NONLINEAR OPTICS

Ch. 5 3rd ORDER NONLINEARITIES

I. **Optical Kerr Effect :** self-focusing, nonlinear Shrödinger equation, self-phase modulation, solitons
II. **Four-wave Mixing**
III. **Raman Scattering :** spontaneous and stimulated Raman scattering, Raman amplification, Raman laser
IV. **Brillouin Scattering :** spontaneous and stimulated Brillouin scattering

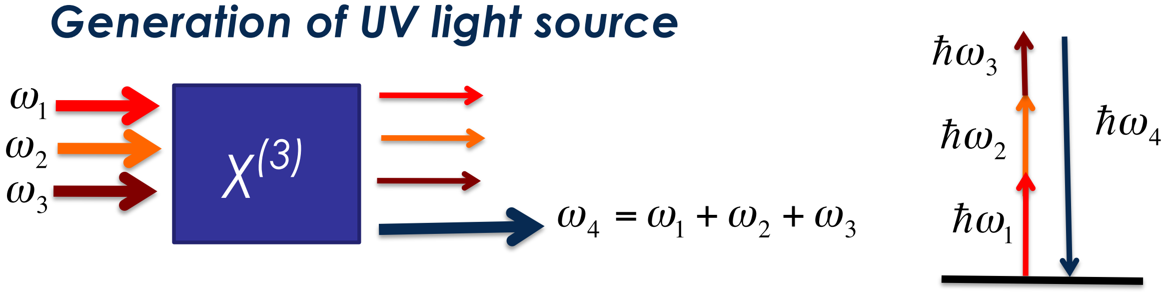
II - Four-wave Mixing

Interaction between 4 waves at $\omega_1 \quad \omega_2 \quad \omega_3 \quad \omega_4$

With : $\omega_4 = \omega_1 + \omega_2 + \omega_3$

$$\vec{P}_{NL}(\omega_4) = D^{(3)}\varepsilon_0 \underline{\underline{\chi}}^{(3)}(\omega_4;\omega_1,\omega_2,\omega_3)\vec{e}_1\vec{e}_2\vec{e}_3 E(\omega_1)E(\omega_2)E(\omega_3)$$

- ***Generation of UV light source***

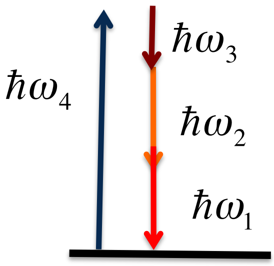


Creation of one photon at ω_4, by means of the simultaneous annihilation of one photon at ω_1, one photon at ω_2 and one photon at ω_3

Phase matching condition to be fullfiled : $\vec{k}_4 = \vec{k}_1 + \vec{k}_2 + \vec{k}_3$

II - Four-wave Mixing

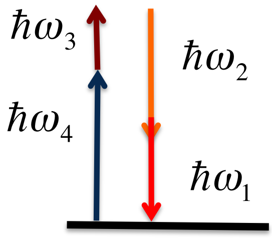
- ***Generation of IR light source***



$$\omega_4 = \omega_1 + \omega_2 + \omega_3$$

$$\vec{k}_4 = \vec{k}_1 + \vec{k}_2 + \vec{k}_3$$

Annihilation of one photon at ω_4, accompanied by the simultaneous creation of three photons, resp. at ω_1, ω_2 and ω_3



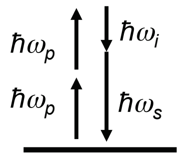
$$\omega_4 + \omega_3 = \omega_1 + \omega_2$$

$$\vec{k}_4 + \vec{k}_3 = \vec{k}_1 + \vec{k}_2$$

II - Four-wave Mixing

- **Degenerate FWM**

The annihilation of simultaneously two pump photons leads to the creation of one signal photon and one idler photon

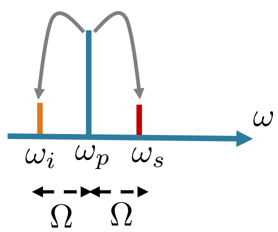


$$\hbar\omega_p + \hbar\omega_p = \hbar\omega_s + \hbar\omega_i \qquad \Delta\beta = 2\beta_p - \beta_s - \beta_i$$

Nonlinear polarization terms @ ω_s $\quad P_{NL}^{(3)}(\omega_s) = 3\epsilon_0\chi^{(3)}E(\omega_p)E(\omega_p)E(-\omega_i)$

@ ω_i $\quad P_{NL}^{(3)}(\omega_i) = 3\epsilon_0\chi^{(3)}E(\omega_p)E(\omega_p)E(-\omega_s)$

Energy transfer from the pump to sideband frequencies



$$\beta(\omega) = \beta_0 + \beta_1\Delta\omega + \frac{\beta_2}{2}\Delta\omega^2 + \cdots$$

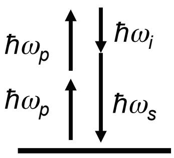
Phase matching term : $\boxed{\Delta\beta \simeq -\beta_2\Omega^2}$

Second order dispersion coef.

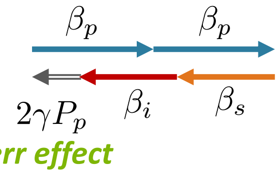
II - Four-wave Mixing

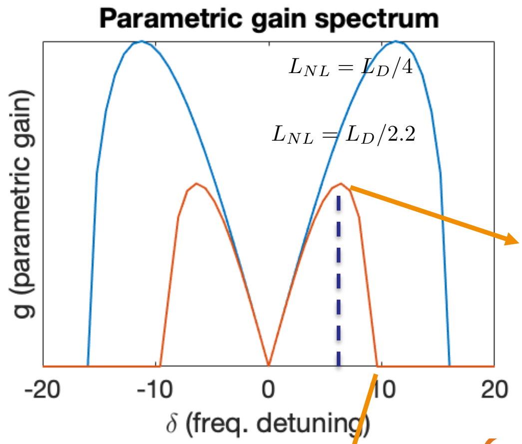
- **Degenerate FWM**

A more extensive analysis shows that the phase matching condition is modified by the refractive index change induced by the optical Kerr effect
=> Details : lecture notes §5.3.2



Phase matching condition





➜ For $n_2>0$, amplification in the <u>anomalous dispersion regime</u> $\beta_2 < 0 \quad \gamma > 0$

$$\Omega_{max}/\sqrt{2}$$

Phase matching condition $\Delta\beta = |\beta_2|\Omega^2 = 2\gamma P_0$

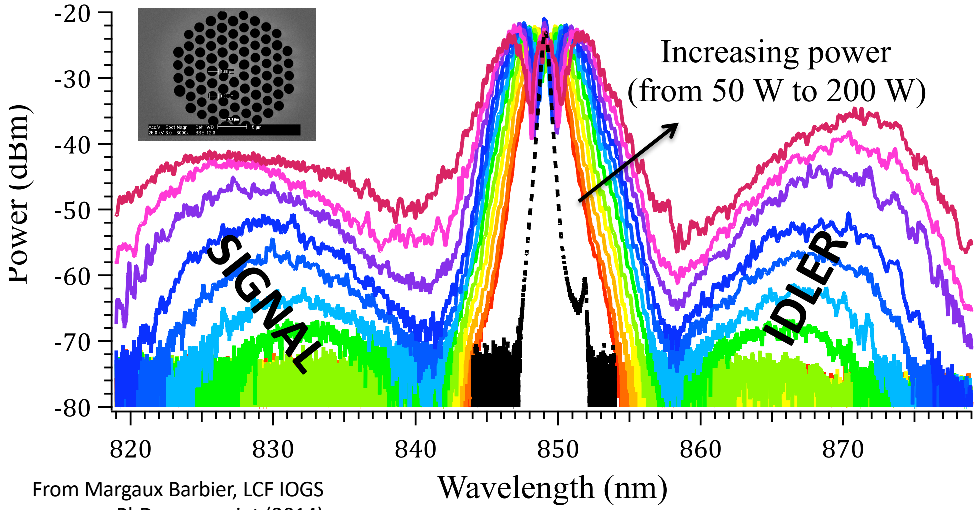
$$\Omega_{max} = \sqrt{\frac{4\gamma P_0}{|\beta_2|}}$$

