Nonlinear Optics

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Introduction

The phenomena of nonlinear optics are widely varied, both as to the nature of the nonlinear process and the conditions under which they occur. To give some examples; with high power lasers generation of optical harmonic frequencies up to the 20th harmonic have been demonstrated, so that a 1.06μm laser has generated radiation at ^{53}nm[1]; by exploiting the long interaction length and small core diameter available in an optical fibre a stimulated scattering process (stimulated Brillouin scattering) has been observed with a continuous laser of submilliwatt power output [2]; through the phenomenon of an intensity dependent refractive index in an optical fibre, optical pulses have been compressed temporally to a duration as short as 8fs [3]; by means of nonlinear optical phase-conjugation, lensless imaging can be achieved with submicron resolution [4]. These are just a few examples, which indicate the range of phenomena. The book by Shen gives a comprehensive coverage of the main phenomena [5]. In a short course the subject matter must necessarily cover a limited range and the following topics have been chosen: harmonic generation, frequency mixing, parametric oscillation and amplification; stimulated Brillouin scattering (SBS); stimulated Raman scattering (SRS); self-focussing and self-phase modulation (SPM); phase-conjugation;nonlinear effects in optical fibres, in particular SBS,SRS and SPM. An aim of the course is to give an elementary discussion of each

of the processes, with some typical order of magnitude estimates to indicate efficiency, threshold etc.

Optical Harmonic Generation, frequency mixing,optical parametric oscillation and amplification.

Optical second harmonic generation, first observed in 1961 by Franken et al [6] is considered as the observation which marks the birth of nonlinear optics. The influence of an incident electric field E in a medium is to induce a polarisation P in the medium of the form

$$P = \epsilon_0\chi^{(1)}E + \epsilon_0\chi^{(2)}E^2 + \epsilon_0\chi^{(3)}E^3 + \ldots \qquad (1)$$

The first term gives rise to linear optical phenomena, and the first nonlinear term $\epsilon_0\chi^{(2)}E^2$ is responsible for second harmonic generation and so-called three-wave interactions of the form

$$\omega_1 \pm \omega_2 = \omega_3 \qquad (2)$$

where two waves, of frequency ω_1, ω_2 interact in the nonlinear medium to produce a third wave ω_3, which is either the sum $\omega_1 + \omega_2$ or difference $\omega_1 - \omega_2$ of the two incident waves. Second harmonic generation is a particular case of this where ω_1 and ω_2 are equal and their sum is generated. The nonlinear susceptibility $\chi^{(2)}$ is non-zero only for media which lack a centre of inversion which in practice restricts it to crystal materials. For the three-wave process indicated in (2) to lead to efficient generation of ω_3 it is

necessary for three waves to be phase-matched, ie. to have their wave vectors $\underline{k}_1, \underline{k}_2, \underline{k}_3$, satisfy the following equation

$$\underline{k}_1 \pm \underline{k}_2 = \underline{k}_3 \qquad (3)$$

The effect of dispersion would normally prevent (3) from being satisfied. However in a birefringent crystal, with one or more wave propagating as extraordinary wave and the other as ordinary wave, it is possible, given sufficient birefringence to use the birefringence to offset the dispersion and thus satisfy (3). This need for a large birefringence further restricts the list of possible candidates for efficient nonlinear generation. Other requirements on the medium are good optical quality, good transmission at the frequencies $\omega_1, \omega_2, \omega_3$ and good resistance to optically induced damage. Given this demanding lists of requirements it is not surprising that the list of useful nonlinear crystals is short. Byer [7] tabulates the relevant parameters of some of the best materials. Typically it is found that crystal lengths of the order of 1cm to a few cm are needed, and incident intensities of a few Mw/cm^2 to a few hundred Mw/cm^2 are needed for efficient conversion, whether it be harmonic generation, sum frequency generation, or difference frequency generation. With these techniques, with a limited number of nonlinear crystals such as ADP, KDP, $LiNbO_3$ and $LiIO_3$, dye lasers have had their tuning range extended to cover the range ~200nm to 4μm. In this case the tunability is provided by the dye laser and the tuning capability is then shifted to a different spectral region by mixing with another laser or, in the case of harmonic generation with itself.

