Simultaneous phase matching and internal interference of two second-order nonlinear parametric processes

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Abstract: We demonstrate the simultaneous generation and internal interference of two second-order parametric processes in a single nonlinear quadratic crystal. The two-frequency doubling processes are Type 0 (two extraordinary fundamental waves generate an extraordinary second-harmonic wave) and Type I (two ordinary fundamental waves generate an extraordinary second-harmonic wave) parametric interactions. The phase-matching conditions for both processes are satisfied in a single periodically poled grating in LiNbO3 using quasi-phase-matching (QPM) vectors with different orders. We observe an interference of two processes, and compare the results with the theoretical analysis. We suggest several applications of this effect such as polarization-independent frequency doubling and a method for stabilizing the level of the generated second-harmonic signal.

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References and links


1. Introduction
Cascading of several parametric processes in a single nonlinear quadratic crystal results in many new phenomena in nonlinear optics including the generation of large nonlinear phase shifts [1], cascaded generation of the third and forth harmonics, polarization switching, generation of solitary waves [2], etc. Among these cascaded interactions, an important role is played by two simultaneous frequency doubling processes. Generally, these processes can be divided into two large groups: (i) the processes that share one or both fundamental waves and (ii) the processes where two independent fundamental waves, e.g. two orthogonally polarized waves (A and B), generate one second-harmonic (SH) wave. The first group of the parametric processes has been studied extensively, both theoretically and experimentally [3-7]. The second group of parametric processes, which we can denote symbolically as `AA-S: BB-S’ has been studied only theoretically. The first theoretical proposal to employ this kind of parametric process was described in Ref. [8]. Later, in Ref. [9] it was shown that, as a result of the second-order cascaded interaction, one of the fundamental waves propagates in a waveguide created by a SH beam, and the conditions for soliton propagation can be realized. Grechin and Dmitriev [10] studied theoretically these parametric interactions from the viewpoint of improving efficiency of the second-harmonic generation (SHG) process. Calculations of domain structures for dual-phase-matched SHG at a fundamental wavelength of 964 nm are presented in Ref. [11]. However, to the best of our knowledge, no experimental results have been reported so far which deal with the coexistence of these two parametric processes in a single nonlinear quadratic crystal. The main purpose of this paper is to demonstrate that in one-dimensional periodically poled structures we can realize suitable conditions for the experimental observation of this novel type of the double-phase matched (DPM) parametric interactions which can result in an interference of the SH waves generated by two simultaneous frequency-doubling processes. We discuss the power exchange between two fundamental waves as a result of the cascaded third-order four-wave mixing. We compare our theoretical analysis with experimental results and demonstrate an excellent agreement.

The paper is organized as follows. In Sec. 2 we discuss the phase-matching conditions and introduce our theoretical model. In Sec. 3 we present details of the sample preparation and describe our experimental setup. Section 4 presents our experimental results and discussions as well as a comparison with the theory. Some possible applications are discussed in Sec. 5

2. Theoretical background
Simultaneous generation of two kinds of second-harmonic parametric processes is not a trivial task because two phase-matching (PM) conditions, $\Delta k_1=0$ and $\Delta k_0=0$ for the Type I and Type 0, respectively, should be satisfied simultaneously. Lithium niobate (LiNbO 3) is a popular
material for implementing quadratic nonlinear processes due to its good nonlinear response, large transparency window covering the visible and near-mid infrared, its availability as a cheap and high quality material, and its associated technologies such as proton exchange waveguides and periodic poling for QPM. For QPM of a DPM process in poled LiNbO₃, two phase-matching conditions must be simultaneously satisfied. The first condition is for SHG with ordinary fundamental waves (y-polarized), which we denote as \( Y_{1Y1-Z2} \). The second condition is for SHG with extraordinary (z-polarized) fundamental waves, which we denote as \( Z_{1Z1-Z2} \). The QPM conditions for these processes is 
\[
\Delta k_i = k_{z2} - 2k_{y1} - m_i G
\]
and
\[
\Delta k_o = k_{z2} - 2k_{z1} - m_o G \, ,
\]
where \( m_i \) and \( m_o \) are the orders of the QPM vector, \( G = 2\pi/\Lambda \) and \( \Lambda \) is the period of the QPM grating. Both the processes generate extraordinary (z-polarized) SH waves.

