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Devices Based on Second-Order Non-Linear Effects in Glass Fibres

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Abstract

Techniques for the generation of second-order non-linearities in optical fibres are described. Applications to non-linear frequency-optic modulation via the Pockels effect is demonstrated.

Introduction

Photodarkening or polarisation effects in glasses have been known long before the advent of non-linear optics. They have been attributed glass defects and have been the subject of intensive investigations with the object to produce radiation hardened[1] and UV-transmitting[2] fibres. Not until Hill et al[3] demonstrated that refractive index gratings may be written into optical fibres the potential for applications of glass defects to optical fibre devices such as narrow-band filters or reflectors was realised. Until recently, however, it was assumed that second-order non-linear phenomena are not observable in glasses. This was concluded from their inversion symmetry, which does not allow the presence of a second-order non-linear susceptibility and leaves the third-order susceptibility as the non-linear coefficient of lowest order[4].

It was therefore that considerable interest was generated by the discovery of efficient second-harmonic generation in optical fibres by Osterberg and Margulis[5]. In this, high intensity infra-red light is launched into and optical fibre over a period of several hours, which leads to the gradual growth of a second-order non-linearity $\chi^{(2)}$ and the generation of frequency-doubled light. Phase-matching between the fundamental and SH-wave is then achieved via a spatial modulation of $\chi^{(2)}$ of the correct period as postulated by Farries et al[6]. Stolen and Tom[7] attributed the generation of the non-linearity of a defect poling mechanism self-induced by third-order optical rectification between the fundamental and the SH-wave. Recently Fermann et al[8] have observed that a weak second-order non-linearity may also be induced by launching only high-intensity blue light into a fibre. Optical fibre poling with applied external dc-electric fields was first demonstrated by Bergot et al[9] by the simultaneous excitation and orientation of defect centres. In this, high intensity blue light was employed for defect excitation and the poling fields were applied in special fibres with internal electrodes developed by Li et al[10]. Li and Payne[11] have demonstrated that strong electric fields on their own are sufficient to create a Pockels effect in fibres, but without an electronic contribution to the second-order non-linearity[12]. The potential of externally poled fibres to efficient non-linear frequency mixing was demonstrated by Fermann et al[13] by employing mode interference gratings (MIGs) for phase-matching. MIGs allow a good exploitation of the second-order non-linearity and long interaction lengths for the non-linear process.

So far a full explanation of the mechanisms behind the photorefractive and poling effects has not been given. However, two models exist for the material processes behind the poling mechanism. The first model attributes the induced dipole non-linearity to an ordered trapping of holes at Ge E′ centres generated by two-photon absorption induced breaking of Ge-Ge or Ge-Si bonds[14]. The second model assumes that GeO molecules exist interstitially in the glass matrix, which contract when exited (by single or multi-photon absorption) and orient under the influence of a strong poling field[15]. Recently Payne[16] has attributed the related generation of index changes to frozen-in space charge fields generated by diffusion of electrons (excited and retrapped at defect sites) away from points of strong optical illumination.

Here we present a detailed description of the poling technique, where we emphasize the aspect of frequency-doubling and the Pockels effect. We analyse the merits of several phase-matching techniques and measurements of second-order non-linearities are developed as the result of the above analysis. Finally, measurements of poling dynamics are presented and limiting mechanisms are elucidated.

Fibre Designs and Fabrication

For the generation of self-written[5,6] or seeded $\chi^{(2)}$-gratings[7] (i.e. written by interaction between a pump-wave and its SH) standard telecommunications fibres may be used, where the highest conversion efficiencies, i.e. 13% with a pump power of 900W[17], were obtained in fibres with a high GeO_2 doping level and small additions of P_2O_5. The efficient poling of fibres requires dc-electric fields > 100V/um, which are conveniently applied when the fibres comprise internal electrodes. Two designs may be used (Figure 1). Both preform designs typically produce elliptical fibre cores after pulling, where the major axis is aligned perpendicular to the holes. The preforms may be produced by drilling one or two holes along the core axis. The d-shaped is formed by removing one preform side by grinding. Fibre pulling is conventional. Twin-hole fibres allow the application of the strongest electric fields. Recently fields up to 800V/um could be applied in such fibres without the occurance of breakdown[18].

