Suppression of quadratic cascading in four-photon interactions using periodically poled media

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We demonstrate theoretically that periodically poled materials with a suitable poling period can strongly reduce all $\chi^{(2)}:\chi^{(2)}$ cascading during $\chi^{(3)}$ third-harmonic generation and optical parametric amplification in noncentrosymmetric crystals: the calculated reduction is, respectively, 3 and 6 orders of magnitude along the X axis of $KTiOPO_4$. This is then an original and promising way to achieve efficient pure cubic optical frequency conversion, for the study of three-photon quantum properties.

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Nonlinear optical interactions produce nonclassical states of light, such as squeezed states in the case of second-order interactions. Third-order optical parametric interactions have the specific interest of generating new classes of photon states, with novel statistics [1] and nontrivial quantum interference pattern in their Wigner function [2]. The study and manipulation of such states would only be possible through efficient four-wave optical parametric interactions, such as resonant or nonresonant three-photon down-conversion $\chi^{(3)}(3\omega=\omega+\omega+\omega)$, for example [2,3]. No experimental demonstration has been achieved up to now, despite several attempts [3]. Due to the very weak amplitude of third-order $\chi^{(3)}$ nonlinear coefficients, efficient frequency conversion is only possible if the four-wave coupling is phase matched in the nonlinear medium. To the best of our knowledge, phase-matched four-wave interactions at wavelengths of interest for quantum optical experiments have only been reported in noncentrosymmetric crystals. In such materials, $\chi^{(2)}$ coefficients are nonzero, so that quadratic interactions can occur; even if they are not phase matched, they might be more efficient than cubic processes because of the relative amplitudes of the $\chi^{(2)}$ and $\chi^{(3)}$ coefficients. As an example, during third-harmonic generation (THG) the photons at 3ω are generated by direct THG, $\chi^{(3)}(\omega+\omega+\omega=3\omega)$, and by quadratic cascading interactions, $\chi^{(2)}(\omega+\omega=2\omega):\chi^{(2)}(\omega+2\omega=3\omega)$ simultaneously, which is detrimental to the study of the specific $\chi^{(3)}$ photon correlations. In βBaB_2O_4, where the largest THG efficiency ($\approx 5\%$) ever was reported, quadratic cascading is more efficient than the phase-matched cubic process: $[\chi^{(2)}:\chi^{(2)}/\chi^{(3)}]^2\approx 160\%$ [4]. The achievement of an efficient pure cubic optical parametric interaction is then still an open issue.

In this paper, we demonstrate that parasitic quadratic interactions can be suppressed by using periodically poled crystals with the suitable period; this will make phase-matched pure $\chi^{(3)}$ interactions possible in noncentrosymmetric crystals. Such periodic materials have become very popular in the past 10 years because quasi-phase-matching (QPM) leads to efficient three-wave optical parametric interactions [5]. In contrast, we propose an original use of these materials, in order to make $\chi^{(2)}$ interactions inefficient. The basic concept is presented in Fig. 1, which reports the nonlinear coefficients involved along the X axis of a periodically poled $KTiOPO_4$ (ppKTP) crystal with the period $\Lambda=2d$, where d is the length of one single domain. A 50% duty cycle is only considered throughout the present study. The second-order coefficients $\chi^{(2)}_{zzz}(=\chi^{(2)}_{33})$ and $\chi^{(2)}_{yyz}=\chi^{(2)}_{yzy}(=\chi^{(2)}_{24})$ are reversed every d, because periodic poling induces a reversal of the Z axis every d, while $\chi^{(3)}_{yyzz}=\chi^{(3)}_{yzyz}=\chi^{(3)}_{yzzy}(=\chi^{(3)}_{24})$ is left unchanged under a domain reversal. Depending on the period Λ, the efficiency of the quadratic cascading interactions will then be lowered or enhanced, always leaving the cubic process efficiency unaffected [6]. Note that for the same reason, QPM for four-wave mixing is not possible with the usual technique of Z-axis reversal and may be achieved by modulating the refractive indices [7], which is less efficient than phase matching. Two examples are considered throughout the paper: THG $\chi^{(3)}(\omega+\omega+\omega=3\omega)$, and optical parametric amplification $\chi^{(3)}(3\omega-\omega-\omega=\omega)$.

