1-1-2003

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**Recommended Citation**

Polyakov, Sergey; Jankovic, Ladislav; Kim, Hongki; Stegeman, George; Carrasco, Silvia; Torner, Lluis; and Katz, Mordechai, "Properties of quadratic multi-soliton generation near phase-match in periodically poled potassium titanyl phosphate" (2003). *Faculty Bibliography 2000s*. 3967.  
https://stars.library.ucf.edu/facultybib2000/3967
Properties of quadratic multi-soliton generation near phase-match in periodically poled potassium titanyl phosphate

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Abstract: The properties of the multi-quadratic-soliton generation process have been investigated both theoretically and experimentally near and on phase-match in non-critically-phase-matched, periodically poled, potassium titanyl phosphate (PPKTP). It was found that multi-soliton generation occurs primarily due to asymmetry in the input beam and at phase-matching. The number of solitons generated depended on the input intensity in a non-trivial way.

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OCIS codes: (190.5530) Pulse propagation and solitons, (190.4410) Nonlinear optics, parametric processes.

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1. Introduction

Quadratic spatial solitons were predicted in the 1970s to consist of multi-frequency waves coupled together via parametric mixing through the second order nonlinearity $\chi^{(2)}$ [1]. They have been studied extensively by inputting a fundamental field in second harmonic generation geometries for which the stationary quadratic soliton consists of in-phase fundamental and harmonic fields with a specific amplitude ratio [1-10]. The usual up- and down-conversion processes ($\omega + \omega \rightarrow 2\omega$ and $2\omega - \omega \rightarrow \omega$ respectively) then produce the appropriate second harmonic field with distance into the crystal. During this process there are non-adiabatic periodic oscillations of both the fundamental and harmonic fields which lead to the generation of radiation fields that are emitted into cones about the propagation axis [11,12]. Since the generation of quadratic solitons in bulk media by inputting only a fundamental is typically 50% or less efficient, a great deal of electromagnetic radiation is available in these cones [13]. The simplest second harmonic generation geometry for investigating multi-soliton generation is a non-critically-phase-matched (NCPM) one [7,8]. The experimental investigations discussed here used a periodically poled KTP crystal with co-polarized fundamental and harmonic fields.

It has been shown numerically that there are two mechanisms which break the cylindrical symmetry of these radiation fields and can lead to multiple solitons forming out of these radiation cones [11,12]. One occurs when asymmetric beams are incident, leading to spatially anisotropic up- and down-conversion processes and hence to multiple solitons being generated [12]. A second is the anisotropic diffraction that is a consequence of the directional anisotropy in the refractive index in planes orthogonal to the propagation direction and leads to multi-soliton generation along directions of minimum diffraction [11]. It was shown that both the effects can interfere either constructively or destructively [12]. Furthermore, the range of values of the phase-mismatch for which multi-solitons are efficiently generated is found to be small. Finally, the dependence of the number of solitons generated is demonstrated experimentally to have a complex input intensity dependence. Contrary to intuition, it does not increase indefinitely with intensity, rather multisoliton generation only occurs in a band of input powers. Here we report a detailed study of the process.

2. Experimental aspects

The fabrication and nonlinear characterization of these PPKTP samples is well-documented elsewhere [14-16]. Periodic poling with a period of 8.99\,\mu m along the crystal’s c-axis results...
in a first order effective nonlinearity of 9.5pm/V for NCPM at 1064nm at a temperature of 42.7°C for propagation along the a-axis (x-axis) [14-16]. For the 1 cm (=L) long crystal used, the SHG bandwidth has been measured to be 0.2 nm, in good agreement with [14]. This crystal is biaxial so that a low intensity, initially circularly symmetric beam propagating in the bulk material diffracts at a different rate along the “b” and “c” axis and becomes elliptical on propagation.

Fig. 1. Experimental setup. The prism (inset) can be moved into the path of the beam to alter the direction of the beam asymmetry (but not its shape).