An alternative means of producing a tunable output is to use the phenomenon of optical parametric amplification. Consider two waves ω_1, ω_2 mixing to produce their difference ω_3, in a phase-matched interaction

$$\left.\begin{aligned} \omega_1 - \omega_2 &= \omega_3 \\ \underline{k}_1 - \underline{k}_2 &= \underline{k}_3 \end{aligned}\right\} \quad (4)$$

The generated wave ω_3 can then mix with ω_1, to generate ω_2, since (4) can simply be rewritten

$$\left.\begin{aligned} \omega_1 - \omega_3 &= \omega_2 \\ \underline{K}_1 - \underline{k}_3 &= \underline{k}_2 \end{aligned}\right\} \quad (5)$$

So wave ω_3 helps to generate ω_2 and wave ω_2 helps to generate ω_3. Thus both ω_2 and ω_3 are amplified as they pass through the medium, ie. the presence of the high frequency wave ω_1 (the pump) leads to amplification of the low frequency pair of waves (called signal/ idler waves) whose frequencies add to give the pump frequency. Of all possible signal/idler combinations it is the pair for which the phase-matching condition ($\underline{k}_1 + \underline{k}_2 = \underline{k}_3$) is satisfied that has the highest amplification. Given the presence of amplification, and of noise at the signal and idler frequencies, by constructing a resonator which provides feedback, the signal and idler waves can be made to

grow from noise, just as a conventional laser resonator allows growth from spontaneous emission. Such a device is known as an optical parametric oscillator (OPO). The nonlinear medium can also be used simply as an amplifier (for an injected signal) without the need for mirrors - in this case it is known as an optical parametric amplifier (OPA). Where sufficient gain is available on a single pass through the medium, the OPA can give a superradiant output in which noise is amplified rather than an injected signal.

The operating characteristics of optical parametric oscillators has been discussed at length by Byer [7]. Relatively few materials have been able to operate successfully since, unlike harmonic generation and difference frequency generation, it is necessary to reach a threshold before any significant generationoccurs and the intensity required for this is uncomfortably close to the material damage threshold. However $LiNbO_3$, pumped by a 1.06μm NdYAG laser has proved a successful OPO giving tuning of the signal and idler output from ~1.4μm to 4μm [8]. Typically however the pump power required for this is several megawatts. More recently a visible parametric oscillator based on urea, a recently developed nonlinear crystal, shows promise for wide tunability in the visible region of the spectrum. Such a device could provide a serious competitor to dye lasers, offering advantages of an extremely wide tuning range without the degradation problems exhibited by dyes. While OPO's pumped by pulses from Q-switched lasers (pulse durations from ~10ns to few 10's of ns) have had their performance constrained by optical damage, the situation is rather different for mode-locked (ie. subnanosecond) pump pulses.

Much higher intensities can be withstood by the crystal, thus much higher parametric gains are possible. Single pass gains of $\sim e^{30}$ are readily achieved, sufficient to allow amplification from noise to a level comparable to the pump intensity itself. This provides an attractive way of generating short pulses, eg. on the picosecond scale, over wide tuning ranges in the visible and near infrared. Recent results from this approach are reported by Kaiser and coworkers [9,10,11] and include the successful use of infrared materials such as $AgGaS_2$ despite their disappointingly low damage thresholds when used with long pulses.

Three wave interactions, with their requirement of a crystalline nonlinear medium, immediately impose a limit on the maximum frequency that can be generated as a consequence of the UV absorption shown by solids. This limitation can be overcome using a gaseous medium, but then $\chi^{(2)}$ is zero and the next higher order susceptibility $\chi^{(3)}$ must be used. This susceptibility is responsible for four wave mixing processes such as

$$\omega_1 \pm \omega_2 \pm \omega_3 = \omega_4 \qquad (6)$$

for which the phase-matching condition is

$$k_1 \pm k_2 \pm k_3 = k_4$$

Details of the results from such four wave mixing experiments are found in [5, 12, 13]. Requirements differ from three-wave mixing in

two main ways. First, the susceptibility is generally lower, so that either much higher powers must be used, or the laser frequencies are tuned to resonances (often two-photon) of the medium to resonantly enhance the susceptibility. Second, the phase-matching relies on anomalous dispersion effects (one wave above a resonance frequency), either in the nonlinear medium itself or a buffer gas added to the medium.

