Cascading, quasi-phase-matching and induced Kerr effects

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Abstract

A strong cascading effect in quadratic nonlinear materials requires that the interaction between the fundamental wave and its second harmonic is phase-matched. Quasi-phase-matching is a technique, in which the relative phase is adjusted at regular intervals using a structural periodicity built into the medium. Potentially it allows for phase-matching in any material over a wide range of wavelengths using the largest component of the nonlinear susceptibility. The periodic structure induces cubic nonlinear Kerr effects, that can alter the properties of the material, e.g. in terms of solitons and CW switching.

Introduction

Nonlinear optics is traditionally discussed in terms of the separate effects of quadratic and cubic nonlinearities. For example, quadratic or $\chi^{(2)}$ nonlinearity is well-known for phenomena such as frequency conversion and parametric amplification, while cubic nonlinearity usually is associated with frequency degenerate processes, such as an intensity-dependent refractive index, self-focusing, solitons, four wave mixing, etc.

Although the cubic nonlinearity can be used for frequency conversion (third harmonic generation, electric field induced second harmonic generation, etc.), it is generally not very effective and consequently not often used for this purpose. In contrast, the quadratic nonlinearity can effectively mimic cubic properties and even support spatial solitons. In these phenomena the important effect is the self-induced modulation of the amplitude and phase of the fundamental beam(s).

Clearly the physics of such self-action effects requires two successive quadratic or second order processes in order for the net output to be back at the input frequency (ω). This can occur via up-conversion ($\omega+\omega\rightarrow 2\omega$), better known as second harmonic generation (SHG), followed by down-conversion ($2\omega-\omega\rightarrow\omega$), or via down-conversion ($\omega-\omega\rightarrow 0$), better known as optical rectification, followed by up-conversion ($\omega+0\rightarrow\omega$). It is the successive nature of the processes needed to modify the fundamental beam that led to the term 'cascading' for this class of effects and to the symbolic representation $\chi^{(2)}:\chi^{(2)}$, as the effects are proportional to $\left(\chi^{(2)}\right)^2$.

The two key features of cascading were predicted in the first decade of nonlinear optics. The existence of a nonlinear phase shift in the fundamental wave (FW) during SHG was first discussed by Ostrovskii in 1967 [1] and the existence of solitons was predicted in 1974 by Karamzin and Sukhorukov [2] (Incidentally, Sukhorukov's son will this year start his Ph.D. at the Optical Sciences Centre (OSC) at the Australian National University (ANU), working on $\chi^{(2)}$-effects as his father). However, the experimental work on cascading, starting in the late 1960s, was primarily concerned with measuring the interference between $\chi^{(3)}$- and $\chi^{(2)}$-related contributions far from the SHG wavevector-matching condition $\Delta k=2k_1-k_2=0$, where k_1 and k_2 are the wavevectors of the fundamental and second harmonic (SH), respectively. In this so-called cascading limit, where the crystal length L is much larger than the coherence length $L_c=\pi/\Delta k$, it is straightforward to reduce the coupled equations for SHG to a single equation for the FW with a cubic self-phase modulation term, whose coefficient is proportional to $\left|\chi^{(2)}\right|^2/\Delta kL$. In general the cascaded contributions were therefore small, since ΔkL was large.

It was not until the late 1980s and early 1990s that experiments were made sufficiently close to the phase-match condition to show a nonlinear phase-shift of the FW in excess of π with a dominant contribution from cascading [3]. These experiments proved that cascading is a strong effect near phase-matching and led to a tremendous interest in its application in all-optical phenomena (see [4] for a review). A comprehensive literature now exists of experimental and theoretical studies of cascading. In particular, a significant part of the theoretical work on solitons has been done in Australia at the OSC, ANU, since 1996 in strong collaboration with the Danish groups of Peter L. Christiansen at the Institute of Mathematical Modelling (IMM), the Technical University of Denmark, and Jens Juul Rasmussen at the Optics and Fluid Dynamics Department, Risø National Laboratory.

Here I consider one of the most important issues for obtaining a strong cascaded nonlinearity: Phase-matching. One of the most promising techniques for achieving phase-matching is the so-called quasi-phase-matching (QPM) technique, by which a grating is built into the medium to compensate for the mismatch. An important point about QPM, which was not realized until recently [5], is that it induces effective cubic nonlinearities, which affects the cascading process.

