# Combined stimulated Raman scattering and continuum self-phase modulations

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A theory describing the combined effects of stimulated Raman scattering and continuum self-phase modulation is developed. As may be expected, the effects are not simply additive. Calculations are presented which determine the interaction of these effects in various limits.

## I. INTRODUCTION

Two effects of interest in understanding the propagation of intense ultrafast optical pulses through matter are stimulated Raman scattering[1] and continuum self-phase modulation.[2,3] In this paper we discuss the combined operation of these two effects.

As a strong optical pulse traverses a medium, energy is transferred from a primary mode, which is at the laser frequency, to other modes of the system. In the case of stimulated Raman scattering (SRS) the shift in frequency takes place in units of the vibrational frequencies of the medium. Thus the primary mode becomes depleted and the various Stokes modes are intensified. When spectral broadening occurs, it occurs in a quantized manner.

In the case of self-phase modulation (SPM), however, the repopulation of the spectral intensity takes place in a more gradual manner. Owing to the nonlinearity of the medium, the pulse heterodynes against itself and gradually increases its spectral width. There is a continuum of frequencies produced in this process. Supercontinuum generation spanning the visible and infrared region was first observed by Alfano and Shapiro when intense picosecond laser pulses were passed through liquids and solids.[3]

Since both the stimulated Raman scattering and self-phase modulation spread the spectral width of the pulse, one can ask if they occur independently of each other or if they compete and interfere. The answer is, perhaps, obvious, but has not been adequately studied in the literature. Clearly, if the SRS effect is effective in transferring energy to the Stokes branches very rapidly, SPM will not occur to any significant extent. On the other hand if SPM is the dominant effect, a Stokes shift of the primary mode must be replaced by a Stokes shift of a broadened primary mode, and the spectrum will lose its discrete character. Thus the two effects influence each other and should be studied simultaneously. Our goal here will be to calculate the extent of this interaction in varying limiting cases.

In the next section we introduce a simplified model which captures the essence of both processes. We will limit our attention to one-dimensional propagation, even though the transverse character of the pulse is needed in order to explain some phenomena (such as self-focusing). Likewise, dispersion effects will be neglected, as these mainly affect the spatial evolution of the pulse. A number of other simplifying assumptions will be made to bring the equations to a tractable level.

## II. THEORY

The system of interest consists of an electromagnetic field interacting with the molecular vibration (phonon) field of a liquid. The electromagnetic field $E$ consists of a laser pulse at frequency $\omega$ and a Stokes field at frequency $\omega-\sigma$, where $\sigma$ is the phonon frequency. The dielectric constant is of the form $\epsilon=\epsilon_0+\epsilon_2E^2+\gamma Q$ where $\gamma$ determines the coupling of the phonon field $Q$ to the electromagnetic field. The effective oscillator mass density associated with the phonons is denoted by $\mu$. We assume phase matching and neglect anti-Stokes and higher Stokes fields. The phonons are assumed to be decaying in time with a rate $\Gamma$. The basic equations describing the system of interest are

$$\frac{\partial a_1}{\partial \eta}=i[\beta(|a_1|^2+2|a_2|^2)a_1+\varphi a_2], \tag{1}$$

$$\frac{\partial a_2}{\partial \eta}=i[\beta(|a_2|^2+2|a_1|^2)a_2+\varphi^* a_2], \tag{2}$$

$$\frac{\partial \varphi}{\partial \xi}=-\varphi+i\delta a_2^* a_1, \tag{3}$$

where $a_1$ denotes the envelope of the vector poten-

tial for the laser pulse, $a_2$ is the corresponding quantity for the Stokes wave, and $\varphi$ is proportional to the phonon amplitude. In Eqs. (1)–(3) we have let $\xi = \Gamma(t - z/v)$, where $v$ is the phase velocity of light, $\eta = \Gamma z/v$, and defined two parameters $\beta = 3\epsilon_2\omega^3/\Gamma\epsilon_0 c^2$ and $\delta = \gamma^2\omega/16\pi\mu\sigma\epsilon_0 c^2\Gamma^2$.