Stimulated Raman Scattering

The physical process responsible for stimulated Raman scattering is an oscillating polarisability induced in the nonlinear medium. An example of this is provided by a molecular gas (H_2 gas is a commonly used Raman medium) in which molecular vibration provides the oscillating polarisability. The relationship between the polarisation P of the medium, and an applied optical field E is,

$$P = \alpha E \qquad (7)$$

where α is the polarisability. To account for the oscillating polarisability we can write $\alpha = \alpha_0 + \alpha_1 \mathrm{Cos}\omega_R t$ where ω_R is the vibration frequency. If the incident field has the form $E\mathrm{Cos}\omega_p t$ it can be seen that P has oscillating components at both the Stokes frequency $\omega_s = \omega_p - \omega_R$ and the anti-Stokes frequency $\omega_{as} = \omega_p + \omega_R$. Such radiation is generated as spontaneous scattered light. If however the molecule is now subjected to incident fields at both ω_p and ω_s, the molecular vibration is strongly driven at frequency ω_p

$-\omega_s$, since this is resonant with the vibration frequency. Thus a positive feedback situation exists in which Stokes radiation together with pump radiation drive the vibration,and pump radiation scattered by the vibration generates more Stokes radiation, which drives the vibration more strongly, and so on. Thus starting from noise, ie. spontaneously generated Stokes radiation, a strong Stokes beam can build up, with power comparable to that of the pump itself. Under steady state conditions the intensity of a Stokes wave as it propagates through the Raman medium, collinear with the pump wave, is given by

$$I_s(z) = I_s(o)\exp(g_R I_p z) \qquad (8)$$

The distance z is the length travelled in the medium, I_p is the pump intensity, g_R is the Raman gain coefficient and $I_s(o)$ is the noise intensity at the pump input end of the medium. When the exponent $g_R I_p z$ reaches a value of ~20-30 the Stokes intensity can be comparable to the pump intensity. This defines the threshold condition, ie. that

$$g_R I_p \ell \simeq 25 \qquad (9)$$

where ℓ is the length of the Raman medium.

Values of g_R vary rather widely, but we quote here values appropriate to the media that have proved important as Raman media; for H_2 gas g_R has the value $2.4 \times 10^{-11}/\lambda_s(\mu m)$ m/W where λs is the Stokes wavelength

in micrometers; for fused silica g_R has the value 1×10^{-13}m/w for a pump wavelength of 1.06μm (Stokes wavelength ~1.1μm). The much smaller g_R for fused silica does not prevent a low threshold from being achieved when the medium in the form of an optical fibre since a very large value of $I_p\ell$ can be achieved even for a low pump power.

A rough estimate of the threshold pump power is now made for the case of H_2. First we note that if a diffraction limited pump beam is used, the product of I_p and length ℓ is at best $\sim P_p/\lambda_p$ where P_p is the pump power. This follows from the argument that a pump beam focussed to a waist of spot size w_0, hence area πw_0^2 and intensity $P_p/\pi w_0^2$, has essentially constant value over the length of a confocal parameter, $2\pi w_0^2/\lambda$, but diminishes rapidly at greater distances from the waist. Thus $I_p\ell \sim P_p/\lambda_p$. Using this result, the relation (9) and the quoted value for g_R gives a threshold pump power of the order of megawatts for a 1.06μm pump (λ_s~1.9μm since the Raman shift ω_R corresponds to 4155cm^{-1}). For a non-diffraction limited beam (having a divergence M times greater than for the diffraction limit) the threshold is M times greater. The importance of high quality laser beams is emphasised by this result. A threshold pump power of megawatts is readily achieved by typical pulsed dye lasers, thus offering a simple technique for extending the tuning range of a dye. In fact the tuning range is greatly extended by virtue of the fact that multiple Stokes shifts (and antiStokes shifts) can occur once the threshold for first Stokes generation has been reached. The generated frequencies, are ω_{s1}, $\omega_{s2}, \ldots \omega_{sn}$ where

$$\omega_{sn} = \omega_p - n\omega_R \tag{10}$$

and $$\omega_{asn} = \omega_p + n\omega_R \tag{11}$$

The origin of these additional frequency shifts is a four-wave interaction involving initially just ω_p and ω_{s1}, for example

$$\omega_p + \omega_p - \omega_{s1} = \omega_{as1} \tag{12}$$

$$\omega_{s1} - \omega_p + \omega_{s1} = \omega_{s2} \tag{13}$$

and so on.

These processes require the corresponding phase-matching condition to be satisfied, eg.

$$\underline{k}_p + \underline{k}_p - \underline{k}_{s1} = \underline{k}_{as1} \tag{14}$$

whereas the initial generation of first Stokes radiation is automatically phase-matched.

