

Quantum theory of degenerate four-wave mixing and Raman scattering in fibers

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Four-wave mixing and Raman scattering in fibers have been extensively studied theoretically and experimentally. We show that a quantum-theory-based analytical description gives good predictions for quite general experiments involving small signal compared to the undepleted pump beam. It also leads to well known limiting cases. Finally, we show original agreement between experimental data and our theory.

Raman scattering and degenerate four-wave mixing (FWM) are phenomena usually known as third order ($\chi^{(3)}$) nonlinear processes. Nevertheless, as FWM is a quasi-instantaneous process, Raman scattering is a delayed one because of the implication of coupling to molecular vibration (phonon). In the past, these two parts of the nonlinear response were often considered separately (see for instance [1]), but the fact that both should be considered together has been pointed out by Bloembergen and Shen [2] in early studies. This was confirmed by experiments : the temporal dispersion which is a key parameter in FWM was shown to play an important role in Raman scattering as well. Indeed, the gain of the Raman Stokes wave can be affected by the dispersion, leading to extinction of Raman scattering when there is zero phase mismatch [3], or to exponential growth of the Raman anti-Stokes wave [4] which in the naive approach is exponentially damped. This late effect explains blue detuning of supercontinuum [5, 6].

A quantum description of light propagation in optical fibers, including both the instantaneous Kerr non linearity and the Raman scattering in a dispersive fiber, has been developed [7] and used to quantify noise limits on squeezing, noise in $\chi^{(3)}$ parametric amplifiers [8], noise in coherent anti-Stokes Raman scattering, and noise in photon pair generation experiments [9]. In the present work we apply this quantum theory, using the formulation of [8], to describe the spontaneous growth of Raman Stokes and anti-Stokes waves. Our analytical predictions are compared to experimental results reported in [4, 5] which had previously been fitted with hybrid quantum/classical formulae, or numerical calculations.

We consider a continuous pump with complex amplitude $\sqrt{P} e^{i\phi(x,t)}$, where P is the power flowing through the fiber and $\phi(x,t) = -\omega_p t + [k_p + \gamma P]x$. In this expression, ω_p is the pump frequency and k_p is the (linear) wave number. Self-phase modulation gives a nonlinear contribution γP to the total wave number. Here γ is the third-order nonlinearity

parameter of the fiber [1]. The quantum field operator

$$A(x, t) = e^{i\phi(x, t)} \int_0^\infty \left(\sqrt{\frac{\hbar\omega_s}{2\pi}} A_s(\Omega, x) e^{i\Omega t} + \sqrt{\frac{\hbar\omega_a}{2\pi}} A_a(\Omega, x) e^{-i\Omega t} \right) d\Omega + \text{h.c.} \quad (1)$$

describes the perturbations around this stationary solution through the combined effect of Raman scattering and four-wave mixing. These are composed of symmetrically detuned Stokes and anti-Stokes photons with respective frequencies $\omega_s = \omega_p - \Omega$ and $\omega_a = \omega_p + \Omega$, and wave numbers k_s and k_a . The operators $A_s(\Omega, x)$ and $A_a(\Omega, x)$ are *destruction operators* for Stokes and anti-Stokes photons. Their equations of motion can be deduced from the quantum theory of light propagation and solved analytically (so long as the perturbation field is small compared to the pump) [8]. Once $A_s(\Omega, x)$ and $A_a(\Omega, x)$ are known, any physical quantity can be computed. Here we are concerned by the mean *spectral photon-flux densities*

$$f_{s,a}(\Omega, x) = \lim_{\varepsilon \rightarrow 0} \frac{1}{2\pi\varepsilon} \times \int_{\Omega-\frac{\varepsilon}{2}}^{\Omega+\frac{\varepsilon}{2}} \int_{\Omega-\frac{\varepsilon}{2}}^{\Omega+\frac{\varepsilon}{2}} \langle A_{s,a}^\dagger(\Omega_1, x) A_{s,a}(\Omega_2, x) \rangle d\Omega_1 d\Omega_2. \quad (2)$$

Using the approach of [8], we find ($f_+ \equiv f_s$ and $f_- \equiv f_a$)

$$f_\pm(\Omega, x) = \frac{1}{2\pi} \frac{|\chi(\Omega)|^2}{|\kappa(\Omega)|^2} |\sinh(\kappa(\Omega)x)|^2 + \frac{|\text{Im}[\chi(\Omega)]|}{\pi} \rho_\pm(\Omega, x) \left(n(\Omega) + \frac{1}{2} \pm \frac{1}{2} \right) \quad (3)$$

$$\text{with } \rho_\pm(\Omega, x) = \int_0^x dx' \left| \cosh(\kappa(\Omega)x') \pm i \frac{\Delta k(\Omega)}{2\kappa(\Omega)} \sinh(\kappa(\Omega)x') \right|^2 \quad (4)$$

