Cascaded fourth-harmonic generation in a single nonlinear crystal

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We investigate theoretically the process of phase-matched fourth-harmonic generation in a single quadratic crystal. In the case of a plane-wave fundamental or weak focusing, the results have an analytical form. The optimal focusing position of the fundamental beam depends on the values of the mismatches for each of the two steps of the second-order cascading: doubling of the fundamental frequency and doubling of the frequency of the generated second-harmonic wave. It is shown that the optimized scheme for the cascaded single-crystal fourth-harmonic generation requires specially chosen mismatches of the two steps and focusing into the center of the nonlinear media. © 2005 Optical Society of America

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1. INTRODUCTION

The tendency for miniaturization and design of compact integrated optical devices stimulates the research and development of single-crystal devices for efficient third-, fourth- and fifth-harmonic generation. Multistep cascading as a method for high-order harmonic generation in single nonlinear media has been the subject of intensive investigations for the past three decades. For example, third-harmonic generation in a single quadratic crystal has been studied both theoretically and experimentally in a number of publications reviewed in Ref. 1. Recently several papers reported single-crystal third-harmonic generation with efficiency exceeding 20%.[2–4] Fifth-harmonic generation in a single cubic medium has also been studied both experimentally and theoretically in a number of publications, e.g., Refs. 5–10. In Refs. 11–13 we investigated the generation of the third and fifth harmonics by cascading two phase-matched processes in a single nonlinear medium with a focused fundamental beam.

Another important example of multistep cascading interaction, which could have significant application, is the single-crystal fourth-harmonic generation (FoHG) based on second-order cascading. There are two possible cascades of second-order processes that will lead to FoHG.

(a) The second-harmonic field is the only intermediate field. The generation of the fourth harmonic involves two frequency-doubling processes: $\omega+\omega=2\omega$ and $2\omega+2\omega=4\omega$.

(b) Both second-harmonic and third-harmonic fields are intermediate fields. The generation of the fourth harmonic involves frequency doubling of the fundamental, followed by two sum-frequency-generation processes: $\omega+\omega=2\omega, 2\omega+\omega=3\omega$, and $3\omega+\omega=4\omega$.

In both cases the fourth-harmonic amplitude will be proportional to $(\chi^{(2)})^3$. Which of the two cases will be more effective depends on the phase-matching conditions. Obviously, the first one is easier to be realized in that only two phase-matching conditions have to be fulfilled simultaneously. Here we will consider the interaction $\omega+\omega=2\omega$ and $2\omega+2\omega=4\omega$.

The first experiment with FoHG in a single crystal is reported in the research of Akhmanov *et al.*,[14] in which they investigated FoHG in a lithium formiate crystal. It was concluded that the generated fourth harmonic is due to the simultaneous action of two- and three-step cascaded processes with required quadratic, cubic, and direct fourth-order nonlinearities. Single-crystal FoHG is reported in $CdGeAs_2$,[15] $LiNbO_3$,[16–18] and KTP[19] with efficiency well below 1% in that one of the steps is far from the phase-matched condition. As shown in Refs. 20–23, if both steps are phase matched, efficiency close to 100% can be expected. Some possibilities for double-phase-matched single-crystal FoHG by using homogeneous quasi-phase-matched (QPM) structures are shown in Refs. 24 and 25. Double-phase-matched schemes for FoHG in two-dimensional nonlinear photonics crystals are proposed in recent studies.[26–28] However, in spite of the published papers, there is no systematic investigation of the phase-matched conditions and efficiency for single-crystal FoHG either for a plane wave or for a focused fundamental beam.

Here in this study we present theoretical analysis of the phase-matching conditions and the efficiency for single-crystal FoHG for both weak and strong focusing conditions in nondepleted approximation. The analytical formula valid for weak focusing can be easily adopted for the plane-wave case. We demonstrate that double phase matching can be achieved with the so-called phase-reversed QPM structure.