For the fundamental wavelength (1064.5 nm) generated from the Nd:YVO₄ laser used in these experiments, we find two suitable periods in periodically poled LiNbO₃ for simultaneous PM of two SHG processes: (a) the period 45.77 µm at 179.53°C with \( m_1 = 1 \) and \( m_2 = 7 \), and (b) the period of 32.03 µm at 242.1°C with \( m_1 = 1 \) and \( m_2 = 5 \). In this paper, we present the experimental results for a 45.77 µm (\( m_1 = 1, m_2 = 7 \)) periodically poled grating such that the DPM process is \( Y_{1Y1-Z2} \) (1st order): \( Z_{1Z1-Z2} \) (7th order).

Equations for plane waves that describe the process of generating one and the same second harmonic wave from two differently polarized fundamental waves (in our case, polarized orthogonally along the Y and Z axes in LiNbO₃), assuming no losses for the fundamental and SH waves and neglecting possible temporal delay effects, have the following form [8]:

\[
\frac{dA}{dx} = -i\sigma_1 A^* \exp\left(-i\Delta k_1 x\right)
\]
\[
\frac{dS}{dx} = -i\sigma_2 A^2 \exp\left(i\Delta k_1 x\right) - i\sigma_4 B^2 \exp\left(i\Delta k_0 x\right)
\]
\[
\frac{dB}{dx} = -i\sigma_3 S B^* \exp\left(-i\Delta k_0 x\right)
\]

where \( A \) denotes the fundamental field polarized along \( y \) axis, and \( B \) denotes the fundamental field polarized along \( z \) axis. \( S \) is the second harmonic field, which in LiNbO₃ is polarized along the \( z \) axis. Furthermore, \( \sigma_1 = \frac{2\pi d_{31}}{\lambda_1 n_y} f_1 \), \( \sigma_2 = \frac{2\pi d_{31}}{\lambda_2 n_z} f_1 \), \( \sigma_3 = \frac{2\pi d_{33}}{\lambda_3 n_z} f_2 \), and \( \sigma_4 = \frac{2\pi d_{33}}{\lambda_2 n_z} f_2 \), where \( d_{33} = d_{zz} \) and \( d_{31} = d_{yz} \) are the LiNbO₃ nonlinear coefficients, \( f_1 = 2/(m_1 \pi) \), \( f_2 = 2/(m_2 \pi) \) are the QPM reduction factors. The mismatch parameters \( \Delta k_i \) and \( \Delta k_0 \) defined above depend strongly on the temperature. For the numerical calculations with this system of Eqs. (1-3), we replace \( f_1 \) and \( f_2 \) with \( f_1 = f_2 = F(x) \) (the poling function), describing the actual shape of the QPM grating. The phase mismatches \( \Delta k_i \) and \( \Delta k_0 \) have been calculated using Sellmeier data from [12].

The set of Eqs. (1-3) can become uncoupled under certain conditions. First, if we consider the low-power (small signal) regime, the depletion of two fundamental waves can be neglected and Eq. (2) can be solved analytically for the SH field. The solution for this case can directly describe the effect of the internal interference of the two generated SH fields. The resulting solution of Eq. (2) with respect to the squared amplitude of the SH wave (proportional to the intensity) is:

