Generation and amplification of Raman Stokes and anti-Stokes waves

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We present general analytical expressions of Stokes and anti-Stokes spectral photon-flux densities that are spontaneously generated by a single monochromatic pump wave propagating in a single-mode optical fiber. We validate our results by comparing them with experimental data. Limiting cases of the general expressions corresponding to interesting physical situations are discussed. © 2008 Optical Society of America

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The third order nonlinearity seen by light propagating in optical fibers is composed of two parts: a quasi instantaneous contribution coming from the electronic response, and a delayed Raman response coming from the coupling to molecular vibrations (phonons). The influence of these two terms on the propagation of light are often considered separately, see for instance the treatment in [1]. But in many cases they must be treated collectively, together with the influence of dispersion which plays an essential role, particularly when the phase matching conditions are (even approximately) satisfied.

That a holistic approach is necessary was first understood by Bloembergen and Shen [2], and confirmed experimentally in [5], that the Raman Stokes and anti-Stokes waves are spontaneously generated by a single monochromatic pump wave propagating in a single-mode optical fiber. We validate our results by comparing them with experimental data. Limiting cases of the general expressions corresponding to interesting physical situations are discussed. © 2008 Optical Society of America

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, \( \omega_p \) is the pump frequency and \( k_p \) is the (linear) wave number. Self-phase modulation gives a nonlinear contribution \( \gamma P \) to the total wave number. Here \( \gamma \) is the third-order nonlinearity parameter of the fiber [1].

The quantum field operator

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

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) [12].

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

\[
f_{s,a}(\Omega, x) = \lim_{\epsilon \to 0} \frac{1}{2\pi \epsilon} \int_{\Omega - \epsilon}^{\Omega + \epsilon} \int_{\Omega - \epsilon}^{\Omega + \epsilon} \langle A_{s,a}(\Omega_1, x) A_{s,a}^\dagger(\Omega_2, x) \rangle d\Omega_1 d\Omega_2. \tag{2}
\]

Using the approach of [12], we find

\[
\begin{align}
f_s(\Omega, x) &= \frac{1}{2\pi} \frac{|\chi(\Omega)|^2}{|\kappa(\Omega)|^2} \left| \sinh(\kappa(\Omega)x) \right|^2 \\
&\quad + \frac{\left| \text{Im}[\chi(\Omega)] \right|}{\pi} \rho_+(\Omega, x) \ (n(\Omega)+1) 
\end{align} \tag{3a}
\]
\[ f_a(\Omega, x) = \frac{1}{2\pi} \left| \frac{\chi(\Omega)}{\kappa(\Omega)} \right|^2 \sinh(\kappa(\Omega)x) \]
\[ + \frac{\text{Im}[\chi(\Omega)]}{\pi} \rho_-(\Omega, x) n(\Omega), \]

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

The photon fluxes depend on three basic parameters: the pump power \( P \), the detuning \( \Omega \) and the fiber temperature \( T \). The pump power and the non-linear response of the fiber are combined as \( \chi(\Omega) = \gamma P \left[ (1 - f_s) + f_s \chi_r(\Omega) \right] \), where the nonlinearity is decomposed into an instantaneous (electronic) part and a retarded (molecular or Raman) one. \( f_s \approx 0.18 \) is the fraction of the total nonlinearity due to the Raman scattering and \( \chi_r(\Omega) \) is the normalized spectral Raman response \( (\text{Re}[\chi_r(\Omega)] = 1, \text{Im}[\chi_r] < 0) \). The complex parameter \( \kappa(\Omega) = |\kappa(\Omega)|^2 - \Delta k(\Omega) \chi(\Omega)|^{1/2} \) controls the rate of the Stokes and anti-Stokes waves. It depends both on \( \chi(\Omega) \) and on the linear phase-mismatch \( \Delta k(\Omega) = k_s + k_a - 2k_p \). When the fiber is pumped sufficiently far away from the zero dispersion wavelength, the phase-mismatch \( \Delta k(\Omega) \) is well approximated by \( \beta_2 \Delta \Omega^2 \), where \( \beta_2 \) is the group-velocity dispersion parameter. The real part of \( \kappa(\Omega) \) is chosen positive so that it can be interpreted as the amplification gain for small signals at frequencies \( \omega \pm \Omega \). Finally, the temperature \( T \) influences the photon fluxes through the factor
\[ n(\Omega) = (\exp(\hbar \Omega/k_B T) - 1)^{-1} \]

related to the phonon population.

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 \((|\kappa|x \to 0)\) and the stimulated amplification and wave-mixing limit \((\text{Re}[\kappa|x \to \infty])\). 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
\[ f_s \approx \frac{1}{\pi} \text{Im}[\chi] x (n + 1) \]
\[ f_a \approx \frac{1}{\pi} \text{Im}[\chi] x n \]

up to the first order in \( |\kappa|x \). This contribution to the total photon flux is called the spontaneous Raman scattering.

