Module 4 : Third order nonlinear optical processes

Lecture 30 : Stokes anti-Stokes coupling

Objectives

In this lecture you will learn the following

- The coupling of Stokes and anti-Stokes fields.
- The effect of phase matching on the Raman gain and condition for the efficient generation of anti-Stokes wave.

Stokes anti-Stokes coupling

In our discussion of stimulated Stokes Raman scattering, we have seen that the Stokes wave is amplified at the cost of the pump ware. Coupling between the Stokes and pump wave is mediated by a third order nonlinear optical mixing process.

The corresponding non linear susceptibility was shown to be

$$\chi_R(\omega_s) = N\frac{\varepsilon_0}{6m}\frac{\left(\frac{\partial\alpha}{\partial Q}\right)_e^2}{\left(\omega_m^2-(\omega_p-\omega_s)^2+2i\gamma(\omega_p-\omega_s)\right)} \tag{30.1}$$

In its derivation sign of $\omega_p-\omega_s$ was nonspecific.

Next we examine the spatial evolution of the anti-stokes wave propagating through the medium in the presence of the pump field. This process naturally will be governed by the nonlinear polarization induced in the medium by the interaction of the pump wave, material excitation $\hbar\omega_m$ and the anti-stokes field as derived from the classical driven and damped oscillator model in the previous lecture. The same is given below

$$P_{NL}(z,\omega_{as},t) = \left[P_{NL}(z,\omega_{as})e^{-i\omega_{as}t}+c.c.\right]$$

$$= N\frac{\varepsilon_0^2}{m}\left(\frac{\partial\alpha}{\partial Q}\right)_e^2\left[\frac{E_pE_s^*}{(\omega_m{}^2-\Omega^2-2i\gamma\Omega)}e^{i(Kz-\Omega t)}+c.c.\right]\left(E_pe^{i(k_pz-\omega_pt)}+E_se^{i(k_sz-\omega_st)}+c\cdot c\right) \tag{30.2}$$

The corresponding nonlinear susceptibility for the induced anti-Stokes Raman scattering can be readily written as

$$\chi_R(\omega_{as}) = \frac{B}{\left(\omega_m^2-\left(\omega_p-\omega_{as}\right)^2+2i\gamma\left(\omega_p-\omega_{as}\right)\right)} \tag{30.3}$$

Where

$$B = N\frac{\varepsilon_0}{6m}\left(\frac{\partial\alpha}{\partial Q}\right)_e^2 \tag{30.4}$$

Since

$$\omega_p-\omega_s = -\left(\omega_p-\omega_{as}\right) \tag{30.5}$$

It is easy to see that

$$\chi_R(\omega_{as}) = \chi_R^*(\omega_s) \tag{30.6}$$

Near Raman resonance the expression of the susceptibility can be simplified as

$$\chi_R(\omega_{as}) = -\frac{B}{2\omega_m(\omega_{as}-(\omega_p+\omega_m)+i\gamma)} \tag{30.7}$$

Note that at the Raman resonance $\omega_{as} = \omega_p + \omega_m$, it is purely imaginary and positive definite as shown in its frequency dispersion in figure 30.1