2. SOLUTION FOR PEAK AMPLITUDES

The effect of FoHG, as a result of simultaneous action of the processes of second-harmonic generation and sum-frequency mixing of the second-harmonic waves, is described by the following system of differential equations, derived with the assumption of zero absorption of all interacting waves:

$$\left(\frac{\partial}{\partial z}+\frac{i}{2k_1}\Delta_\perp\right)A_1=-i\ \sigma'_1A_2A_1^*\exp(-i\Delta k_1z),$$

$$\left(\frac{\partial}{\partial z}+\frac{i}{2k_2}\Delta_\perp\right)A_2=-i\ \sigma_1A_1^2\exp(i\Delta k_1z)$$

$$-i\sigma'_2A_4A_2^*\exp(-i\Delta k_2z),$$

$$\left(\frac{\partial}{\partial z}+\frac{i}{2k_4}\Delta_\perp\right)A_4=-i\sigma_2A_2^2\exp(i\Delta k_2z). \quad (1)$$

Here A_1, A_2, and A_4 denote the complex amplitudes of the fundamental, second-harmonic, and fourth-harmonic waves, $\Delta_\perp$ stands for the operator $(\partial^2/\partial x^2)+(\partial^2/\partial y^2)$, $\sigma_1=2\pi d_{\mathrm{eff,I}}/\lambda_1n_2$, $\sigma_2=4\pi d_{\mathrm{eff,II}}/\lambda_1n_4$, $\sigma'_1=(n_2/n_1)\sigma_1$, and $\sigma'_2=(n_4/n_2)\sigma_2$. Phase-mismatch parameters are $\Delta k_1=k_2-2k_1-G_1$ and $\Delta k_2=k_4-2k_2-G_2$, where G_1 and G_2 are two QPM vectors that can be used for achieving the phase matching, where k_j are the wave vectors of the waves involved in the process. For birefringence phase matching, $G_1=0$ and $G_2=0$. The high-order nonlinearities $\chi^{(3)}$ and $\chi^{(4)}$ are neglected because their contribution is rather small when we work in conditions close to double and triple phase matching for the second-order processes.

For not high input fundamental intensities, the effects of depletion of the fundamental wave and second-harmonic wave due to FoHG can be neglected, and system (1) reduces to

$$\left(\frac{\partial}{\partial z}+\frac{i}{2k_1}\Delta_\perp\right)A_1=0,$$

$$\left(\frac{\partial}{\partial z}+\frac{i}{2k_2}\Delta_\perp\right)A_2=-i\sigma_1A_1^2\exp(i\Delta k_1z),$$

$$\left(\frac{\partial}{\partial z}+\frac{i}{2k_4}\Delta_\perp\right)A_4=-i\sigma_2A_2^2\exp(i\Delta k_2z). \quad (2)$$

System (2) allows some of the results to be obtained in an analytical form that will help to gain physical insight into the process of cascaded FoHG.

We solve system (2) by assuming only one input beam—the fundamental that has a Gaussian spatial distribution:

$$A_1(x,y,z)=A\ u_1(x,y,\xi_1). \quad (3)$$

A is the electric field amplitude at the center of the focal spot $(0,0,z_0)$, and ξ_1 is defined below, after Eqs. (5a) and (5b). With these notations the distribution of the fundamental field at the entrance of the nonlinear medium ($z=0$) is

$$A(x,y,0)=A_{\mathrm{in}}\exp\left[-(x^2+y^2)\left(\frac{1}{w_{\mathrm{in}}^2}-\frac{ik_1}{2R_1}\right)\right], \quad (4)$$

where the amplitude, beam radius, and wave-front curvature at the nonlinear medium entrance are $A_{\mathrm{in}}=[w_{01}^2/w_{\mathrm{in}}^2-i(b_1/2R_1)]A_0$, $w_{\mathrm{in}}^2=w_{01}^2[1+4(z_0^2/b_1^2)]$, $R_1=(z_0+b_1^2/4z_0)$, respectively.

To simplify system (2), we apply the procedure of the trial solution as described in Refs. 29 and 30. In a nondepletion approximation, a fundamental beam will preserve its Gaussian form. Moreover, the generated second and fourth harmonics will be Gaussian beams as well but with different focal spot radii:

$$A_2(x,y,z)=S(z)u_2(x,y,\xi_2), \quad (5a)$$

$$A_4(x,y,z)=F(z)u_4(x,y,\xi_4), \quad (5b)$$

with

$$u_j(x,y,\xi_j)=\frac{1}{1-i\xi_j}\exp\left[-\frac{x^2+y^2}{w_{0j}^2(1-i\xi_j)}\right],\quad j=1,2,4.$$