Stokes and anti-Stokes photons are emitted independently. A Stokes photon emission is always accompanied by the emission of a phonon of energy \( \hbar \Omega \), in contrast to the anti-Stokes process for which conservation of energy requires a phonon to be absorbed. For this reason, the anti-Stokes process in inhibited when the phonon population is zero (low temperature limit). Using Eq. (5), one finds that \( \lim_{|\kappa|x \to 0} f_a/f_s = \exp[-\hbar \Omega/(k_B T)] \) in accordance with the usual formulation theory of spontaneous Raman scattering in fibers (see [5] and references within). Upon adding the extra term \( |\chi|^2 x^2/(2\pi) \) to Eqs. (3a) and (3b) one accounts for the spontaneous four-photon scattering process in which a Stokes photon and an anti-Stokes photon are emitted together after the absorption of two pump photons. This is the process 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” (see [14,15] for a more thorough discussion). It is interesting to note that linear dispersion has no impact on the values of the photon fluxes up to the second order in \( |\kappa|x \) since they do not depend on \( \Delta k \). However, the validity of the Maclaurin expansion is restricted to small values of \( \Delta k \) because \( |\kappa| \to (\Delta k/2) \) for fixed pump power and large \( \Delta k \) values.

The asymptotic behavior for \( \text{Re}[\kappa|x \to \infty \text{ is also simple since we keep only the exponentially growing terms in Eqs. (3) :} \]
\[ f_s \sim \frac{e^{2\text{Re}[\kappa]}}{8\pi} \left( \frac{|\chi|^2}{|\kappa|^2} + \frac{|\text{Im}[\chi]|}{\text{Re}[\kappa]} \frac{|\kappa + i \Delta k|^2}{|\kappa|^2} (n + 1) \right) \]
\[ f_a \sim \frac{e^{2\text{Re}[\kappa]}}{8\pi} \left( \frac{|\chi|^2}{|\kappa|^2} + \frac{|\text{Im}[\chi]|}{\text{Re}[\kappa]} \frac{|\kappa - i \Delta k|^2}{|\kappa|^2} n \right). \]

Note that \( |\kappa + i \Delta k| > |\kappa - i \Delta k| \) 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-Stokes to Stokes fluxes at the peak of the Raman gain \( \Omega/(2\pi) = 13.2 \text{ THz} \) at \( T = 300 \text{ K} \) 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. (3), both in the normal and anomalous

Fig. 1. Ratio of anti-Stokes to Stokes photon fluxes, at the peak of the Raman gain, as a function of \( \gamma P/\Delta k \).
dispersion regimes, and we have
\[ f_s \approx f_a \sim \frac{e^{2\text{Re}[\kappa]x}}{8\pi} \frac{|\chi|^2}{|\kappa|^2}. \] (8)

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 \( \kappa \approx |\text{Im}[\chi]| + i(\Delta k/2 + \text{Re}[\chi]) + o(|\chi/|\Delta k|) \) holds and
\begin{align*}
  f_s &\sim \frac{e^{2|\text{Im}[\chi]|x}}{2\pi} (n + 1) \quad (9a) \\
  f_a &\sim \frac{e^{2|\text{Im}[\chi]|x}}{2\pi} \frac{|\chi|^2}{\Delta k^2} (n + 1). \quad (9b)
\end{align*}

The ratio of anti-Stokes to Stokes fluxes is then given by \( f_a/f_s = |\chi|^2/\Delta k^2 \). It is worth noting that it is independent of temperature. This simple ratio was first exhibited in [5]. However, it should be noted that its origin is more complicated than the argument given in [5] since the different processes of pair creation and phonon emission/absorption all contribute to it.

Even though Eqs. (7), (8) and (9) are valid for arbitrary detunings \( \Omega \), we now focus on \( \Omega/2\pi = 13.2 \) THz (corresponding to the Raman peak) in order to compare theory to experiments.

Experimental Stokes and anti-Stokes spectral photon-flux densities in the \( \gamma P/|\Delta k| \ll 1 \) regime are available from [5]. They are plotted in Fig. 2 as a function of the pump power \( P \). For the pump powers that are considered \( \gamma P/|\Delta k| \leq 0.032 \). As seen from the figure, the asymptotic formula \( \Theta \) [dark grey curves] apply for \( P \geq 300 \) W only. At lower pump power the condition \( \text{Re}[\kappa]x > 1 \) is no longer satisfied. For very low pump powers, one can however apply the formula \( \Phi \) [light grey curves] since \( |\kappa|x \ll 1 \). Outside these limiting cases Eqs. \( \Psi \) reproduce very well the spectral photon-flux densities that are observed experimentally [black curves].

In [6], experimental data are obtained for a photonic crystal fiber with \( \gamma = 150 \) W\(^{-1}\)km\(^{-1}\) and \( \beta_2 = 7 \) ps\(^2\)km\(^{-1}\) (normal dispersion). Measurements have been carried out with \( x = 3 \) m and \( P = 90 \) W, as well as with \( x = 0.7 \) m and \( P = 400 \) W. In both cases \( \text{Re}[\kappa]x \gg 1 \) but since \( \gamma P/|\Delta k| \) is close to one neither Eqs. \( \Theta \) nor Eqs. \( \Phi \) can be used to compute the ratio \( f_a/f_s \). Our result \( \Theta \) should be used instead. It predicts the ratio \( f_a/f_s = 0.028 \) for \( P = 90 \) W and \( f_a/f_s = 0.16 \) for \( P = 400 \) W \((T = 300 \) 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 [6] 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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