Amplification factor
$$G_{max}(z) \simeq 1 + \sinh^2(\gamma P_p z) = 1 + \sinh^2(\Phi_{NL}(z))$$

II - Four-wave Mixing

- **Degenerate FWM**

➜ Optical Parametric Fluorescence Effect



From Margaux Barbier, LCF IOGS PhD manuscript (2014)

➜ spontaneous generation of signal and idler photons : application in quantum optics (generation of photon pairs)

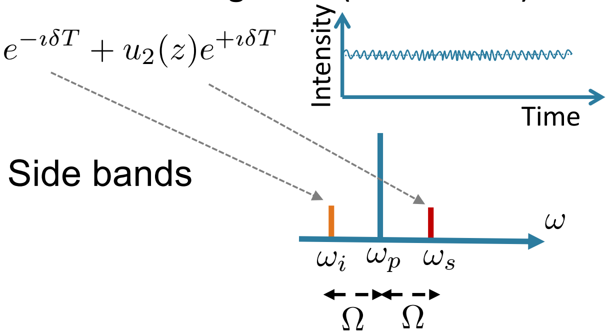
II - Four-wave Mixing

• Degenerate FWM ➜ Modulation instability

Standard model to describe the energy transfer between a CW pump and fluctuations (treated as perturbations)

Nonlinear propagation of the following wave (CW + noise) :

$$u(z,T) = u_0(z) + u_1(z)e^{-\imath\delta T} + u_2(z)e^{+\imath\delta T}$$



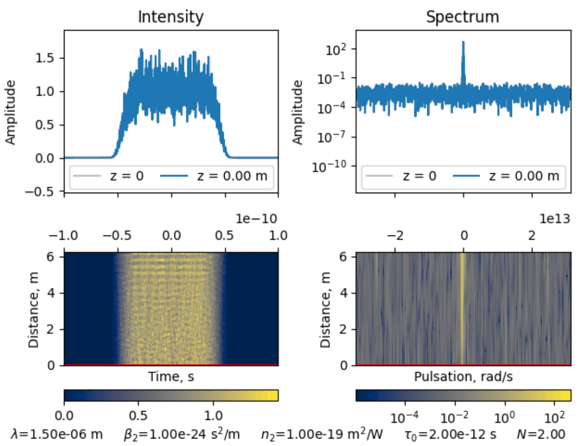
➜ For n_2>0 and in the <u>anomalous dispersion regime</u>, an amplification of the intensity fluctuations is expected (as illustrated by the following simulations)

II - Four-wave Mixing

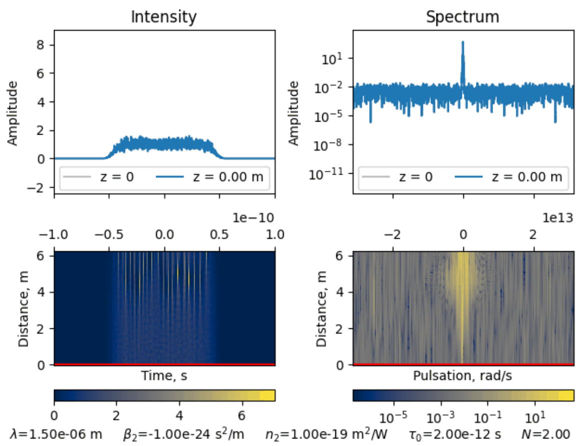
Modulation Instabilities

Evolution of a quasi-CW pulse through a 3rd order nonlinear waveguide

$\beta_2 > 0$ et $\gamma > 0$.



$\beta_2 < 0$ et $\gamma > 0$

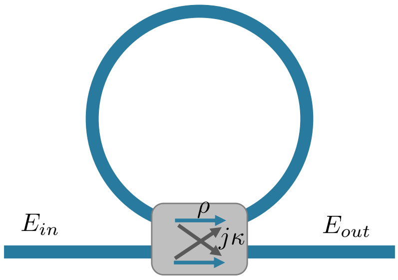


From, M. Dyatlov, LP2N - IOGS

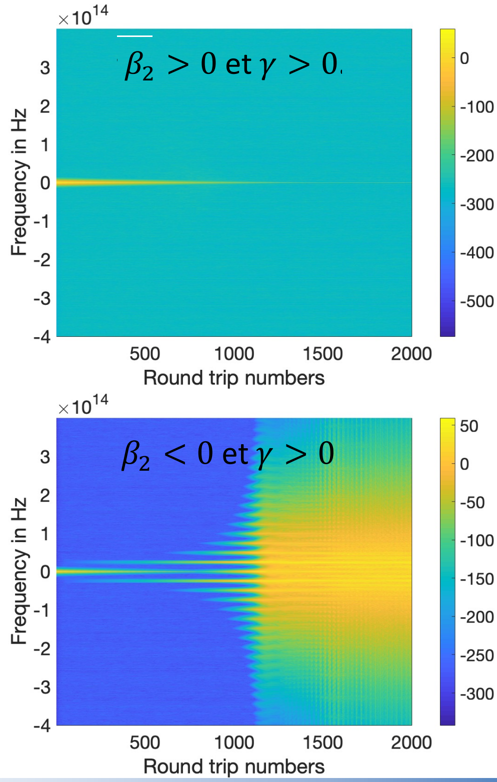
➜ Depending on the dispersion regime, nonlinear propagation can lead to modulation instability in intensities