The experimental layout is shown in Fig. 1. An EKSPLA, 10 Hz, Nd:YAG laser operating at 1064 nm was used for all of the experiments described here. The prism can be moved into the laser beam’s path which effectively reflects any asymmetry of the input beam about the plane containing the beam path and the z-axis [12]. Both 25ps (bandwidth = 0.14 nm) and 50ps (bandwidth=0.11 nm) pulses were used and the very good quality output beam from the laser was further improved by spatial filtering to give beam quality factors $M^2 = 1.0$. The input beam waist ($1/e^2$ of intensity) at focus was about 17 μm which corresponds to 6.4 diffraction length in the PPKTP sample. For both input and output pulses, their energies were monitored on a shot-to-shot basis by energy detectors, as well as their spatial distribution by cameras. The typical shot-to-shot variation in the pulse energies was ±2%, rms.

3. Numerical analyses

KTP (and hence PPKTP) is a biaxial crystal. For a-axis (x in equations below) propagation, a planar cut of the index ellipsoid for c-axis (z in equations below) polarization yields an ellipse, i.e. an incident circular beam becomes elliptical on propagation. Thus the coupled mode equations appropriate to describing quadratic soliton generation in this bulk, optical medium are [17]

$$i \frac{\partial A_1}{\partial x} + \left( D_{11} \frac{\partial^2 A_1}{\partial z^2} + D_{12} \frac{\partial^2 A_2}{\partial y^2} + \frac{\alpha_c(\omega)}{2} |A_1|^2 A_1 + \frac{\alpha_c(\omega)}{2} |A_2|^2 A_2 - \Gamma A_2^* A_1^* \right) \exp(-i\Delta k x) = 0 \text{ (1)}$$

$$i \frac{\partial A_2}{\partial x} + \left( D_{21} \frac{\partial^2 A_2}{\partial z^2} + D_{22} \frac{\partial^2 A_1}{\partial y^2} + \frac{\alpha_c(\omega)}{2} |A_2|^2 A_2 + \frac{\alpha_c(\omega)}{2} |A_1|^2 A_1 - \Gamma A_1^* A_2^* \right) \exp(-i\Delta k x) = 0 \text{ (2)}$$
where $A_1$ and $A_2$ are the fundamental ($\omega$) and harmonic ($2\omega$) amplitudes and the $\alpha_2$ are the two photon absorption (TPA) coefficients (which are known to be large primarily at the harmonic wavelength). In these equations $D_{11}, D_{12}, D_{21}$ and $D_{22}$ stand for the diffraction of a fundamental wave (FW, first index is 1) and second harmonic (SH, first index is 2) along the $z$ (second index is 1) and $y$ (second index is 2) axes respectively, $\Gamma$ is proportional to the coefficient of quadratic nonlinearity and $\Delta k$ is the wavevector mismatch between the fundamental ($k_i, i=1$) and second harmonic ($k_i, i=2$) wavevectors, i.e. $\Delta k = 2k_1-k_2$. Note that since these equations have different diffraction coefficients along the beam transverse dimensions $z$ and $y$ they do not preserve circular symmetry, as opposed to an isotropic medium in which $D_{11}=D_{12}$ and $D_{21}=D_{22}$.

A form of the Beam Propagation Method (BPM) with the 4th order Runge-Kutta scheme for solving of the nonlinear part was used to numerically calculate the evolution of a CW and a fully 3D spatio-temporal input beam under the influence of the equations above [11,12]. The typical numeric windows were 512x512 with 20x20 grids per beam size and 256x256x256 with 40x40x40 grids per beam size and pulse width for the CW and (3+1)D simulations respectively. In addition, the windows sizes and grid densities were varied to verify the absence of mesh and boundary effects. The total energy was found to be conserved in the calculations. For the input fundamental beam, an ellipse with a Gaussian shape of the form $A_i(z,y) = A_i \exp[-z^2/w_1^2 - y^2/w_2^2]$ was assumed. Here $w_1\neq w_2$ for an elliptical input beam and $w_1=w_2$ for a circular symmetric beam.

