\varphi_2>0$. The maximum power transfer from the strong wave to the weak occurs when $2\varphi_0-\varphi_1-\varphi_2=\frac{1}{2}\pi$. The last term in (3), the cross coupling term, may also contribute additional weak-wave retardation or advancement, depending on the phases of the waves. However, for maximum energy transfer the cross coupling term does not contribute to a change in the phase velocity of either weak wave. Note that in order to phase match in the forward direction (i.e., $\vec{k}_0$, $\vec{k}_1$, and $\vec{k}_2$ are collinear), the weak-wave retardation must be cancelled by an advancement due to the cross coupling term. To achieve this, however, the phases must be chosen so that $2\varphi_0-\varphi_1-\varphi_2=\pi$, in which case the energy transfer is zero. Because of the weak-wave retardation, the weak waves in the maximum-energy-transfer case travel at angles

$$\Theta_{\mathrm{opt}}=\pm\left(\frac{2\Delta k}{k_0}\right)^{1/2}=\pm\left(\frac{2\Delta n}{n_0}\right)^{1/2}=\pm\left(\frac{\epsilon_2|\mathcal{E}_0|^2}{2\epsilon_0}\right)^{1/2} \qquad (6)$$

with respect to the strong wave. Note that this angle is equal to the critical angle.[2]

The threshold condition is found from the requirement that for net gain in either weak wave

$$\mathcal{P}_i \geq (n_0 c/8\pi l_i)|\mathcal{E}_i|^2 \quad (i=1,2), \qquad (7)$$

where l_i is the decay length. For the case of resonators, $l_i=L_i(1-R_i)^{-1}$, where L_i is the cavity length and R_i is the reflectivity of the mirrors. Combining these conditions for both weak waves, we find that the threshold is

$$\Delta n=\frac{\epsilon_2|\mathcal{E}_0|^2}{4n_0}\geq\frac{1}{4\pi}\left(\frac{\lambda_0{}^2}{l_1 l_2}\right)^{1/2}. \qquad (8)$$

The gain per unit length is $g=4\pi\Delta n/\lambda_0$, where λ_0 is the wavelength of the incident light in vacuo.

A typical threshold, evaluated with $l_1=l_2=10$ cm, $\epsilon_2=7.5\times10^{-11}$ esu (Kerr effect in CS_2), and $\lambda_0=6943$ Å, is 9 MW/cm^2. The gain at an intensity of 100 MW/cm^2 is 1.1 cm^{-1}, which is comparable with the stimulated Raman gain of 1 cm^{-1} for CS_2 for the same laser intensity.[4] For this intensity the optimum angle of emission Θ_{opt} is 2.8×10^{-3} rad.

The above analysis can easily be generalized to the case where the two weak waves E_1 and E_2 have different frequencies ω_1 and ω_2, where $\omega_0-\omega_1=\Omega=\omega_2-\omega_0$. In the expression for the threshold (8) and the gain, $\lambda_0{}^2$ is replaced by $\lambda_1\lambda_2$. This implies the possibility of a four-photon parametric oscillator[1,5] whenever there exists a real $\chi^{(3)}=\epsilon_2/4\pi$.

Stimulated light-by-light scattering and its associated weak-wave retardation strongly influences stimulated Rayleigh-wing scattering, a consideration which has not been included in previous treatments.[6] The fact that both scattering processes have large gains in the near-forward direction, where Stokes-anti-Stokes coupling can occur, requires a simultaneous treatment of the two processes. Using the coupled wave approach,[7] let us investigate the interaction of the three waves E_0, E_1, and E_2 given by (1). As before we assume that $\mathcal{E}_0 \gg \mathcal{E}_1, \mathcal{E}_2$, so that the weak waves affect only negligibly the strong wave in the process of interaction. Hence $\mathcal{E}_0$ will be considered a constant.

The specific form of intensity-dependent dielectric change responsible for stimulated Rayleigh-wing scattering, assuming a simple orientational relaxation time τ, is[8,9]

$$\Delta\epsilon=\frac{\epsilon_2}{2}\left\{|\mathcal{E}_0|^2+\left(\frac{\mathcal{E}_0{}^*\mathcal{E}_1}{1+i\Omega\tau}e^{i\Omega t-i\vec{q}\cdot\vec{r}}+\frac{\mathcal{E}_0{}^*\mathcal{E}_2}{1-i\Omega\tau}e^{-i\Omega t+i\vec{q}\cdot\vec{r}}+\mathrm{c.c.}\right)\right\}, \qquad (9)$$

