3.0 The Theory of Non Linear Effects in Optical Fibres.

3.1 Introduction.

Various non linear effects has been observed when high intensity light passed through optical fibres or in bulk materials. The non linear effects commonly observed are stimulated Raman scattering (SRS), stimulated Brillouin scattering (SBS), four photon mixing (FPM), second harmonic generation (SHG) and many others. The interaction of a light field with a macroscopic scattering medium containing no magnetisation and no current due to free charges may be described by Maxwell's equation as follows [2]:

$$\nabla \times E = -\frac{1}{c}\frac{\partial H}{\partial t} \qquad (3.1)$$

$$\nabla \times H = \frac{1}{c}\frac{\partial D}{\partial t} \qquad (3.2)$$

$$\nabla \cdot D = 0 \qquad (3.3)$$

$$\nabla \cdot H = 0 \qquad (3.4)$$

The relationship between the electric displacement vector D and the electric field vector E is :

$$D = E + 4\pi P \qquad (3.5)$$

where P is the electric polarisation in the medium. Substituting equation (3.5) into equation (3.2) and eliminating H yields the wave equation:

$$\nabla \times (\nabla \times E) + \frac{1}{c^2}\frac{\partial^2 E}{\partial t^2} = - \frac{4\pi}{c^2}\frac{\partial^2 P}{\partial t^2} \tag{3.6}$$

We can assume that the electric field and the polarisation are described by the summation of plane waves having slowly varying amplitudes i.e. :

$$E(\omega_i) = \frac{1}{2}\left[E(\omega_i)e^{-i\omega_i t} + E(\omega_i)e^{i\omega_i t}\right] \tag{3.7}$$

$$P(\omega_i) = \frac{1}{2}\left[P^L(\omega_i)e^{-i\omega_i t} + P^L(\omega_i)e^{i\omega_i t}\right]$$

$$\frac{1}{2}\left[P^{NL}(\omega_i)e^{-i\omega_i t} + P^{NL}(\omega_i)e^{i\omega_i t}\right] \tag{3.8}$$

(Note : L denotes linear and NL denotes non linear.)

For low to moderate light intensity, the polarisation P is proportional to the electric field E and the proportionality constant is given by the linear susceptibility of the medium, χ

$$P = \chi^1 E \tag{3.9}$$

When the intensity of light become sufficiently intense, the second order and the third order non-linear susceptibility becomes significant, as such the above formula can now be written as follows :

$$P = \chi^1 E + \chi^2 EE + \chi^3 EEE + \ldots\ldots \tag{3.10}$$

The first term describes processes such as direct absorption and spontaneous Raman scattering, the second term is a non-linear term that describes phenomena like second harmonic generation and finally the third term describes processes like third harmonic generation, stimulated Raman scattering and intensity dependent refractive index.

3.2 Raman scattering

Raman scattering is the interaction of light photons with the molecules vibrational or rotational levels. This inelastic interaction leaves the molecule in an excited state, with an annihilation of a pump photon, $h\nu_p$ and an emission of a photon of a lower frequency, which is known as the Stokes frequency $h\nu_s$. The frequency difference between the pump and the Stokes is known as the Raman shift. The Raman shift is independent of the exciting photon because the molecule in state | a >, when excited by a light photon, would first be excited to a virtual state prior to going to the final state | b >. The difference between | a > and | b > is known as the Raman shift.

3.2.1 Spontaneous Raman Scattering

Spontaneous Raman scattering is a two photon scattering process that usually leaves the molecules in an excited vibrational or rotational level. As in all spontaneous scattering, the photon emitted has no coherency with the pump photon. This process is schematically represented in figure 3.1. If the pump is sufficiently intense, it is possible to re-excite the molecule once again from excited state | b > , before the molecule decays to the ground state | a > , to another intermediate level as depicted in figure 3.1b. Thus, it becomes possible for a second line to appear which is shifted to a higher frequency than that of the pump source. This higher frequency is known as the Anti-Stokes lines. Anti-Stokes lines proliferates from Stokes transition as such we would expect to see more Stoke lines than Anti-Stoke lines.