Generation of Non-Linearities

In the following we summarise the response of optical fibres to defect excitation light, a poling field and excitation poling. Defect excitation light on its own leads to induced losses[2] and refractive index changes[3,19]. Resonant defect excitation occurs at a wavelength of 480nm via two photon absorption into the 240nm absorption band of germanosilicate glass[2]. A similar resonance was observed for the small $\chi^{(2)}$ $(2\Omega=\Omega+\Omega)$ permanently induced by defect excitation along[8,12]. Figure 2 shows the dependence of the $\chi^{(2)}$ $(2\Omega=\Omega+\Omega)$ on defect excitation wavelength. In this a number of fibre samples (GeO_2 concentration = 18 mole%) was exposed to pulsed blue light (1.7GW/cm^2 peak intensity and 250W/cm^2 average intensity) for a time period of 5 minutes each and the permanently induced $\chi^{(2)}$ $(2\Omega=\Omega+\Omega)$ was measured by SHG.

A poling field on its own induces a permanent Pockels effect (a non-linearity of the form $\chi^{(2)}$ $(2\Omega=\Omega+\Omega)$. The induced Pockels coefficient as a function of poling field is shown in Figure 3. In this the fibre was exposed to a strong poling field of a given value for a time period of 10 minutes and subsequently the Pockels coefficient was measured via electrooptic modulation with a weak field. No permanent $\chi^{(2)}$ $(2\Omega=\Omega+\Omega)$ can be induced by using a poling field on its own. A small semi-permanent $\chi^{(2)}$ $(2\Omega=\Omega+\Omega)$ may however be induced[20] when high-intensity infrared light is simultaneously launched into a fibre. This non-linearity decays within minutes after switching the poling field off.

Excitation poling generates a permanent $\chi^{(2)}$ $(2\Omega=\Omega+\Omega)$ in the fibre. The dependence of the induced $\chi^{(2)}$ $(2\Omega=\Omega+\Omega)$ on poling field and time is shown in Figure 4 where a cw excitation intensity of 6mW/um^2 at a wavelength of 488nm was used. In this fibre no saturation of the induced $\chi^{(2)}$ with applied poling field strength was observed. Clearly more measurements about the behaviour of the induced $\chi^{(2)}$ for poling fields >150V/um in fibres of
different compositions are needed to establish accurate $\chi^{(2)}$ -saturation levels[9]. The intensity and time dependence of the induced non-linearity is shown in Figure 5, where pulsed blue light at 480nm was used for defect excitation.

Poled fibres may easily be bleached optically be using high-intensity blue/green light or thermally be exposing the fibres to temperatures > 200°C. Subsequently they may be repoled. Figure 6 is a measurement of the bleaching characteristics of an induced second-order non-linearities with time even at room temperature is observed. However, in case of the Pockels effect the decay was found to be less than 10% over a time period of 3 months.

Phase-Matching Techniques

Apart from the Pockels effect, efficient second-order non-linear frequency mixing depends on effective phase-matching[21], by employing externally-induced $\chi^{(2)}$-gratings or by using mode-interference grating (MIGs)[13]. When no external fields are used the $\chi^{(2)}$-gratings may be written by coherent interaction between an infrared pump-wave and its SH[6]. By taking advantage of fibre birefringence these grating can be used for SHG at wavelengths other than the writing wavelength[22]. However, here we restrict the discussion to SHG in excitation poled fibres.