4. Multi-soliton generation: On phase-match
4.1 Simulations
Steady-state (stationary) solutions of Eqs. (1) and (2) have been obtained numerically in the absence of TPA, for bulk media by a number of authors in the limit of isotropic diffraction [18]. For the more relevant anisotropic diffraction case, the quadratic solitons in general have an elliptical shape, are stable, and form a two parameter family, that is two parameters are needed to specify the solitons and their properties [17]. One describes diffraction anisotropy and the other is related to both wavevector mismatch and the peak intensity (nonlinearity). This means that for a given experimental geometry (which specifies $\Delta k$ and anisotropy), increasing the peak intensity of, for example, the fundamental leads to a decrease in the width of both soliton components. This narrowing of the soliton’s field distribution with increasing peak intensity is very similar to that found with Kerr solitons [19].

Experimentally, as discussed in the introduction, the solitons are excited by inputting the fundamental beam only and relying on $\chi^{(2)}$ parametric processes to generate the appropriate harmonic component. This is quite different from inputting the stationary solutions which contain in-phase fundamental and harmonic components of a specific amplitude ratio. Simulations have shown that both the fundamental and harmonic oscillate with distance with the peak oscillations dying out with distance from the input facet [9,10]. Radiation is emitted during these oscillations, and, as the peak intensity is increased, progressively more of the input intensity appears in the radiation cones emitted at small angles from the incidence axis [11,12].

Shown in Fig. 2 is the calculated evolution of the fundamental and harmonic beams for the case of PPKTP for an input intensity about three times that of the single soliton’s threshold intensity (which was established experimentally to be $=3.4 \text{ GW/cm}^2$ for a beam waist of about 17 $\mu$m) [8]. Assumed here is a perfectly circular input beam. Although in actuality they are slightly elliptical, the cuts of the radiation cones orthogonal to the incident beam propagation axis appear approximately as circles with specific radii centered on the beam axis. Note that self-trapped beams are formed on the second radiation cone. It is clear that the solitons coalesce from intense radiation fields far from the central on-axis soliton. Also, the
trajectories of the two satellite solitons diverge with the net transverse momentum conserved. Slow coalescence of side solitons is a typical but not the only possible outcome. In some cases, determined by the material parameters and beam intensities, the complex beam propagation may result in appearance of two side solitons much closer to the beam axis which get the energy of the central feature, yielding only two solitons at the output.

Multiple solitons are also emitted when the input beam has an elliptical shape in a medium in which diffraction is spatially isotropic about the propagation direction. It has been shown in reference 17 that in certain limits this case can be recast mathematically into the anisotropic diffraction case. In PPKTP the diffraction anisotropy is about 11% for the FW \((D_{11}/D_{12})\) and 13\% \((D_{21}/D_{22})\) for the SH. Therefore, to a good approximation \(D_{11}D_{22}=D_{12}D_{21}\) and hence a rescaling of one or both of the transverse co-ordinates can return the equations to circular symmetry. This rescaling also affects a perfectly circular incident beam transforming it into an elliptical beam in the rescaled co-ordinates. Naturally, the reverse operation, i.e. rescaling of the coordinates to bring an elliptical input beam to a circular shape would introduce an effective diffraction anisotropy. Therefore within this approximation, the evolution of the multiple solitons patterns for an elliptical incident beam with its axes being different by 5\% in an optically isotropic medium will be same as for anisotropic diffraction with a circular symmetric incident beam with a difference in the ratio of major to minor diffraction coefficients of about 11\%. Such a beam asymmetry is very small and difficult to achieve and control experimentally. Hence with typical input beams one expects the beam asymmetry to dominate the multi-soliton generation process. In the absence of anisotropic diffraction, the solitons are aligned along the major axis of the input beam. When both mechanisms are present, the anisotropic diffraction of PPKTP and beam shape contributions can either interfere constructively or destructively, depending on the detailed circumstances [12]. Note that it is possible for the two effects to cancel each other and hence for multi-soliton generation to be suppressed. However, for materials with anisotropic diffraction far from the quasi-isotropic limit discussed here the interference of these two effects is more complex and cannot be characterized as simply “constructive” or “destructive”.

Fig. 2. (456 KB) Simulation of the spatial evolution of three soliton generation on phase match with a high input intensity fundamental beam (three times higher than single soliton threshold 3.4GW/cm², where the input intensity of 1 a.u. in the simulations corresponds to approximately 14GW/cm²). Left-hand-side the fundamental; Right-hand-side the harmonic. The propagation distance between successive frames is about 0.1 in units of diffraction lengths. Simulation parameters: \(D_{11}=0.0943\), \(D_{12}=0.0847\), \(D_{21}=0.0478\), \(D_{22}=0.0423\), \(\alpha_{1,2}=0\), input intensity=0.73 a.u., and the Eqs. (1), (2) are normalized to set \(\Gamma=0.01\).