II - Four-wave Mixing

Modulation Instabilities in microcavities

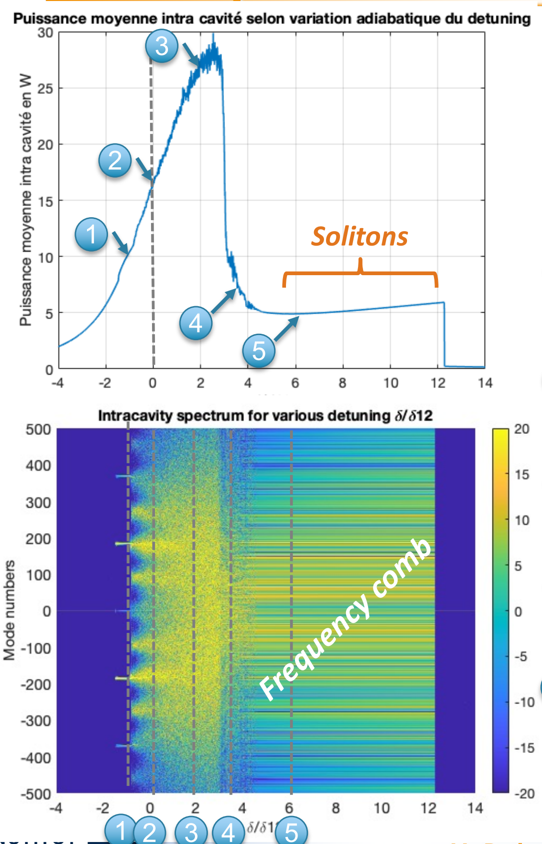


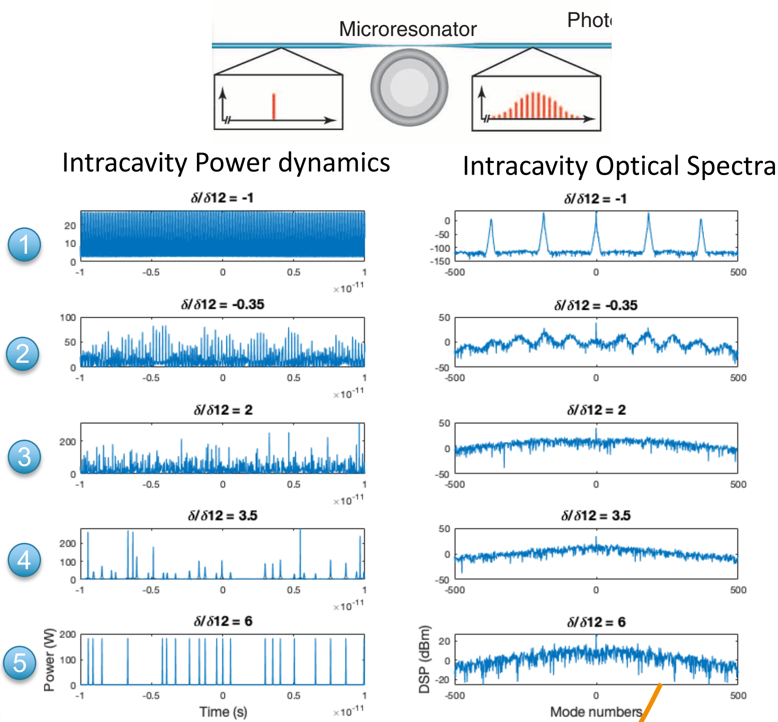
Intracavity spectrum evolution with the round trip numbers



➜ **The modulation instability can lead to the generation of a periodic frequency comb**

Frequency Comb Generation in nonlinear µresonators – Numerical simulations

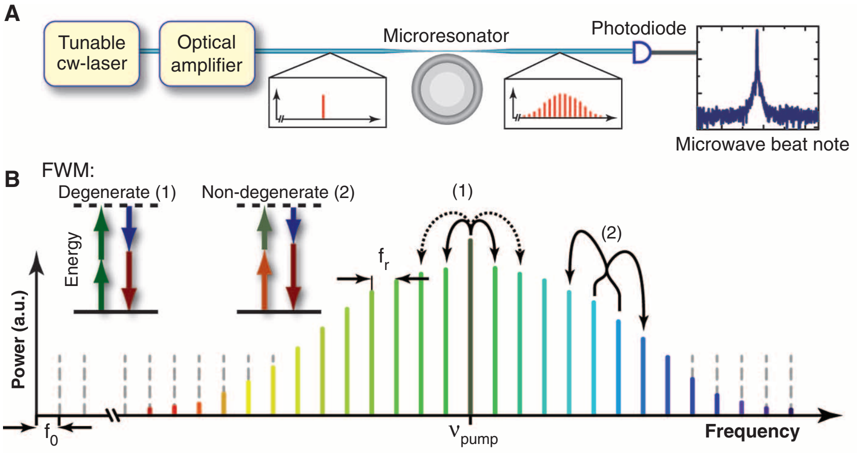




Generation of a Frequency comb in a microresonator pumped by a CW laser

II - Four-wave Mixing

- **Generation of optical Frequency Combs in nonlinear micro-cavities**

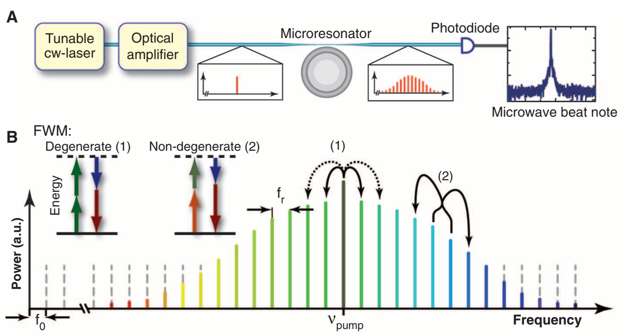


From Kippenberg, Science (2011)

Frequency comb generation in a CW pumped nonlinear µcavity
➜ Parametric Frequency conversion
- Degenerate FWM
- Followed by non-degenerate FWM

II - Four-wave Mixing

- **Generation of optical Frequency Combs in nonlinear micro-cavities**



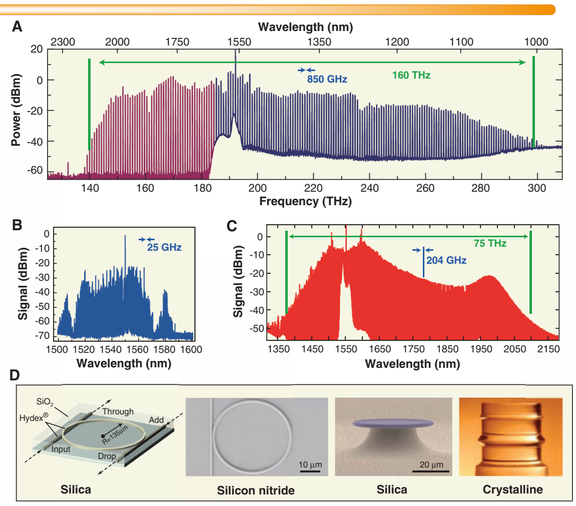
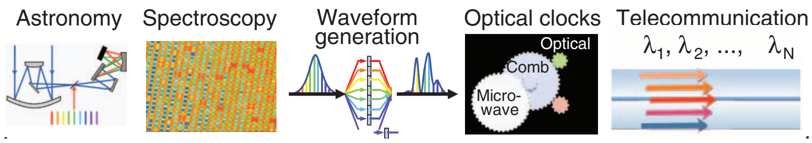


Fig. 3. Microresonator-based frequency combs. **(A)** Spectrum of an octave-spanning frequency comb generated using a silica microtoroidal resonator (*24*). **(B)** An optical frequency comb generated using a crystalline CaF_2 resonator with a mode spacing of 25 GHz (*27*). **(C)** Optical spectrum covering two-thirds of an octave (with a mode spacing of 204 GHz) generated using an integrated SiN resonator (*31*). **(D)** Experimental systems in which frequency combs have been generated (from left to right): Silica waveguides on a chip (Hydex glass) (*32*), chip-based silicon nitride (SiN) ring resonators (*30*) and waveguides, ultrahigh Q toroidal microresonators (*24*) on a silicon chip, and ultrahigh *Q* millimeter-scale crystalline resonators (*27*).

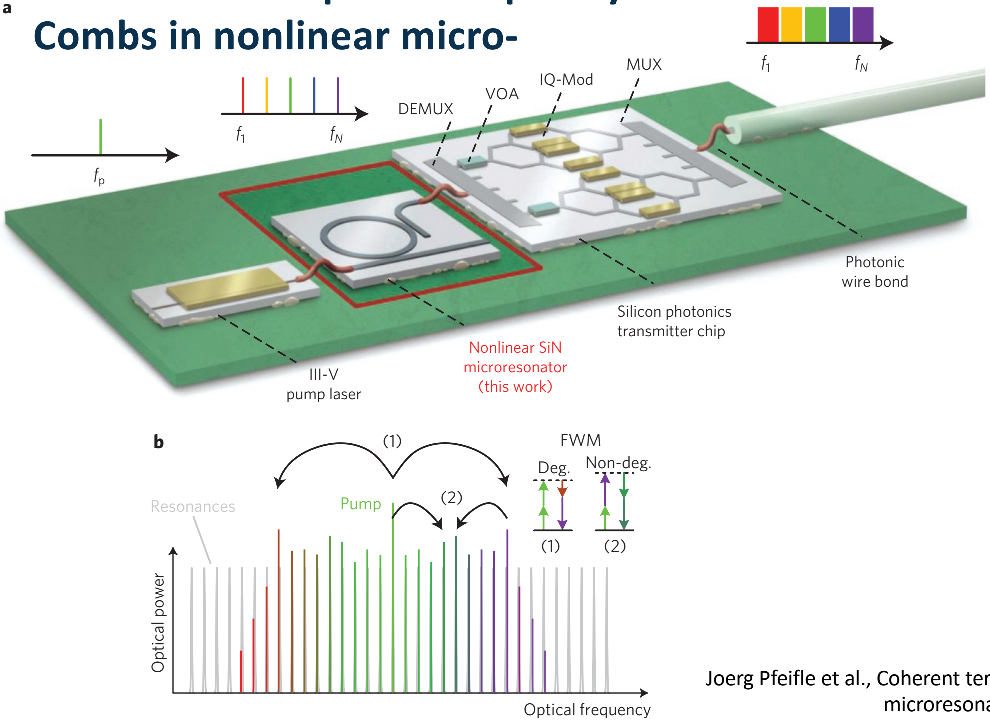
Applications :