The boundary conditions at $z = 0$ are that the input laser pulse $a_1$ is given by

$$a_1(\xi, \eta)|_{\eta=0} = F(\xi), \tag{4}$$

and the Stokes wave $a_2$ is due to noise

$$a_2(\xi, \eta)|_{\eta=0} = f(\xi). \tag{5}$$

In principle we can also specify a boundary condition for $\varphi$, but this is unnecessary if we make the following simplifying assumption. Let us take the input pulse to be of the form

$$F(\xi) = F_0 \exp[-(\xi/\Gamma\tau)^2], \tag{6}$$

where $\tau$ is the pulse duration and $F_0$ is the amplitude of the vector potential ($F_0 = cE_0/\omega$). If the relaxation rate is rapid compared with the time scale over which $a_1$ and $a_2$ vary then Eq. (3) may be replaced by

$$\varphi = i\delta a_2^* a_1. \tag{3'}$$

Inserting this into Eqs. (1) and (2) and expressing the fields in polar form $a_j = \rho_j^{1/2}\exp(i\phi_j)$ gives the Volterra-Lotka[4] equations

$$\frac{\partial\rho_1}{\partial\eta} = -2\delta\rho_1\rho_2 \tag{7}$$

and

$$\frac{\partial\rho_2}{\partial\eta} = +2\delta\rho_1\rho_2. \tag{8}$$

The phases are $\phi_1 = \beta\eta(\rho_1 + 2\rho_2)$ and $\phi_2 = \beta\eta(\rho_2 + 2\rho_1)$. Equations (7) and (8) describe the predator-prey relationship between the Stokes and incident pulses. In the case where $\rho_2 \ll \rho_1$ the solution to the above equations yields

$$a_1 \approx F(\xi)\exp[i\beta\eta F^2(\xi)] \tag{9}$$

and

$$a_2 \approx f(\xi)\exp[i(2\beta - i\delta)\eta F^2(\xi)]. \tag{10}$$

These envelopes may be Fourier analyzed to obtain information regarding the predicted experimental spectrum.

The frequency spectrum of the electromagnetic field is proportional to

$$I(\omega) = \left| \int_{-\infty}^{\infty} d\xi (a_1 e^{i(\omega-\omega_1)\xi/\Gamma} + a_2 e^{i(\omega-\omega_1+\sigma)\xi/\Gamma}) \right|^2. \tag{11}$$

The two terms will contribute independently since $F$ and $f$ are incoherent. We shall focus our attention on the Raman term first. From Eqs. (10) and (6) one obtains

$$I_R = \left| \int_{-\infty}^{\infty} d\xi f(\xi)\exp i[\eta(2\beta - i\delta)F_0^2 e^{-2(\xi/\Gamma\tau)^2} + (\omega - \omega_1 + \sigma)\xi/\Gamma] \right|^2. \tag{12}$$

Let us neglect the time dependence of $f$ and expand the exponential in a power series:

$$I_R = \left| f\tau\Gamma \sum_{n=0}^{\infty} \frac{[i\eta F_0^2(2\beta - i\delta)]^n}{n!} \left(\frac{\pi}{2n}\right)^{1/2} e^{-(\omega-\omega_1+\sigma)^2\tau^2/8n} \right|^2. \tag{13}$$

For large $\eta$ the important contributions to this sum stem from large values of $n$, so the sum may be replaced by an integral and Stirling's approximation may be employed. Thus,

$$I_R = \left| \int_0^{\infty} \frac{f\tau\Gamma}{2} \frac{dn}{n} \exp\left(\Phi(n) - \frac{(\omega - \omega_1 + \sigma)^2\tau^2}{8n}\right) \right|^2, \tag{14}$$

where

$$\Phi(n) = n - n\ln n + n\ln[i\eta F_0^2(2\beta - i\delta)]. \tag{15}$$

The point where $\Phi$ is maximum is given by

$$n_0 = i\eta F_0^2(2\beta - i\delta). \tag{16}$$

We first assume that the frequency shift satisfies the inequality

$$\left| \frac{(\omega - \omega_1 + \sigma)^2\tau^2}{8n_0} \right| \ll |n_0|, \tag{17}$$

so the exponential in Eq. (14) may be simplified by retaining only the exponent $\Phi(n)$. Then, from the method of steepest descents, we find

$$I_R = \frac{\pi}{2}\left(\frac{f\Gamma\tau}{F_0}\right)^2 \frac{1}{\eta} \frac{e^{2\eta\delta F_0^2}}{(4\beta^2 + \delta^2)^{1/2}}. \tag{18}$$