Given these additional frequency shifts, and given the large shift available in H_2 ($4155cm^{-1}$, the highest molecular vibrational frequency), extremely wide tuning ranges can be achieved simply by focussing the pump light into a high pressure hydrogen cell. Wavelengths ranging from 140nm to 20μm have been generated in H_2. Given the rather high threshold for SRS in H_2 when the pump beam is

simply focussed in the gas, it is worth noting that a threshold reduction of at least an order of magnitude can be achieved by confining the pump beam and the H_2 gas in a hollow glass capillary waveguide.[13, 14]

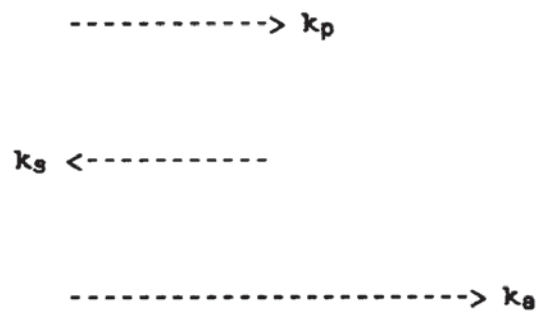
Stimulated Brillouin Scattering

An incident light wave of frequency ω_p (the 'Pump') is scattered by an acoustic wave. This scattered light wave is Doppler shifted to some frequency ω_s since the acoustic wave acts as a moving diffraction grating. The scattered light and incident light together form a 'standing wave' (actually the 'standing wave' moves slowly since the two light waves are not of the same frequency). The 'standing wave' leads, via electrostriction, to an increase in density at the antinodes and a decrease at the nodes. In fact this spatially periodic, moving structure of alternate high and low density coincides with the acoustic wave from which the initial scattering started, so a positive feedback situation exists which can start from the acoustic waves present in the medium due simply to thermal exitation. The incident light wave produces a scattered wave which via the 'standing wave' formed then drives the acoustic wave - hence stronger scattering occurs and so on. The process occurs most strongly for a backward scattered light wave and forward travelling acoustic wave. The frequencies and wave vectors are k_p, ω_p (pump), k_s, ω_s (the scattered wave or Stokes wave) and k_a,ω_a (acoustic wave) and satisfy

$$\omega_p = \omega_s + \omega_a \qquad (15)$$

$$\underline{k}_p = \underline{k}_s + \underline{k}_a \qquad (16)$$

where $\underline{k}_p$, $\underline{k}_s$ and $\underline{k}_a$ are as shown below



The frequency shift ω_a is given by

$$\omega_a = 2\pi\nu_a = \frac{4\pi n v a}{\lambda_p} \qquad (17)$$

where v_a is the acoustic velocity, n is the refractive index and λ_p the pump wavelength. Taking the values for liquid CS_2 [5]; the frequency shift is ~6GHz, for a pump wavelength of ~600nm and the steady Brillouin gain coefficent g_B ~10^{-9}m/W. The Stokes wave intensity $I_s(Z)$ under steady state conditions obeys a relation similar to that for SRS (equation (8)):

$$I_s(z) = I_s(o) \exp (g_B I_p z) \qquad (18)$$

where z is now measured in the opposite direction to the direction of pump propagation. The SBS threshold in CS_2 is some two orders of magnitude less than the SRS threshold in H_2, ie. ~10kW, since g_B is ~two orders greater than g_R. Since the Stokes shift is so small

(~four orders less than the Raman shift in H_2), SBS does not find application like SRS as a means of frequency shifting. In many respects SBS as a process has nuisance value and methods for suppressing are adopted. One such method is to use a broad bandwidth pump, $\Delta\nu_p \gg \Delta\nu_B$ where $\Delta\nu_B$ is the Spontaneous Brillouin linewidth, since the value of g_B is thereby reduced by a factor $\frac{\Delta\nu_B}{\Delta\nu_p}$. Hence the SBS threshold (which also requires the condition $g_B I_p z \sim 25$) is increased by the factor $\frac{\Delta\nu_p}{\Delta\nu_B}$. Stimulated Brillouin scattering does however find use in that it gives a particularly simple means of achieving phase-conjugation.