From Kippenberg, Science (2011)

II - Four-wave Mixing

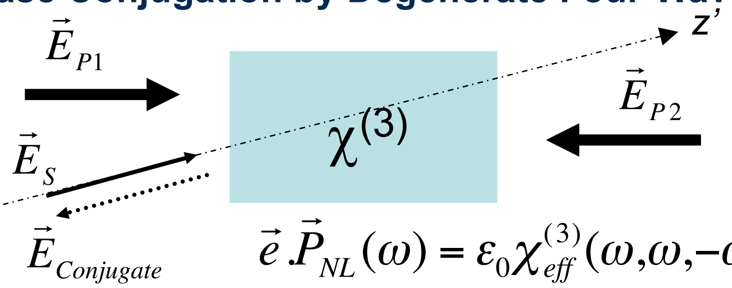
- **Generation of optical Frequency Combs in nonlinear micro-**



Joerg Pfeifle et al., Coherent terabit communications with microresonator Kerr frequency combs
NATURE PHOTONICS | VOL 8 | MAY 2014

II - Four-Wave Mixing

Phase Conjugation by Degenerate Four-Wave Mixing



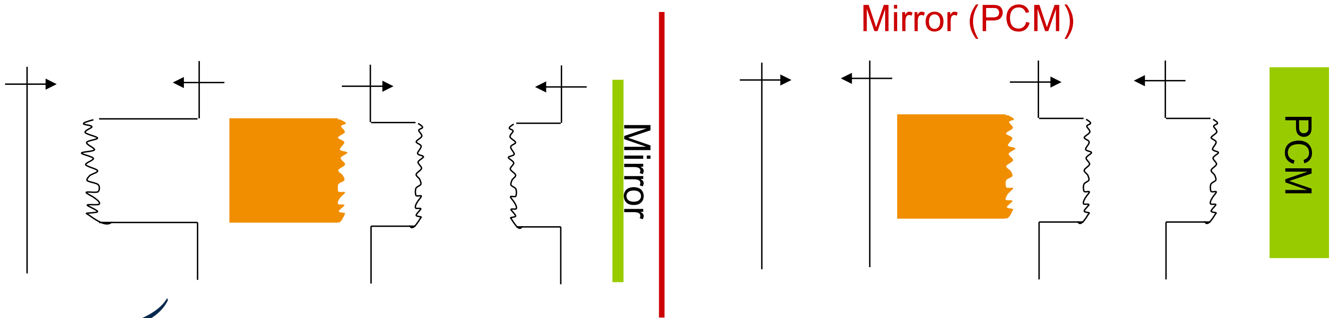
$$\vec{e}.\vec{P}_{NL}(\omega) = \varepsilon_0 \chi^{(3)}_{eff}(\omega,\omega,-\omega) A_{P1} A_{P2} A_S^* e^{-ikz'}$$

Generation of a conjugate wavefront

Automatic phase-matching with the conjugate wave

Reflection on a Mirror

Reflection on a Phase Conjugate Mirror (PCM)



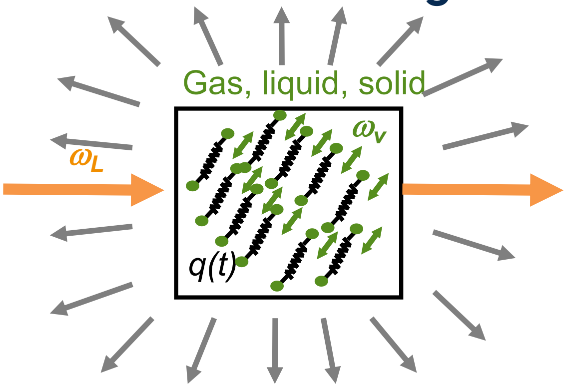
NONLINEAR OPTICS

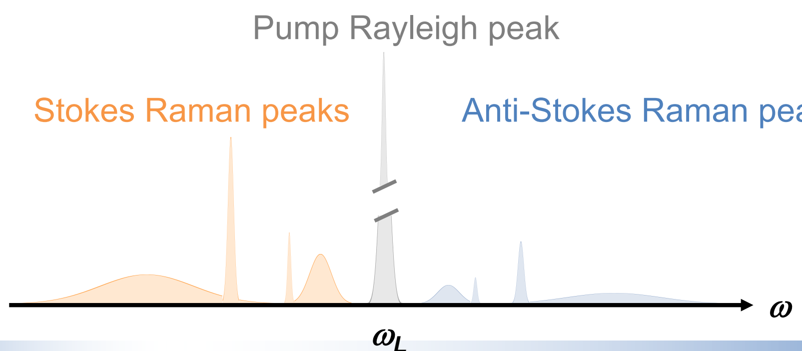
Ch. 5 3rd ORDER NONLINEARITIES

I. **Optical Kerr Effect :** self-focusing, nonlinear Shrödinger equation, self-phase modulation, solitons
II. **Four-wave Mixing**
III. **Raman Scattering :** spontaneous and stimulated Raman scattering, Raman amplification, Raman laser
IV. **Brillouin Scattering :** spontaneous and stimulated Brillouin scattering