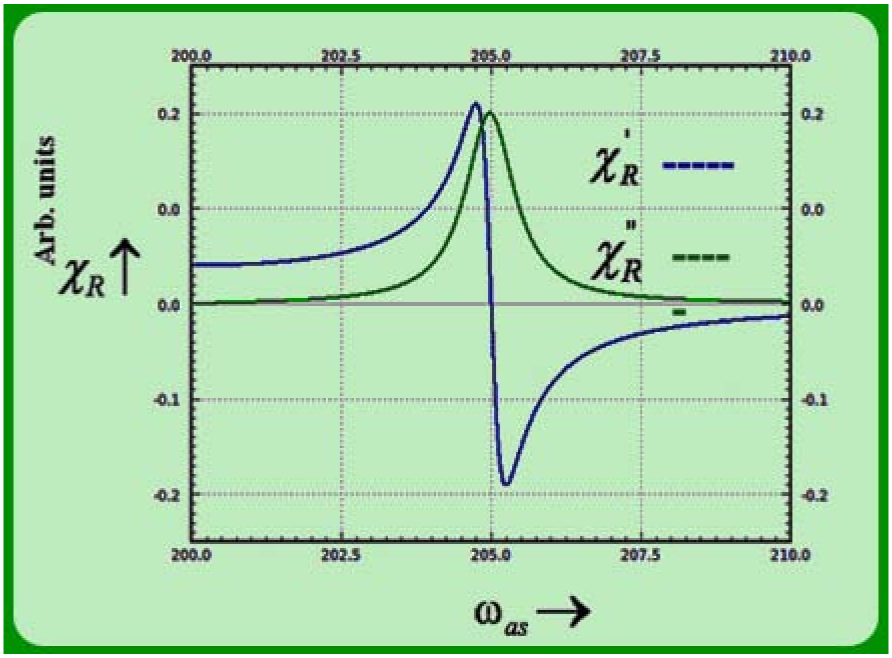

Figure 30.1 Frequency dispersion of the Real and imaginary parts of anti-Stokes Raman Susceptibility

We assume that the pump field remains constant – the weak conversion limit.

In analogy to the equation (30.29), the growth of anti–Stokes wave will then be described by

$$\frac{\partial E_{as}}{\partial z} = -i\frac{\omega_{as}}{2n_{as}c}\chi_R(\omega_{as})\left|E_p\right|^2 E_{as} = -\alpha_{as}\left|E_p\right|^2 E_{as} \tag{30.8}$$

where

$$\alpha_{as} = -3i\frac{\omega_{as}}{n_{as}c}\chi_R(\omega_{as}) \tag{30.9}$$

is the field attenuation coefficient for the induced anti-Stokes Raman scattering

At resonance, $\alpha_{as} = \frac{\omega_{as}\cdot B}{n_{as}\gamma c}$ and it is positive definite $\Longrightarrow$ anti-Stokes field is attenuated.

However, it has been demonstrated that the anti-Stokes wave is generated with intensity comparable to that of the Stokes wave, at least under certain conditions. An inspection of the equation (30.2) reveals that this arises from the following additional contribution to the $P_{NL}(\omega_{as})$ which depends on the amplitude of the Stokes wave.

$$P_{NL}(\omega_{as}) = \frac{3\varepsilon_0 A E_p^2 E_s^* e^{i(2k_p-k_s)z}}{\omega_m^2-(\omega_p-\omega_{as})^2+2i\gamma(\omega_p-\omega_{as})} \tag{30.10}$$

(Remember $\omega_{as} = 2\omega_p - \omega_s$)

Here

$$A = N\frac{\varepsilon_0}{3m}\left(\frac{\partial\alpha}{\partial Q}\right)_e^2 = 2B \tag{30.11}$$

The corresponding nonlinear susceptibility for this four wave-mixing of $\omega_p;\omega_s;\omega_{as}$ frequencies is

$$\chi^{(3)}\left(-\omega_{as};\omega_p,-\omega_s,\omega_p\right) = \chi_{sasc}\left(\omega_{as}\right) = \frac{A}{\left(\omega_m^2-\left(\omega_p-\omega_{as}\right)^2+2i\gamma\left(\omega_p-\omega_{as}\right)\right)} \tag{30.12}$$

Thus

$$\chi_{sasc}\left(\omega_{as}\right) = 2\chi_R\left(\omega_{as}\right) \tag{30.13}$$

Total source polarization for anti-Stokes wave is thus

$$P_{NL}\left(\omega_{as}\right) = 6\varepsilon_0\chi_R\left(\omega_{as}\right)\left|E_p\right|^2 E_{as}e^{ik_{as}z} + 3\varepsilon_0\chi_{sasc}\left(\omega_{as}\right)E_p^2E_s^*e^{i\left(2k_p-k_s\right)z} \tag{30.14}$$

In a similar way there is a contribution to Stokes polarization that depends upon the amplitude of the anti-Stokes wave.
Total polarization for the stokes wave then is

$$P_{NL}\left(\omega_s\right) = 6\varepsilon_0\chi_R\left(\omega_s\right)\left|E_p\right|^2 E_se^{ik_{as}z} + 3\varepsilon_0\chi_{sasc}\left(\omega_s\right)E_p^2E_{as}^*e^{i\left(2k_p-k_s\right)z} \tag{30.15}$$

Once again

$$\chi_{sasc}\left(\omega_s\right) = 2\chi_R\left(\omega_s\right) = \chi_{sasc}^*\left(\omega_{as}\right) \tag{30.16}$$

