Raman / Brillouin scattering

The previous nonlinear process, harmonics, sum/difference frequency generation... are all elastic scattering process.

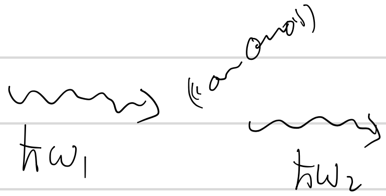
Which means there's no energy exchange between light and matter.

However, inelastic process was discovered way before elastic process.

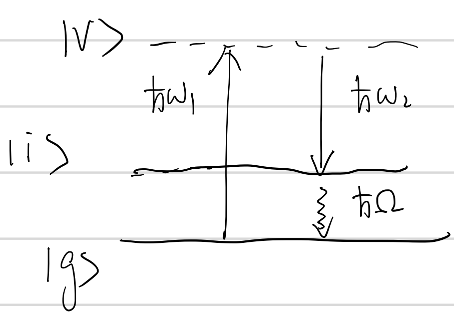
In this type of process, photon has to exchange energy with the material

Depending on where the energy goes, there are many different type of scattering.

1°. Raman Scattering : photon exchanges energy with molecular vibration/rotation modes.



Energy diagram:



The first Raman effect discovered was spontaneous Raman effect, where

$$\omega_1 = \omega_2 + \Omega$$

Because Ω is related to the molecular features, Raman effect has become popular in spectroscopy.

Raman Spectroscopy (commercilized).

Now many groups are working on very advanced Raman spectroscopy either for high resolution, or high sensitivity.

Video: Sunny, Xiaoliang, Xie, Harvard University.

Question: is Raman second / third nonlinear effect?

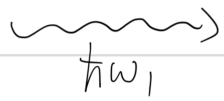
Easy guess: has to be third for a small frequency shift.

$$\frac{\partial E_2}{\partial z} = \dots |E_1|^2 E_2 \quad ; \quad \text{You can't have } E_1 E_2,\ E_1 E_2^*, \text{ etc.}$$

Their frequency doesn't match.

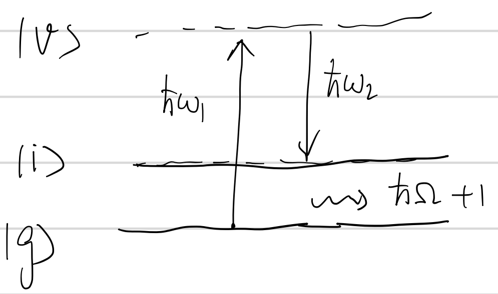
More detailed analysis can be found in Nonlinear Optics.

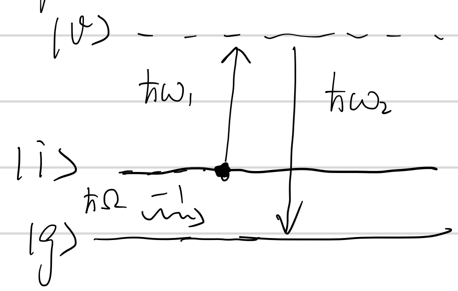
Spontaneous vs. Stimulated scattering.



The incident photon can take / give some energy from the material therefore the output photon can have high (anti-Stokes) or lower (Stokes) frequency

Let's look at these two process:





This is Stokes process.
Give energy to molecules

$\hbar\omega_1 \longrightarrow \hbar\omega_2 + \hbar\Omega$

This is anti-Stokes process:
Take $\hbar\Omega$ energy from molecules

$\hbar\omega_1 + \hbar\Omega \longrightarrow \hbar\omega_2$

What is the ratio of Stokes photon and anti-Stokes photon?

$$\frac{N_i}{N_g} = e^{-\hbar\Omega/KT} ;$$

For anti-Stokes process, you need the molecules to be at $|i\rangle$ state, while stokes process: at ground-state.