THIRD-HARMONIC GENERATION

THG $\chi^{(3)}(\omega^e+\omega^e+\omega^o=3\omega^o)$ is phase matched along the X axis of KTP for the fundamental wavelength $\lambda_\omega=1620$ nm; o and e refer to ordinary and extraordinary polarizations, respectively. Among possible cascades, two dominate: $\chi^{(2)}(\omega^e+\omega^o=2\omega^o):\chi^{(2)}(\omega^e+2\omega^o=3\omega^o)$ and $\chi^{(2)}(\omega^e+\omega^e=2\omega^e):\chi^{(2)}(\omega^o+2\omega^e=3\omega^o)$. A maximum efficiency of 2.4% was achieved with the cascading ratio $[(\chi^{(2)}:\chi^{(2)})/\chi^{(3)}]^2\approx 13\%$ [8,9]. The complex amplitude of

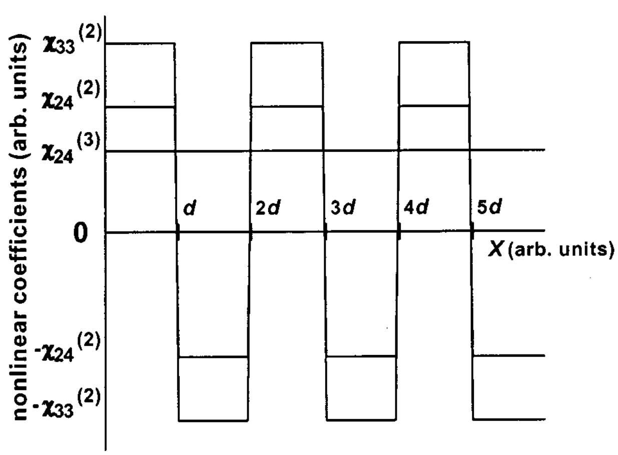

FIG. 1. Quadratic and cubic nonlinear coefficients as a function of the propagation distance X in the ppKTP.

the third-harmonic electric field along the direction of propagation X is deduced from the set of wave equations of the collinear phase-matched THG under the undepleted pump approximation [8,10]:

$$\frac{\partial E(\omega^e,X)}{\partial X} \approx \frac{\partial E(\omega^o,X)}{\partial X} \approx 0,$$

$$\frac{\partial E(2\omega^o,X)}{\partial X} \approx j\kappa^o_{2\omega}\chi^{(2)}_{24}(2\omega)E(\omega^o,0)E(\omega^e,0) \times \exp(-j\Delta k^{o\text{-}eo}_{\mathrm{SHG}}X),$$

$$\frac{\partial E(2\omega^e,X)}{\partial X} \approx j\kappa^e_{2\omega}\chi^{(2)}_{33}(2\omega)[E(\omega^e,0)]^2 \exp(-j\Delta k^{e\text{-}ee}_{\mathrm{SHG}}X), \tag{1}$$

$$\frac{\partial E(3\omega^o,X)}{\partial X} \approx j\kappa^o_{3\omega}\chi^{(2)}_{24}(3\omega)\{E(\omega^e,0)E(2\omega^o,X) \times \exp(-j\Delta k^{o\text{-}eo}_{\mathrm{SFG}}X) + E(\omega^o,0)E(2\omega^e,X) \times \exp(-j\Delta k^{o\text{-}oe}_{\mathrm{SFG}}X)\} + j\kappa^o_{3\omega}\chi^{(3)}_{24}(3\omega) \times [E(\omega^e,0)]^2E(\omega^o,0)\exp(-j\Delta k_{\mathrm{THG}}X).$$

$\kappa^{o,e}_{\omega_i} = \pi/\eta^{o,e}_{\omega_i}\lambda_{\omega_i}$ with $\eta^{o,e}_{\omega_i}$ is the refractive index; the phase mismatches of the quadratic processes are $\Delta k(\omega_a = \omega_b + \omega_c) = k_a - k_b - k_c$, where $k_i = \eta^{o,e}_{\omega_i}\omega_i/c$ is the wave-vector modulus. For the phase-matched cubic THG, $\Delta k_{\mathrm{THG}} = k^o_{3\omega} - 2k^e_{\omega} - k^o_{\omega} = 0$. The refractive indices and nonlinear coefficients of KTP are given in Ref. [8]. The input electric fields are nil at 2ω and 3ω, and $|E(\omega^e,0)| = \sqrt{2}|E(\omega^o,0)|$ according to the considered phase-matched set of polarizations. For a nonpoled crystal, the analytical integration of Eq. (1) gives the total effective coefficient [8]:

$$\chi^{\mathrm{tot}}_{\mathrm{eff}}(3\omega) = \chi^{\mathrm{casc}}_{\mathrm{eff}}(3\omega) - \chi^{(3)}_{24}(3\omega)$$

$$\text{with } \chi^{\mathrm{casc}}_{\mathrm{eff}}(3\omega) = \frac{\pi\chi^{(2)}_{24}(3\omega)}{\lambda_{2\omega}}[H^{o\text{-}eo}_{\mathrm{SHG}} + H^{e\text{-}ee}_{\mathrm{SHG}}]$$

$$H^{o\text{-}eo}_{\mathrm{SHG}} = \frac{\chi^{(2)}_{24}(2\omega)}{\eta^o_{2\omega}\Delta k^{o\text{-}eo}_{\mathrm{SHG}}} \text{ and } H^{e\text{-}ee}_{\mathrm{SHG}} = \frac{\chi^{(2)}_{33}(2\omega)}{\eta^e_{2\omega}\Delta k^{e\text{-}ee}_{\mathrm{SHG}}}. \tag{2}$$

The coherence lengths of the two SHG's are $l_o = \pi/|\Delta k^{o\text{-}eo}_{\mathrm{SHG}}| = 24.6\ \mu\mathrm{m}$, and $l_e = \pi/|\Delta k^{e\text{-}ee}_{\mathrm{SHG}}| = 14.1\ \mu\mathrm{m}$. The first factor in Eq. (2), $\chi^{\mathrm{casc}}_{\mathrm{eff}}$, is responsible for the generation of the third harmonic by the two cascading interactions $E^{\mathrm{casc}}(3\omega^o,X)$, while the second one, $\chi^{(3)}_{24}$, accounts for the cubic process generating $E^{\mathrm{cubic}}(3\omega^o,X)$. The cascading is constructed all along the propagation in the crystal, because $\Delta k^{o\text{-}eo}_{\mathrm{SHG}} + \Delta k^{o\text{-}eo}_{\mathrm{SFG}} = \Delta k^{e\text{-}ee}_{\mathrm{SHG}} + \Delta k^{o\text{-}oe}_{\mathrm{SFG}} = \Delta k_{\mathrm{THG}} = 0$ [8]; the corresponding intensity, $I^{\mathrm{casc}}(3\omega^o,X) \propto |E^{\mathrm{casc}}(3\omega^o,X)|^2$, increases continuously, as shown in Fig. 2(a). In the same way, the intensity generated by the phase-matched cubic process, $I^{\mathrm{cubic}}(3\omega^o,X) \propto |E^{\mathrm{cubic}}(3\omega^o,X)|^2$, increases as a function of L^2. An important point to note is that $\chi^{\mathrm{casc}}_{\mathrm{eff}}$ and $\chi^{(3)}_{24}$ have opposite contributions in Eq. (2), so that the generated total third-harmonic electric field is lower than the one in the cubic process alone.

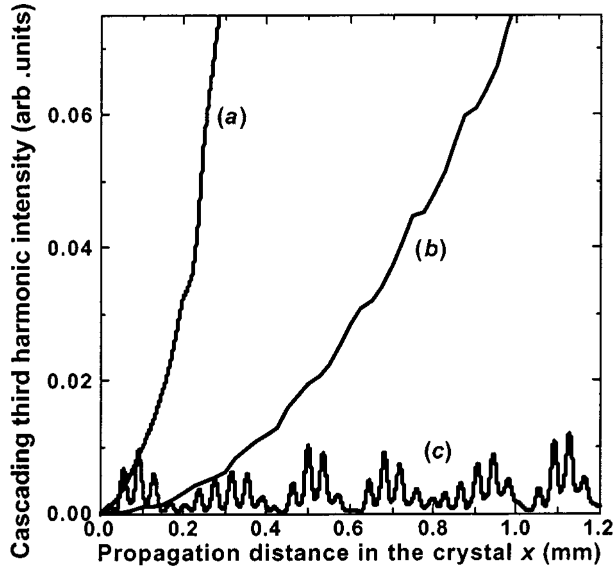

FIG. 2. Third-harmonic intensity generated by the quadratic cascading processes along the propagation in the crystal for different poling periods: nonpoled crystal (a), $d = 6\ \mu\mathrm{m}$ (b), and $d = 37\ \mu\mathrm{m}$ (c), $\lambda_{\omega} = 1620$ nm for all curves.