Using SVEA, the spatial evolution of the stokes and anti-stokes field is then given by the following pair of differential equations

$$\frac{d}{dz}E_s = -\alpha_sE_s + \kappa_sE_s^*e^{i\Delta kz} \tag{30.17}$$

and

$$\frac{d}{dz}E_{as} = -\alpha_{as}E_{as} + \kappa_{as}E_{as}^*e^{i\Delta kz} \tag{30.18}$$

where the abbreviations α_j, and κ_j for j = s, as stand for the nonlinear absorption and coupling coefficients defined as below, respectively.

$$\alpha_s = -\frac{-3i\omega_s}{n_sc}\chi_R\left(\omega_s\right)\left|E_p\right|^2 \tag{30.19}$$

$$\alpha_{as} = -\frac{-3i\omega_{as}}{n_{as}c}\chi_R\left(\omega_{as}\right)\left|E_p\right|^2$$

$$= \alpha_s^*\left(\frac{n_s\omega_{as}}{n_{as}\omega_s}\right) \tag{30.20}$$

and

$$\kappa_s = 3i\frac{\omega_s}{n_sc}\chi_{sasc}\left(\omega_s\right)E_p^{\ 2} \tag{30.21}$$

$$= \alpha_s e^{2i\phi_p}$$

$$\kappa_{as} = 3i\,\frac{\omega_{as}}{n_{as}c}\,\chi_{sax}\left(\omega_{as}\right)E_p^{\;2}$$

$$= \alpha_s^*\left(\frac{n_s\omega_{as}}{n_{as}\omega_s}\right)e^{2i\phi_p} \tag{30.22}$$

$\Delta\vec{k} = 2\vec{k}_p - \vec{k}_s - \vec{k}_{as}$ is the wave vector mismatch

Equations (30.17) and (30.18), describe the coupled dynamics of the Stokes and anti-Stokes fields. The coupling of the two is facilitated by $\Delta k \sim \alpha_s$ in the second term on the R.H.S. For large $\Delta\vec{k}$, only the first term is significant and the two equations decouple. In this case as usual, the Stokes field is amplified and the anti-Stokes field is attenuated.

The consequence of their coupled nature can be understood in terms of the simple quantum physical picture as follows.

Subsequent to the creation of the stimulated Stokes photon upon interaction with the pump and Stokes field, the molecule is available in the higher excited state. This energy is immediately utilized by the pump for the creation of anti-Stokes photon with the energy the energy $\hbar\omega_{as}$. This way, the Stokes and anti-stokes coupling is established and the Stokes and anti-stokes fields are emitted with comparable intensity. Since the molecule does not change its final state, the generation of Stokes and anti -Stokes photons under this coupling is a parametric process.

The energy balance is given by

$$2\hbar\omega_p = \hbar\omega_s + \hbar\omega_{as} \tag{30.23}$$

In a parametric process the momentum conservation must be fulfilled i.e. $\Delta\vec{k} = 0$ or

$$2\overrightarrow{k_p} = \overrightarrow{k_s} + \overrightarrow{k_{as}} \tag{30.24}$$

Where $\overrightarrow{k_p}$, $\overrightarrow{k_s}$, $\overrightarrow{k_{as}}$ are the propagation rectors

for the pump, Stokes and anti-Stokes waves, respective. Their magnitudes are given by

$$\left|\vec{k}_p\right| = \frac{\omega_p n_p}{c},\left|\vec{k}_s\right| = \frac{\omega_s n_s}{c},$$

$$\text{and } \left|\vec{k}_{as}\right| = \frac{\omega_{as} n_{as}}{c} \tag{30.25}$$

Here n_p, n_s and n_{as} are the refractive indices at pump, Stokes and anti Stokes frequencies.

Since the frequencies of the three fields lie very close, we can expand their refractive indices in the neighborhood of ω_p

$$n_s = n_p - \omega_m \frac{dn}{d\omega} \tag{30.26}$$

and

$$n_{as} = n_p + \omega_m \frac{dn}{d\omega} \tag{30.27}$$

Thus

$$\left|\vec{k}_s\right| + \left|\vec{k}_{as}\right| = 2\left|\vec{k}_p\right| + \frac{2}{c}\omega_m^2 \frac{dn}{d\omega} \quad (30.28)$$

∵ in the region of normal dispersion

$$\frac{dn}{d\omega} > 0$$
$$\left|\vec{k}_s\right| + \left|\vec{k}_{as}\right| > 2\left|\vec{k}_p\right| \quad (30.29)$$

$\Rightarrow$ conservation of momentum cannot be satisfied with parallel propagation vectors. The equation (30.24) constitutes a non degenerate triangle as shown in figure 30.2.