In general, the $\chi^{(2)}$ induced by excitation poling may be expanded into a power series of the defect excitation intensity I, the GeO_2 (or possibly another dopant) concentration in the fibre, g, and the poling field strength E_{dc}. Assuming that there is only one dominant term in the expansion, we may write

(1)

where δ, β and α are constants. We verified experimentally[8] that $\alpha \approx 1$ for $E_{dc} < 150$V/um in our fibres. No exact estimates for β and δ are yet available. The results from section 3 indicate that $0.5 < \beta < 1$. Here we assume that β is unity for simplicity. Thus $\chi^{(2)}$-grating may be induced in fibres by modulating one of the three parameters in Equation (1). In general the $\chi^{(2)}$ induced in the fibre will then be of the form

(2)

where $G = 2\pi/\Lambda$ is the grating vector and Λ the grating period. Efficient frequency conversion is obtained only under phase-match conditions, i.e. when

(3)

or

(4)

where the first expression refers to modal phase-matching and the second expression refers to phase-matching with the help of grating. Here we have neglected the influence of refractive index gratings[23] on phase-matching, since it is easily shown that they are orders of magnitude less efficient than $\chi^{(2)}$-gratings for phase-matching second-order non-linear processes in fibres.

Modal Phase-Matching

Modal phase-matching is achieved by using the z-independent part of $\chi^{(2)}$ in the fibre. The first term on the right hand side of Equation (2) is then relevant. Assuming that the fundamental wave is propagating in the ψ_p-mode and the SH is propagating in the ψ_{sh}-mode, the conversion efficiency as a function of z is written as[13]

(5)

where

I is the intensity of the pump wave, n the refractive index at the pump wavelength λ and in deriving Equation (5) we used SI units and the definition of non-linear coefficients according to Shen[24]. The overlap integral O in Equation (5) is defined as

(6)

where χ_0 is the average $\chi^{(2)}$ over the effective core area A. It follows immediately that a fundamental wave in the LP_{01}-mode can only couple to a SH LP_{0n}-mode. Further, since $\chi^{(2)}$ is related to defects of the germanosilicate glass matrix and almost zero in pure silica[25], for a germania doped step-index fibre $\chi^{(2)}$ also follows a step profile[21]. The overlap integral for the coupling of a fundamental wave in the LP_{01} -mode to a SH-wave in the LP_{02}-mode for a weakly guiding fibre[26] is shown in Figure 7 as a function of the V-value[26] of the fundamental wave, assuming both a step profile and a uniform $\chi^{(2)}$. The overlap integral remains always small, which indicates that the available non-linearity is poorly exploited in this case.

Phase-Matching with Internally-Written Mode-Interference Gratings

A better exploitation of the available non-linearity than by modal phase-matching is possible by using MIGs. If the fibre is slightly multi-moded at the defect excitation wavelength, mode interference forms a spatially periodic intensity distribution in the fibre. Defects are then preferentially excited at the antinodes and when a poling field is applied at the same time a $\chi^{(2)}$-grating structure corresponding to the mode-interference pattern is formed. The phase-match condition is now given by Equation (4), where $G=k_a - k_b$ and k_a, k_b are the propagation constants of a mode pair at the defect excitation wavelength. The conversion efficiency in this case is again given by Equation (5), where the overlap integral from Equation (6) is obtained by replacing $\chi^{(2)}$ (x,y) with

(7)

where ψ_a, ψ_b are the transverse field distributions of the mode pair used to write the grating. The overlap thus effectively comprises fiber modes, i.e. two blue, two infrared and one SH. The overlap as a function of germania concentration and the required core-radius to obtain phase-matched SHG with a pump-wave at 1064nm is shown in Figure 8. Here it is assumed that the MIG was written by mode-interference between the LP_{01} and the LP_{11} mode at 488nm, the Sh is propagating in the LP_{11} mode and the pump in the LP_{01} mode. We note that the overlap approaches unity, which means that the non-linearity is effectively exploited. However, when the fibre propagates n modes at the excitation wavelength, the overlap decreases at least as 2/n (for equal mode excitation), since only a fraction of the blue light power is propagating in the correct mode pair.

