Introduction to $\chi^{(2)}$ processes

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Abstract

This paper gives an introduction to $\chi^{(2)}$ processes from an historical point of view, highlighting some of the major developments that have contributed to the enormous enhancement, of around thirteen order of magnitude, in frequency conversion efficiency per unit pump power, since the earliest harmonic generation experiment. Current developments are also briefly reviewed, with emphasis on the recent achievements and prospects of quasi-phase-matched materials.

The field of nonlinear optics is generally considered to have its foundation in the experiment of Franken at al in 1961 [1], in which they generated the optical harmonic of light from a ruby laser, (second harmonic generation, SHG), by passing it through a quartz crystal. Expressing the polarisation, P, induced in a medium by an applied field, E, as a power series in E,[2],

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

one identifies the second harmonic generation process with the first nonlinear term, i.e. the quadratic term $\epsilon_0\ \chi^{(2)}\ E^2$, whose magnitude is defined by the second order nonlinear susceptibility, $\chi^{(2)}$ and is only non-zero in non-centrosymmetric media. While the seminal importance of Franken's experiment cannot be overstated, it is interesting to note that the earliest experiments that explicitly revealed the existence of the quadratic term, are those of Pockels in 1893, in which he described what is now known as the linear Pockels effect. This effect involves one field at optical frequency and the other a d.c. field. The technology of the late 19th century was already able to produce high enough d.c. field strengths to allow the Pockels effect to be observed. It was only with the invention of the laser in 1960, that sufficiently high optical field strengths became available to allow demonstration, in the following year, that the quadratic term gave observable effects, i.e. optical frequency conversion, with all-optical input fields.

It is also of historical interest to note that an early attempt to achieve optical frequency conversion was even made by Newton after he first separated the colours of the solar spectrum. In his own words (Optics, 1704), 'when one sort of Ray hath been well parted from those of other kinds, it hath afterwards obstinately retained its colours, notwithstanding utmost endeavours to change it'. With our vantage point it is interesting to speculate how Newton might have explained colour changes had they occurred, since he was in fact a staunch advocate of the particle model of light.

The conversion efficiency from fundamental (694nm) to second-harmonic (347nm) in Franken's experiment was rather low ~10^{-8} (10^{-6} %). Expressed as a conversion efficiency per Watt of fundamental power, the figure is ~ 10^{-10} %/W. Such have been the developments in harmonic generation techniques since then that conversion efficiencies are now approaching ~10^{3} %/W, corresponding to a conversion efficiency of 10% for 10mW of power, the power level available from a cheap diode laser. So, harmonic generation has rapidly developed from the realm of exotic curiosity to the area of commercial commodity. A simple equation relating the second harmonic power to the fundamental power can be used to indicate the reasons why conversion efficiencies were initially so low and have subsequently been increased by so much, by ~13 orders of magnitude in fact.

The second harmonic power $P_{2\omega}$, generated by a beam of fundamental light of power, P_{ω}, in a beam area A, passing through a length L of nonlinear medium is given by

$$P_{2\omega} \propto \frac{d^2 P_{\omega}^2 L^2}{A} \tag{2}$$

where, following common practice, we use the nonlinear coefficient d ($d = \chi^{(2)}/2$) in place of $\chi^{(2)}$. In deriving this equation it is assumed that there is negligible dispersion between fundamental and harmonic waves over the length L of the medium, i.e. that the harmonic emission from dipoles over the entire length of the medium are in phase and hence add cumulatively. In fact material dispersion, i.e. the fact that the refractive indices for fundamental, n_ω, and harmonic, $n_{2\omega}$, have different values, implies a rather short length, the so-called coherence length, $\ell_{coh} = \lambda_\omega / 4(n_{2\omega} - n_\omega)$, over which this addition can occur cumulatively. Where the crystal length exceeds ℓ_{coh}, successive cancellation and addition of the harmonic fields from each coherence length means that the maximum generated harmonic power does not exceed that given by equation (2) with L replaced by ℓ_{coh}. The small value of ℓ_{coh} (~10μm) in Franken's experiment was the major factor leading to the small observed conversion efficiency. Other factors were (i) the rather small value of d (0.3 pm/V), whereas a number of recently developed materials have d values of between one and two orders of magnitude greater than that of quartz. The choice of quartz was made on the basis of being a readily available, non-centro-symmetric optical material. (ii) The use of a non-diffraction-limited laser beam, resulting in a significantly larger value of A than ideally possible.