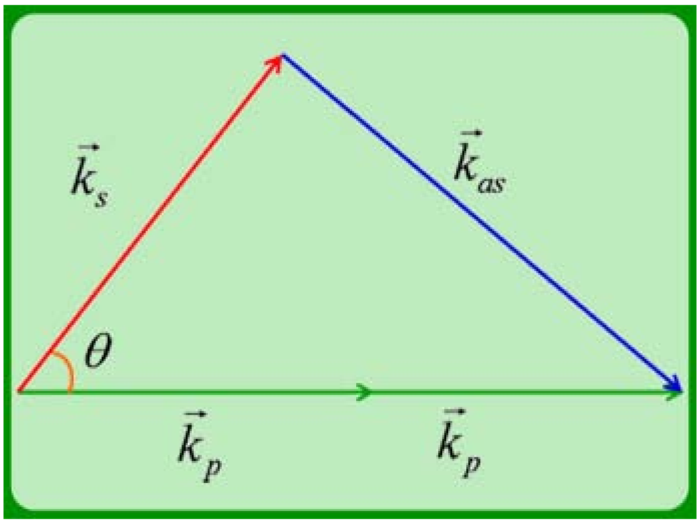

Figure 30.2 Non-degenerate triangle of wave vectors to satisfy momentum conversation

Consequently, Stokes and anti-Stokes lights are emitted in the form of conical shells about the direction of the pump wave with definite aperture angle. Let us now the coupled equations in some more detail.

We can write equation (30.17) in the form

$$\left(\frac{d}{dz} + \alpha_s + i\frac{\Delta k}{2}\right) E_s e^{-i\frac{\Delta kz}{2}} = \kappa_s E_{as}^* e^{i\frac{\Delta kz}{2}} \quad (30.30)$$

and the complex, conjugate of equation (30.18) as

$$\left(\frac{d}{dz} + \alpha_{as}^* - i\frac{\Delta k}{2}\right) E_{as}^* e^{i\frac{\Delta kz}{2}} = \kappa_{as}^* E_s e^{-i\frac{\Delta kz}{2}} \quad (30.31)$$

we can use equation (30.31) to eliminate E_{as}^* from equation (30.20) and get a second order differential equation instead of two coupled first order equations.

$$\left(\frac{d}{dz} + \alpha_{as}^* - i\frac{\Delta k}{2}\right)\left(\frac{d}{dz} + \alpha_s + i\frac{\Delta k}{2}\right) E_s e^{-i\frac{\Delta kz}{2}} = \kappa_s \kappa_{as}^* E_s e^{-i\frac{\Delta kz}{2}} \quad (30.32)$$

using the trial solution

$$E_s = E_s(0) e^{i\Delta kz/2} e^{gz} \quad (30.33)$$

in equation (30.32), one gets a quadratic equation

$$\left(g + \alpha_{as}^* - i\frac{\Delta k}{2}\right)\left(g + \alpha_s + i\frac{\Delta k}{2}\right) = \kappa_s \kappa_{as}^* \quad (30.34)$$

Here g represents the growth rate.
Equation 30.34 has following solutions.

$$g_{\pm} = -\frac{1}{2}\left(\alpha_s + \alpha_{as}^{*}\right) \pm \frac{1}{2}\left[\left(\alpha_s - \alpha_{as}^{*} + \mathrm{i}\,\frac{\Delta k}{2}\right)^2 + 4\kappa_s\kappa_{as}^{*}\right]^{\frac{1}{2}} \tag{30.35}$$

The field solutions for the signal and idler can be written

$$E_s(z) = \left(E_s^{+}e^{g+z} + E_s^{-}e^{g-z}\right)e^{\frac{\mathrm{i}\Delta kz}{2}} \tag{30.36}$$

and

$$E_{as}^{*}(z) = \left(E_{as}^{+}e^{g+z} + E_{as}^{-}e^{g-z}\right)e^{-\frac{\mathrm{i}\Delta kz}{2}} \tag{30.37}$$

$\Rightarrow$ E_s and E_{as} waves couple together to become two composite modes with gain coefficients g_+ and g_-, respectively. This case is reminiscent of case #4 in Lecture 19. The degree of coupling depends upon $\frac{\Delta k}{\alpha_s}$. The coefficients E_s and E_{as} are determined by the initial conditions. The real parts of gain coefficients correspond to the growth rates and the imaginary parts to the phase change rates.