In Eqs. (3), (5a), and (5b), $\xi_j=2(z-z_0)/b_j$, with b_j as the confocal parameters of the beams involved, $b_j=k_jw_{0j}^2$, where w_{0j} is the focal spot radius. The amplitudes S and F also correspond to the center of the focal spot of the second and fourth harmonics, respectively. The spot radii are connected with the relations $w_{0j}=w_{01}/\sqrt{j}$, see Refs. 29 and 30, and this allows us to assume that $b_4\cong b_2\cong b_1=b$ (neglecting a slight difference in the index of refraction of the three waves) and $\xi_4\cong\xi_3\cong\xi_1=\xi$. Substituting Eqs. (3), (5a), and (5b) in system (2) and noting that

$$(u_1)^2=\frac{u_2}{(1-i\xi)},$$

$$(u_2)^2=\frac{u_4}{(1-i\xi)}, \quad (6)$$

we come to the following system of first-order differential equations, which is equivalent to system (2) but easier to solve,

$$\frac{\partial A}{\partial z}=0,$$

$$\frac{\partial S}{\partial z}=-\frac{i\sigma_1A^2}{1-i\xi}\exp(i\Delta k_1z),$$

$$\frac{\partial F}{\partial z}=-\frac{i\sigma_2S^2}{1-i\xi}\exp(i\Delta k_2z). \quad (7)$$

The straightforward solution of system (7) gives

$$S(\xi)=-\frac{i\sigma_1A^2b}{2}\exp(i\Delta k_1z_0)\int_{\beta_1}^{\xi}\frac{\exp(i\Delta k_1b\tau/2)}{1-i\tau}\mathrm{d}\tau, \quad (8)$$

$$F=\frac{i\sigma_1^2\sigma_2A^4b^3\exp[iz_0(2\Delta k_1+\Delta k_2)]}{8}\int_{\beta_1}^{\beta_2}\frac{\exp(i\Delta k_2b\xi/2)}{1-i\xi}$$

$$\times\left[\int_{\beta_1}^{\xi}\frac{\exp(i\Delta k_1b\tau/2)}{1-i\tau}\mathrm{d}\tau\right]^2\mathrm{d}\xi, \quad (9)$$

where the limits of integrations are $\beta_1=-m(1+2l)$ and $\beta_2=m(1-2l)$, with m as the strength of focusing $m=L/b$

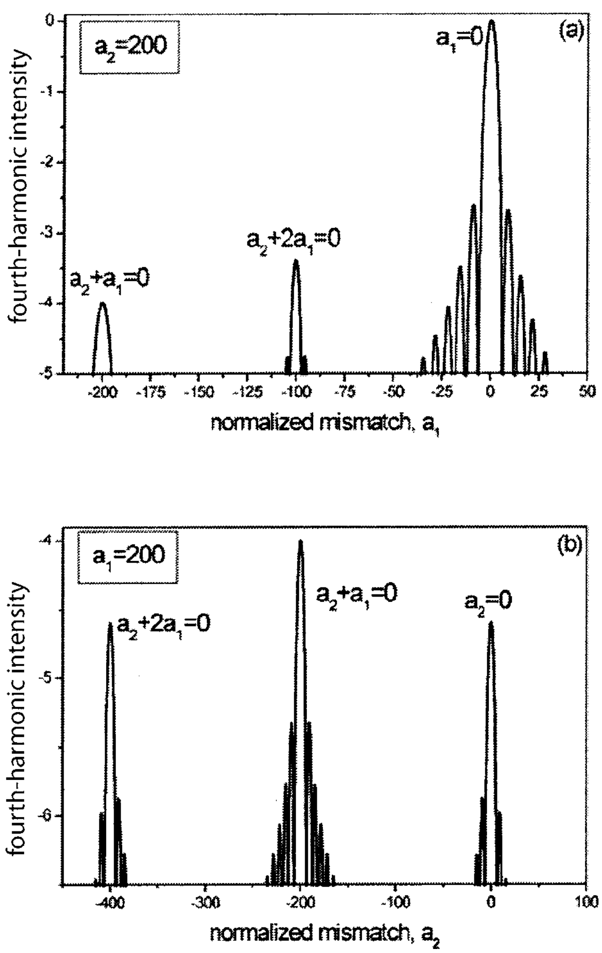

Fig. 1. Relative intensities of the fourth-harmonic wave at different phase-matching conditions in 'a logarithmic' scale. All intensities are normalized to the case of phase matching for second-harmonic generation ($a_1=0$). (a) $a_2=200$ and (b) $a_1=200$.

(L is the length of the nonlinear medium) and l as the dimensionless parameter indicating the position of the focus, $l=(2z_0-L)/2L$.

The efficiency of the FoHG is defined as

$$\eta_{4\omega}=\frac{\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}A_4A_4^{*}\mathrm{d}x\mathrm{d}y}{\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}A_1A_1^{*}\mathrm{d}x\mathrm{d}y}.$$

Equation (9) can be solved analytically in a weak focusing approximation and in a plane-wave limit. For arbitrary focusing, Eq. (9) has to be solved numerically in Subsection 2.B.