Phase-conjugation

Phase-conjugation is defined as the process in which the phase of the output wave is complex conjugate to the phase of the input wave. If we express the input wave as

$$E(\underline{R},t) = 1/2\ [E(\underline{R})\exp[-i\omega t]+c.c] \tag{19}$$

where $$E(\underline{R}) = \left|E(\underline{R})\right|\exp[i\Phi(R)] \tag{20}$$

then the phase conjugate wave $E_{conj}(\underline{R},t)$ is

$$E_{conj}(\underline{R},t) = 1/2\ [E^*(\underline{R})\exp[-i\omega t]+cc] \tag{21}$$

This phase conjugate wave is the exact reversal of the input wave. If

such a wave is generated it has the particularly interesting property of reversing, or 'undoing' any aberrations that were imposed on the input beam as the phase conjugate beam returns through the aberrator. This has numerous applications [15,16]; one in particular is the possibility of amplifying a laser beam without aberration by passing the beam through the amplifier, then generating the phase conjugate beam and passing it back through the amplifier, whereupon it emerges amplified but with the aberrations caused by the first pass completely removed on the second pass. The phase conjugate beam can be generated by a number of nonlinear optical processes, including degenerate four-wave mixing and SBS (In fact SBS only approximates to a phase-conjugate - clearly it cannot be exact since the back scattered Stokes wave is not quite the same frequency as the input (pump) wave.

Degenerate four-wave mixing is a special case of four-wave mixing where all four frequencies are the same. If we consider the form taken by the polarisation $P = \varepsilon_0 \chi^{(3)} E^3$ where the three fields are of the form, $E_1 \exp i(\omega t - kz)$, $E_2 \exp i(\omega t + kz)$ ie. counterpropagating plane waves in the +z and-z direction, and a third, input wave $E_3 \exp i(\omega t - \underline{k}.\underline{r})$, then one contribution to a polarisation P at frequency ω will be of the form

$$P = \varepsilon_0 \chi^{(3)} E_1 E_2 E_3 \exp i[(\omega t - kz) + (\omega t + kz) - (\omega t - \underline{k}.\underline{r})]$$

$$= \varepsilon_0 \chi^{(3)} E_1 E_2 E_3 \exp i(\omega t + \underline{k}.\underline{r}), \qquad (22)$$

and thus polarisation will therefore radiate a wave which is the phase

conjugate of wave3. Thus the output wave is the phase conjugate of the input wave. The requirement for this experimental arrangement is then simply two counterpropagating plane pump waves and an input wave which overlaps with these waves in a nonlinear medium which displays an intensity dependent refractive index (since this is the physical meaning of the $\chi^{(3)}$ nonlinearity giving rise to a polarisation at the frequency ω of the three incident waves.) An intensity dependent refractive index can arise from a number of physical causes - orientation effects in liquids, saturation effects, thermal effects, photo- refractive effects, electrostrictive effects, and all of these have been used to demonstrate phase-conjugation. In addition SBS and SRS show approximate phase-conjugation, with extremely good quality phase-conjugation being achieved in practice via SBS [see [16]]. A simple explanation of the phase-conjugation behaviour arising from SBS can be given as follows: for the highest gain in the SBS process to be achieved the three interacting waves, pump, Stokes, and acoustic wave must have identical shape of wavefront so that the maximum interaction between the waves occurs. Since the Stokes wave is backward travelling and yet has the same wavefront shape as the forward travelling input wave (pump) it follows that, apart from the fact that there is a small frequency difference between the pump and Stokes wave, the Stokes wave is the phase conjugate of the pump wave. The simplicity of phase-conjugation via SBS is very attractive since it merely involves focussing the pump beam into the Brillouin medium. However sufficient pump power must be available to reach threshold and this may be anywhere between a few tens of kilowatts to a few megawatts. Degenerate four wave mixing generally involves a more

complicated experimental set up but on the other hand can operate at much lower power levels eg. a few milliwatts in photorefractive materials [17]

Nonlinear optical effects in optical fibres

The threshold condition for SBS and SRS, namely $g_B I_p \ell > \sim 25$, and $g_R I_p \ell > \sim 25$ can be achieved for very low powers in optical fibres as a result of the small core area (a few micrometers diameter for a single mode fibre) and the large length ℓ available. The manifestation of nonlinear effects in fibres has other significant differences from results in bulk media (ie. an unguided geometry). Thus dispersion can play an important role since it can become significant over the long interaction length available and the guided geometry allows nonlinear response to behave in an ideal 'plane-wave' fashion, eg. with the entire fibre mode subject to the same self phase modulation despite the fact that the mode has a nonuniform intensity profile and hence a spatially nonuniform response of the nonlinear medium.
Some general references on nonlinear optical effects in fibres are given in 18-21.