Quasi-phase-matching

Consider unseeded SHG, in which the incident FW, with frequency ω and wavelength λ, is linearly polarized along one of the crystal axes. In the crystal the FW interacts with the $\chi^{(2)}$-susceptibility and generates a SH wave at the frequency 2ω. The FW travels with a velocity determined by the index of refraction $n_1=n(\omega)$, whereas the velocity of the SH wave is determined by $n_2=n(2\omega)$. In general $n_2\neq n_1$ because of dispersion in the material, so that the fundamental and SH waves travel at different phase velocities. Since the direction of power flow between them is determined by their relative phase, the continuous phase slip due the difference in phase velocity leads to a periodic alternation of the direction of the power-flow and a repetitive growth and decay of the SH intensity. This situation is illustrated by curve B in Fig.1.

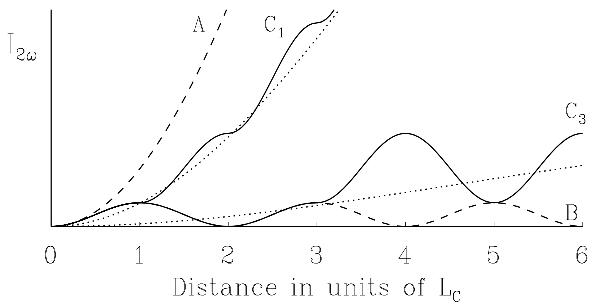

Fig.1. Evolution of SH intensity during unseeded SHG under conditions for type I phase-matching, obtained numerically from the analytical solution, as expounded in [6]. Dashed curves A and B show phase-matched and non phase-matched interaction in a uniform crystal, respectively. Solid curves C_1 and C_3 show first-order and third-order QPM, respectively. Dotted lines indicate the equivalent lowest-order evolution, which can be found analytically.

The distance over which the relative phase of the two waves changes by π is the coherence length $L_c = \pi / \Delta k = \lambda / 4(n_1 - n_2)$, which is also the half-period of the growth and decay cycle of the SH. If the refractive indices are matched, the SH field grows linearly with distance, and thus the intensity grows quadratically, as shown by curve A in Fig.1. This condition is termed phase-matching. Traditionally the techniques to achieve phase-matching have been divided into two types: In type I the incident FW is polarized along one of the crystal axes. In type II, the birefringence of an anisotropic material is used, and thus the incident FW has a polarization component along both the ordinary and extraordinary axis.

A fundamentally different method of generating continuous growth of the SH intensity is to invert the relative phase between the waves after each coherence length, where the SH intensity has reached its maximum in a cycle. The phase is thus reset periodically so that on average, the proper phase relationship is maintained. This has led to the name 'quasi-phase-matching' (QPM) for this technique of achieving phase-matching, which is typically discussed as type I.

First-order QPM, in which the relative phase is inverted every coherence length, is illustrated by curve C_1 in Fig.1, and third-order QPM, with phase-inversion every $3L_c$, is illustrated by curve C_3. Clearly, on average growth of the SH intensity can be achieved by any odd-number QPM. However, first-order leads to the most rapid growth and hence the largest conversion efficiency. Note that even order QPM can also occur when alternating the number of coherence lengths after which the phase is reversed. For example, second-order QPM can be obtained by periodically alternating between L_c and $3L_c$.

One way to invert the phase is to change the sign of the nonlinear $\chi^{(2)}$-coefficient. In the early days of QPM this was done by forming alternating stacks of thin wafers of the nonlinear crystal, rotating alternate wafers by 180°. A more practical approach in ferroelectric crystals involves forming regions of periodically reversed spontaneous polarization. Such regions of one sign of the $\chi^{(2)}$-coefficient are termed 'domains', in analogy with the ferroelectric domains that creates them.

The above explanation of QPM is a purely "space-domain" description. It is mathematically more convenient, and often helps intuition, to describe the effect in Fourier space, where the domain structure is a grating in the nonlinear coefficient, with a certain grating wavevector. In this context, phase-matching occurs when the wavevector of the QPM grating equals the wavevector mismatch Δk. Additionally it is easily recognized from the Fourier point of view that any periodic structure of the nonlinear coefficient, which possesses a proper spatial wavevector component, can accomplish QPM. Thus, complete sign reversal is not required, but only a modulation is necessary, with sign reversal being a special (and the most efficient) case.