A. Weak Focusing and Plane-Wave Limits

With weak focusing ($m \ll 1$) the limits of integration in Eq. (9) will be much smaller than 1 and, as a result, when the focus is in the center of the crystal ($z_0=L/2$), we have

$$F(L)=\frac{(\sigma_1)^2\sigma_2}{(a_1)^2}A^4b^3\exp(-3im)\left\{\frac{2\exp[im(a_2+a_1)]-1}{(a_2+a_1)}-\frac{\exp(ima_2)-1}{a_2}-\frac{\exp[im(a_2+2a_1)]-1}{(a_2+2a_1)}\right\}, \quad (10)$$

where $a_j=\Delta k_j b+2$.

In the plane-wave limit ($m\rightarrow 0$), we can set $ma_j \rightarrow \Delta k_j L$. Then the plane-wave version of Eq. (10) has the following form:

Table 1. Expressions for the Fourth-Harmonic Field $F(L)$ for Both the Weak Focusing Limit and the Plane-Wave Approximation

Phase Matching	Plane-Wave Approximation $F(L)$=	Weak Focusing Limit $F(L)$=	Relative Intensity Δk_1 Fixed Δk_2 Tuning	Relative Intensity Δk_2 Fixed Δk_1 Tuning
$a_1\approx 0$		$\{-3iF_0\exp[im(a_2-6)/2]/a_2m\}\times[\sin(a_1m/2)/(a_1m/2)]^2$		$9/(a_2)^2$
$\Delta k_1L\approx 0$	$\{-3iF_0\exp(i\,\Delta k_2L/2)/\Delta k_2L\}\times[\sin(\Delta k_1L/2)/(\Delta k_1L/2)]^2$			$9/(\Delta k_2L)^2$
$a_2\approx 0$		$\{[-3F_0\exp(-i3m)]/(a_1m)^2\}\times\{[\sin(a_2m/2)]/(a_2m/2)\}$	$9/(a_1)^4$	
$\Delta k_2L\approx 0$	$\{-3F_0\exp(-i\Delta k_1L)/(\Delta k_1L)^2\}\times[\sin(\Delta k_2L/2)/(\Delta k_2L/2)]^2$		$9/(\Delta k_1L)^4$	
$a_2+2a_1\approx 0$		$\{-3F_0\exp[im(a_2+2a_1-6)/2]/(a_1)^2m^2\}\times\{\sin[(a_2+2a_1)m/2]/(a_2+2a_1)m/2\}$	$9/(a_1)^4$	$144/(a_2)^4$
$\Delta k_2+2\Delta k_1\approx 0$	$\{-3F_0\exp[i(\Delta k_2+2\Delta k_1)L/2]/(\Delta k_1L)^2\}\times\{\sin[(\Delta k_2+2\Delta k_1)L/2]/(\Delta k_2+2\Delta k_1)L/2\}$		$9/(\Delta k_1L)^4$	$144/(\Delta k_2L)^4$
$a_2+a_1\approx 0$		$\{6F_0\exp[im(a_2+a_1-6)/2]/(a_1)^2m^2\}\times\{\sin[(a_2+a_1)m/2]/(a_2+a_1)m/2\}$	$36/(a_1)^4$	$36/(a_2)^4$
$\Delta k_2+\Delta k_1\approx 0$	$\{6F_0\exp[i(\Delta k_2+\Delta k_1)L/2]/(\Delta k_1L)^2\}\times\{\sin[(\Delta k_2+\Delta k_1)L/2]/(\Delta k_2+\Delta k_1)L/2\}$		$36/(\Delta k_1L)^4$	$36/(\Delta k_2L)^4$
$a_2=0$; $a_1=0$		$F_0\exp(-i3m)$	1	1
$\Delta k_2=0$; $\Delta k_1=0$	$F_0=i\{(\sigma_1)^2\sigma_2/3\}A^4L^3$		1	1