Note that $\chi^{(2)}$-gratings may also be written by externally modulating either the poling field[27] or the defect excitation intensity[19]. Coherence lengths up to 1cm have been demonstrated in such devices and it has been suggested that coherence lengths up to 10cm should be achievable by using optimised fibre designs[27], which is similar to the coherence lengths that have been obtained with MIGs. Therefore externally-written gratings may eventually lead to higher conversion efficiencies than MIGs, since they allow the propagation of both the pump and SH-wave in the fundamental mode and therefore always ensure an overlap near unity.

Experimental Demonstration of Phase-Matching

Figsures 9 and 10 show the measured SH-signal as a function of pump-wave-length obtained in a modally phase-matched fibre (fibre 1) and a fibre with a MIG (fibre 2). The parameters of these fibres are given in table (1). The overlap integral for fibre 2 was crudely estimated to be about 3%. The overlap integral is small since a large number of modes were propagating at the defect excitation wavelength. A better estimate of the overlap integral can be made in a fibre that is only double-mode at the defect excitation wavelength and single-mode at the pump wavelength. However, despite this uncertainty there is remarkably good agreement for the magnitude of the induced $\chi^{(2)}$ in the two fibres. The value for the second-order non-linearity in fibre 1 is only one tenth of that for KDP[24], which has a value of $9.8*10^{-13}$ (m/V).

Note also that the position of the phase-match peaks may be estimated from an analysis of the dispersive properties of the fibres[13]. Since the fibres used here have strongly elliptical cores, it is then useful to resort to a simple slab-waveguide model[28]. The theoretical positions of the phase-match peaks given in table (1) were calculated using a slab-waveguide model and the measured values for the core area, core ellipticity and a measurement of the refractive index profile. Again a good agreement between the measured and theoretically estimated position of the phase-match peaks is observed.

Limiting Mechanisms

The efficiency of second-order non-linear processes in fibers is governed by the size of the overlap integrals, the interaction length and the magnitude of the induced non-linearity and absorption. At high optical pump powers and long interaction lengths the onset of third-order non-linear interactions leads to a further limitation. However, here we consider second-order non-linear interactions with only moderate pump powers and short interaction lengths where third-order non-linear interactions may be neglected.

The size of the induced non-linearity for the Pockels effect and SHG is strongly dependend on the poling field strength. Since the poling fields that may be applied to side-channel fibres[18] (i.e. 800V/um close to the theoretical breakdown limit for pure silica glass[29]) are a factor of 2-5 larger than the poling fields that were used in the present series of experiments, a further increase in the magnitude of the induced non-linearities by poling seems possible.

The interaction lengths for the Pockels effect are limited only by the magnitude of the induced absorption and the lengths of the electrodes that may be incorporated into optical fibres. Typically this leads to maximum interaction lengths of several metres. In the case SHG the temporal coherence of the pump source and the writing source (i.e. when using MIGs) and spatial nonuniformities limit the possible interaction lengths. The decrease of the visibility of a MIG due to the finite spectral width of the writing laser may be obtained from an analysis as presented by Tom et al.[30]. The result is that for typical spectral widths of Argon lasers (≈0.2 cm^{-1}), coherence lengths> 2m may be achieved with MIGs. The dominant limiting mechanism in this case are small fibre nonuniformities which prevent coherent wave propagation between the pump and SH-wave for lengths>10cm. Further, the refractive index gratings that accompany the MIG writing process[8,23] lead to mode coupling between the modes at the excitation wavelengths. This causes the phase of the exciting modes to advance continuously along the fibre (compared to mode propagation without a MIG) and thus also limits possible interaction lengths.

Measurement of Second-Order Non-Linearities in Fibres

Here we describe measurements of the non-linear coefficients involved in SHG and in the Pockels effect. In this we neglect any off-diagonal terms for $\chi^{(2)}$, since only a $\chi^{(2)}{}_{111}$ tensor element aligned parallel[21,22] (or antiparallel[31]) to the poling field was found to be induced by poling.