The photon fluxes depend on four basic parameters: the detuning Ω , the linear phase-mismatch $\Delta k(\Omega) = k_s + k_a - 2k_p$, the non linearity $\chi(\Omega) = \gamma P [(1 - f_r) + f_r \chi_r(\Omega)]$ including $f_r = 18\%$ of Raman response which is dependent of the detuning through the normalized spectral Raman response ($\text{Re}[\chi_r(0)] = 1$, $\text{Im}[\chi_r] < 0$), and the temperature T through the *phonon population* $n(\Omega) = (\exp(\hbar\Omega/k_B T) - 1)^{-1}$. The complex parameter $\kappa(\Omega) = [-(\Delta k(\Omega)/2)^2 - \Delta k(\Omega)\chi(\Omega)]^{1/2}$ controls the growth rate of the Stokes and anti-Stokes waves. The real part of $\kappa(\Omega)$ is chosen positive so that it can be interpreted as the *amplification gain* for small signals at frequencies $\omega_p \pm \Omega$. The integral in Eq. (4) can be carried out exactly, leading to somewhat cumbersome expressions. Here however, we prefer to focus on two physically important limits : the *spontaneous scattering limit* ($|\kappa|x \ll 1$) and the *stimulated amplification and wave-mixing limit* ($\text{Re}[\kappa]x \gg 1$). The first one is of great importance for photon-pair generation, while the second one applies to supercontinuum generation. Considering the limit of small $|\kappa|x$, Eqs. (3) give

$$f_\pm \approx \frac{1}{\pi} |\text{Im}[\chi]| x \left(n + \frac{1}{2} \pm \frac{1}{2} \right) \quad (5)$$

up to the first order in $|\kappa|x$. Using expression of $n(\Omega)$, one finds that $\lim_{|\kappa|x \rightarrow 0} f_a/f_s = \exp[-\hbar\Omega/(k_B T)]$ in accordance with the usual theory of spontaneous Raman scattering in fibers (see [4] and references within). Upon adding the extra term $|\chi|^2 x^2 / (2\pi)$ to Eqs. (5) one accounts for the *spontaneous four-photon scattering* process which is used for photon-pair generation in fibers. Since it is only of second order in $|\kappa|x$, spontaneous

Raman scattering always plays a detrimental role in photon-pair generation experiments and is referred to ‘‘Raman noise’’[9].

The asymptotic behavior for $\text{Re}[\kappa]x \gg 1$ is also simple since we keep only the exponentially growing terms in Eqs. (3) :

$$f_{\pm} \sim \frac{e^{2\text{Re}[\kappa]x}}{8\pi} \left(\frac{|\chi|^2}{|\kappa|^2} + \frac{|\text{Im}[\chi]|}{\text{Re}[\kappa]} \frac{|\kappa \pm i\frac{\Delta k}{2}|^2}{|\kappa|^2} \left(n + \frac{1}{2} \pm \frac{1}{2} \right) \right) \quad (6)$$

Note that $|\kappa + i\frac{\Delta k}{2}| > |\kappa - i\frac{\Delta k}{2}|$ which implies that the Stokes wave is always stronger than the anti-Stokes wave, as expected. In Fig. 1 we have plotted the ratio f_a/f_s of the anti-

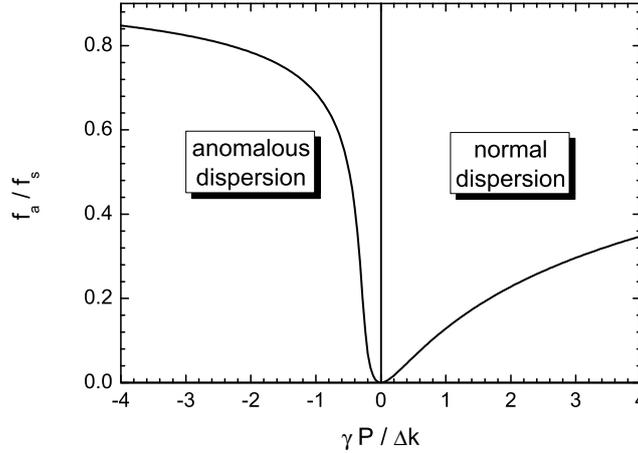


Figure 1: Ratio of anti-Stokes to Stokes photon fluxes, at the peak of the Raman gain, as a function of $\gamma P/\Delta k$.