For the ppKTP situation of Fig. 1, the nonlinear coefficients appear in system (1) as

$$\chi^{(2)}_{ij}(\omega_l,X) = \begin{cases} +\chi^{(2)}_{ij}(\omega_l) \text{for } 2md \leqslant X \leqslant (2m+1)d \\ -\chi^{(2)}_{ij}(\omega_l) \text{for } (2m+1)d \leqslant X \leqslant (2m+2)d, \\ ij = 24 \text{ or } 33 \end{cases} \tag{3}$$

$$\chi^{(3)}_{33}(\omega_l,X) = +\chi^{(3)}_{33}(\omega_l) \quad \forall X,$$

where m is an integer, and $d(=\Lambda/2)$ is the length of one domain. The integration of Eqs. (1) with coefficients (3) gives the total field at the third-harmonic pulsation coming out of the crystal, $E^{\mathrm{tot}}(3\omega^o,X) = E^{\mathrm{casc}}(3\omega^o,X) + E^{\mathrm{cubic}}(3\omega^o,X)$.

The analytical calculation of the generated second-harmonic fields given in the Appendix allows us to understand the role of the poling period. From each relation (A1), if the domain length $\Lambda/2$ is equal to an odd multiple of the considered l_c, the efficiency is maximum, which is the basic principle of QPM [5], and which is not interesting in our case. In contrast, if $\Lambda/2$ is an even multiple of l_c, the generated intensity at 2ω oscillates as a function of X and remains

TABLE I. Ratio of the cascading over cubic intensities, r, and normalized total third-harmonic intensity, both at the exit of an $L=1$-mm-long sample with the period Λ.

$d=\Lambda/2$ (μm)	4	6	37	Nonpoled
r (%)	0.163	1.10	0.0199	12.8
$I^{\rm tot}(3\omega^o,L)$ (arb. units)	2.62	2.96	2.49	1.00

very low, as for a non-phase-matched interaction in a nonpoled crystal. This is also the case if $\Lambda/2$ is short compared to l_c. The total third-harmonic field is given by relation (A2). The generated cascading intensity depends on the relative value of $\Lambda/2$ with respect to both coherence lengths l_e and l_o. Because $\Delta k_{\rm SHG}^{o\text{-}oe}$ and $\Delta k_{\rm SHG}^{e\text{-}ee}$ have opposite signs, the two complex sums $S_{o\text{-}eo}$ and $S_{e\text{-}ee}$ in Eq. (A2) are π-out-of-phase. As a consequence, if $\kappa_{2\omega}^{o}\chi_{24}^{(2)}(2\omega)S_{o\text{-}eo}$ and $\kappa_{2\omega}^{e}\chi_{33}^{(2)}(2\omega)S_{e\text{-}ee}$ have equal amplitudes, the total field generated by the cascaded quadratic processes in nil; this is the case for $d=37\ \mu$m which is illustrated in Fig. 2: the intensity calculated along the direction of propagation in a ppKTP with that poling period remains very low throughout the crystal. As discussed above, a second way to suppress the quadratic processes is to choose d much shorter than both l_o and l_e, as can be seen for $d=6\ \mu$m in Fig. 2. In that case, the second-order nonlinear coefficients are reversed before the cascading interactions can experience efficient construction.

The reduction of the cascading $\chi^{(2)}$ processes also leads to a second advantage: because $\chi_{\rm eff}^{\rm casc}$ and $\chi_{24}^{(3)}$ have opposite contributions, the suppression of the cascading gives a larger absolute value for $\chi_{\rm eff}^{\rm tot}$. The generated third-harmonic intensity at the exit of the ppKTP, $I^{\rm tot}(3\omega^o,L)$, is then increased whenever compared to the nonpoled crystal. The values in Table I show that the enhancement factor may be up to 3.

These calculations concern phase-matched THG. If the fundamental wavelength is slightly different from the phase-matched value, the efficiencies of the cubic interaction and cascaded quadratic processes are reduced in a similar manner, because $\Delta k_{\rm THG}=\Delta k_{\rm SHG}^{o\text{-}eo}+\Delta k_{\rm SFG}^{o\text{-}eo}=\Delta k_{\rm SHG}^{e\text{-}ee}+\Delta k_{\rm SFG}^{o\text{-}oe}$ [8]. As a consequence, the spectral acceptance bandwidth calculated in a ppKTP crystal is equal to the one in a nonpoled KTP sample, $L\Delta\lambda=11$ nm cm along the x axis for $\lambda_\omega=1620$ nm. For the same reason, the cascading interactions are efficiently reduced over that complete bandwidth.