In contrast to the anisotropic diffraction case for which the diffraction parameters are fixed by the material’s wavelength and propagation direction, the beam ellipticity is a parameter which can be varied. The threshold for multi-soliton generation depends on the
magnitude of this ellipticity. Finally, note that the larger the ellipticity, the closer to the input boundary multiple solitons start to form [12]. Conversely, the smaller the net anisotropy (ellipticity + diffraction anisotropy), the longer the crystal needs to be for multiple solitons to be observed.

The input intensity dependence of the number of solitons generated reflects the dominant mechanism operative, how far from the input beam axis the solitons are formed and the forces between the solitons. The CW prediction for a circularly shaped beam input into the anisotropic PPKTP is shown in Fig. 3. For a given crystal length, there is a well-defined threshold (for a well-defined beam asymmetry and/or anisotropy) for the onset and disappearance of multi-soliton generation. In-phase solitons are formed near the incident beam axis and they experience attractive forces and energy exchange with both the central soliton and each other due to their small initial separation. They are also formed with the transverse momentum associated with the diverging radiation ring. Thus their subsequent dynamics involves a trade-off between this initial transverse momentum and the attractive forces. As a result, it is even possible in the CW case for the central soliton to be eliminated completely and all of the self-trapped energy to appear in the satellite solitons. When the divergence angle of the satellite solitons is positive, they continue to diverge indefinitely [11]. As the input intensity increases, the attractive forces become stronger and the net divergence becomes weaker. A negative divergence angle means that the additional solitons are now converging from their coalescence point towards, but have not yet reached, the propagation axis for the length of crystal chosen. For large enough incident intensities the attractive forces are strong enough to force all of the localized beams to collapse back to the center soliton.

Inclusion of 2PA into the equations results in a larger intensity range over which three, apparently self-trapped beams can be observed for a specific crystal length. Since these beams decay and broaden with propagation distance, their definition as solitons can be questioned. Furthermore, the beam waists in the presence of 2PA are larger than its absence, in better agreement with the experimental values. If the 2PA contribution is too large, it can also affect other properties of multisoliton generation, such as the spatial alignment of the output pattern.

Fig. 3. The divergence angle (a.u.) and number of solitons predicted by CW simulations after 5 diffraction lengths as a function of input intensity (in arbitrary units). A gaussian beam input with $w_1 = w_2$ was assumed with the anisotropic diffraction appropriate for PPKTP.

This strong dependence on the input intensity reflected in Fig. 3 implies that, if the excitation is made with pulsed light, different parts of the pulse will undergo different dynamical routes [12]. An example of a full 3D spatio-temporal simulation for a given input intensity and beam ellipticity, at 10 diffraction lengths is shown in Fig. 4. Each shot of the
movie corresponds to the field spatial distribution taken at different times (sketch in the inset) along the pulse.

Fig. 4. (172 KB) FW spatial field for different temporal snapshots at 10 diffraction lengths for an elliptical input beam with \( w_1/w_2=1.07 \). The inset sketches the shape of the temporal profile and the moving dot shows the location of the temporal slice shown.

The stability of the satellite solitons and their monotonic divergence with distance was checked by continuing the beam propagation for many tens of diffraction lengths. This rules out any interpretation of the observed structures in terms of multi-hump solitons for the current excitation conditions [21].

4.2 Experiments

Experiments were performed on PPKTP. First it was verified that either single or multi-soliton generation can be observed in this sample, depending on the input beam conditions. The results in Fig. 5(a) showed experimentally single soliton generation for incident peak intensities up to 20 GW/cm\(^2\). The input beam ellipticity for this case was about 5\(\pm\)2\%. Furthermore, the orientation of this ellipticity was appropriate for reducing the effects of the crystal’s diffraction anisotropy. Based on the simulations, the net asymmetry is too small to produce multisoliton generation for the crystal length employed with the incident peak intensities up to 20 GW/cm\(^2\). Shown for comparison in Fig. 5(b) is a demonstration of multi-soliton generation for an incident beam with a larger ellipticity. Clearly multi-soliton generation occurs.
Fig. 5. Fundamental (FW) and second harmonic (SH) output patterns for two cases. (a) Single soliton generation. (b) Multi-soliton generation for a strongly asymmetric input beam. Insets show the input beams.