Stokes to Stokes fluxes at the peak of the Raman gain ($\Omega/(2\pi) = 13.2$ THz) at $T = 300$ K as a function of $\gamma P/\Delta k$. In these circumstances, $n = 0.14$ and $\chi = \gamma P (0.82 - i 0.25)$ for silica optical fibers. When $\gamma P/|\Delta k| \gg 1$ pair creation dominates over Raman scattering in Eqs. (6), both in the normal and anomalous dispersion regimes, and we have $f_s \simeq f_a \sim \frac{e^{2\text{Re}[\kappa]x}}{8\pi} \frac{|\chi|^2}{|\kappa|^2}$. However f_a/f_s tends much faster to 1 in the anomalous dispersion regime than in the normal dispersion regime, as is apparent from Fig. 1. When the opposite limit $\gamma P/|\Delta k| \ll 1$ is considered, the approximation $\kappa \simeq |\text{Im}[\chi]| + i(\Delta k/2 + \text{Re}[\chi]) + o(|\chi/\Delta k|)$ holds and

$$f_s \sim \frac{e^{2|\text{Im}[\chi]|x}}{2\pi} (n+1) \quad \text{and} \quad f_a \sim \frac{e^{2|\text{Im}[\chi]|x}}{2\pi} \frac{|\chi|^2}{\Delta k^2} (n+1). \quad (7)$$

The ratio of anti-Stokes to Stokes fluxes is then given by $f_a/f_s = |\chi|^2/\Delta k^2$. This simple ratio was already derived in a classical context [2, 4].

In order to compare theory to experiments, we now focus on $\Omega/(2\pi) = 13.2$ THz (corresponding to the Raman peak). Experimental Stokes and anti-Stokes spectral photon-flux densities in the $\gamma P/|\Delta k| \ll 1$ regime are available from [4]. They are plotted in Fig. 2 as a function of the pump power P such as $\gamma P/|\Delta k| \leq 0.032$. As seen from the figure, the asymptotic formula (7) [dark grey curves] apply for $P \geq 300$ W (i.e. $\text{Re}[\kappa]x \gg 1$) while formula (5) [light grey curves] apply for $P \leq 50$ W (i.e. $|\kappa|x \ll 1$). Outside these limiting cases Eqs. (3) reproduce very well the spectral photon-flux densities that are observed experimentally [black curves].

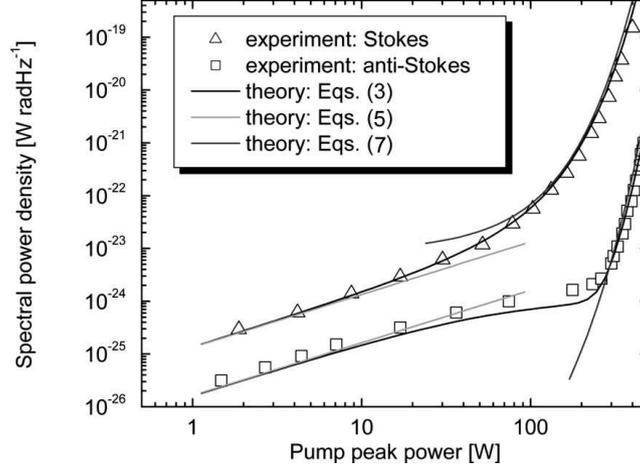


Figure 2: Evolution of the Stokes and anti-Stokes spectral photon-flux densities with the pump power P . The parameters are: $\gamma = 23 \text{ W}^{-1}\text{km}^{-1}$, $x = 2.85 \text{ m}$, $\beta_2 = 50 \text{ ps}^2\text{km}^{-1}$ and $T = 293 \text{ K}$. The experimental data are from [4]. (In the theoretical curves, we took into account the shape of the pump pulses in the same way as in [4]).

In [5], experimental data are obtained for a photonic crystal fiber with $\gamma = 150 \text{ W}^{-1}\text{km}^{-1}$ and $\beta_2 = 7 \text{ ps}^2/\text{km}$ (normal dispersion). Measurements have been carried out with $x = 3 \text{ m}$ and $P = 90 \text{ W}$, as well as with $x = 0.7 \text{ m}$ and $P = 400 \text{ W}$. In both cases $\text{Re}[\kappa]x \gg 1$ but since $\gamma P/|\Delta k|$ is close to one so we have to use our result (6) to compute the ratio f_a/f_s . It predicts the ratio $f_a/f_s = 0.028$ for $P = 90 \text{ W}$ and $f_a/f_s = 0.16$ for $P = 400 \text{ W}$ ($T = 300 \text{ K}$). These are in good agreement with the measured ratios ($f_a/f_s = 0.016$ and 0.22 respectively) given the uncertainty on the experimental parameters in [5] and probable polarization effects.

In summary we have developed an analytic theory to account for the growth of the Stokes and anti-Stokes waves from the combined effects of pair creation and Raman scattering. The results are in good agreement with earlier experimental observations. They should find applications in the optimization of supercontinuum sources based on long pump pulses.

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