For SBS in a fused silica fibre the relevant physical parameters for a pump wavelength of 1.55μm (a region of interest for optical communication) are $v_a = 5960 ms^{-1}$, $n = 1.44$ and hence from (17), $\nu_a =$ 11.1GHz.[22]. The Brillouin gain coefficient g_B ia given by

$$g_B = \frac{2\pi n^7 p^2}{c\lambda^2 \rho v_a \Delta\nu_B} \qquad (23)$$

where n is the refractive index, p is the longitudinal elasto optic

coefficient ρ is the medium density, $\Delta\nu_B$ is the linewidth (FWHM) for spontaneous Brillouin scattering. Given g_B, and the length of fibre, allows the threshold pump intensity I_p (and hence power, knowing the fibre core dimensions) to be calculated from $g_B I_p \ell \gtrsim 25$. $\Delta\nu_B$ varies as λ_p^{-2} [23], with $\Delta\nu_B$ having the value 38.4MHz for λ_p = 1.0μm. g_B is thus independent of λ_p and the threshold pump intensity is likewise independent of pump wavelength. The value of g_B for fused silica is 4.5×10^{-11} m/w. This figure implies very low threshold powers, in the milliwatt region if single mode fibres (ie.having core diameters of a few μm) of a few kilometer length are used. Experiments confirm this [22,24]. To achieve such a low threshold requires that the scattering process reach steady state conditions, ie. that the linewidth $\Delta\nu_p$ of the pump laser be less than $\Delta\nu_p$. If $\Delta\nu_p \gg \Delta\nu_B$ then the gain is reduced relative to the steady state value by the factor $\Delta\nu_B/\Delta\nu_p$. This is equivalent to replacing $\Delta\nu_B$ in (23) by $\Delta\nu_p$.

This feature provides a means for suppressing SBS, for example by using short pump pulses which consequently have a large bandwidth $\Delta\nu_p$. Thus, although in the steady state, the SBS threshold is lower than the stimulated Raman scattering (SRS) threshold, by using short pump pulses , of a few nanoseconds or less, the SBS threshold can be increased to a level above the SRS threshold, which still shows a steady state response even for short pulses.

Finally we note that extremely low SBS thresholds can be achieved in a low loss resonator configuration. Stokes et al [25] resonated both the pump wave (from a 633 nm He-Ne laser) and the Stokes wave in an

all-fibre ring resonator, and achieved a threshold pump power of just 0.56mw.

Stimulated Raman scattering in optical fibres has the threshold condition $g_R I_p \ell > \sim 25$. The peak value of g_R in fused silica for a 1μm pump wavelength, λ_p, is 1 x 10^{-13}m/w with a λ_p^{-1} dependence. (Strictly the wavelength dependence is λ_s^{-1} where λ_s is the Stokes wavelength, but since the frequency shift Ω_R is much less than ω_p typically, a λ_p^{-1} dependence is a good approximation). The shift ω_R corresponds to $\sim 450 cm^{-1}$, but the Raman spectrum extends from zero shift to $\sim 1000 cm^{-1}$ with a full width half maximum of $\sim 250 cm^{-1}$[18]. Different glasses have somewhat different linewidths and frequency shifts, but generally within a factor of two of the values quoted here. The Raman gain coefficient is thus less than the steady state Brillouin gain coefficient by more than an order of magnitude. So although in principle the Raman threshold would be easily reached with cw pump powers of ~1w or less, in practice SBS threshold may be reached first if the pump linewidth is narrow. This can suppress SRS. To avoid this situation a broad band pump may be used since for pump pulses of spectral bandwidth $\Delta\nu_p \gg \Delta\nu_B$, the steady state Brillouin gain is reduced by a factor $\Delta\nu_B/\Delta\nu_p$ whereas SRS gain still corresponds to the steady state value.

One way to provide a broad bandwidth pump is to use short pump pulses, and given that $\Delta\nu_B \sim 38$MHz at 1.06μm it can be seen that for $\Delta\nu_B/\Delta\nu_p \ll 10$ one requires pump pulses of less than ~1nsec. With short pulses

the effects of group velocity dispersion in the fibre become significant, the interaction length in the fibre is limited by the mismatch between pump and Stokes group velocities, leading to the pump and Stokes pulses becoming separated after a sufficient length of fibre. The difference δt in the times taken for pump and Stokes pulses to travel a length L of fibre is

$$\delta t = \frac{L}{c} D \left(\frac{\lambda_p - \lambda_s}{\lambda} \right) \qquad (24)$$

where $D = \lambda^2 d^2 n/d\lambda^2$ is the material dispersion.