using equation (30.19) and (30.20)

$$g_{\pm} = -\frac{1}{2}\alpha_s\left(1 - \frac{n_s\omega_{as}}{n_{as}\omega_s}\right) \pm \frac{1}{2}\left[\left\{\alpha_s\left(1 + \frac{n_s\omega_{as}}{n_{as}\omega_s}\right) + \mathrm{i}\Delta k\right\}^2 - 4\alpha_s^2\,\frac{n_s\omega_{as}}{n_{as}\omega_s}\right]^{\frac{1}{2}} \tag{30.38}$$

using $\frac{n_s\omega_{as}}{n_{as}\omega_s} \approx 1$, we get

$$g_{\pm} = \pm\left[\mathrm{i}\,\alpha_s\Delta k - \left(\frac{\Delta k}{2}\right)^2\right]^{\frac{1}{2}} \tag{30.39}$$

It can be seen here that the coupled gain $g_{\pm} = 0$ for $\Delta k = 0$
$\Rightarrow$ under the perfect phase matching condition neither the Stokes nor the anti-Stokes wave will grow because the anti -stokes wave couples so strongly to the stokes wave that the gain for the Stokes wave is cancelled out by the loss of the anti-Stokes wave. The real parts of $g_{\pm}$ are shown in figure 30.3. In this Figure, we can see that if the mismatch is large, the coupling is weak. In this condition, the stokes wave experiences growth and the anti -stokes experiences loss. As the phase mismatch decreases, the gain is suppressed. Significant anti-Stokes generation is expected when $\Delta k \sim \alpha_s$.

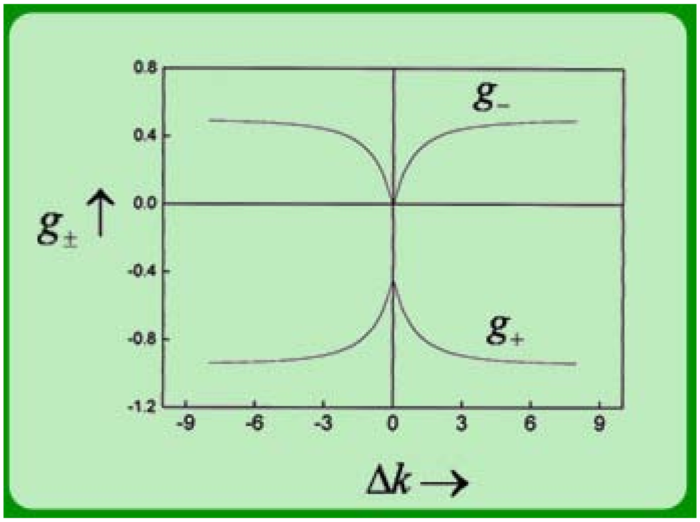

Figure 30.3

In the spontaneous Raman effect anti-Stokes wave is also generated. Do we expect the amplification of an anti-Stokes wave when it propagates through Raman active medium? We will address this question in the next lecture.

Note :
In Raman effect with molecular vibration, a localized molecule changes its state and hence no phase matching is necessary in this case. In case of Raman scattering in crystals, optical phonons which are the collective excitation of the entire system and behave as if they have finite energy and momentum. Conservation of energy and momentum must be obeyed. In particular for all wave vectors parallel

$$\vec{k}_p = \vec{k}_s + \vec{k}_m \qquad (30.40)$$

Then from equations (30.26) and (30.40)

$$\omega_m = \omega_p \left[1 - \frac{n_p}{n_s} \right] \pm \frac{c k_m}{n_s} \qquad (30.41)$$

$\Rightarrow$ By appropriate choice of $\frac{n_p}{n_s}$ in a birefringent crystal values of ω_m and hence ω_s can be changed by appropriately choosing one wave to be ordinary and other one to be extraordinary.

We thus obtain a Stokes wave that can be tuned continuously and amplified by stimulated scattering. It can thus be a method to generate coherent infrared radiation.

Recap

In this lecture you have learnt the following

- The coupling of Stokes and anti-Stokes fields.
- The effect of phase matching on the Raman gain and condition for the efficient generation of anti-Stokes wave. It has been shown that for perfect phase matched Raman gain is completely suppressed and neither of the two fields grow and significant anti-Stokes generation takes place when phase wave vector mismatch is comparable to the Stokes field attenuation factor.