From the previous results, we evaluate the contribution of the cascading processes as $r=I^{\rm casc}(3\omega^o,L)/I^{\rm cubic}(3\omega^o,L)$ where L is the crystal length. As can be seen from relation (A3), the ratio r does not depend on the incident pump intensity because the efficiencies of the cascading and cubic interactions are both proportional to $I^2(\omega^e,0)I(\omega^o,0)$. For a nonpoled sample, relation (A5) gives $r=12.8\%$ and it does not depend on the crystal length. This ratio r is plotted in Fig. 3 as a function of the period Λ for a 1-mm-long ppKTP along the x axis. This curve has been smoothed for better clarity, because the infinite sums of rapidly oscillating functions in relation (A3) lead to a less continuous shape of the curve. As detailed above, r is maximum if d is an odd multiple of l_o or l_e, while r is reduced for other particular values of d, e.g., $d=37\ \mu$m. Nevertheless, the variations of r become less important when d increases, because the crystal tends to a nonpoled one. The main results deduced from Fig. 3 are summarized in Table I: the cascading contributions can be reduced to less than 0.1%, which is three orders of magnitude lower than for a nonpoled crystal. As an example, for a 3-mm-long ppKTP with the period $\Lambda=8\ \mu$m, an incoming fundamental intensity of 20 GW/cm^2 will lead to a conversion efficiency of 9.17%, and a cascading ratio $r=0.164\%$. Such intensities are easily achieved with picosecond pulses [9], and would be very favorable for the study of specific three-photon quantum correlations.

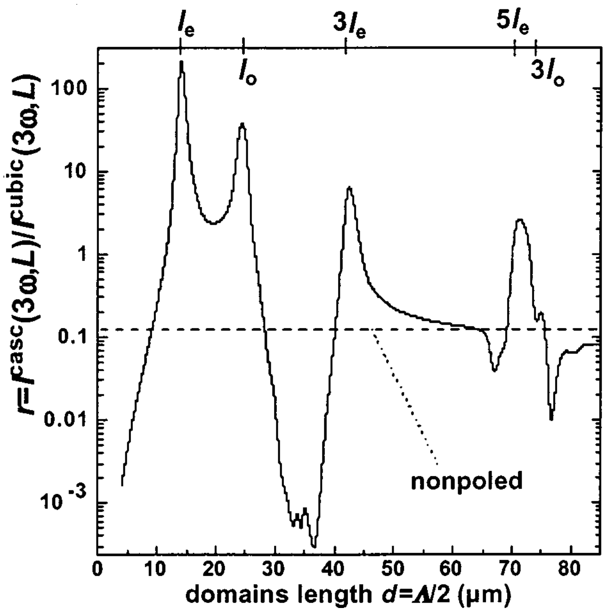

FIG. 3. Ratio of the third-harmonic intensities generated by the cascading processes and the cubic one, as a function of the domains length, calculated at the exit of a 1-mm-long ppKTP crystal, for $\lambda_\omega=1620$ nm.

OPTICAL PARAMETRIC AMPLIFICATION

The previous phase-matching condition along the x-axis of KTP also allows us to perform optical parametric amplification (OPA): $\chi^{(3)}(3\omega^o-\omega^e-\omega^e=\omega^o)$. In that case, two beams are incident on the nonlinear crystal: the pump with electric field $|E(3\omega^o,0)|$, and the signal field $|E(\omega^e,0)|$. The input field at ω^o is nil. The cubic interaction consists in an amplification of the signal field at ω^e and generation of the idler at ω^o, while the pump field at $3\omega^o$ is depleted. The different interacting electric fields at the exit of the crystal are deduced from a set of wave equations very close to Eq. (1). The generation of the idler field, at ω^o, is also possible by the same two cascading quadratic processes, $\chi^{(2)}(3\omega^o-\omega^e=2\omega^o):\chi^{(2)}(2\omega^o-\omega^e=\omega^o)$ and $\chi^{(2)}(\omega^e+\omega^e=2\omega^e):\chi^{(2)}(3\omega^o-2\omega^e=\omega^o)$, and the analysis developed for THG may be reproduced in the present case. In contrast, the situation is simpler for the amplification of the signal field at ω^e, which is mainly associated to a single