\[
|S|^2 = A^2 \text{sinc}(\Delta k_1 L/2) \exp\left(-i\phi L/2\right) + \frac{gm_1 d_{31}}{m_2 d_{33}} B^2 \text{sinc}(\Delta k_0 L/2) \left(\frac{\Delta d_{33}}{\lambda_2 n_z} L\right)^2
\]
where \( \phi(T) = \Delta k_t - \Delta k_i = 2\pi \left[ 2(n_x - n_y)/\lambda + (m_x - m_y)/\Lambda \right] \). At the QPM temperature required for DPM, \( \phi(T) = 0 \) since \( \Delta k_0 = 0 \) and \( \Delta k_i = 0 \). If \( A \) and \( B \) are obtained with a half wave plate (HWP) producing a linearly polarized fundamental rotated at angle \( \theta \), then the amplitude of the two orthogonal fundamental components will be \( A = A_0 \cos 2\theta \) and \( B = A_0 \sin 2\theta \) for an initial amplitude of \( A_0 \). A quarter wave plate (QWP) situated after the HWP can introduce a \( \pi/2 \) phase shift between the two fundamental waves. This is introduced in Eq. (4) by the parameter \( g \) which is \( g = -1 \) with the QWP and \( g = +1 \) without the QWP. In Fig. 1(a) the dependences of the second harmonic intensity on the HWP angle are shown for the cases of \( g = \pm 1 \), and for two different ratios of the contributions from both fundamental waves based on the upper and lower limits for the published values of the nonlinear coefficients \( (d_{33}/d_{31}) = 0.5 \) and \( d_{33}/d_{31} = 0.9 \). It is clearly seen how the internal interference of the two SH waves after the introduction of QWP causes the generated SH signal to be reduced to zero when both processes are generating a SH waves with equal amplitude but out of phase with \( \pi \). Assuming a 50 % duty cycle the HWP the angle at which the SH signal will be reduce to zero will be \( \theta = \frac{1}{2} \arctan \left( \frac{m_x d_{31}}{m_y d_{31}} \right) \). The curves with and without QWP will have alternate trends if the signs of the two nonlinearities are opposite, thus the effect can be used as a method for measurement of the relative sign of the nonlinearities involved. Also the ratio between the maximum and minimum SH output on the curves generate with no QWP can be used for estimation of the ratio of the relevant nonlinear components.

Fig. 1. Simulations of the generated the second-harmonic output for Z:Z1-Z2:Z1 double phase matching in LiNbO3 for the two different ratios of effective nonlinearities \( d_{33}/d_{31} = 0.5 \) (top) and \( d_{33}/d_{31} = 0.9 \) (bottom). The second-harmonic output is plotted against: (a) HWP angle which changes a ratio between the amplitudes of two fundamental components; (b) temperature detuning near the exact phase-matching temperature for the frequency-doubling processes (179.53°C). The cases when two fundamental waves are in phase (\( g = +1 \)) or shifted out-of phase (\( g = -1 \)) are shown by blue-dash or red-solid lines, respectively.

In Fig. 1(a) we are at the ideal phase-matching conditions so the phase factor is \( \phi(T) = 0 \). If the processes are not perfectly phase matched such that \( \Delta k_i, \Delta k_0 \neq 0 \) then \( \phi(T) = 0 \) and will have a finite value and affect both the phase and the efficiency of the process described by Eq. (4). In Fig. 1(b) the second harmonic intensity is plotted as a function of temperature tuning with the HWP rotation angle fixed at 22.5°. The two different ratios of the contributions from both fundamentals are shown. The interference effects are present and lead to an increase or
decrease of the net SH signal at the peak of the temperature curve. In the high power regime it is no longer possible to neglect the depletion of the fundamental waves. In this case the set of Eqs. (1-3) is solved numerically with the poling function \( F(x) = \text{signum}(\sin(2\pi x / A)) \).

Depending on the signs of the two nonlinearities (or the phase shift between the two fundamental fields) one can observe a collective contribution of both fundamentals to the SH efficiency, or the exchange of energy between the fundamental fields. The resulting dependence of the intensity of the two fundamental waves and the SH signal is shown in Fig. 2 as a function of the normalized crystal length \( L / L_n \) (with \( L_n = 1 / (\sigma_i A_i) \)). Figure 2(a) shows the case of the same signed nonlinearities with \( \pi/2 \) shifted fundamentals such that \( g = -1 \) (or equivalently differently signed nonlinearities with fundamentals in phase) and Fig. 2(b) shows the same sign nonlinearities with fundamentals in phase such that \( g = +1 \) (or equivalently different signed nonlinearities with \( \pi/2 \) shifted fundamentals). The power exchange between the fundamental waves in Fig. 2(a) is the result of a third order process due to \( \chi^{(2)} : \chi^{(2)} \) cascading. This cascaded third order process is analogous to a four wave mixing process. The equivalent process can be symbolically marked as B:AB*A. To illustrate this interpretation let us assume that one of the pumps (B) is much weaker than the other and neglect the depletion only for the stronger fundamental wave. Then for the fundamental wave B at exact temperature for DPM we have:

\[
B(L) = \Re[\beta(0)\exp\left(-\frac{1}{2}\sigma_3\sigma_2 A^2 L^2\right)] + i\Im[\beta(0)\exp\left(\frac{1}{2}\sigma_3\sigma_2 A^2 L^2\right)]
\]  

(5)

For the small signal regime, or short crystal lengths when arguments of the exponents are small Eq. (5) is reduced to:

\[
B(L) = B_o + \sigma_3\sigma_2 B_o A A^2 L^2
\]  

(6)

where \( \sigma_i, \sigma_3 \) is proportional to the product, \( d_{33}^{(2)} d_{31}^{(2)} \) proving that the change of the amplitude of the fundamental wave B is due to a third order process driven by \( \chi^{(2)} : \chi^{(2)} \) cascading. For the case where B is not weak compared to A we can numerically integrate the system of Eq. (1-3) to gain insight into the parametric process taking place. Figure 2 demonstrates the case where A and B are initially of equal amplitude.
3. Experimental setup and samples

The experimental setup is shown in Fig. 3. The laser used is a Nd:YVO₄ laser with a wavelength of 1064.5 µm, producing 10 ns pulses at 10 kHz and an average power of ~ 400 mW. A temperature controller with accuracy ±0.1°C was used for changing the temperature of the periodically poled lithium niobate (PPLN) sample. The input fundamental beams were focused inside the crystal with a 50 mm focal length singlet lens. The PPLN samples were fabricated from 500 µm thick, z-cut LiNbO₃ with a rapid prototyping method utilizing laser micro-machined topographical electrode geometries [13, 15]. In brief, 3mW average power from the 1kHz output of a Spectra Physics ‘Hurricane’ system delivered using a 10x objective lens and 6 passes per-scribe at a feed-rate of 50 mm/min was used to produce ~25 µm deep scribes in the crystal surface with an aspect better than 1.5. Two different period gratings were fabricated in the same sample, one with 45.75 µm and the other 45.79 µm. These periods were fabricated within the resolution of the translation stages, and the values have been inferred from the temperature tuning data in the following section. The poling was carried out using the electrolyte poling cell method with a saturated NaCl solution. Two consecutive 100 ms, 13.2 kV pulses were applied across the crystal to achieve good quality poling. The applied field is below the coercive field for the bulk crystals, 13.3 kV (26.6 kV/mm), so that domain reversal was restricted to the regions beneath the scribes and the width was determined by the depth of the scribes and the applied field. PPLN samples with 100 domains (4.6 mm long) and a duty cycle of ~ 35 % were produced using this technique. The mean value of the duty cycle is very close to the optimum for 7th order QPM (36%), and causes a decrease in the efficiency of the Type I SHG process of about 15 %.