Raman Scattering

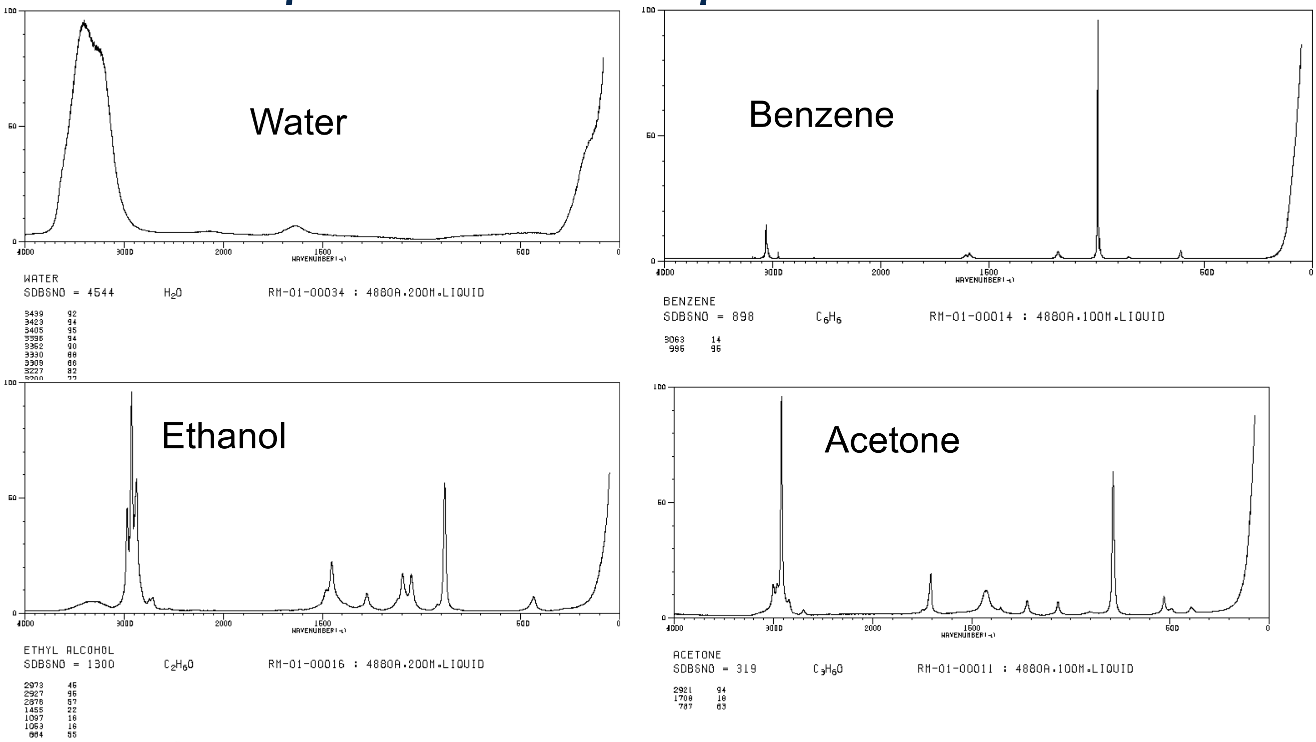
- ***Spontaneous Raman Scattering***





III - Raman Scattering

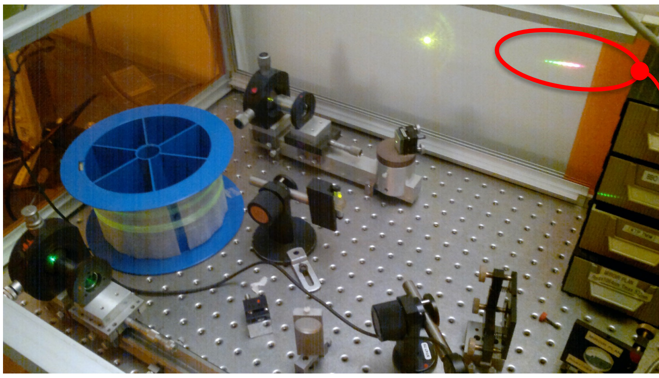
- ***Raman spectra of various liquids***



III - Raman Scattering

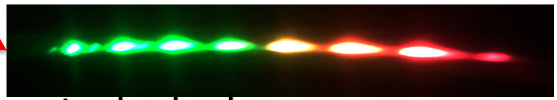
III - Raman Scattering

- ***Raman scattering in a silica based optical fibre***



Output Optical Spectrum

Crédit Photo : T. Claude, A. Volte (2015)

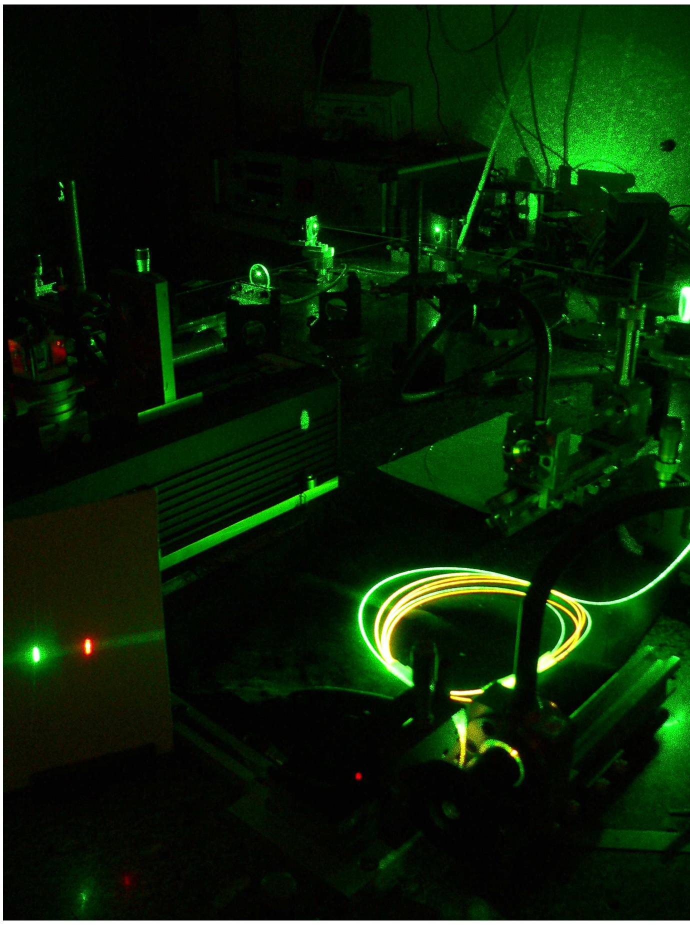


...

λ_{pump} λ_{s1} λ_{s2} λ_{s3} ...

532 nm

Cascaded Raman Effect



Scattering

- ***Raman scattering in a hollow-core photonic crystal fibre filled with ETHANOL***

(Sylvie LEBRUN, LCF)

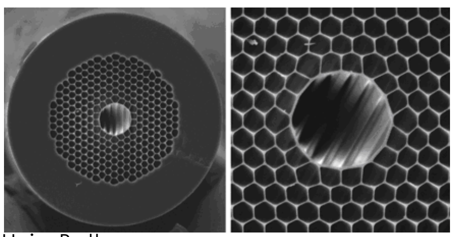
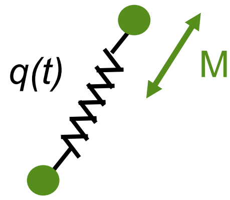


Photo : Univ. Bath

III - Raman Scattering

- ***Spontaneous Raman Scattering - Microscopic origin***



$$q - q_0 = q_1 \cos(\omega_v t)$$

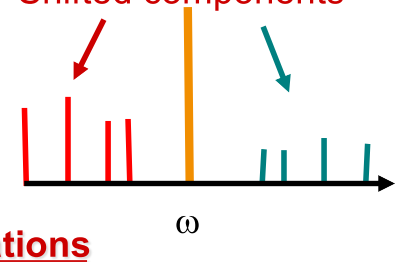
Polarizability of the molecule $\alpha = \alpha_0 + \left(\frac{\partial \alpha}{\partial q}\right)_0 (q - q_0)$

Incident monochromatic wave : $E = E_0 \cos(\omega t)$

Induced dipole : $p = \alpha\ E$ (α : polarizability of the molecule)

$p(\omega)$ $p(\omega + \omega_v)$ $p(\omega - \omega_v)$

AntiStokes **Stokes**

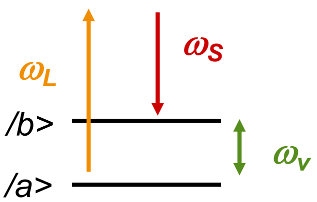


Origin of the process : <u>polarizability fluctuations</u>

III - Raman Scattering

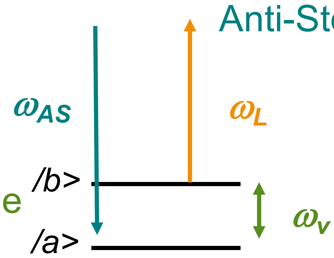
- ***Raman Scattering - Energy level diagrams***

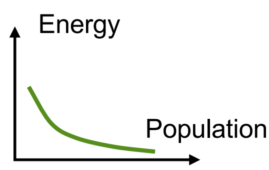
Stokes component



"optical" phonon
Vibrational mode of the molecules

Anti-Stokes component





Raman Stokes scattering :
Initial state for the medium = \a>
Creation of one Stokes photon ω_S is accompanied by the annihilation of one photon ω_L
Final state for the medium = \b>