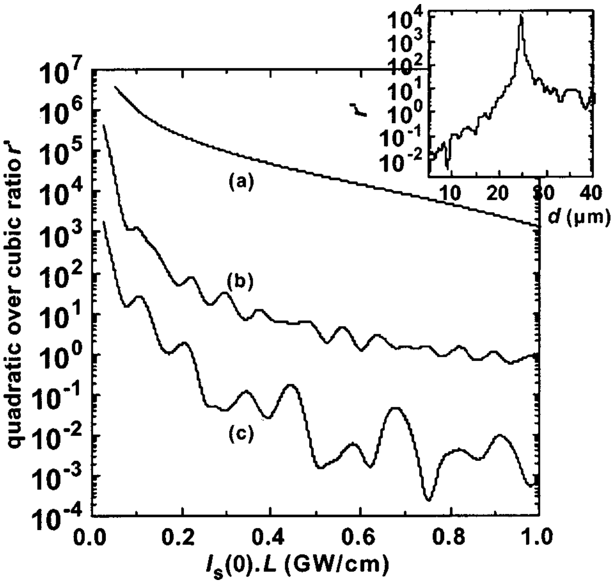

FIG. 4. Amplification of a wave of wavelength $\lambda_{\omega}=1620$ nm for a pump wavelength $\lambda_{3\omega}=540$ nm. Ratio of the quadratic over cubic contributions, r', as a function of $I(\omega^e,0)L$, for different poling periods: nonpoled (a), $d=37$ μm (b), $d=9$ μm (c). Inset graph: same ratio, as a function of the domains length $d(=\Lambda/2)$, for $I(\omega^e,0)L=0.5$ GW/cm.

quadratic parasitic amplification $\chi^{(2)}(3\omega^o-\omega^e=2\omega^o)$, because the other quadratic interactions involve weaker fields. The total signal electric field at the exit of the crystal, $E^{\text{tot}}(\omega^e,L)$, is deduced from the numerical integration of the coupled differential equations, by considering nonlinear coefficients configuration given in Fig. 1 and with a nil input electric field at $2\omega^o$. Similarly to the usual three-wave OPA, the signal gain is defined as $G=|E^{\text{tot}}(\omega^e,L)/E(\omega^e,0)|^2-1$. In order to calculate the relative contributions of the quadratic and cubic interactions, we independently integrate the set of coupled equations with the cubic terms only, which gives the emerging signal field $E^{\text{cubic}}(\omega^e,L)$. The signal field generated inside the crystal by the cubic process is then $E^{\text{cubic}}(\omega^e,L)-E(\omega^e,0)$. So the signal field generated inside the crystal by the quadratic process is $E^{\text{quadr}}(\omega^e,L)=E^{\text{tot}}(\omega^e,L)-E^{\text{cubic}}(\omega^e,L)$. The ratio of the quadratic to cubic contributions is then $r'=|E^{\text{quadr}}(\omega^e,L)/(E^{\text{cubic}}(\omega^e,L)-E(\omega^e,0))|^2$ or equivalently $r'=|(E^{\text{tot}}(\omega^e,L)-E^{\text{cubic}}(\omega^e,L))/(E^{\text{cubic}}(\omega^e,L)-E(\omega^e,0))|^2$.

An analytical expression of r' is given in relation (A6) for the limit of small gain. This equation shows that the incoming pump intensity has no effect on the ratio r', because the efficiency of both $\chi^{(2)}$ and $\chi^{(3)}$ interactions is proportional to $I(3\omega^o,0)$. However, the crystal length and the incident signal intensity play a crucial role in that case. The ratio r' is plotted in Fig. 4 as a function of $I(\omega^e,0)L$, which is the relevant parameter according to relation (A6), and to the numerical integration in the case of larger gain. For a nonpoled KTP crystal, r' is very large in the case of short crystal or low initial signal intensity, because the quadratic amplification is associated to a nonlinear coefficient nine orders of magnitude larger than that of the cubic amplification. On the other hand, the $\chi^{(2)}$ interaction is non-phase-matched, so that its efficiency does not increase with the crystal length, contrary to that of the $\chi^{(3)}$ process. Furthermore, the cubic process increases as $I^2(\omega^e,0)$, while the efficiency of the quadratic interaction depends on $I(\omega^e,0)$. Correspondingly, r' is a decreasing function of $I(\omega^e,0)L$. However, that ratio remains larger than 10^3 over the entire considered range of the parameter, which forbids the use of a nonpoled KTP crystal in order to achieve a purely cubic interaction.