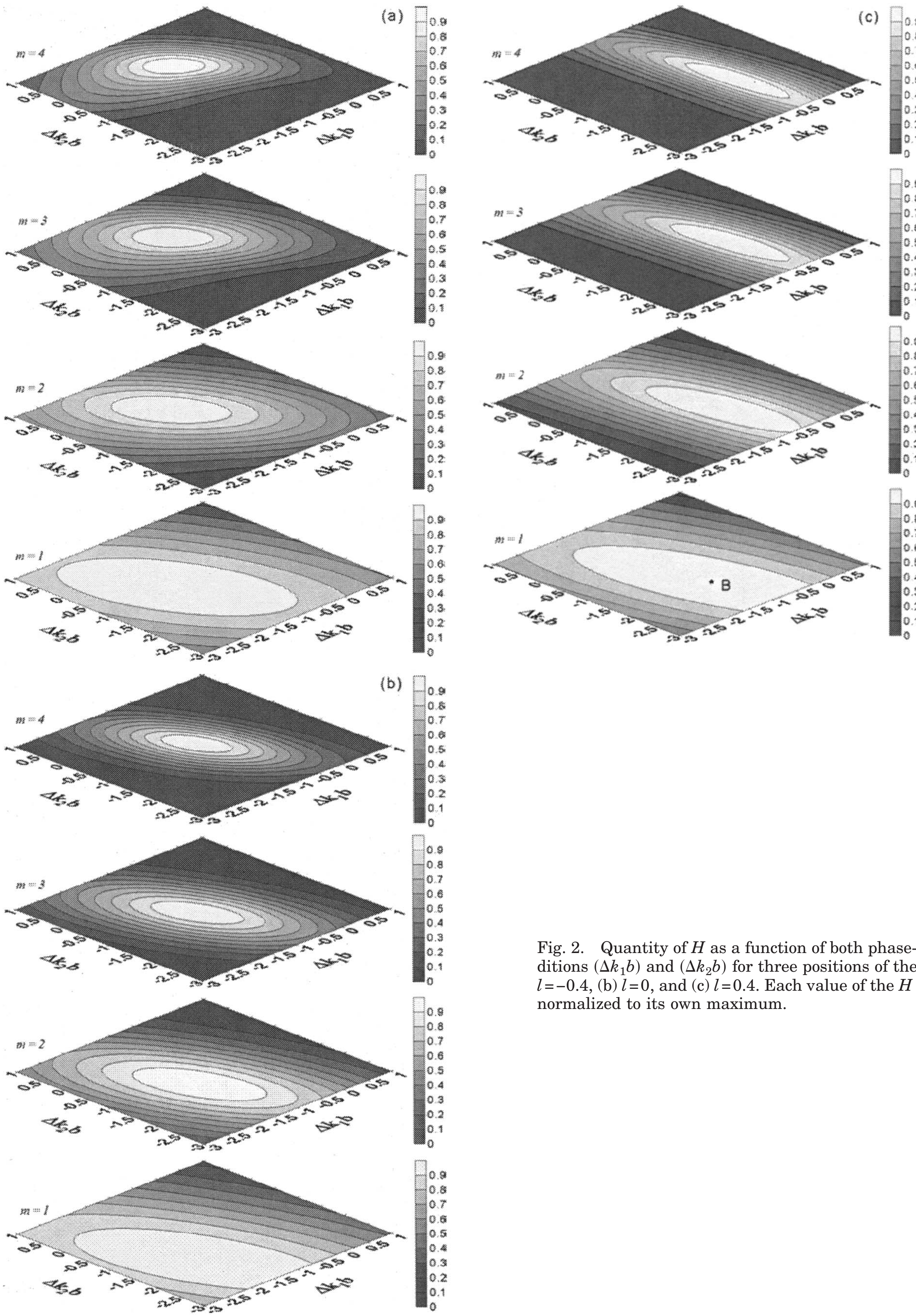

Fig. 2. Quantity of H as a function of both phase-matching conditions $(\Delta k_1 b)$ and $(\Delta k_2 b)$ for three positions of the focal spot: (a) $l=-0.4$, (b) $l=0$, and (c) $l=0.4$. Each value of the H distribution is normalized to its own maximum.