An elegant solution to the problem of increasing the coherence length was reached simultaneously by Giordmaine[3] and Maker et al [4], who showed that material birefringence, which provides a difference of refractive index for ordinary and extraordinary polarisation could be used to cancel the index difference due to dispersion and so equalise n_ω and $n_{2\omega}$, i.e. the process is 'phase-matched'. Thus, with $n_{2\omega} = n_\omega$, the coherence length is essentially infinite and the length L in equation (2) can then be the actual length of material. The consequent increase in L is of around three orders of magnitude, thus giving nearly six orders of magnitude increase in conversion efficiency. This phase-matching capability instantly made many prospects for exploitation of $\chi^{(2)}$ both realisable and practical. Meanwhile another approach to the phase-matching requirement had been proposed by Armstrong et al [5] in their seminal paper on nonlinear optical interactions. Their suggested scheme, now known as quasi-phase-matching involved the use of a medium, structured so that after each coherence length of the medium the sign of the nonlinearity is reversed (see fig.1), so that in effect the phase error accumulated over a coherence length is reset to zero by the sign reversal, thus enabling the harmonic emission from each successive coherence length to add in phase. At the time of publication of this idea, no simple means existed for creating such a structure, and it needed some thirty years before a really practical and elegant implementation emerged. This is now dramatically transforming the prospects for numerous applications which exploit $\chi^{(2)}$. More will be said of this later.

Returning to equation (2) we now briefly discuss the question of how the frequency conversion efficiency can be enhanced by reducing the area A. A detailed discussion of optimum focussing has been given by Boyd and Kleinman [6]. Consideration of the diffraction behaviour of an unguided light beam of finite transverse extent shows that for a length L of medium, the minimum area that can be maintained essentially constant over the length L corresponds to the so-called confocal focussing condition, for which $A \sim L\lambda$ where λ is the wavelength of light in the medium. A beam focussed to this size is close to the optimum for frequency conversion efficiency [6]. This result applies for a diffraction-limited beam, i.e. one whose beam quality factor M^2, is unity. (The quantity M^2 specifies the factor by which the beam divergence exceeds that of a diffraction-limited beam.) For a general value of M^2, the minimum area is $\sim M^2 L\lambda$, so the optimum frequency conversion efficiency is reduced by a factor $1/M^2$. This provides an illustration of a general rule that pump beam quality is of paramount importance in nonlinear optics.

The expressions given above for minimum beam area are relevant to the case where there is no double-refraction, i.e. where the Poynting vector and the wave normal are in the same direction. In general however, birefringence phase-matching, involving some general angle θ_m of propagation relative to the optic axis, will result in double refraction, with the extra-ordinary and ordinary beams walking off from each other by some angle ρ. The minimum beam size in the walk-off plane should then be ~$L\rho$, to avoid separation of ordinary and extraordinary beams. In practice this can pose a severe limitation on the minimum spot size and hence area A. The discovery of the nonlinear material $LiNbO_3$ proved to be a great boon in this context as it allowed '90°-phase-matching', (θ_m = 90°), hence no double-refraction, for various wavelengths of interest (in particular for frequency doubling of the 1μm radiation produced by Nd lasers). Access to 90°-phase-matching (or 'non-critical-phase-matching', as it is also called) has been a major contributor to the process of reducing the pump powers required for efficient frequency conversion. Potassium titanium phosphate (KTP) which allows nearly non-critical phase-matching for SHG from ~1μm, and lithium triborate (LBO) which allows, by virtue of its large change of refractive index with temperature, a very wide range of non-critically phase-matched wavelengths, have both become prominent nonlinear materials, due in large part to this 90°-phase-matching feature. Such is the gain in efficiency from the tight focussing allowed with non-critical phase matching that efficient frequency conversion can be achieved with cw pump lasers. Thus intracavity frequency doubling using KTP and LBO (see e.g. [33]), now allows generation of cw green light at multiwatt level from a Nd laser, thereby offering an all-solid-state alternative to the Argon laser. Quasi-phase-matched material, such as periodically-poled lithium niobate (PPLN), is a particularly noteworthy example of a material that allows tight focussing since it involves propagation perpendicular to the optic axis, and thus provides non-critical phase-matching whatever the frequencies of the interacting waves. Also, unlike birefringence phase-matching, quasi-phase-matching allows the interacting waves to have the same polarisation, thus giving access to a significantly larger nonlinear coefficient (the 'diagonal' element d_{33} in $LiNbO_3$) than that (d_{31}) which is available in birefringence phase-matching. With these benefits the possibility then exists, as recently demonstrated ([7], to be discussed later) of highly efficient and reliable operation of parametric oscillators driven by a cw pump.