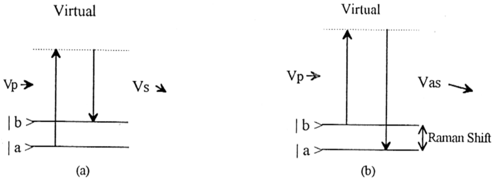

Figure 3.1 **A schematic representation of spontaneous Raman scattering (a) Stokes and (b) Anti-Stokes**

3.2.2 Stimulated Raman Scattering

When the light intensity becomes sufficiently intense, population inversion between the virtual state and the excited state | b > can achieve sufficiently high volume for stimulated Raman scattering to occur. In stimulated Raman scattering, the third order non-linear susceptibility mixes the pump and the Stokes component to produce stimulated Stokes which are coherent to the pump. Taking the cube of equation 3.7 gives:

$$[E(\omega_s)]^3 = \frac{6}{8} E(\omega_s)E(\omega_p)E(\omega_{-p})\, e^{-i(\omega_s + \omega_p - \omega_p)t} + \text{c.c.} \qquad (3.11)$$

Taking the non linear terms in equation (3.8) gives:

$$P(\omega_s) = \tfrac{1}{2} P^{NL}(\omega_s)e^{-i\omega_s t} + \tfrac{1}{2} P^{NL}(\omega_s)e^{i\omega_s t} \qquad (3.12)$$

Equating equation (3.11) and equation (3.12) gives:

$$\tfrac{1}{2}P^{NL}(\omega_s)e^{-i\omega_s t} = \tfrac{6}{8}\varepsilon_o\chi(\omega_s+\omega_p - \omega_p)E(\omega_s)E(\omega_p)E(\omega_{-p})e^{-i(\omega_s+\omega_p-\omega_p)t}$$

Knowing that $\omega_s = \omega_s + \omega_p - \omega_p$. This gives :

$$P^{NL}(\omega_s) = \tfrac{3}{2}\varepsilon_o\chi(\omega_s + \omega_p - \omega_p)E(\omega_s)E(\omega_p)E(\omega_{-p}) \qquad (3.13)$$

Similarly the Anti-Stokes components are also mixed via the third non linear susceptibility to produce coherent Anti-Stokes lines. Expanding the cube of equation (3.7) gives:

$$[E(\omega_{as})]^3 = \tfrac{6}{8} E(\omega_{-s})E(\omega_p)E(\omega_p)e^{-i(-\omega_s+\omega_p+\omega_p)t} + \text{c.c} \qquad (3.14)$$

Taking the non linear terms in equation (3.8) gives:

$$P(\omega_{as}) = \tfrac{1}{2} P^{NL}(\omega_{as})e^{-i\omega_{as}t} + \tfrac{1}{2} P^{NL}(\omega_{as})e^{i\omega_{as}t} \qquad (3.15)$$

Equating equation (3.14) and (3.15) gives:

$$\tfrac{1}{2}P^{NL}(\omega_{as})e^{-i\omega_{as}t} = \tfrac{6}{8}\varepsilon_o\chi(-\omega_s + \omega_p + \omega_p)E(\omega_{-s})E(\omega_p)E(\omega_p)e^{-i(-\omega_s+\omega_p+\omega_p)t}$$

knowing that an anti-Stokes and Stokes photons is created at the annihilation of two pump photon, hence we get $\omega_{as} = -\omega_s + \omega_p + \omega_p$ this gives:

$$P^{NL}(\omega_{as}) = \frac{3}{2}\,\varepsilon_o \chi\,(-\omega_s + \omega_p + \omega_p) E(\omega_{-s}) E(\omega_p) E(\omega_p) \qquad (3.16)$$

Both equation (3.13) and (3.16) represents the stimulated generation of Stokes and Anti-Stokes respectively due to the third order non linear susceptibility.

3.2.2.1 Amplification of Stokes and absorption of Anti-Stokes.

An Anti-Stokes frequency injected together with a pump into a non linear medium will be absorbed where as a Stokes frequency injected along with a pump will be amplified, we per Jones W.J. et al. 1964 [17]. A semi-classical description of the above absorption of the Anti-Stokes lines and amplification of Stokes lines are given in Yariv 1989 [46] and Barrett J.J 1981 [2] respectively.

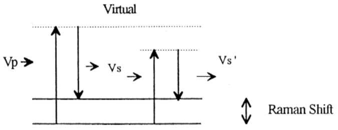

Figure 3.2 **A schematic representation of multiple Stokes generation, shown here is the first order Stokes Vs, pumping the second order Stokes Vs '.**

When the Stokes signal becomes sufficiently intense, this Stokes can in turn act as a new pump source to pump higher order Stokes lines at lower frequencies, this can be schematically represented in figure 3.2. This new second order Stokes $V_{s'}$ will also be Raman shifted from the first order Stokes by 450 cm^{-1} and shifted from the pump by a total of 900 cm^{-1}, each order will be shifted in multiples of 450 cm^{-1} from the pump. When the second order Stokes becomes sufficiently intense, it in turn will become a pump source to pump the third order Stokes and the third will be the pump for the fourth and so on. The relation between the non-linear polarisation and the third order electric field, can be represented by taking the cube of equation (3.7) as follows :

$$E(\omega_{s'}) = \tfrac{6}{8}\, E(\omega_s)E(\omega_s)E(\omega_{-p})e^{-i(\omega_s+\omega_s-\omega_p)t} + c.c. \qquad (3.17)$$

taking the non-linear terms of equation (3.8) gives:

$$P(\omega_{s'}) = \tfrac{1}{2}\, P^{NL}(\omega_{s'})e^{-i\omega_{s'}t} + \tfrac{1}{2}\, P^{NL}(\omega_{s'})e^{i\omega_{s'}t} \qquad (3.18)$$