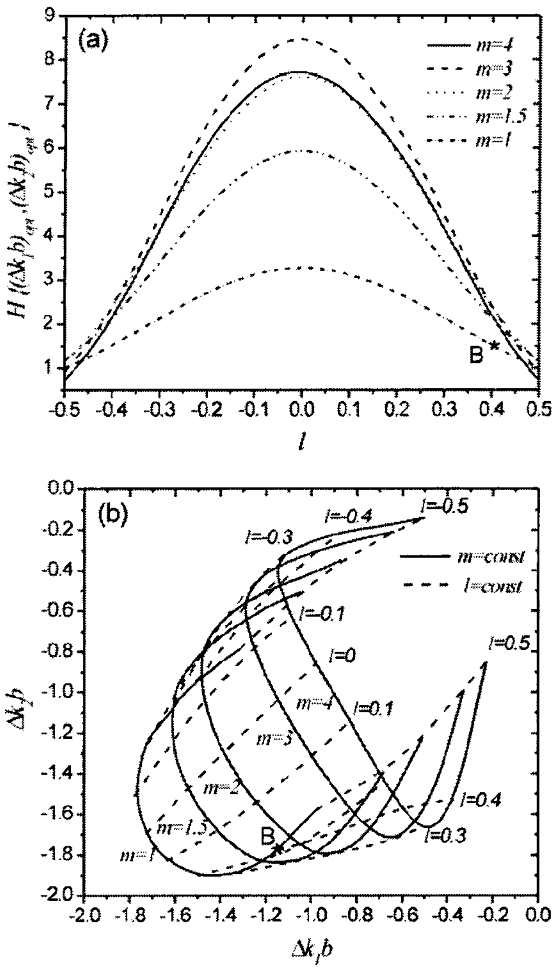

Fig. 3. (a) Quantity of H calculated at optimal phase-matching conditions as a function of the position of focusing l and (b) optimal phase matching for both steps for several values of focusing m and the position of focusing.

$$F(L)=\frac{(\sigma_1)^2\sigma_2}{(\Delta k_1)^2}A^4\left\{\frac{2\exp[i(\Delta k_2+\Delta k_1)L]-1}{(\Delta k_2+\Delta k_1)}-\frac{\exp(i\Delta k_2L)-1}{\Delta k_2}-\frac{\exp[i(\Delta k_2+2\Delta k_1)L]-1}{(\Delta k_2+2\Delta k_1)}\right\}. \tag{11}$$

Analyzing Eqs. (10) and (11), we find four possibilities for phase-matched generation with a single-crystal cascaded fourth harmonic.

(a) Phase matching for the first step $\omega+\omega=2\omega$. As can be seen from Fig. 1 this phase matching gives maximum efficiency among the cases when only one phase-matching case is close to be satisfied. The tuning curve is peaked when $a_1=\Delta k_1 b+2=0$ and the required deviation from exact phase matching is $\Delta k_1 L=-2m$. In the plane-wave case the phase-matching condition is $\Delta k_1 L=0$.

(b) Phase matching for the second step $2\omega+2\omega=4\omega$. The maximum is obtained when $a_2=\Delta k_2 b+2=0$, which corresponds to the deviation $\Delta k_2 L=-2m$. In the plane-wave case the phase-matching condition is $\Delta k_2 L=0$.

(c) Phase matching for the direct FoHG $\omega+\omega+\omega+\omega=4\omega$ when $a_2+2a_1=(k_4-4k_1)b+6=0$, which corresponds to the deviation $(\Delta k_2+2\Delta k_1)L=-6m$. In the plane-wave case the phase-matching condition is $(\Delta k_2+2\Delta k_1)L=(k_4-4k_1)L=0$.

(d) Phase matching for the FoHG by FWM $2\omega+\omega+\omega=4\omega$ when $a_2+a_1=(k_4-2k_1-k_2)b+4=0$, which corresponds to the deviation $(\Delta k_2+\Delta k_1)L=-4m$. It can be seen that different phase-matching conditions are a linear combination of the two main phase matchings $\Delta k_1 L$ and $\Delta k_2 L$, respectively, a_1 and a_2. In addition, note that, following Eq. (11), in cases (c) and (d) the FoHG process is also a result of cascading $\chi^{(2)}$ processes, in spite of the fact that the phase-matching conditions correspond to high-order processes.

In Table 1 are shown the expressions of the fourth-harmonic field for the different phase-matching conditions for both the weak focusing limit and the plane-wave approximation. On Fig. 1 is shown an example of their relatives intensities. The intensities are normalized to the case of the FoHG at $\Delta k_1 L\approx 0$ ($a_1\approx 0$). If we compare the widths of the tuning curves among the different single phase-matching conditions, we can note that the width of the tuning curves for the phase matching around $\Delta k_1 L\approx 0$ ($a_1\approx 0$) is narrower than the others.