The physics of QPM was outlined already in 1962 by Armstrong et al.,[7] but only recently have experimental difficulties been overcome and stable techniques been developed (see[8] for a review). The currently most mature techniques for QPM are electric field poling (now possible at room temperature) [9], and ion-exchange [10] of ferroelectric materials such as $LiNbO_3$ and KTP. The use of QPM in polymers is attractive because of the high nonlinear coefficients of these materials, but has proven difficult to control. However, recent results with QPM achieved through alternating domains of polymer and linear materials (i.e. no sign reversal), are promising. [11]

As can be seen in Fig.1 the SH intensity generally grows more slowly, even using lowest-order QPM, than it does with birefringent phase matching, where the phase-mismatch is fixed at zero throughout the length of the crystal. The strength of QPM lies in its ability to accomplish noncritical phase-matching, which would otherwise be impossible, through the introduction of an additional wavevector. Thus QPM has the following advantages compared with more conventional techniques:

- No restrictions as regards material or polarization
- Use of largest $\chi^{(2)}$ tensor component
- Matching at any wavelength and temperature
- Increased angular acceptance bandwidths

It should be noted that phase-matching may also be achieved by modulation of the linear susceptibility. However, this technique has been difficult to implement, because the amplitude of the linear index modulation must be comparable to the dispersion in order to achieve efficient conversion. [12]

Induced Kerr effects

So far QPM has mainly been discussed in terms of simply matching the wavevectors of the fundamental and SH waves thereby enabling efficient SHG and CW switching, known to be possible at exact phase-matching. In 1996 the $\chi^{(2)}$-group at OSC in Australia started working on how the QPM grating affects the properties of spatial solitons. In doing so we discovered that independent of the geometry (bulk media, planar or channel waveguides) QPM not only leads to phase-matching, but also induces cubic nonlinear terms such as self- and cross-phase modulation (SPM and XPM) in the effective averaged dynamical

equations [5]. Let me briefly explain the physics behind this induced cubic nonlinearity and discuss some examples of the effects, it can introduce.

Consider a QPM crystal as sketched in Fig.2, in which the $\chi^{(2)}$

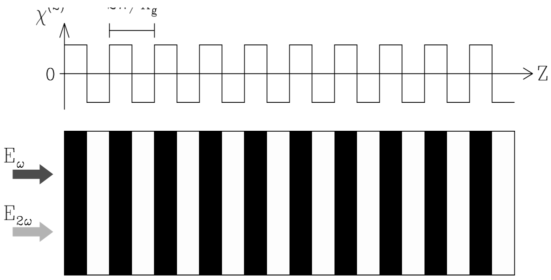

Fig.2. Sketch of a crystal with the typical square QPM modulation of the $\chi^{(2)}$ nonlinearity.

susceptibility is modulated by a periodic grating with domain length π / k_g. The evolution of the amplitude of the FW, $E_\omega(z)$, and the SH, $E_{2\omega}(z)$, is governed by the equations (see [4,7])

$$i\frac{dE_\omega}{dz} = g(z)\chi E_\omega {}^* E_{2\omega} e^{-i\Delta k z} = 0 \tag{1}$$

$$i\frac{dE_{2\omega}}{dz} = g(z)\chi E_\omega{}^2 e^{i\Delta k z} = 0 \tag{2}$$

where χ is proportional to the effective nonlinear coefficient. The grating is described by the function $g(z)$ of amplitude 1, which has an infinite Fourier spectrum of spatial harmonics of the grating wavevector k_g

$$g(z) = \sum_n g_n e^{ink_g z} \tag{3}$$

where $g_{2n} = 0$ and $g_{2n+1} = 2 / i\pi(2n+1)$ for the square function shown in Fig.2.