Equating equation (3.17) and (3.18) gives :

$$\tfrac{1}{2}\, P^{NL}(\omega_{s'})e^{-i\omega_{s'}t} = \tfrac{6}{8}\varepsilon_o\chi(\omega_s+\omega_s-\omega_p)E(\omega_s)E(\omega_s)E(\omega_{-p})e^{-i(\omega_s+\omega_s-\omega_p)t}$$

Given that $\omega_{s'} = 2\omega_s - \omega_p$, this gives :

$$P^{NL}(\omega_{s'}) = \tfrac{3}{2}\,\varepsilon_o\chi\,(\omega_s+\omega_s-\omega_p)\,E(\omega_s)E(\omega_s)E(\omega_{-p}) \qquad (3.19)$$

Equation (3.19) is the relation between non-linear polarisation and the electric field for the generation of the second order Stokes from the first order Stokes. Multiple Stokes generation was observed by experiments carried out by pumping a silica fibre with laser light from a Nd-YAG laser at near infra-red at 1064 nm and at frequency doubled 532 nm. A detail discussion of the experiment result is discussed in section 4.2. Multiple Stokes generation was also observed by Barbosa 1982 [1] up to the 10th order and also by Gabriagues 1989 [11], who observed multiple Stokes generation up to the 6th order. Observation of higher order by Barbosa was made possible by increasing the sensitivity of the receiver by ten times.

3.2.3 Raman Gain Spectrum.

Figure 3.3 shows the Raman gain spectrum for silica dioxide - the basic building block for optical fibre. This Raman gain spectrum was obtained from Stolen R.H. et. al. 1973 [38]. This Raman gain shows distinctive peaks around 450 cm^{-1} and 800 cm^{-1} with a shoulder around 600 cm^{-1}. If a laser light is incident upon this material, the first stimulated Raman scattering line would occur at 450 cm^{-1} away from the incident laser source and the second stimulated Raman scattering line would occur at 800 cm^{-1} away from the incident laser source. A detail experimental results will be given in section 4.2. Table 3.1 shown the Raman shift for common materials given by Gabriagues 1989 [11] and Bloembergen 1967 [3]. Table 3.1 shows that the Raman shifts for fibres is dependent on the relative index difference between the core and the cladding of the fibre.

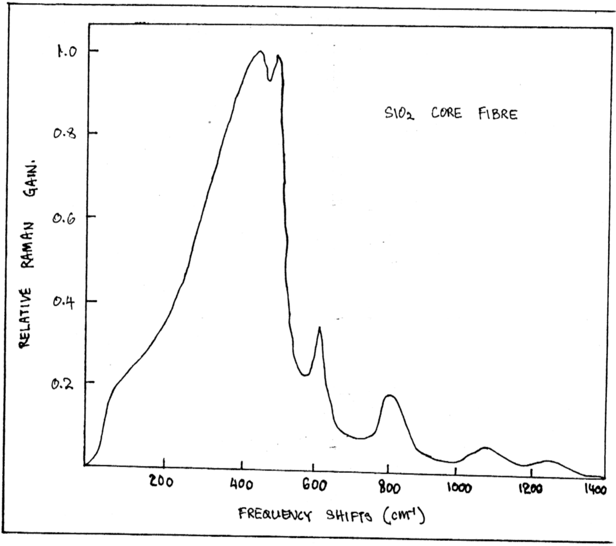

Figure 3.3 Shows the relative Raman Gain vs. frequency shift in silica dioxide.