Given a pump pulse of duration δt, equation (24) yields the maximum length L of fibre over which the pump and Stokes pulses remain overlapping. Typical values for the visible spectrum [26], where D = 0.08 for λ = 500nm, yield a length of 100m for a ~1nsec pulse.

An interesting experimental situation where dispersion effects have been eliminated is reported by Chraplyvy et al [27] who observed SRS from D_2 gas dissolved in a silica fibre. here the Raman shift from a 1.06μm pump gave a large wavelength shift to ~1.56μm (~3000cm^{-1} shift). By careful choice of the fibre parameters however it was possible to arrange that the group velocity passed through a minimum value between the pump and Stokes wavelengths and thus gave essentially matched group velocities for λ_p and λ_s.

Numerous examples can be given of the use of SRS in fibre to generate a wide range of near infrared wavelengths by multiple Stokes scattering. Here the first Stokes wave can act as pump for a second

scattering process and so on producing as many as ten shifts of ~400cm^{-1} each. References to this application of SRS are found in [28].

Four-wave parametric mixing in fibres involves the generation of an wave at frequency ω_4, from sums and differences of three frequencies, $\omega_1, \omega_2, \omega_3$.

ie. $\omega_1 \pm \omega_2 \pm \omega_3 = \omega_4$ (25)

The nonlinear susceptibility responsible for such a process is $\chi^{(3)}$ and has its origin in a nonlinear response of the electrons to an applied field. $\chi^{(3)}$, unlike $\chi^{(2)}$, can be non-zero in a centrosymmetric medium such as glass. $\chi^{(2)}$ the nonlinear susceptibility, which gives rise to second harmonic generation in noncentrosymmetric media, such as the glass of the media, is expected to be zero in glass. (However recent experiments in fibres have shown that second harmonic generation can occur [29,30]; the explanation for this observation is tentative but it could arise from a quadrupole interaction [11] rather than a dipole interaction or it could rise from anisotropy in the glass as a result of strain[30]).

The process indicated in (25) has been given various names - four-wave parametric mixing (three waves interact to generate a fourth), four-photon mixing, stimulated four-photon mixing and, rather misleadingly, three-wave sum frequency mixing. A common manifestation of the process indicated in (25) is with just one frequency ω_p input

to the medium, ($\omega_1 = \omega_2 = \omega_p$ in (25), and with the frequencies generated, one lower than ω_p (commonly called the Stokes wave, ω_s), and one an equal amount higher than ω_p (called the anti Stokes wave ω_{as})

$$\omega_p + \omega_p = \omega_s + \omega_{as} \qquad (26)$$

The Stokes and anti Stokes waves are not provided as input but undergo parametric amplification from noise. The Stokes and anti Stokes output powers can approach that of the pump, indicating that very large parametric amplification ($\sim e^{25}$) can readily occur. In fact the parametric gain is typically comparable to, and usually somewhat greater than the Raman gain [31].

For the four-wave mixing process to lead to efficient generation of new frequencies it is necessary for the process to be phase-matched, ie. that the propagation constants for the waves β_1 etc. satisfy

$$\beta_1 \pm \beta_2 \pm \beta_3 = \beta_4 \qquad (27)$$

Generally, dispersion of the medium would preclude the possibility of this exact phase-matching, however by allowing different frequencies to propagate in different modes, the mode dispersion can be used to offset the material dispersion. A detailed discussion of this is given by Stolen [32] and Stolen and Bjorkholm [31]. Phase matching has been achieved for large Stokes shifts, up to $\sim 4000 cm^{-1}$ [32], which is an order of magnitude greater than the shift produced by SRS.

A disadvantage of this technique for phase-matching (compared with phase-matching in bulk media) is that the generated radiation may not be in the lowest order mode and that the phase-matching condition cannot be easily tuned, thus the generated frequencies are essentially fixed for a given fibre and pump wavelength . With the use of birefringent fibre the birefringence can be used to provide some compensation for the dispersion, as commonly used in harmonic generation in crystals. In this way phase-matching has been achieved in a monomode fibre [34] by allowing different waves to have different polarisations. This has the advantage that the generated radiation can have an ideal mode configuration, with the output from the fibre being close to a TEM_{00} profile. By additionally applying stress to a birefringent fibre to modify the birefringence it has been shown that a useful degree of tuning of the generated frequency can be achieved [35].