When both main phase matchings are simultaneously fulfilled, the conversion into the fourth harmonic is maximal. In the situation of exact double phase matching, the field of the fourth harmonic in a plane-wave approximation is[28]

$$F_0=i\frac{(\sigma_1)^2\sigma_2}{3}A^4L^3.$$

Introducing normalized efficiency in total efficiency per watts and per square centimeter units for the first step and second step, respectively, as $\eta_{0,1}$ and $\eta_{0,2}$ we get for the efficiency of FoHG[1]

$$\eta_{4\omega}=\frac{\eta_{0,1}^2\eta_{0,2}}{9}P_1^3L^6.$$

Efficiency of the single-crystal $\chi^{(2)}$-based FoHG depends on the sixth power of the length and the third

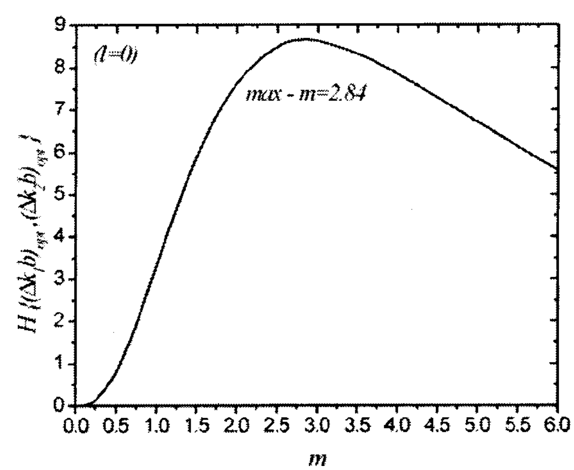

Fig. 4. Quantity of H as a function of the strength of focusing m for focusing in the center of the nonlinear media.

power of the pump and can be easily estimated with known efficiencies for the separated steps. For pump intensities at which the depletion effect of the fundamental and second-harmonic wave can not be neglected, system (1) has to be solved numerically. For this range of pump intensities, 100% conversion of the fundamental to the fourth-harmonic wave is possibly independant of th ratio of the nonlinear coupling coefficients σ_2/σ_1.[20–23] This behavior strongly contrasts with the $\chi^{(2)}$-based cascaded third-harmonic generation in single nonlinear media in which 100% conversion is possible only for a specific ratio of nonlinear coupling coefficients.[1]

B. Arbitrary Focusing

In this subsection we consider the arbitrary value of the strength of focusing m, still keeping the nondepletion approximation for the fundamental and second-harmonic beam. The efficiency conversion in the fourth harmonic is calculated from Eq. (9) and is found to be

$$\eta_{4\omega}=\frac{2\sigma_1^4\sigma_2^2 n_4 L^3 P_1^3}{\epsilon_0^3 c^3 \lambda_1^3 n_1} H(m,l,\Delta k_1,\Delta k_2), \qquad (12)$$

where

$$H(m,l,\Delta k_1,\Delta k_2) = \frac{1}{m^3}\left|\int_{\beta_1}^{\beta_2}\frac{\exp(i\Delta k_2 b\xi/2)}{1-i\xi}\left[\int_{\beta_1}^{\xi}\frac{\exp(i\Delta k_1 b\tau/2)}{1-i\tau}\mathrm{d}\tau\right]^2\mathrm{d}\xi\right|^2.$$

The dependence of H on the four parameters describing the system (strength of focusing m, position of focusing l, and conditions for phase matching of both steps $\Delta k_1 b$ and $\Delta k_2 b$) was investigated. For maximum FoHG efficiency it is necessary to tune both mismatches to their optimal values $(\Delta k_1,\Delta k_2)=(\Delta k_{1,\mathrm{opt}},\Delta k_{2,\mathrm{opt}})$

Table 2. Optimal Phase Matchings and Corresponding Values of H for Focusing in the Center of the Nonlinear Medium for Different Values of the Ratio $m=L/b$

m	$(\Delta k_1 b)_{\mathrm{opt}}$	$(\Delta k_2 b)_{\mathrm{opt}}$	H
1	−1.73	−1.70	3.32
1.5	−1.55	−1.48	6.05
2	−1.39	−1.30	7.78
3	−1.15	−1.04	8.64
4	−0.98	−0.86	7.87

This can be seen on the contour plots shown on Figs. 2(a)–2(c) calculated for four different strengths of focusing. Figure 2(a) is calculated for focusing position $l=-0.4$, close to the input face of the nonlinear medium. Figure 2(b) illustrates the two-dimensional phase-matching curves when focusing is in the center of the nonlinear medium, $l=0$, whereas Fig. 2(c) is calculated for focusing position $l=0.4$, close to the output face of the nonlinear medium. Each efficiency distribution is normalized to its own maximum. Figures 3(a) and 3(b) allow one to obtain the maximum H for arbitrary values of these four parameters. Figure 2(a) shows the dependence of H on the strength of focusing m and the position of focusing l. The optimal $(\Delta k_1 b)_{\mathrm{opt}}$ and $(\Delta k_2 b)_{\mathrm{opt}}$ for each point of Fig. 3(a) can be found from Fig. 3(b). For example, as can be seen from Fig. 3(a), point B ($m=1$, $l=0.4$, with $H=1.5181$) corresponds to the following optimal phase mismatches $(\Delta k_1 b)_{\mathrm{opt}}=-1.15(\Delta k_2 b)_{\mathrm{opt}}=-1.78$, as can be seen from Fig. 3(b). The optimal phase matchings, for the most important case of focusing in the center of the nonlinear crystal are given in Table 2.