The use of periodically poled crystals also leads to a large modification of the ratio r', as can be seen from the graph (inset) in Fig. 4: r' is maximum for a domains length d $(=\Lambda/2)=24.6$ μm, which is equal to the coherence length of the quadratic process $l_o=\pi/\Delta k_{\text{SFG}}^{o\text{-}eo}=24.6$ μm; in contrast, $d=9$ μm leads to a strong reduction of r', which can reach six orders of magnitude with respect to the nonpoled crystal. Similar to the case of SHG, a large reduction of r' is obtained with d much shorter than l_o. As an example, a 10-mm-long ppKTP crystal with the poling period Λ $(=2d)=18$ μm leads to a gain $G=9.4\%$ and a quadratic ratio $r'=0.1\%$, for incoming intensities 20 GW/cm^2 and 1 GW/cm^2 at the pump and signal wavelengths, $\lambda_{\omega}=1620$ and $\lambda_{3\omega}=540$ nm, respectively.

On the other hand, the two cascading quadratic processes are involved in the generation of the idler beam ω^o in the same interaction. According to the above example of THG, two ways exist for the reduction of the cascading ratio in that case: a poling period so that both cascading processes achieve destructive interference, or a very short period. As a consequence, the suitable poling period should be different from the optimum one defined for the amplification of the signal beam. So, in OPA, the poling period has to be chosen according to the beam that will be considered for the quantum measurements, and a simultaneous reduction of the quadratic contributions to both idler and signal beams might only be obtained with a very short period. A possible alternative approach could use more complicated aperiodic structures with a duty cycle different from 50%, which were shown to enable the simultaneous optimization of two interactions [11].

CONCLUDING REMARKS

Periodically poled KTP crystals specifically affect the efficiency of the quadratic interactions and leave the cubic process unchanged. With a proper poling period, cascading quadratic contributions during the phase-matched THG are reduced by three orders of magnitude, and become less than 0.1%. The benefit is even larger in OPA, where the quadratic process is reduced by six orders of magnitude. The suitable poling periods calculated in the present study, $\Lambda(=2d)=8$, $\Lambda=74$ or $\Lambda=18$ μm are fully compatible with the usual poling technique. Actually, it has been demonstrated that KTP can be poled with a period as short as $\Lambda=3$ μm, and that homogeneous and regular poling can be achieved with $\Lambda=9$ μm [12]. This technique should then constitute an original and promising way to achieve efficient pure cubic optical parametric interactions.

The calculation of the quadratic contribution to the efficiency of a $\chi^{(3)}$ optical parametric oscillator is not in the scope of our paper. Nevertheless, it should be noted that in this case, since the quadratic interactions involve additional wavelengths with respect to the cubic process, they can be made nonresonant with a proper choice of the reflectivities of the mirrors, which would certainly reduce their contribution whenever compared to the resonant cubic interaction.

APPENDIX

For the above two SHGs ($\omega^e+\omega^o=2\omega^o$) and ($\omega^e+\omega^e=2\omega^e$), with the coherence lengths $l_o=\pi/|\Delta k_{\mathrm{SHG}}^{o\text{-}eo}|$ and $l_e=\pi/|\Delta k_{\mathrm{SHG}}^{e\text{-}ee}|$, respectively, the $2\omega^o$ and $2\omega^e$ harmonic fields generated by a quasi-phase-matched interaction may be expressed analytically from the equations relative to $\partial E(2\omega^o,X)/\partial X$ and $\partial E(2\omega^e,X)/\partial X$ in system (1), and by expanding relation (3) as a Fourier series [5]. In the undepleted pump approximation, the integration leads to

$$|E(2\omega^o,X,\Lambda)|=\kappa_{2\omega}^o\chi_{24}^{(2)}(2\omega)E(\omega^o,0)E(\omega^e,0)X \times\sum_{m=-\infty}^{+\infty}\mathrm{sinc}\left(\frac{m\pi}{2}\right)\mathrm{sinc}\left[\left(\frac{2m}{\Lambda}-\frac{1}{l_o}\right)\frac{\pi X}{2}\right]$$

$$|E(2\omega^e,X,\Lambda)|=\kappa_{2\omega}^e\chi_{33}^{(2)}(2\omega)E(\omega^e,0)E(\omega^e,0)X \times\sum_{m=-\infty}^{+\infty}\mathrm{sinc}\left(\frac{m\pi}{2}\right)\mathrm{sinc}\left[\left(\frac{2m}{\Lambda}-\frac{1}{l_e}\right)\frac{\pi X}{2}\right] \quad \text{(A1)}$$

with $\mathrm{sinc}(x)=\sin(x)/x$.