Self phase modulation

The refractive index of any medium reveals a dependence on the intensity of the incident light when sufficiently high intensities are involved. Usually it is sufficient to consider only the first nonlinear term, so the refractive index n is written as

$$n = n_0 + n_2 \langle \underline{E}. \underline{E} \rangle \qquad (28)$$

Where $\langle \underline{E}.\underline{E} \rangle$ indicates a time average and $\underline{E}$ is the applied optical field. n_2 is referred to as the nonlinear index. Another name for

this intensity dependent index is the optical Kerr effect. For a linearly polarised wave $\langle \underline{E}.\underline{E} \rangle$ is 1/2 E^2 where E is the peak field amplitude and this is related to the intensity I by

$$I = \epsilon_0 cn \langle \underline{E}.\underline{E} \rangle . \qquad (29)$$

The value of n_2 (in MKS units) for fused silica is $\sim 10^{-22}$ m^2/v^2. Values of n_2 are frequently quoted in e.s.u., the conversion factor between MKS and e.s.u. being 9 x 10^8, so that for fused silica, n_2 $\sim 1.10^{-13}$ e.s.u. Significantly higher values for n_2 are found in other glasses [36], up to ~40 times greater in SF59. The origin of n_2 in glasses for phenomena on a rapid time scale (say less than nanoseconds) is mainly the nonlinear response of the electronic motion. On longer time scales electrostriction can play a significant role and for liquids effects due to molecular reorientation can dominate in n_2, even down to picosecond time scales.

One of the most important manifestations of n_2 in bulk media is the phenomenon of self-focussing. Self focussing effects are less important in fibres, although they could be significant in multimode fibres. Self phase-modulation is an important consequence of n_2 which shows itself readily in fibres. It refers to the fact that an optical wave as it propagates through a medium obeying (28) causes a change of index and hence of its own phase relative to a wave for which E is vanishingly small. It results in a sweep of frequency being imposed on a pulse as a consequence of the time varying intensity and hence time varying refractive index. Some idea of the magnitude of such an

effect can be obtained by considering a pulse of duration T and peak intensity I, for which the maximum rate of change of n is from (28) and (29), $n_2 I/(\varepsilon_0 cnT)$ and hence the frequency shift is $2\pi n_2 I\ell/(\varepsilon_0 \lambda cnT)$ after traversing a length ℓ of medium. A significant frequency shift would be of the same order as the spectral width of the pulse itself, ie. $\sim\frac{1}{T}$ assuming a transform limited pulse. So the intensity that would cause such a shift is $\lambda\varepsilon_0 cn/(2\pi n_2 \ell)$ and for a 1km length of fibre of 5μm core diameter this implies a power of only 100mW at 1μm wavelength. This estimate illustrates the cumulatively large effect that a small change in index can have over a length of fibre.

When long lengths of fibre are involved it is necessary to consider the simultaneous effect of dispersion and self-phase modulation. The effects are quite different depending on whether normal (positive) dispersion applies or amomalous (negative) dispersion. A simple notion of the effect of dispersion can be seen by considering the sign of frequency change occurring in a pulse. At the leading edge of the pulse, I is increasing and since n_2 is positive (generally) it follows that the frequency is decreased, ie. the leading edge of the pulse becomes redder. The trailing edge becomes bluer. With normal dispersion the red light travels faster than blue, so the pulse spreads. This continues progressively as the leading edge continues to get redder. The result is a long pulse with an almost linear chirp of frequency from low to high frequency. By passing such a temporally and frequency broadened pulse through a negatively dispersive optical system the blue light can be made to catch up with the red light and thus compress the pulse. Compression to a duration equal to the reciprocal of the spectral width of the chirped pulse can be achieved

in this way. This technique has attracted considerable interest in recent years [37-41] and has been developed to the point of a commercially available compressor [42]. The negatively dispersive medium used to compress the chirped pulse is usually a pair of diffraction gratings [43]. Compressions of a factor of 100 are readily achieved and the shortest pulse duration of ~8 femtoseconds has been achieved in this way [44].

By contrast a negatively dispersive fibre leads on it own to pulse compression as the reddened leading edge of the pulse travels more slowly than the bluer trailing edge. Soliton pulses can form under appropriate conditions and these, along with the so-called Soliton laser are a very active area of current research [45-48].

Other uses of the intensity dependent refractive index have also been suggested. One example relies on the fact the polarisation state of radiation in a birefringent fibre will, as a result of n_2, be dependent on the intensity. This offers the possibility of intensity discrimination [49] and optical pulse reshaping [50] by allowing the change of polarisation state to be converted, via transmission through a polariser, into amplitude change.

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