As a last comment on the question of reducing the area of the interacting beams, the benefit of using a waveguide geometry should be noted. In principle, a waveguide can have lateral dimensions of the order a few times the wavelength of the propagating light, so that A is reduced from the best possible value in a bulk medium ($\sim L\lambda$) to a few times λ^2, an enhancement of the order of L/λ. This factor can easily give an enhancement of three orders of magnitude to the conversion efficiency. While the use of a waveguide geometry in nonlinear media has a long pedigree, (see [8] and the extensive list of references therein), it has really begun to flourish relatively recently when combined with the quasi-phase-matching technique. The way was led with $LiNbO_3$, since not only did it lend itself to a practical waveguide fabrication technology, but it also offered a means for producing the periodic sign reversal of the nonlinear coefficient, so that the principle suggested by Armstrong et al [5] began to have a practical realisation (see [34] for a survey of this topic). The combination of high nonlinearity, tight confinement of the interacting waves, and ability to phase-match for any interacting frequencies, has led to great interest in its potential for compact blue light sources via SHG from infrared diode lasers. It is this route that has led to the very high efficiencies of ~1000 %/W mentioned earlier.

Once the technique of birefringence phase-matching had been demonstrated in 1962, the route to high conversion efficiency was open. The use of Q-switching, to provide high pump-power was also an important factor in achieving high efficiency. Then with high conversion efficiency achieved for second harmonic generation, the reverse process of optical parametric amplification and oscillation (OPA, OPO) became accessible. Early theoretical papers had indicated the feasibility of achieving optical parametric oscillation [10,11]. A useful indicator to available parametric gains is based on the fact [12] that if in SHG, a specific conversion efficiency η is achieved, for some given pump power P, then if instead a wave at the harmonic frequency is used as a pump, with the same power P, the parametric gain for the subharmonic would also equal η. So, since resonator losses of a few per cent were readily achievable, it followed that once harmonic conversion efficiencies of a few per cent were reached, the converse process of optical parametric oscillation should also be achievable.

The first optical parametric oscillator was demonstrated by Giordmaine and Miller [13], using $LiNbO_3$ as the nonlinear medium, pumped by the second harmonic of a Q-switched Nd laser. The achievement of extensive tunability was a major attraction, unrivalled at that time as tunable coherent sources, such as dye lasers, had not yet appeared on the scene. Thus the early days of parametric oscillators were optimistic days. Eventually however, this optimism later turned to disillusion, and it is only in recent years that the OPO has firmly re-established itself. The reasons for this chequered history are many. The earliest OPOs were doubly-resonant oscillators (DRO), with the virtue of a low threshold power (a cw threshold as low as 45mW was achieved in 1968, [14]), but with the problem of extreme frequency instability [15]. The instability problem could be overcome by using a singly resonant oscillator (SRO), as first demonstrated by Bjorleholm [16], but the penalty to be paid here was a higher threshold, which, for the nonlinear materials available at the time was uncomfortably close to the damage threshold. At the same time, dye lasers had now appeared allowing tunability (albeit limited, mainly to the visible spectrum) to be obtained in a simple and reliable manner. So the scale of effort on OPOs was greatly reduced as their early promise was perceived as being difficult to attain in practice.