4. Experimental results and discussions

Figure 4 shows the overlapping experimental PM curves together with the theoretical curves for the two frequency doubling processes: (i) Type I – Y₁Y₁-Z₂ (1st order) and (ii) Type 0 – Z₁Z₁-Z₂ (7th order). The theoretical curves are rescaled to match the maximum of the Type I SHG process. In the calculations we use the data for \(d_{33} = -25.3\) pm/V and \(d_{31} = -4.6\) pm/V from [14]. The full width half maximum (FWHM) of the experimental curves (1.7°C for Type I and 5.5-9.5°C for Type 0) can be compared with the theoretical ones (1.3°C for Type I and 4.1°C for Type 0 for L = 4.6 mm). The width of the Type 0 process is broader than the theoretical PM curve, which results from some imperfections in the grating caused by the poling process. We note that the Z₁Z₁-Z₂ frequency doubling process is much more sensitive to the imperfections in the grating, because of the 7th order QPM used. Figure 4 shows the experimental phase-matching temperature curves, taken separately launching only one of the fundamental polarizations in the crystal for each measurement. In the grating with the 45.75 µm period the peak PM temperatures for the Type I and Type 0 processes are separated by ~
3.5°C, which corresponds to a deviation of about 50 nm from the exact period required for DPM. The 45.79 µm period grating deviates from the exact DPM period by only 6 nm. The measured SH power for the Type I process for 350 mW input power reaches 21 mW, which corresponds to 6 % external (measured net efficiency) and 8.2 % internal efficiency for this process, (where the internal efficiency is calculated from the external efficiency, allowing for the Fresnel losses at the surfaces which may be recovered if anti-reflection coatings are used). The sufficient overlap in the PM curves demonstrated in these measurements allowed us to study the effects of simultaneous action of two frequency doubling processes by launching both components of the fundamental beam (Y and Z polarized) into the sample. Both fundamental fields are launched simultaneously into the crystal by using a HWP in front of the linearly polarized laser source. Changing the relative amplitudes of the two fundamental polarization components was realized by rotating the HWP. With the addition of a QWP, inserted after the HWP, we were able to introduce a $\pi/2$ phase shift between the two fundamentals. Consequently both the Type I and Type 0 SHG process were active in the PPLN, producing second harmonic fields that can interfere constructively or destructively. The sample temperature for the HWP rotation experiments was chosen to correspond to the peak conversion of the Type I process. The results of these measurements together with the theoretical curves are shown in Fig. 5.

![Fig. 4](image)

**Fig. 4.** Experimental (dots) and theoretical (lines) temperature phase-matching curves for SHG Type I (blue) and SHG Type 0 (red) in the 45.75 µm and 45.79 µm period gratings. 4(a) 45.79 µm grating with $\sim$6 nm deviation from the ideal calculated grating for DPM of the two processes. 4(b) 45.75 µm grating with $\sim$50 nm deviation from the ideal calculated grating for DPM of the two processes.

For the 45.79 µm sample it can be seen that the two processes are very close to exact double phase matching ($\phi = 0$), and the two generated SH fields interfere constructively when the two fundamental fields are in phase and destructively for the case of $\pi/2$ phase shifted fundamentals. This behavior indicates that the two nonlinear coefficients $d_{33}$ and $d_{31}$ have the same sign, in agreement with previous measurements [12,14]. For the other grating with period 45.75 µm the Type 0 process is not exactly phase matched. We estimate its normalized mismatch to be $\Delta k_0 L = -1.14\pi$ at the point of exact PM for the Type I process that is equivalent to introducing phase shift between the two SH fields equal to $\phi = -1.14\pi$ [Eq. (4)]. Existence of this phase shift causes reverse behavior of the two SH interference curves. The theoretical curves derived using the phase mismatch indicated by the temperature tuning curves plotted in Fig. 4 show good agreement with the experimental ones.
In another experiment the temperature tuning curves when components of both fundamentals are launched in the crystal were recorded. This experiment was performed for the 45.79 µm grating in a similar fashion to the individual temperature PM in Fig. 4. The HWP was set to 22.5° (equal amplitude for both fundamental waves) and then the SHG was monitored as a function of temperature for the cases of QWP in and out of the setup. In this case the phase matching curves exhibit features due to the interference of the two SH processes. Results of these measurements and the corresponding theoretical curves are shown in Fig. 6. Analyzing the curves presented in Figs. 5 and 6 we notice that internal interference depends on (i) the relative sign and magnitude of the nonlinearities involved, (ii) the relative phase between two fundamental waves and (iii) the distance “…ΔkL” between two processes. SH interference will be difficult to observe if the mismatch between the two processes is larger than 2π.

![Fig. 5](image1.png)

**Fig. 5.** Theoretical (lines) and experimental (dots) relationship between the SH power and the HWP angle which is controlling the input polarization, such that both the Y, Y, Z and Z, Z, Z processes are present. Blue traces indicate linear polarizations where only the HWP is in use. Red traces indicate elliptical/circular polarizations where a QWP is used in tandem with the HWP. 5(a) for QPM grating with 6 nm deviation from the ideal calculated grating for DPM of the two processes. 5(b) for QPM grating with 50 nm deviation from the ideal calculated grating for DPM of the two processes.