So roughly; $\dfrac{I_S}{I_{Anti\text{-}S}} = e^{\hbar\Omega/KT}$;

Usually: $\dfrac{\Omega}{2\pi} \sim$ few THz for heavy molecular bond.

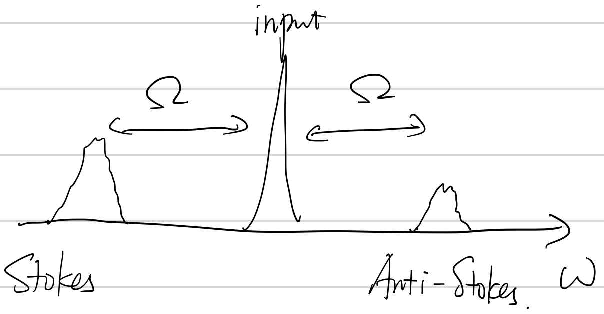
$SiO_2 \sim 13 THz$; $h/KT \sim 0.16 \times 10^{-12}$ s
Room temperature

$$\therefore \quad \frac{\hbar\Omega}{KT} = \frac{h \cdot \frac{\Omega}{2\pi}}{KT} = 0.16\times10^{-12} \times 13\times10^{+12}$$

$$= 2$$

$$\therefore \quad \frac{I_S}{I_{Anti-S}} \sim e^{-2} = 0.135.$$

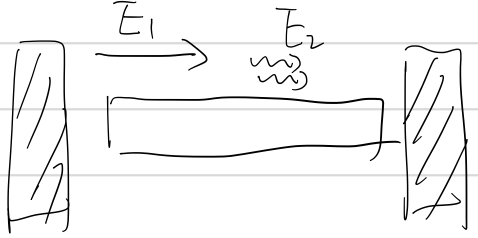
Some useful number to remeber: $KT \sim \hbar\Omega$ when $\frac{\Omega}{2\pi} = 6\,THz$



Stimulated emission:

Stimulated emission means the previous generated photon/phonon will trigger more photon/phonon.

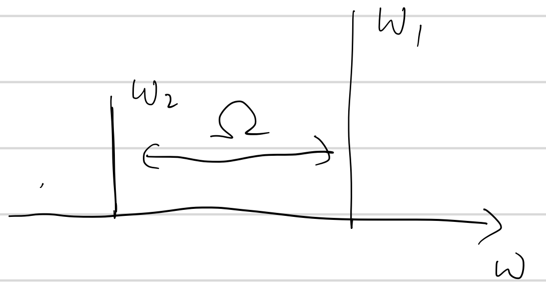
Stokes is easier than anti-Stokes.



For simplicity, work with only two frequencies:

$$\frac{dE_2}{dt} = g_2 |E_1|^2 E_2 - \frac{1}{\tau} E_2$$

$$\frac{dE_1}{dt} = -g_1 |E_2|^2 E_1 - \frac{1}{\tau} E_1$$



How do we get to here?

People have found that (experimentally)

$$\frac{d}{dz} I_{\omega_2} \propto n_{\omega_1}(n_{\omega_2}+1)$$

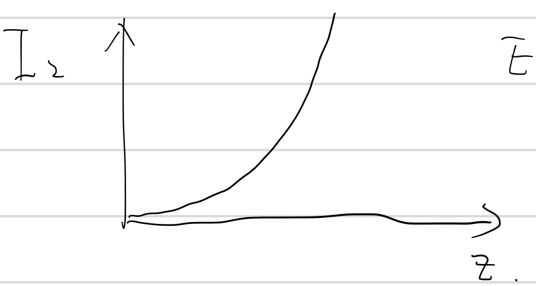
$$\Rightarrow \frac{d|\tilde{E}_2|^2}{dz} \propto |\tilde{E}_1|^2 |\tilde{E}_2|^2$$