Second-Harmonic Generation

The non-linear coefficient governing SHG is best measured by an indirect comparative method. In this $\chi^{(2)}(2\Omega=\Omega+\Omega)$ is compared to $\chi^{(3)}(2\Omega=\Omega+\Omega)$, which is the third-order non-linear coefficient governing electric-field induced SHG[4,27] (ESHG) and is well known in the field of non-linear optics. Here we used measurements[32] of the intensity-dependent refractive index to obtain the value $\chi^{(3)}=2.75*10^{-22}$ (m/V)^2 for our fibres.

Practically, before excitation poling a fibre a poling field E_{dc} on its own is applied and the SH-conversion efficiency η_{pol} due to the permanently induced non-linearity is now measured again at the centre of the modal phase-match peak (due to small optically induced refractive index changes the centre of the phase-match peak typically shifts by < 1nm between these two measurements). The non-linearity induced by poling is then given by

(8)

where we assumed that all optical waves were aligned parallel to the poling field and O_{ESHG} and O_{pol} are the overlap integrals for the two processes. The magnitude of the overlaps as a function of the V-value of the pump wave (in the LP_{01} mode) and the SH-wave (in the LP_{02} mode) was given in Figures7, where O_{ESHG} corresponds to the uniform $\chi^{(2)}$ and O_{POL} to the step-profile $\chi^{(2)}$. Note that the longitudinally uniform part of $\chi^{(2)}$, i.e $\chi^{(2)}$o as shown in Equation (2) is now measured. Further, no information about the interaction length is required and O_{ESHG}/O_{POL} is an accurately defined ratio close to unity. The values of $\chi^{(2)}$ for fibre 1 in Table 1 was measured by this method. Note that in this we found a shift of -0.27nm in the position of the phase-match peak after poling.

The non-linearity induced by poling may equally be estimated from a direct measurement of the SH-conversion efficiency at a given pump power. For modally phase-matched fibres this is a viable technique, since only the coherence length has to be obtained from an additional measurement. It was shown that both measurement techniques are in good agreement[21]. For the case of fibres phase-matched with MIGs, however, a high uncertainty exists about the magnitude of the overlap integrals, which means that only upper limits for the induced non-linearity may be accurately determined.

The Pockels Effect

In the following we assume again that all optical waves are aligned parallel to the dc-fields and the induced non-linearity. The Pockels effect leads to a phase-shift of a propagating wave in the presence of an electric field. Thus the Pockels coefficient may be measured by standard techniques involving the electrooptic modulation of light[11,31]. In this the phase shift induced by an applied electric field E_{dc} in the presence of a Pockels coefficient r_{11} is given by

(9)

where O_p is the intensity overlap integral for the Pockels effect and l is the interaction length. In addition to r_{11} a Kerr coefficient s_{11} which is not measurably changed by the poling process (a small change in the refractive index is observable, however[31]) is always present in the fibre. The phase shift induced by s_{11} is given by

(10)

where the overlap integral $O_k \approx 1$ in standard fibres s_{11} was measured by electrooptic modulation as $s_{11}=1.0*10^{-22}$ $(m/V)^2$ at a wavelength of 633nm both for GeO_2 and P_2O_5-doped silica fibres[11,31]. We may interpret the Pockels effect as stemming from an induced internal electric field, which biases the Kerr effect and thus leads to the linear dependence of the phase shift on the modulating field[31]. Combining eqs. (9) and (10) we obtain for the internal field

(11)

We may then define a poling efficiency η as the ratio of the induced internal field over the applied external poling field[31]

(12)

The highest value[31] for r_{11} was obtained as $r_{11}=2.7*10^{-15}$ (m/V) for a poling field of 150 V/um at a poling temperature of 300°C. This then results in a poling efficiency of 9%. If a poling efficiency of 50% could be achieved for a poling field of 800 V/um, this would result in a Pockels coefficient of $r_{11}=8*10^{-14}$