Now the view of OPOs has changed dramatically, back to one of great optimism. New materials have played an important part in this. For example BBO and LBO have very high damage thresholds so that the problem of operating close to the damage threshold has been removed. Advances in lasers have also played a role with good laser beam quality, whose importance was mentioned earlier, now being the norm rather than the exception. The availability of short pulses (picosecond and femtosecond) has also made a major contribution. Since the parametric gain is determined by the instantaneous power level and furthermore the damage threshold intensity for short pulses increases with decreasing pulse duration, it follows that parametric processes are particularly well suited to short pulse pumping. For example, it is readily possible to pump a parametric amplifier with an intense enough short pulse to achieve a gain of $\sim 10^{15}$ and hence efficient optical parametric generation (OPG) from parametric noise in a single pass through the crystal. This type of device, pioneered by Piskarskas and co-workers (see e.g. [17] for a recent example) provides a very simple arrangement for achieving wide tunability of intense short pulses. Such a scheme does however, require a very intense pump pulse, which until recently has implied rather large laser systems. One way of reducing the power requirement, to a level compatible with a modest pump laser, is to use synchronous pumping of an OPO by a train of mode-locked pump pulses. This approach, first demonstrated by Burneika et al [18], has since been extended to pumping with a cw-mode-locked train (first report of a SRO device by Edelstein et al [19]) and pumping with all-solid-state mode-locked pump lasers. The fact that the gain is determined by the peak power of the mode-locked pulses, has meant that lasers of quite modest average cw power are sufficient to drive an SRO device, so that all-solid-state schemes are possible using a cw diode-pumped Nd laser as the pump source (see e.g. [20] for a KTP device, [21] for a LBO device).

Thus the major revival of interest in OPOs in the late 1980s had already led, by the early 1990s, to the point where a number of commercial devices had been developed. Prospects for further developments have since been greatly enhanced by the arrival of quasi-phase-matched material, in particular periodically poled lithium niobate. The ability to achieve periodic poling in $LiNbO_3$ by a simple application of an electric field to a photolithographically patterned surface had been the initial step, first applied to a waveguide [22] and then to a bulk medium [35], with subsequent developments allowing poling, through samples of up to ~1mm in thickness. Such a thickness is large enough for use with infrared wavelengths in bulk (unguided) geometry over several centimetres of length. The large nonlinearity (up to ~20pm/V), wide spectral transmission (~0.4 to 4.8μm), and ability to phase-match, simply by choice of period, for any interacting wavelengths, make PPLN an extremely attractive nonlinear material. Examples of recent achievements with this material are:

(i) cw single-resonant oscillation, pumped at 1064nm and giving multiwatt output at wavelengths of 3μm and longer [7].

(ii) efficient cw generation of 473nm light by frequency doubling a 946nm Nd laser [23]

(iii) multigrating PPLN device giving OPO tuning from 1.36 - 4.83 μm[24]

(iv) efficient (up to 65%) SHG to the green, using a picosecond pulse source at 1047nm [25]

(v) low threshold synchronously pumped OPOs [26]

(vi) efficient SHG and OPG using subpicosecond pulses from an erbium-doped fibre laser system [27], thus bringing the OPG within reach of modest pump lasers.

All of the above results are early results with much scope for further development, indicating very strongly that PPLN and other quasi-phase-matched materials will play an important role in the future exploitation of $\chi^{(2)}$.

In this paper the main emphasis has been given to the development of sources involving the use of $\chi^{(2)}$ for frequency conversion. Even for this limited area of application of $\chi^{(2)}$ it has not been possible to give comprehensive coverage as space limitations have had to be observed. So we conclude with a brief mention of some other areas not specifically concerned with source development where effects based on $\chi^{(2)}$ are growing in importance. Naturally all of the developments that have contributed so much to the advances in sources based on $\chi^{(2)}$, in particular the availability of new materials, have their benefits carry over into these other areas.