where $\vec{q}$ and Ω have been defined previously. This dielectric constant modulates the total field $E=E_0+E_1+E_2$ to produce polarization source terms which drive the wave equation as follows:

$$-k_x^{\,2}\mathcal{E}_1 + 2ik_{1z}\frac{d\mathcal{E}_1}{dz} = -\frac{\epsilon_2\omega_1^{\,2}}{2c^2}\{|\mathcal{E}_0|^2\mathcal{E}_1 + \mathcal{E}_0^{\,2}\mathcal{E}_2^{\,*}\}\times\frac{1}{1+i\Omega\tau}, \tag{10}$$

$$-k_x^{\,2}\mathcal{E}_2^{\,*} - 2ik_{2z}\frac{d\mathcal{E}_2^{\,*}}{dz} = -\frac{\epsilon_2\omega_2^{\,2}}{2c^2}\{|\mathcal{E}_0|^2\mathcal{E}_2^{\,*} + \mathcal{E}_0^{\,*2}\mathcal{E}_1\}\times\frac{1}{1+i\Omega\tau}, \tag{11}$$

where we have chosen

$$k_{iz}^{\,2} - \frac{\epsilon_0\omega_i^{\,2}}{c^2} - \frac{\epsilon_2|\mathcal{E}_0|^2}{2c^2}\omega_i^{\,2} = 0 \quad (i=1,2), \tag{12}$$

and where we have assumed that $\mathcal{E}_1$, $\mathcal{E}_2$ are slowly varying functions of z, the direction of propagation of the strong wave (a good approximation when $k_x, g \ll k_0$). The boundary conditions are determined at the plane $z=0$. Because of wave-vector matching, $k_x = k_{1x} = -k_{2x}$. Substitution of trial solutions $\mathcal{E}_1 = \mathcal{E}_{10}e^{i\gamma z}$ and $\mathcal{E}_2^* = \mathcal{E}_{20}^*e^{i\gamma z}$ gives eigenvalues

$$\gamma_\pm = \pm\frac{\Theta k_0}{2}\left(\Theta^2 - \frac{\epsilon_2|\mathcal{E}_0|^2}{\epsilon_0}\frac{1}{1+i\Omega\tau}\right)^{1/2}, \tag{13}$$

where $\Theta = k_x/k_0$ and where we have assumed $\Omega/\omega_0 \ll 1$. The gain is given by $g = -2\,\mathrm{Im}\gamma_-$, the maximum of which for all Ω occurs at Θ_{opt} given by (6). The gain at the optimum angle is

$$g_{\mathrm{opt}} = (k_0\epsilon_2|\mathcal{E}_0|^2/2\epsilon_0)(1+\Omega^2\tau^2)^{-1/2}$$
$$= (4\pi/\lambda_0)\Delta n(1+\Omega^2\tau^2)^{-1/2}, \tag{14}$$

which gives the result given after (8) when $\Omega = 0$. It should also be noted that when $\Omega = 0$, Eq. (13) reduces to a result found recently by Bespalov and Talanov.[10] A plot of gain versus angle for various values of $\Omega\tau$ is given in Fig. 1. At large angles the anti-Stokes wave is decoupled from the Stokes wave, and hence we expect that the Stokes gain becomes independent of angle as is verified by the figure as well as by the asymptotic form of (13) for large angles,

$$g = -2\,\mathrm{Im}\gamma_- = k_0\frac{\epsilon_2|\mathcal{E}_0|^2}{2\epsilon_0}\frac{\Omega\tau}{1+(\Omega\tau)^2}. \tag{15}$$

Although our above approximations are not valid at very large angles ($k_x \approx k_0$), Eq. (15) is nevertheless the correct expression at very large angles for the gain in the direction of propagation. The maximum gain for the degenerate or near-degenerate case ($\Omega\tau \ll 1$) in (14) is twice as large as the maximum stimulated Rayleigh-wing gain without coupling to the anti-Stokes wave [$\Omega\tau = 1$ in (15)]. This is due to the fact that for Rayleigh-wing scattering, $\mathrm{Max}\,\mathrm{Im}\epsilon_2 = \frac{1}{2}\mathrm{Max}\,\mathrm{Re}\epsilon_2$. The opposite ratio holds for the usual Raman effect, where the linewidth is much smaller than the Raman frequency. This arises from the fact that the frequency dependence of the real and imaginary parts of the nonlinear susceptibility $\chi^{(3)}$ in Rayleigh-wing scattering is interchanged from that for Raman scattering. Clearly, from the figure, the gain is maximum near $\Theta = 0$, implying that the coupling to the anti-Stokes wave actually enhances the Stokes gain in contrast to the stimulated Raman case,[7] an inelastic process, where the coupling to the anti-Stokes wave near the phase-matched angle greatly depresses the gain. The maximum in the gain in the figure is due to light-by-light scattering (an elastic process).