Raman anti-Stokes scattering :
Initial state for the medium = \b>
Creation of one anti-Stokes photon ω_{AS} is accompanied by the annihilation of one photon ω_L
Final state for the medium = \a>

Boltzmann distribution

$$exp\left(\frac{-\hbar\omega_{ab}}{k_B T}\right)$$

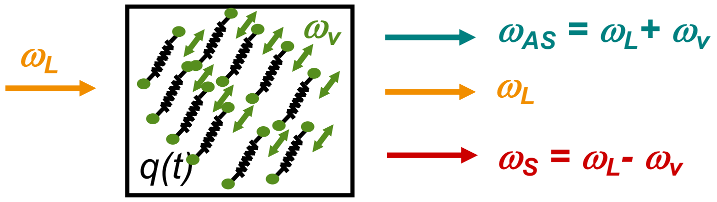
Since $\hbar\omega_{ab} \gg k_B T$

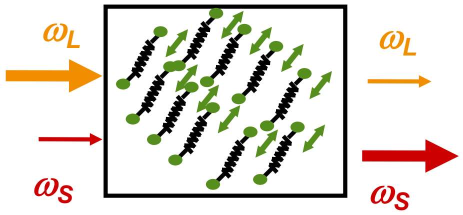
Stokes scattering is predominant

III - Raman Scattering

- ***Stimulated Raman scattering - Classical behavior***

Molecules in vibration





- The beating between the wave components ω_L ***and*** ω_S strengthens (drives) a collective and coherent vibration of the molecules @ ω_L
- Consequence : amplification of the signal @ ω_S

Energy required to drive the dipole oscillation : $W = \frac{1}{2}\langle \mathbf{p}(z,t).\mathbf{E}(z,t)\rangle$

Time average

$$\mathbf{p} = \alpha \mathbf{E} \quad \Rightarrow \quad W = \frac{1}{2}\alpha\langle \mathbf{E}^2(z,t)\rangle$$

III - Raman Scattering

- ***Stimulated Raman scattering - Classical behavior***

Driven force applied onto the oscillator :

$$F = \frac{dW}{dq} = \frac{1}{2}\frac{d\alpha}{dq}\langle \mathbf{E}^2(z,t)\rangle$$

Applied fields : $E = E_L \cos\omega_L t + E_S \cos\omega_S t$

Equation of motion of a classical harmonic oscillator :

$$\boxed{\frac{d^2q}{dt^2} + 2\gamma\frac{dq}{dt} + \omega_v^2 q = \frac{F(t)}{m}}$$

$$E(z,t) = A_L e^{i(k_L z - \omega_L t} + A_S e^{i(k_S z - \omega_S t} + CC$$

$$\frac{d^2q}{dt^2} + 2\gamma\frac{dq}{dt} + \omega_v^2 q = \frac{1}{m}\frac{d\alpha}{dq}[A_L A_S^* e^{i(Kz-\Omega t)} + CC]$$

$$K = k_L - k_S$$
$$\Omega = \omega_L - \omega_S$$

III - Raman Scattering

- ***Stimulated Raman scattering - Classical behavior***

$$\frac{d^2q}{dt^2} + 2\gamma\frac{dq}{dt} + \omega_v^2 q = \frac{1}{m}\frac{d\alpha}{dq}[A_L A_S^* e^{i(Kz-\Omega t)} + CC]$$

Driven solution of the form $\quad q = q(\Omega)e^{i(Kz-\Omega t)} + CC$

solution $\quad q(\Omega) = \dfrac{\frac{1}{m}\left(\frac{d\alpha}{dq}\right) A_L A_S^*}{\omega_v^2 - \Omega^2 - 2i\Omega\gamma}$

Expression of the macroscopic polarization

$$P(z,t) = N\alpha(z,t)E(z,t) = N[\alpha_0 + \left(\frac{d\alpha}{dq}\right) q(z,t)]E(z,t)$$

$$P_{NL}(z,t) = N\left(\frac{d\alpha}{dq}\right)[q(\Omega)e^{i(Kz-\Omega t)} + CC].[A_L e^{i(k_L z-\omega_L t} + A_S e^{i(k_S z-\omega_S t} + CC]$$

$$\boxed{P_{NL}(\omega_S) = \epsilon_0\chi_R^{(3)}(\omega_S;\omega_L,-\omega_L,\omega_S)|A_L|^2 A_S e^{ik_S z}}$$

III - Raman Scattering

- ***Stimulated Raman scattering - Classical behavior***

$$P_{NL}(\omega_S) = \epsilon_0\chi_R^{(3)}(\omega_S;\omega_L,-\omega_L,\omega_S)|A_L|^2 A_S e^{ik_S z}$$

$$\boxed{\chi_R^{(3)}(\omega_S) = \frac{\frac{N}{\epsilon_0 m}\left(\frac{d\alpha}{dq}\right)^2}{\omega_v^2 - (\omega_L-\omega_S)^2 + 2i(\omega_L-\omega_S)\gamma}}$$

Susceptibility expression shows that in a resonance case i.e. $\omega_v = \omega_L - \omega_S$

$\chi_R^{(3)}(\omega_S) =$ negative imaginary

III - Raman Scattering

- ***Stimulated Raman scattering***

Nonlinear polarization calculation ***@ ω_S (ω_{AS}) et ω_L*** solving the equation of motion of a classical harmonic oscillator ***:***

$$P_{NL}(\omega_S) = \varepsilon_0\, \chi_R^{(3)}(\omega_S;\omega_L,-\omega_L,\omega_S)|A_L|^2 A_S\, e^{ik_S z}$$

$$P_{NL}(\omega_L) = \varepsilon_0\, \chi_R^{(3)}(\omega_L;\omega_S,-\omega_S,\omega_L)|A_S|^2 A_L\, e^{ik_L z}$$

Susceptibility expression shows that in a resonance case i.e. $\omega_v = \omega_L - \omega_S$

$\chi_R^{(3)}(\omega_S) = \chi_R^{(3)}(\omega_L)^* =$ negative imaginary

Coupled equations :

$$\begin{cases} \dfrac{\partial A_S}{\partial z} = g_R |A_L|^2 A_S \\ \dfrac{\partial A_L}{\partial z} = -\dfrac{\omega_L}{\omega_S} g_R |A_S|^2 A_L \end{cases}$$

Stokes wave amplification

Pump wave depletion

with $g_R = \dfrac{3\omega_S}{2nc}\chi_R^{(3)}(\omega_S)$

III - Raman Scattering

- ***Raman amplification :***

$$\begin{cases} \dfrac{dP_S}{dz} = -\alpha_S P_S + \dfrac{\gamma_R}{A_{eff}} P_L P_S \\ \dfrac{dP_L}{dz} = -\alpha_L P_L - \dfrac{\gamma_R}{A_{eff}} P_S P_L \end{cases}$$

with $\gamma_R = \dfrac{2\, g_R}{2nc\varepsilon_0}$ Raman Gain of the medium in m.W^{-1}

Aeff : effective mode area of the optical fiber

Solution in the undepleted pump approximation

$$P_L(z) = P_L(0)\, e^{-\alpha_L z}$$

$$\boxed{P_S(z) = P_S(0)\, \underbrace{e^{-\alpha_S z}\, \overbrace{e^{\frac{\gamma_R}{A_{eff}} P_L(0) L_{eff}}}^{\text{“ON-OFF” Gain}}}_{\text{Net Gain}}}$$