As an example of an area commanding much current attention, we mention the so-called 'cascaded $\chi^{(2)}$' effect (see e.g.[28]) in which interest centres on the phase changes (intensity-dependent phase-changes) induced in the fundamental wave by the process of harmonic generation and the reverse process of regeneration of fundamental from the harmonic. These intensity-dependent phase-changes can play a role analogous to that of self-phase-modulation due to $\chi^{(3)}$, i.e. the effects due to cascaded $\chi^{(2)}$ can be thought of as arising from some 'effective $\chi^{(3)}$'. Thus one has a whole gamut of possible uses, mirroring uses already developed in $\chi^{(3)}$ materials, but with the added benefit that a number of existing $\chi^{(2)}$ materials offer larger 'effective $\chi^{(3)}$' values than available through actual $\chi^{(3)}$ values. Effects having potential uses include optical switching, spatial soliton propagation, self-mode-locking of lasers. For example the latter relates to a scheme, first reported by Stankov [29] in which an intracavity harmonic generator crystal is combined with a resonator mirror that preferentially reflects the harmonic rather than the fundamental. This combination, referred to as a 'nonlinear mirror', favours operation of the laser in such a way as to maximise the harmonic conversion, so that maximum feedback can be achieved followed by reconversion of harmonic to fundamental, as it returns through the nonlinear cyrstal. This 'nonlinear mirror' therefore favours operation of the laser in a mode-locked regime, and it leads to self mode-locking. In its more recent development [30] this technique shows signs of being able to operate at significantly lower power levels than the conventional ($\chi^{(3)}$-based) so-called Kerr lens mode-locking technique.

Other areas of interest in $\chi^{(2)}$ effects include metrology and quantum optics. Their juxtaposition is an interesting reminder of wave-particle duality, in that the metrology applications exploit the possibility of transferring frequency standards to other frequencies, via frequency conversion, based on the fact that exact frequency relations exist between interacting waves (e.g. the second-harmonic frequency is exactly twice that of the fundamental). Hence a wave description of light is implied, whereas the exploitation of $\chi^{(2)}$ in quantum optics is based on the creation and annihilation of pairs of photons, hence a quantum (or particle) description of light is implied.

Given the space limitations, we do not discuss these areas any further and simple quote two references, which serve as examples of the metrology aspects of $\chi^{(2)}$ [31] and the quantum optics aspects of $\chi^{(2)}$ [32], both of which will be covered by following papers in this journal issue.

In summary, this paper has aimed to give a brief introduction to $\chi^{(2)}$ processes, with a historical perspective, and also aimed at setting a context for the papers that followed in the Summer School on '$\chi^{(2)}$, Second Order Nonlinear Optics : From Fundamentals to Applications', which took place at Les Houches, France, 22-26 April 1996. The empahsis in this paper has been on development of sources using frequency conversion via $\chi^{(2)}$. Progress of $\chi^{(2)}$-based sources has been enormous since the first observation of optical harmonic generation,m with around thirteen order of magnitude improvement in conversion efficiency over a thirty-five year period. Clearly things will not stop there. The practical realisation of quasi-phase-matched materials has recently given a major boost to the subject. Quasi-phase-matching methods are being extended to a variety of materials, including semiconductor materials and organic materials where very large $\chi^{(2)}$ values are seen. Quasi-phase-matching in polymers, and even in glass fibres (where despite being centrosymmetric, a significant $\chi^{(2)}$ can be induced by a 'frozen-in' internal field [9]) offer the prospects of convenient waveguide geometry and cheap mass-production. These advances in materials preparation and device fabrication will progressively bring $\chi^{(2)}$ effects within reach of modestly powered pump sources, so that even applications of $\chi^{(2)}$ that are currently regarded as exotic, such as in metrology and quantum optics, can become the standard technology of the future. As the uses of $\chi^{(2)}$ become more widely deployed and more accessible, it also needs to be recognised that, for example, the presence of a $\chi^{(2)}$ material inside a laser cavity adds further behavioural complexity and richness to what is already a complex device. There will be much fascinating laser physics to be unravelled in the future as lasers and $\chi^{(2)}$ materials become more intimately connected.

Figure Captions

Figure 1

Quasi-phase-matching; the ideal scheme in which a nonlinear interaction having a coherence length ℓ_{coh} is allowed to accumulate progressively over the length of the medium, by using a sign reversal of the nonlinear coefficient after each coherence length.

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