Under conditions of three soliton generation, it was then verified that the dominant mechanism responsible for the preferred direction of multisoliton generation was beam asymmetry [12]. The initial indication usually was that the multisoliton pattern was not aligned along a crystal axis. This was verified by sliding the prism into the path of the beam before the sample to effectively reflect the input beam in the plane containing the polarization and propagation axes. It was demonstrated experimentally that the alignment of the resulting soliton pattern mirrored the change in the major ellipse axis. Note that the generation of three solitons first occurred at intensities larger than three times the threshold intensity for single soliton generation.

Figure 6 shows a collage of output patterns from the crystal as a function of increasing input intensity of the fundamental for elliptical input beams with a larger ellipticity than in Fig. 5(a). As expected, beam narrowing occurs first, leading to single soliton generation. Around 15 GW/cm² three solitons first appear in Fig. 6. Note that the three soliton formation disappears at high intensities, although there is still some elongation of the single remaining soliton along the direction in which the multi-solitons occurred. This is in excellent agreement with the theoretical predictions in view of the fact that pulsed lasers were used so that each experiment involves a distribution of intensities in time and no sharp thresholds are expected.
5. Multi-soliton generation: Off phase-match

Numerical simulations show that multi-soliton generation occurs only under conditions that in practice (i.e., taking into account the finite accuracy of the experimental set-up) correspond to phase-matching. Fig. 7 shows the calculated cw evolution of the input beam for $\Delta kL \approx 15\pi$ as a function of distance into the sample for the same beam etc. conditions as used previously in Fig. 2 (which had $\Delta kL = 0$). Clearly, no multi-soliton generation is predicted to occur.

A collage of the experimental patterns obtained at the exit facet of the crystal is shown in Fig. 8. Clear multi-soliton generation occurs only over a small range of positive phase-mismatch. Again, given the pulsed nature of the excitation, the results are in very good agreement with the simulations.

There are good reasons why multi-soliton generation occurs solely near and at the phase-matching condition. The radiation rings by themselves do not in general have the threshold intensity needed to form the solitons. Mutual focusing of the fundamental and harmonic is necessary along arcs of the cone via the usual mechanisms of “beam narrowing” and “cascading” [20]. This requires an exchange of photons between the two frequency components and has a minimum threshold for soliton formation at zero phase-mismatch. This threshold rises with phase-mismatch faster for negative mismatch than for positive mismatch, explaining why the multi-soliton generation is observed for a larger range of $|\Delta kL|$ with $\Delta kL > 0$. 

Fig. 6. (1.33 MB) Multiple shots of the fundamental beam output patterns obtained with increasing the intensity from 0.5 GW/cm$^2$ to 26 GW/cm$^2$. 

#2400 - $15.00 US Received April 18, 2003; Revised May 22, 2003
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Fig. 7. (404 KB) Calculated evolution with propagation distance of the fundamental (left-hand-side) and the harmonic (right-hand-side) at $\Delta kL \approx 15\pi$ under the same conditions as Fig. 2 (for which $\Delta kL=0$).

6. Summary

It has been shown here that the generation of multiple quadratic solitons in periodically poled KTP is a complex process which is a consequence of using a fundamental beam only at the input. In the actual experiments, the additional solitons are formed by the off-axis, anisotropic break-up of the input beam. Because of the pulsed light conditions, the formation of off-axis solitons might occur together with the generation of an on-axis soliton. This leads to a complex behavior involving a minimum intensity threshold for multi-soliton generation and an upper threshold after which only single solitons are obtained. These features were verified experimentally. In addition, it was found that multi-soliton generation occurs only near exact phase-matching.

This research was supported by an Army Research Office MURI on Solitonic Gateless Computing and by the Generalitat de Catalunya and by the Spanish Government through grant TIC2000-1010.