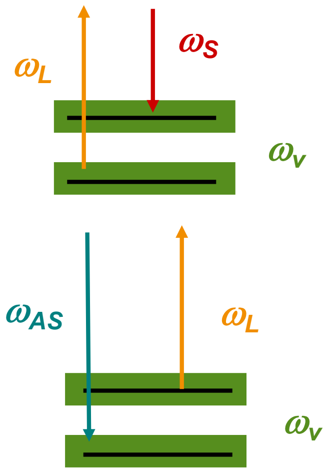
Avec $L_{eff} = \dfrac{1}{\alpha_L}\left(1 - e^{-\alpha_L z}\right)$

Effective length

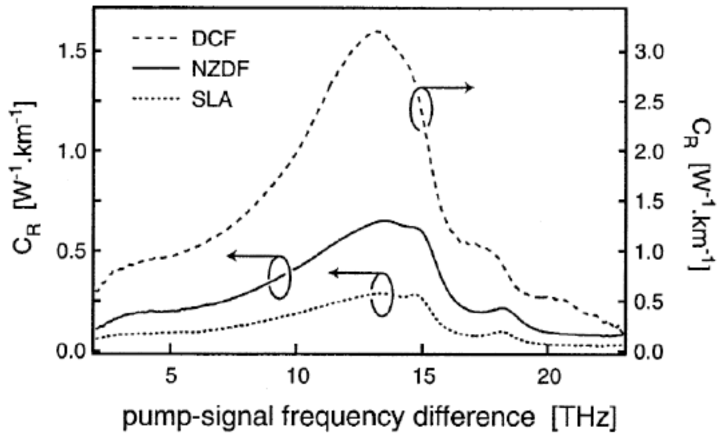
III - Raman Scattering

- ***Example : Raman scattering in silica fiber***

ω_v = 13 THz



Spontaneous Raman Scattering spectra
for various doped silica



Bromage, JLT 22, 79-93 (2004)

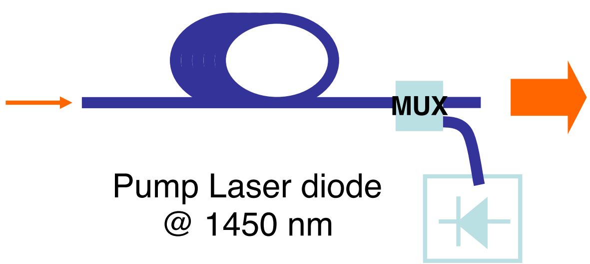
III - Raman Scattering

- ***Raman amplification :***

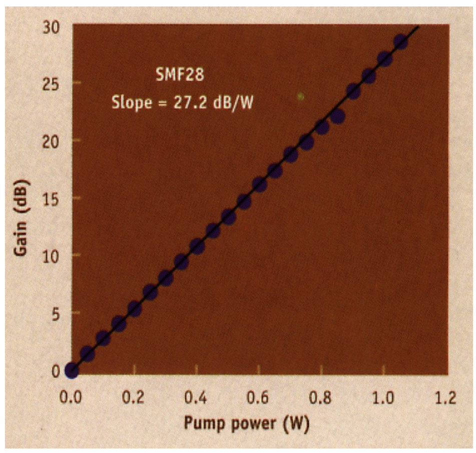
Structure

Stimulated Raman scattering

Standard optical fiber



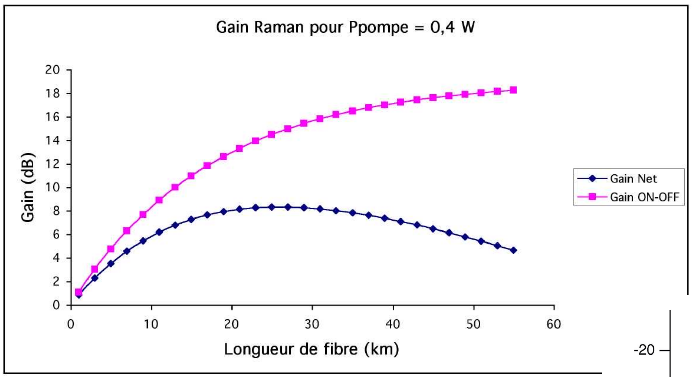
Raman gain amplification



III - Raman Scattering

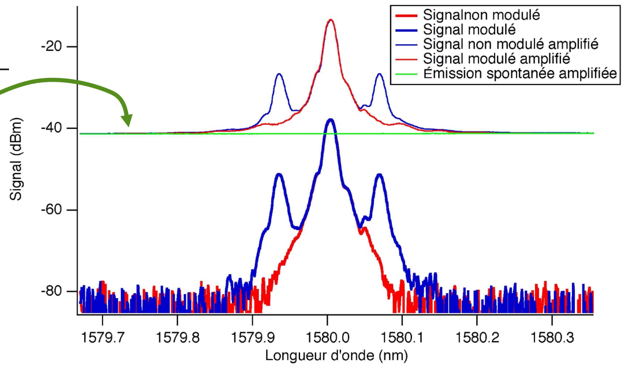
- ***Raman amplification :***

Raman fiber amplification



Input and output optical spectra

Amplified spontaneous emission

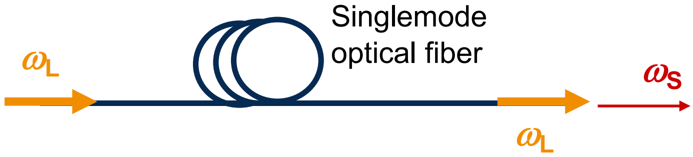


III - Raman Scattering

- ***Amplified spontaneous Raman scattering***

For $P_s(0)=0$, spontaneous Raman scattering

Example :



Calculation of the number of Stokes photons created through the amplification of the photons initially created through spontaneous Raman scattering

$N_s(0)$ = 1 photon per mode

See exercise + [Agrawal, *Nonlinear fiber optics*,Ch8] + [Smith, Appl. Opt. **11**, 2489 (1972)]

Threshold condition : optical power for which $P_s(z) = P_L(z)$

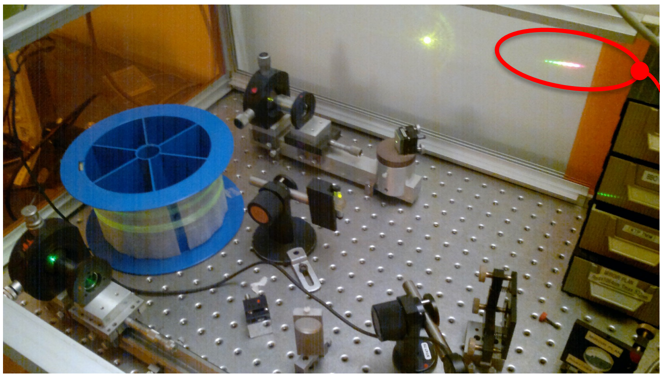
For a lengthy fiber $L_{\text{eff}} \approx 1/\alpha_L \approx 20\ \text{km} \quad @1{,}55\ \mu\text{m}$

$A_{eff} = 50\ \mu m^2$ **$P_{seuil} \approx 600$ mW** (relatively high value)

III - Raman Scattering

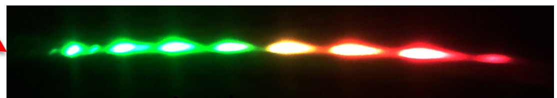
III - Raman Scattering

- ***Raman scattering in a silica based optical fibre***



Output Optical Spectrum

Crédit Photo : T. Claude, A. Volte (2015)