For frequencies such that Eq. (17) is not satisfied the intensity is highly suppressed due to the second term of the exponent in Eq. (14). In this limit we are probing the far spectral wings of the pulse and the spectral intensity drops rapidly. Thus the Raman spectrum is predicted to be rather flat with its intensity given Eq. (18) and to have a half width

$$\Delta\omega_R = (\eta F_0^2/\tau)[8(4\beta^2 + \delta^2)]^{1/2}. \tag{19}$$

The pulse amplitude $F_0$ is related to the energy per unit area $U$ through the relation $F_0 = [cu(8\pi)^{1/2}/\omega^2\tau]^{1/2}$. For pulses of constant energy per unit area we see from Eq. (19) that the Stokes width varies as the inverse square of the primary pulse duration. The broadening is due to a combination of the cross-phase modulation and the Raman terms.

Next let us examine the first term (laser) in Eq. (11). The analysis proceeds along much the same line and we find

TABLE I. Parameters for liquids in a typical laser field (estimated values).

| Quantity | Value in cgs units $CS_2$ | ethanol |
|---|---|---|
| $\epsilon_0$ | 1.7 | 1.3 |
| $\epsilon_2$ | $10^{-11}$ | $10^{-12}$ |
| $\omega$ | $3\times6\times10^{15}$ ($1.9\times10^4$ $cm^{-1}$) | $3.6\times10^{15}$ ($1.9\times10^6$ $cm^{-1}$) |
| $\sigma$ | $1.2\times10^{14}$ (656 $cm^{-1}$) | $5.5\times10^{14}$ (2928 $cm^{-1}$) |
| $\mu$ | 1.3 | 0.81 |
| $\Gamma$ | $9.4\times10^{10}$ (0.5 $cm^{-1}$) | $3.4\times10^{12}$ (18 $cm^{-1}$) |
| $\gamma$ | $10^6$ (estimated) | $10^6$ (estimated) |
| $\tau$ | $6\times10^{-12}$ | $6\times10^{-12}$ |
| $F_0$ | 0.1 | 0.1 |
| $\nu$ | $2.3\times10^{10}$ | $2.6\times10^{10}$ |
| $Z$ | 10 | 10 |
| $\Delta\omega_R$ | $3.8\times10^{15}$ ($2.0\times10^4$ $cm^{-1}$) | $4.4\times10^{14}$ ($2.3\times10^3$ $cm^{-1}$) |
| $\Delta\omega_L$ | $13.3\times10^{14}$ ($7.1\times10^3$ $cm^{-1}$) | $1.5\times10^{13}$ (810 $cm^{-1}$) |

$$I_L(\omega)\approx\pi\Gamma^2\tau^2/\beta\eta \tag{20}$$

for frequencies such that

$$\Delta\omega_L\equiv|\omega-\omega_1|\lesssim 2\beta\eta F_0^2/\tau\,, \tag{21}$$

and $I_L$ drops rapidly for frequencies greater than this value. This is the conventional self-phase-modulation result.

In summary, the theory predicts that the primary laser pulse will undergo self-phase modulation. The Stokes pulse will be parametrically amplified from the noise. The Stokes pulse broadens due to a combination of cross-phase modulation and Raman parametric amplification. The spectral pulse widths increase as the pulses progress down the fluid and grow dramatically as the duration of the pulse is shortened. Theory predicts a ratio of the Stokes spectral width to the primary spectral width given by

$$\frac{\Delta\omega_R}{\Delta\omega_L}=\{8[1+(\delta/2\beta)^2]\}^{1/2}. \tag{22}$$

Thus by the time self-phase modulation has broadened the primary pulse to the point where it overlaps the Stokes line, little trace of a distinct Raman Stokes line should be observed.

As a numerical example, let us compute $\Delta\omega_R$ and $\Delta\omega_L$ for two typical liquids, $CS_2$ and ethanol, in the field of neodymium glass laser. The parameters relevant to such a system are tabulated in Table I, along with the computed values of $\Delta\omega_R$ and $\Delta\omega_L$. These values should be readily observable in an experiment.

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