The asymmetry ratio ρ for Stokes to anti-Stokes at the optimum angle is

$$\rho_{\mathrm{opt}} = \frac{|\mathcal{E}_{10}|^2 - |\mathcal{E}_{20}|^2}{|\mathcal{E}_{10}|^2 + |\mathcal{E}_{20}|^2} = \frac{\Omega\tau}{[1+(\Omega\tau)^2]^{1/2}}. \tag{16}$$

Although the above results are strictly valid for weak beams which affect negligibly the strength of the strong incident beam, it is interesting

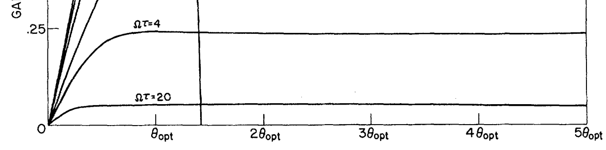

FIG. 1. Plot of gain versus angle for several values of $\Omega\tau$. Using Eq. (13), the gain is given by $g = (1/\sqrt{2})\times k_0\Theta\{[(\delta-\Theta^2)^2 + (\delta\Omega\tau)^2]^{1/2} + (\delta-\Theta^2)\}^{1/2}$, where $\delta = \epsilon_2|\mathcal{E}_0|^2/[\epsilon_0(1+\Omega^2\tau^2)]$. Also $g_{\max} = k_0\epsilon_2|\mathcal{E}_0|^2/(2\epsilon_0)$. The envelope of these curves is found by setting $\Omega\tau = 0$ for $\Theta \leq (\frac{3}{2})^{1/2}\Theta_{\mathrm{opt}}$ and by setting $\Omega\tau = [(4\Theta^2 - 3\epsilon_2|\mathcal{E}_0|^2/\epsilon)/4\Theta^2 - \epsilon_2|\mathcal{E}_0|^2/\epsilon_0)]^{1/2}$ for $\Theta \geq (\frac{3}{2})^{1/2}\Theta_{\mathrm{opt}}$.

to connect the results (when $\Omega = 0$) to the self-focusing and the self-trapping of strong beams, where the exact nonlinear problems have been solved numerically. Beam self-focusing can be considered a self-pumped version of light-by-light scattering where the Fourier components of the beam, traveling close to the forward direction, scatter into components at larger angles and thereby decrease the beam size. For a Gaussian beam of diameter d, we take $k_{xd} = 2/d$ as the dominant Fourier component. Then, if $k_{xd}^2 \ll (\epsilon_2 |\mathcal{E}_0|^2/\epsilon_0) k_0^2$, we obtain from (13) the focusing distance[11] $z_f = 1/(-2\,\mathrm{Im}\gamma_-) = \frac{1}{2}d(\epsilon_2|\mathcal{E}_0|^2/\epsilon_0)^{-1/2}$ as the distance at which we expect marked intensification of the center of the beam.[12]

Self-trapping can be viewed as the steady-state limit of light-by-light scattering, where the various Fourier components lose as much as they gain by scattering and diffracting into each other. Following Bespalov and Talanov,[10] if we set $\Theta_{\mathrm{opt}} = \Theta_{\mathrm{crit}} = k_x/k_0$, where $k_x = 1.22 \times (\pi/d)$, then (6) can be rewritten as[2] $P_{\mathrm{crit}} = (1.22\lambda)^2 c/(128 n_2)$.

Furthermore, the case in which a beam is initially a weak perturbation on a strong background wave can lead to growth and containment of the weak beam and is likely related to the recently observed small-scale trapping.[13] Hence frequency smearing associated with a combination of four-photon and stimulated Rayleigh-wing scattering is likely to occur along with small-scale trapping.

An experimental arrangement for detecting light-by-light scattering could consist of sending a strong laser beam into a medium with a large Kerr effect (e.g., CS_2) accompanied by a weak beam at $+\Theta_{\mathrm{opt}}$ and detecting the generation of another weak beam at $-\Theta_{\mathrm{opt}}$. To demonstrate exponential gain, it would be desirable to detect a threshold in an experimental arrangement utilizing one or two off-axis resonators[14] at $\pm\Theta_{\mathrm{opt}}$.

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