We found out that, in the process of cascaded FoHG in a single crystal by a focused beam, for maximum efficiency the phase matching $\Delta k_1 b$ and $\Delta k_2 b$ have optimal values, different from 0, and these values are different for each m and l. However, for best conversion the optimal position for focusing is in the center of the nonlinear medium. On Fig. 4 we present the dependence of the FoHG efficiency on the focusing parameter m. The optimal strength of focusing for small conversion efficiency is $m=2.84$.

3. DOUBLE-PHASE-MATCHED FOURTH-HARMONIC GENERATION WITH PHASE-REVERSED QUASI-PHASE-MATCHED STRUCTURES

The idea of the phase-reversed QPM structures developed in Ref. 31 can be used for realization of double-phase-matched FoHG in a single quadratic crystal. This structure can be explained as aperiodic QPM grating with QPM period Λ_Q that changes its phase with another period Λ_{ph}. The two periods Λ_Q and Λ_{ph} are the two degree of freedom that allow two different nonlinear processes to be simultaneously phase matched. The designs of the phase-reversed QPM structures as described in Ref. 31 impose an additional condition that the ratio $\delta=\Lambda_{\mathrm{ph}}/2\Lambda_Q$ be an even integer number. The study of the effect of arbitrary $\Lambda_{\mathrm{ph}}/\Lambda_Q$ is not systematically investigated yet, to the best of our knowledge. The exception is a

Table 3. Data for the Phase-Reversed QPM Structures in KTP, $LiTaO_3$, and $LiNbO_3$ by Different Values of the Orders p_1, p_2, q_1, and q_2

Crystal	Orders of QPM Grating				λ_1 (μm)	Λ_Q (μm)	Λ_{ph} (μm)	T (°C)
	p_1	p_2	q_1	q_2				
KTP	1	3	3	1	1.402	5.111	20.440	20
$LiTaO_3$	1	1	5	1	1.622	3.766	22.596	20
	1	5	−1	1	1.622	16.950	67.811	20
$LiNbO_3$	1	1	5	−1	1.724	3.935	23.609	20

publication[32] in which it was found that in the case of cascaded third-harmonic generation the ratio $\Lambda_{ph}/2\Lambda_Q$ could take values different from an integer.

Starting with the two conditions $\Delta k_1 = k_2 - 2k_1 - G_1 = 0$ and $\Delta k_2 = k_4 - 2k_2 - G_2 = 0$, with

$$G_1 = \frac{2\pi}{\Lambda_Q} p_1 + \frac{2\pi}{\Lambda_{ph}} q_1, \quad G_2 = \frac{2\pi}{\Lambda_Q} p_2 + \frac{2\pi}{\Lambda_{ph}} q_2,$$

we find

$$\Lambda_{ph} = \left| \frac{\lambda_1 (q_2 p_1 - q_1 p_2)}{2n_{2\omega}(2p_1 + p_2) - 2n_\omega p_2 - 4n_{4\omega} p_1} \right|,$$

$$\Lambda_Q = \left| \frac{\lambda_1 (q_2 p_1 - q_1 p_2)}{2n_{2\omega}(2q_1 + q_2) - 2n_\omega q_2 - 4n_{4\omega} q_1} \right|,$$

where p_1, p_2 are two orders of the QPM grating and q_1, q_2 are two orders of the phase grating. In Table 3 some examples of calculated periods for double-phase-matched FoHG in congruent $LiNbO_3$,[33] $LiTaO_3$,[34] and KTP[35] are shown. Recently this type of QPM structure has been used for experimental realization of double-phase-matched third-harmonic generation.[32]

4. CONCLUSION

In conclusion, we investigated here the conditions for single-crystal phase-matched fourth-harmonic generation. Efficient FoHG is possible at four different phase-matching conditions. The interaction with fulfilled phase-matching conditions for the second-harmonic-generation process gives maximum efficiency for FoHG (among single phase-matching cases). For highest efficiency, simultaneous phase matching of the two steps is required. The optimal focusing for the double-phase-matched FoHG is in the center of the crystal. An example of double phase matching in $LiNbO_3$, $LiTaO_3$, and KTP with phase-reversed QPM gratings is presented.

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