![Fig. 6](image2.png)

**Fig. 6.** (a). Experimental and (b) theoretical temperature dependences for simultaneous processes Y, Y, Z (1st order) : Z, Z, Z (7th order) in the 45.79 µm grating. The HWP is set so that the fundamental components (Y and Z) are equal. Blue-dash curves show SHG from fundamental waves which are in phase (HWP only), red-solid curves show SHG from fundamental waves which are π/2 out of phase (QWP inline).
5. Possible applications

The investigated scheme can be used for devices where polarization independent SHG is needed. The conceptual device would generate SH for any direction of fundamental input polarization, as long as the phase relations between the fundamental fields do not cause destructive interference of the SH. The output polarization of the generated SH beam will however be fixed parallel to the crystal Z axis. In order to ensure constant SH power for any angle of input polarization the design of the QPM grating will require careful design and fabrication so that the duty cycle factor \( D \) of the grating takes the right value. Indeed, if one takes into account the effective nonlinearity for each of the processes \([16]\),

\[
d_{\text{eff}}^{(1)} \text{ (Type I)} = \frac{2}{m_x \pi} d_{31} \sin (m_x D \pi) \quad \text{and} \quad d_{\text{eff}}^{(0)} \text{ (Type 0)} = \frac{2}{m_z \pi} d_{31} \sin (m_z D \pi)
\]

then the two SH processes will have equal efficiency if \( d_{\text{eff}}^{(1)} \text{ (Type I)} = d_{\text{eff}}^{(0)} \text{ (Type 0)} \), which will require the use of a duty cycle factor that is found from the numerical solution of the equation

\[
\frac{\sin (m_z D \pi)}{\sin (m_x D \pi)} = \frac{d_{31} m_x}{d_{31} m_z}
\]  

Another possible application is generation of stabilized second harmonic radiation. Many applications have very strict tolerances with regards to the allowed deviation from a particular average power. Figure 7 shows a conceptual device that will allow power stabilization of the second harmonic. In this case a HWP is fixed at the position where the two fundamental waves interfere constructively. Using the SH interference we have reported here, a Pockels cell (PC) induces a phase shift \( \varphi(V) \) which can partially reduce the efficiency of the generated SH signal to be at the desired application level. If for some reason the SH signal increases, the feedback signal changes the voltage \( V \) of the Pockels cell in the direction to reduce the level of the generated SH and vice versa. In the light of this application and assuming that both processes are phase-matched simultaneously Eq. (4) will be:

\[
|\alpha|^2 \propto \left| \lambda \exp[i \varphi(V)] + \frac{gd_{31}}{m_d d_{31}} B \right|^2
\]

The modulator, a transverse LiNbO\(_3\) Pockels cell, should be oriented such that its axes coincide with those of the frequency doubling PPLN. In LiNbO\(_3\) the electrooptic coefficients are of substantial different magnitudes, \( r_{33} > r_{31} \), such that any voltage applied to the Pockels cell will predominantly change the phase of one of the polarizations, leading to a voltage-dependent phase difference between the two fundamental waves. The phase induced by the PC will then control the SH power through the dependence in Eq. (8). Also, as we have already noted, another application for the internal interference of the SH fields is to measure the relative sign and relative magnitude of the \( \chi^{(2)} \) tensor components in the QPM crystal.
6. Conclusions

We have observed experimentally a new type of second-order cascaded parametric interaction based on a simultaneous action of two second-harmonic-generation processes in a periodically poled LiNbO$_3$ crystal, the so-called *internal second-harmonic interference*. The same effect is expected to occur in other types of ferroelectric crystals such as KTP and LiTaO$_3$. We have suggested several applications of this novel parametric effect such as the measurement of the sign and relative magnitudes of the second-order nonlinearity, polarization-independent frequency doubling, and stabilized generation of the second harmonic fields.

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