Replacing these two complex field amplitudes in the last equation of system (1), we obtain after a second integration

$$E(3\omega^o,X,\Lambda)=\kappa_{3\omega}^oE(\omega^o,0)[E(\omega^e,o)]^2\{\chi_{24}^{(3)}(3\omega)X+\chi_{24}^{(2)}(3\omega)[\kappa_{2\omega}^o\chi_{24}^{(2)}(2\omega)S_{o\text{-}eo}(X,\Lambda)+\kappa_{2\omega}^e\chi_{33}^{(2)}(2\omega)S_{e\text{-}ee}(X,\Lambda)]\}$$

with

$$S_{o\text{-}eo}(X,\Lambda)=\sum_{m=-\infty}^{+\infty}\left\{\mathrm{sinc}\left(\frac{m\pi}{2}\right)\left[\frac{\exp(jX2\pi m/\Lambda)}{j2\pi m/\Lambda}-\frac{\exp(j\Delta k_{\mathrm{SHG}}^{o\text{-}eo}X)}{j\Delta k_{\mathrm{SHG}}^{o\text{-}eo}}\right]\Big/\left[\pi\left(\frac{2m}{\Lambda}-\frac{1}{l_o}\right)\right]\right\}$$

and

$$S_{e\text{-}ee}(X,\Lambda)=\sum_{m=-\infty}^{+\infty}\left\{\mathrm{sinc}\left(\frac{m\pi}{2}\right)\left[\frac{\exp(jX2\pi m/\Lambda)}{j2\pi m/\Lambda}-\frac{\exp(j\Delta k_{\mathrm{SHG}}^{e\text{-}ee}X)}{j\Delta k_{\mathrm{SHG}}^{e\text{-}ee}}\right]\Big/\left[\pi\left(\frac{2m}{\Lambda}-\frac{1}{l_e}\right)\right]\right\} \quad \text{(A2)}$$

The ratio of the intensities generated by the cascaded quadratic processes and the cubic interaction, respectively, is deduced from the above equation,

$$r(X,\Lambda)=\left(\frac{\chi_{24}^{(2)}(3\omega)|\kappa_{2\omega}^o\chi_{24}^{(2)}(2\omega)S_{o\text{-}eo}(X,\Lambda)+\kappa_{2\omega}^e\chi_{33}^{(2)}(2\omega)S_{e\text{-}ee}(X,\Lambda)|}{\chi_{24}^{(3)}(3\omega)X}\right)^2, \quad \text{(A3)}$$

In the limit of very large poling periods, i.e., when the ppKTP tends to a nonpoled crystal, the two above sums become

$$S_{o\text{-}eo}(X,\Lambda\to\infty)\approx\frac{X}{\Delta k_{\mathrm{SHG}}^{o\text{-}eo}}\quad\text{and}\quad S_{e\text{-}ee}(X,\Lambda\to\infty)\approx\frac{X}{\Delta k_{\mathrm{SHG}}^{e\text{-}ee}}, \quad \text{(A4)}$$

so that the ratio r in Eq. (A3) takes the limit value

$$r(X,\Lambda\to\infty)\to\left(\frac{\chi_{24}^{(2)}(3\omega)\left[\frac{\kappa_{2\omega}^o\chi_{24}^{(2)}(2\omega)}{\Delta k_{\mathrm{SHG}}^{o\text{-}eo}}+\frac{\kappa_{2\omega}^e\chi_{33}^{(2)}(2\omega)}{\Delta k_{\mathrm{SHG}}^{e\text{-}ee}}\right]}{\chi_{24}^{(3)}(3\omega)}\right)^2, \quad \text{(A5)}$$

which is the exactly the value that would be deduced from relation (2).

In the case of optical parametric amplification, assuming small gain and a phase-matched cubic process, a very similar integration leads to the complex amplitudes of the interacting electric fields. It is then possible to deduce the ratio of the contributions of the quadratic and cubic interactions to the amplification of the signal beam:

$$r'(X,\Lambda,E(\omega^e,0))=\left(\frac{\kappa_{2\omega}^o\chi_{24}^{(2)}(3\omega)\chi_{24}^{(2)}(2\omega)|S_{o\text{-}eo}(X,\Lambda)|}{\chi_{24}^{(3)}(3\omega)X|E(\omega^e,0)|^2}\right)^2, \quad \text{(A6)}$$

where $S_{o\text{-}eo}(X,\Lambda)$ is given in Eq. (A2).

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