The same as:

$$\frac{d\tilde{E}_2}{dz} = |\tilde{E}_1|^2 \tilde{E}_2 ; \qquad \frac{d\tilde{E}_2^*}{dz} = |\tilde{E}_1|^2 \tilde{E}_2^*$$

$$\tilde{E}_2^* \frac{d\tilde{E}_2}{dz} + \tilde{E}_2 \frac{d\tilde{E}_2^*}{dz} = 2|\tilde{E}_1|^2 |\tilde{E}_2|^2 .$$

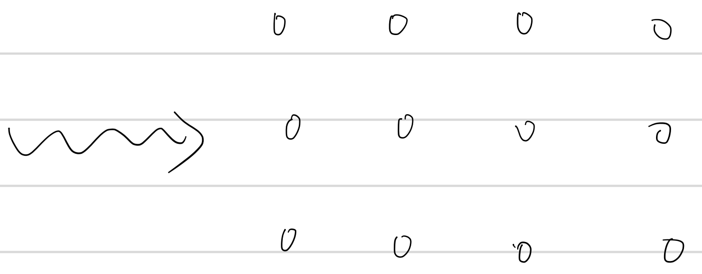
Keep $|\tilde{E}_1| \gg |\tilde{E}_2|$



Exponential: $\frac{dI}{dz} = \alpha \cdot I$.

$$I(z) = I_0 e^{\alpha z}.$$

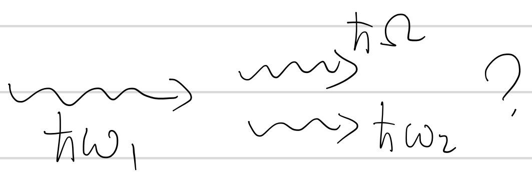
Brillouin Scattering



exchange energy with phonon.

Critical difference. Phonon has large k-vector.

phonons are like photons, they have momentum.



Let's see if this is possible:

Energy conservation:

$$\omega_1 = \omega_2 + \Omega. \qquad (\Omega \sim \text{GHz})$$

Momentum conservation:

$$\frac{\omega_1}{c/n} = \frac{\omega_2}{c/n} + \frac{\Omega}{v} \quad ; \quad v \text{ is phonon speed.}$$

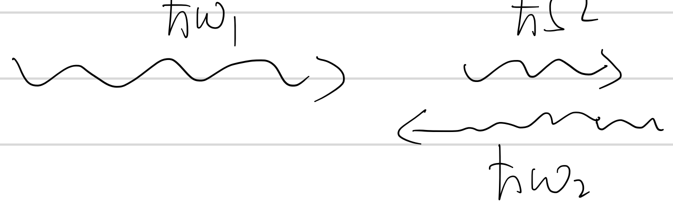
$$\Rightarrow \frac{\omega_1 - \omega_2}{c/n} = \frac{\Omega}{c/n} = \frac{\Omega}{v}.$$

However, speed of light is 3×10^8; phonon is speed of sound, ~ 5000 m/s in solid.

Doesn't work...

So what would happen?

The problem is: the momentum change in photon is too small. To make it huge, you can have backward scattering.



$$\hbar\omega_1 = \hbar\omega_2 + \hbar\Omega$$

$$\frac{\omega_1}{c/n} = \frac{\Omega}{v} - \frac{\omega_2}{c/n} \Rightarrow \frac{\Omega}{v} = \frac{\omega_1+\omega_2}{c/n}$$

$\Omega \ll \omega_1, \omega_2$; $\therefore$ $\Omega \approx \frac{2\omega \cdot v}{c/n}$; $\frac{\Omega}{2\pi} = \frac{2v}{c/n} \cdot \frac{\omega}{2\pi}$

At 1550nm in silica (optical fiber)

$\frac{\omega}{2\pi} = 193.4\text{THz}$; $n = 1.45$; $v = 5600\text{ m/s}$.

$$\therefore \Omega = \frac{2\times 5600\times 1.45}{3\times 10^8} \times 193.4\text{THz} = 10.47\text{ GHz}.$$

This is the frequency difference in a fiber at 1550nm.