(m/V). The Pockels and Kerr effect in conjunction may also be used to measure the direction of the second-order non-linearity induced by poling. Consider the phase shift obtained by a small modulating field E_m in the presence of a bias field E_{bias} due to contributions from the Pockels and Kerr effects

(13)

where we assumed overlap integrals of unity and neglected the terms quadratic in E_m. Now if the Pockels coefficient is parallel to the poling field (assumed positive) a positive bias field enhances the phase-modulation and a negative bias field reduces it. The opposite holds for a Pockels coefficient aligned antiparallel to the poling field. r_{11} was found to be parallel to the poling field for silica fibres doped with P_2O_5 and antiparallel for GeO_2-doped fibres[31].

Summary and Conclusions

Poling of optical fibres was demonstrated with large externally applied dc-fields. It was shown that poling breaks the inversion symmetry of glass and gives rise to large second-order non-linearities. A maximum Pockels coefficient of $2.7*10^{-15}$ (m/V) could thus be induced in a fibre. Electrooptic modulation with total amplitude modulation was obtained with an applied voltage of 1300V in a poled fibre of 50cm length.

An electronic contribution to the second-order non-linearity was induced by excitation poling. A non-linearity of the form $\chi^{(2)}$ $(2\Omega=\Omega+\Omega)$ matching techniques this non-linearity was shown to give rise to efficient SHG. Optically-written second-order non-linear MIGs were shown to be especially advantageous for phase-matching second-order non-linear frequency mixing processes in fibres, since they can give rise to overlap integrals near unity and interaction lengths up to 10cm. A maximum SH conversion efficiency of 1% was obtained in a poled fibre phase-matched with a MIG. A further significant improvement in the performance of poled fibres seems to be possible using optimum fibre designs and poling conditions.

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[GeO_2]	13±1	18±1	mole%
[P_2O_5]	0.5	-	mole%
core area	16±2	9±1	μm^2
aspect ratio	2±0.2	1.7±0.3	
poling field (v/um)	100	140	V/μm
cw excitation intensity	25	5	mW/μm^2
phase-match peak exp.	1.208	1.051	μm
phase-match peak th.	1.7±0.1	1.08±0.1	μm
overlap integral	0.15	0.03	
interaction length	1.5	6	cm
SH-conv. efficiency	0.4	1.0	%
induced $\chi^{(2)}$ ($2\Omega=\Omega+\Omega$)	1.1	2	10^{-13}(m/V)
pump-mode	E_{11}	E_{11}	
SH-mode	E_{31}	E_{12}	
MIG-modes	-	E_{11}/E_{14}	

Table 1 Fibre parameters. The SH-conv. efficiency is normalised for a pump power of 2GW/cm^2. Defect excitation was at 488nm.

Figures

Figure 1 Fibre designs used in poling experiments.

Figure 2 Excitation spectrum of second-order non-linearity induced by blue light alone.

Figure 3 Dependence of induced Pockels coefficient on poling field.

Figure 4 SH-signal as a function of time and poling field for a constant cw defect excitation intensity of 6mW/μm^2 at 488nm as measured in fibre 2.

Figure 5 SH-signal as a function of time and defect excitation power (pulsed excitation) at 480nm for a constant poling field of 140V/μm (for fibre 2).

Figure 6 Bleaching characteristics of excitation poled fibre. Poling parameters are the same as in Figure 4.

Figure 7 Overlap integral (modal phase-matching) for SHG for a pump in the LP_{01} and the SH in the LP_{02} mode as a function of fibre V-value.

Figure 8 Overlap integral (MIG phase-matching) and required core-radius to obtain SHG with a pump at 1064nm in the LP_{01} and the SH in the LP_{11} mode using a MIG written by interference between the same two modes.

Figure 9 SH-power as a function of pump wavelength in modally phase-matched fibre.

Figure 10 SH conversion efficiency as a function of pump wavelength at a pump power of 150W in a fibre phase-matched with MIGs.