The grating introduces a 'reference wavevector', which for *m*th order QPM is simply mk_g. Expanding E_ω and $E_{2\omega}$ in a Fourier series of spatial harmonics of mk_g, we obtain the dynamical equations for the average amplitudes $w = < E_\omega >$ and $v = < iE_{2\omega} >$ [5]

$$i\frac{dw}{dz} = \chi_m w^* v + \gamma_m \left(|w|^2 - |v|^2\right) w = 0 \tag{4}$$

$$i\frac{dv}{dz} = \chi_m w^2 - 2\gamma_m |w|^2 v = 0 \tag{5}$$

where $\chi_m = \chi / (m\pi / 2)$ and $\gamma_m = \chi^2\left(\pi^2 - 8\right) / \left(m\pi^2 k_g\right)$. Diffraction in the transverse *x* and/or *y* direction is easily incorporated by simply adding the corresponding second derivatives.[5] Furthermore, the equations will have the same structure for any QPM modulation, as long as it is periodic, the only change being that the three SPM and XPM terms have different coefficients.[5]

From the lowest-order equations ($\gamma_m = 0$) we see that the average SH amplitude evolves as in phase-matched SHG in a uniform crystal, with the nonlinear coefficient reduced by a factor $m\pi / 2$, which increases with the order of the QPM. This means that the conversion efficieny is reduced by a factor $(m\pi / 2)^2$, which is a well-kown property of QPM.[8] It is beautifully confirmed by curves C_1 and C_3 in Fig.1.

However, the lowest-order equations are obtained by neglecting the coupling between the fundamental mode with wavevector mk_g and higher-order modes with wavevectors $3mk_g$, $5mk_g$ etc. This coupling is not phase-matched, but strongly incoherent, and from the averaged equations (4-5) we see that it induces effective cubic nonlinear Kerr effects in the form of SPM and XPM. However, the SPM term does not appear for the SH and the XPM coefficients are always negative. This merely emphasizes that the induced Kerr effects are of a fundamentally different nature than the conventional Kerr effects inherent in any material. Consequently, the dynamics can be significantly different from the earlier analyzed cases of competing nonlinearities.[12]

It is interesting that such effective cubic nonlinear terms will be induced by any kind of incoherent coupling between modes in $\chi^{(2)}$ media.[13] A simple example is SHG in a waveguide, which is single-moded at the fundamental frequency ω, but supports two modes at 2ω.[13] Incoherent coupling between modes is thus a general physical mechanism that induces cubic nonlinearity.

A: Spatial solitons

In collaboration with the group of Peter L. Christiansen at IMM, in particular C. Balslev Clausen, we have studied the properties of spatial solitons in QPM slab waveguides. The competing quadratic and QPM induced cubic nonlinearities are found to support a novel class of stable solitons. Unlike conventional optical solitons, the QPM solitons have amplitudes that are rapidly varying around a mean value, due to the higher-order modes with wavevectors $3mk_g$, $5mk_g$, etc., introduced by the grating. Since the higher-order modes can be found analytically in terms of the fundamental average mode at k_g,[13] the amplitude and wavenumber of the oscillations can be predicted. Numerical simulations of the original equations (1-2) with diffraction in the transverse *x*-direction (∂_x^2) as shown in Fig.3 confirm the predictions of the amplitude and wavenumber of the QPM solitons and their stability.

An interesting question about QPM solitons is how robust they are towards fluctions in the domain length resulting from imperfect fabrication of the grating. As can be seen from the above calculations, such fluctuations will to lowest order appear as simply a fluctuating residual phase-mismatch in the equations for uniform $\chi^{(2)}$ crystals.[13] Thus an equivalent question would be how fluctuations in the phase-mismatch will affect the conventional $\chi^{(2)}$ solitons.