Materials	Raman shift (cm^{-1})	Remark
SiO_2 (0.9% RID)	420	fibre material
GeO_2 (pure)	424	fibre material
SiO_2 (0.3% RID)	440	fibre material
SiO_2 (1.7% RID)	450	fibre material
SiO_2 (0.37% RID)	450	fibre material
ZBLAN	595	fibre material
Pyridine	992	organic liquid
Toluene	1,004	organic liquid
Calcite	1,075	mineral
$SiO_2 + P_2O_5$ (17% mol)	1,320	fibre material
$SiO_2 + P_2O_5$	1,320	fibre material
Nitrobenzene	1,344	organic liquid
1- Bromonaphthalene	1,368	organic liquid
Cyclohexane	2,852	organic liquid
$SiO_2 + D_2$ (gas in glass)	2,988	fibre material
Benzene	3,064	organic liquid
Hydrogen gas (H_2)	4,155	gas

Table 3.1 **Summary of some published data on Raman shift in common materials. Note that RID is written for the relative index difference between the core and the cladding of the fibre sample.**

3.3 Four Photon Mixing.

Four photon mixing (FPM) or parametric mixing is another non linear effect usually seen when high intensity laser light is coupled into a single mode optical fibre. In comparison to stimulated Raman scattering, four photon mixing offers upconversion as well as downconversion. The range of frequency shifts are broader from a few cm^{-1} up to thousands of cm^{-1}. One drawback of four photon mixing is that this process is not automatically phase match unlike stimulated Raman scattering, and in order to observe four photon mixing, the Stokes and Anti-Stokes photon produced by this process has to be phase matched. There are various was of achieving phase matching and these techniques which will be discussed later on. In a thin sample there is no problem with phase matching but in a thicker sample, there will be some point where the electric field at ω_{as} which will be out of phase with the polarisation produced by mixing $2\omega_p - \omega_s$ so that ω_{as} will be re-absorped. Four photon mixing in a glass fibre absorbs two pump photon and through $\chi_3 E^3$ term in the polarisation expansion, creates one photon higher in frequency, known as the Anti-Stokes wave and one photon lower in frequency, known as the Stokes wave. The third order susceptibility χ_3 is complex, with the real part leading to parametric gain four photon mixing and the imaginary part leading to stimulated Raman scattering. Energy conservation requires that

$$\omega_{as} + \omega_s = 2\,\omega_p \qquad (3.20)$$

In optical fibre, the wavevector mismatch is determined by two factors , the first is the dispersion of the bulk medium of which the guide is made and second is the dispersive characteristic of the wave guide itself. The material and waveguide contribution to Δ k is Δk_m and Δk_w respectively such that, $\Delta k = \Delta k_m + \Delta k_w$. Therefore, phase matching requirement can be written as :

$$\Delta k_m + \Delta k_w = \Delta k = k_s + k_{as} + 2k_p \qquad (3.21)$$

3.3.1 Phase Matching.

Phase matching in silica fibres can be achieved by the following techniques, a brief description on the various techniques and their characteristics are given in table 3.2 , per R.H. Stolen et. al. 1982 [41].

3.3.1.1 Phase matching using fibre modes.

Phase matching can be achieve by having Stokes and Anti-Stokes propagating in different modes in a fibre. Phase matching can be accomplished because the propagation constant for higher order modes is less than that of a lower order mode, Hill K.O. et al 1981, [14]. Phase matching occurs when the pump and Stokes propagates in the lower LP_{01} mode, while the Anti-Stokes propagates in the higher LP_{02} mode.

3.3.1.2 Phase matching using Birefringence.

Phase matching can be accomplished by having the pump propagate in an optical axis and the Stokes and Anti-Stokes propagate in another optical axis that is perpendicular to the axis of the pump waves.

3.3.1.3 Phase matching by divided pump.

Phase matching can be accomplished by dividing the pump between the same two modes as the Stokes and Anti-Stokes, phase matching occurs when both the pump and Anti-Stokes propagates in the LP_{01} mode and the second pump and Stokes propagates in the LP_{02} mode.

3.3.1.4 Phase matching by small frequencies shift.

Phase matching can be accomplished by having both the Stokes and Anti-Stokes generated close to the pump in order of a few cm^{-1} shifts away. The coherence length of the fibre can be quite long if the frequency shift is small, in the region of a few kilometres, [13].

3.3.1.5 Phase matching near the zero dispersion region.

Phase matching can be accomplished by having the pump near the zero dispersion region of the fibre typically near the 1.3 microns or 1.5 microns region. At this region phase matching between the pump, Stokes and Anti-Stokes is achieved spontaneously without the necessity for compensating for material dispersion by using the phase velocity difference between the waveguide modes, [45].

Table 3.2 shows a summary of the frequency shifts and coherent length for all the various phase matching techniques mention earlier.

Phase Matching Techniques	**Frequency shifts (cm^{-1})**	**Coherent length l_c**
Fibre modes	1000 - 2000	10 - 50 cm
Birefringence	100 - 1000	1 - 2 meters
Divided pump	100 - 500	10 meters
Small Frequency shift	1	2 km
Zero dispersion region	1000	6 meters

Table 3.2 **Various phase matching techniques, frequencies shifts and coherent length respectively.**