…

λ_{pump} λ_{s1} λ_{s2} λ_{s3} …

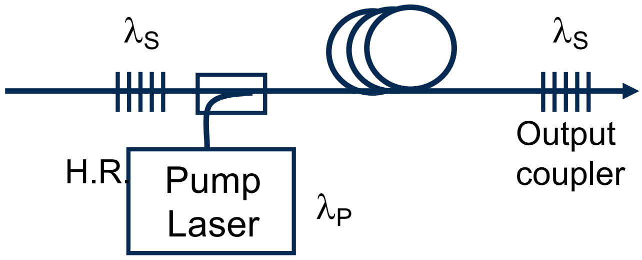
532 nm

Cascaded Raman Effect

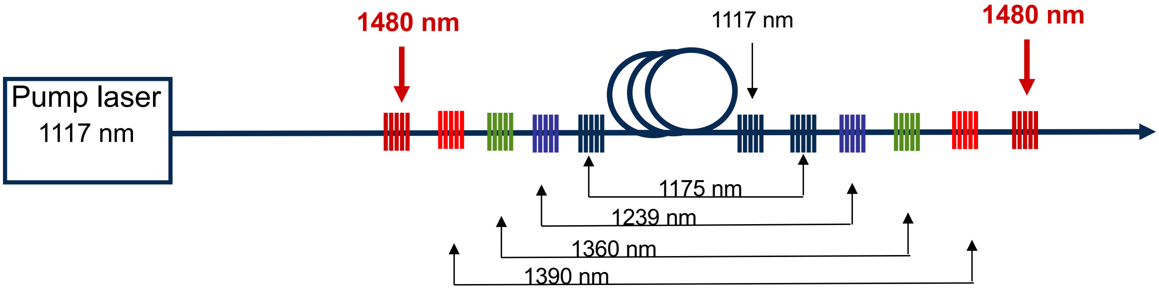
III - Raman Scattering

- ***Raman fiber laser***

$\omega_V = \omega_L - \omega_S$



Cascaded Raman fiber lasers



NONLINEAR OPTICS

Ch. 5 3rd ORDER NONLINEARITIES

I. **Optical Kerr Effect :** self-focusing, nonlinear Shrödinger equation, self-phase modulation, solitons

II. **Four-wave Mixing**

III. **Raman Scattering :** spontaneous and stimulated Raman scattering, Raman amplification, Raman laser

IV. **Brillouin Scattering :** spontaneous and stimulated Brillouin scattering

IV - Brillouin Scattering

• *Origin of the Brillouin scattering*

Inelastic scattering due to the fluctuation of the density of the material

- presence of thermal fluctuations : spontaneous Brillouin scattering
-> scattering of light from acoustic phonons
- density fluctuations reinforced by the beating between two optical waves through electrostriction : stimulated Brillouin scattering

• *Spontaneous Brillouin scattering*

Density fluctuation

$$N = \rho_0 + \Delta\rho\, e^{i(\vec{q}.\vec{r} - \Omega t)} + c.c$$

Average density

Density fluctuation driven by a propagative sound wave

Inelastic scattering onto a sound wave

$\xrightarrow{\omega_L}$ $\boxed{\Delta\rho(\Omega,v)}$

Medium

IV - Brillouin Scattering

Dispersion relation for sound waves

$$\Omega = v\,|q|$$

with v : sound velocity within the medium

Induced macroscopic polarization due to density fluctuations of the medium

$$\vec{P}_L = N(0,\Omega)\,\alpha\,\vec{E}_L(\omega_L)$$

$$P(\omega_L)$$

$$P(\omega_S = \omega_L - \Omega)$$ Stokes wave

$$P(\omega_{AS} = \omega_L + \Omega)$$ Anti-Stokes wave

Phase matching condition

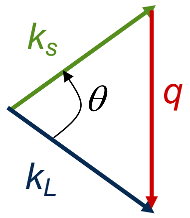
$$\vec{k}_S = \vec{k}_L - \vec{q}$$

$$\vec{k}_{AS} = \vec{k}_L + \vec{q}$$

IV - Brillouin Scattering

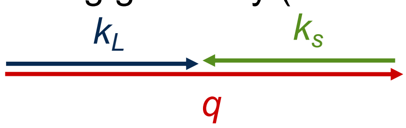
- ***Typical orders of magnitude***

  - amplitude of the acoustic wavevector $|q| > |k|$
  - phase matching condition



$$\Omega_B = 2\frac{v}{c}\omega\sin(\theta/2)$$

The maximum frequency shift is achieved in a backscattering geometry (θ=180°)



  - Brillouin frequency shift = acoustic wave frequency

$$\Omega_B \approx \frac{2\,\omega\, n\, v}{c}$$

Silica case : Ω_B = 12 GHz

  - Linewidth of the Brillouin shift

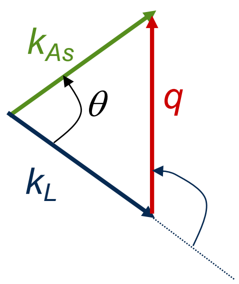
Silica : 10 MHz

IV - Brillouin Scattering

- ***Phase-matching considerations***

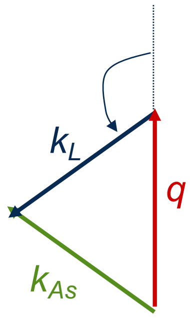
$$|q| > |k|$$

Anti-Stokes Scattering

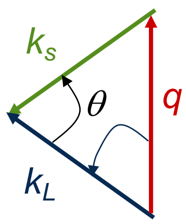


$$\vec{k}_{AS} = \vec{k}_L + \vec{q}$$

$$\left(\vec{k}_L, \vec{q}\right) > \pi/2$$

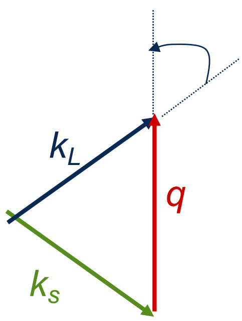


Stokes Scattering



$$\vec{k}_S = \vec{k}_L - \vec{q}$$

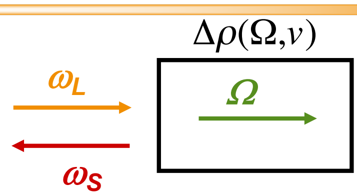
$$\left(\vec{k}_L, \vec{q}\right) < \pi/2$$



IV - Brillouin Scattering

- ***Stimulated Brillouin scattering***

Counterpropgating geometry



CW or quasi-CWregime : Resolution of the coupled optical and sound wave equations shows the following expression of the spatial intensity evolutions for pump and Stokes

For details See [Boyd, Ch.9]

$$\begin{cases} \dfrac{dI_S}{dz} = -g_B I_L I_S + \alpha I_S \\ \dfrac{dI_L}{dz} = -g_B I_L I_S - \alpha I_L \end{cases}$$

Equations similar to SRS, with a difference in the sign (contra-propa. géometry)

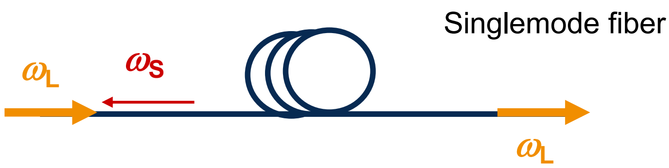
Ex. : silica $g_B = 5\ 10^{-11}$ m/W

IV - Brillouin Scattering

- ***Amplified spontaneous Brillouin scattering***

For $P_s(0)=0$, spontaneous Brillouin scattering

Example :



Calculation of the number of Stokes photons created through the amplification of the photons initially created through spontaneous Brillouin scattering

$N_s(0)$ = 1 photon per mode

Threshold condition : optical power for which $P_s(z) = P_L(z)$

For a lengthy fiber $\quad L_{\mathrm{eff}} \approx 1/\alpha_L \approx 20\ \mathrm{km} \quad @1{,}55\ \mu\mathrm{m}$

$A_{eff} = 50\ \mu m^2$

$P_{seuil} \approx$ 1 mW !!

Lower threshold than Raman process

Very easy to observe in CW regime