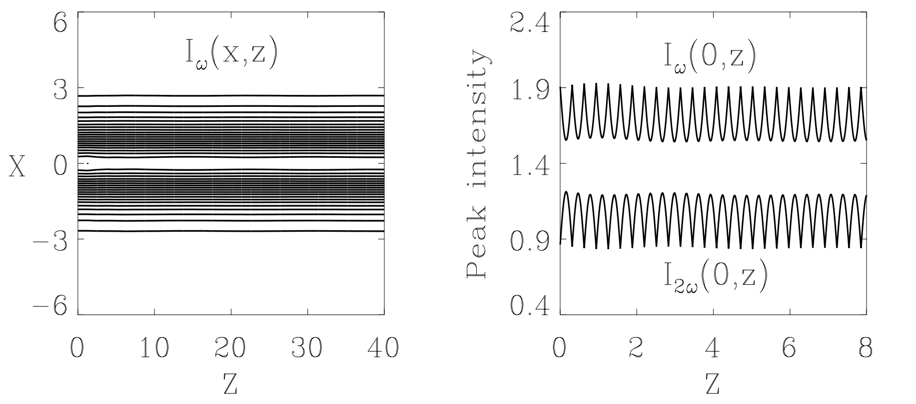

Fig.3. Excitation of a soliton in a slab waveguide with first-order QPM for $k_g = \Delta k = 10$. (a) Contour plot of the intensity of the FW, $I_\omega = |E_\omega(x,z)|^2$, sampled at intervals $4L_c$. (b) Peak intensities of the FW and SH.

Using this relation, our initial numerical investigations showed that random fluctuations of the domain length reduce the phase correlation and act as an effective loss to the QPM solitons.[14] Subsequent more rigorous numerical simulations on the full system by Torner and Stegeman have confirmed this and also shown, that if the fluctuations have long-range correlations, then the solitons are strongly affected and diffract rapidly.[15]

B: Switching properties

In collaboration with the group of Professor Lederer at the Friedrich-Schiller-Universität at Jena, Germany, in particular A. Kobyakov, we have studied the influence of the QPM induced cubic nonlinearity on the amplitude and phase modulation of the FW in channel waveguides.[17] By solving the effective averaged equations analyticaly we have been able to predict configurations for efficient all-optical CW switching.

An example is given in Fig. 4, where we see that a week control signal (difference in input FW intensity cannot be seen with the eye), can lead to a phase-shift of π after one cycle of interaction.[17] The analytical results are beautifully confirmed by the numerical simulations of the dynamical equations (1-2). There are some subtle points about these results. First of all, they require a non-zero residual phase-mismatch, i.e. one has to make the domain period slightly different from the coherence length. Furthermore, this switching property comes at the expense of a rather large holding intensity.[17]

It is interesting that the analytical results of our group are qualitatively the same as numerical results obtained in Australia by Zhao, Town and Sceats in 1995, who studied poled optical fibres with the inherent material cubic nonlinearity taken into account.[16] Thus both the material and induced cubic nonlinearities seem to have comparable effects.

In any case, the introduction of a nonlinear grating in a $\chi^{(2)}$ medium can have interesting and advantageous effects, which are certainly worth studying in more detail. In collaboration with G. Town from the University of Sydney and A. Kobyakov, who will visit the OSC this year for a longer period, we plan to experimentally and theoretically study the effects of cubic nonlinearity in poled fibres in order to determine whether the induced Kerr effects can be strong enough to be detected at all.

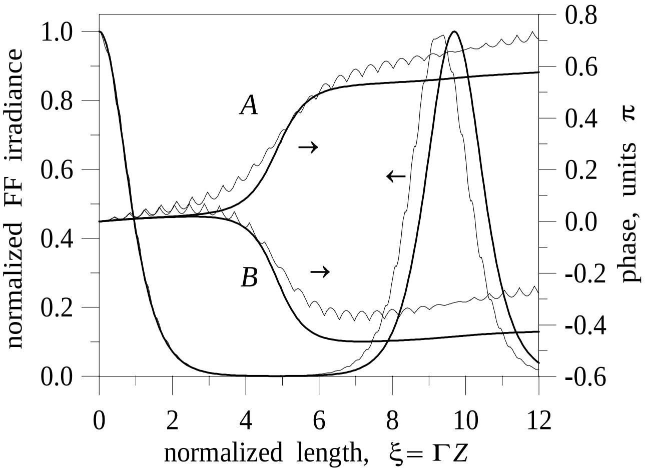

Fig.4. Thick lines: Analytically calculated evolution of the intensity and phase of the FW in a channel waveguide. Thin lines: Results of direct numerical integration.

Acknowledgements

I would like to thank my colleagues Yuri Kivshar, Peter L. Christiansen, Carl Balslev Clausen, Falk Lederer, Andrey Kobyakov and Graham Town for their many contributions to this research into QPM. Part of the work was supported by the Australian Department of Industry, Science and Tourism, grant no. 74.

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