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Double phase-matched cascaded fifth harmonic generation in single nonlinear medium by focussed laser beam

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ABSTRACT Generation of a fifth harmonic by the cascading of two phase-matched third-order processes in a single centrosymmetric nonlinear medium with a focused fundamental beam is investigated theoretically. With the help of analytical and numerical investigations the optimized conditions for maximum conversion into the fifth harmonic are found. In general the optimal position of focusing depends on the values of the mismatches Δk_1 and Δk_2 for both "steps" of the third order cascading ($\omega+\omega+\omega=3\omega$; $3\omega+\omega+\omega=5\omega$). It is shown that for best efficiency this method of fifth harmonic generation requires specially chosen $\Delta k_{1,\text{opt}}$ and $\Delta k_{2,\text{opt}}$ and focusing in the center of the nonlinear media. If the phase matching parameters are fixed and they deviate from the optimal values, then the optimal strength of focusing and position of the focus spot should be calculated according to the analysis presented here.

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1 Introduction

Generation of high order harmonics is an efficient way to obtain coherent radiation in the UV and VUV region when it is needed. Cascading processes play an important role in these types of interactions [1, 2]. This work considers theoretically fifth harmonic generation (FHG) by third order cascading in a single nonlinear medium, with a focused fundamental beam. This investigation is an extension of previous work [3] where the efficiency of cascaded third harmonic generation in a single quadratic crystal in a focussed beam, has been considered.

Theoretical descriptions of the FHG in a single cubic medium has been published by Tomov and Richardson [4], while the first experimental work on fifth harmonic generation in a single centrosymmetic media was reported by Akhmanov et al. [5]. Later, several works contributed to the understanding of the interference of the direct and cascade processes [6–10]. An efficient fifth harmonic can be generated when phase matching exists for the direct process $\omega+\omega+\omega+\omega+\omega=5\omega$. Additionally, the fifth harmonic can be generated through several channels of third-order cascading. For example, those generated in the nonlinear media third harmonic in the first step of possible cascading, can produce the fifth harmonic through one of the interactions $3\omega+\omega+\omega=5\omega$ or $3\omega+3\omega-\omega=5\omega$ as summarized in Table 1.

A common way to increase FHG efficiency is the use of a focused fundamental beam. Conditions for optimal focusing for the direct process and for the case when only one of the steps is phase matched, are considered in [4, 7–9]. The fifth harmonic generation by third order cascading however, is most efficient when both steps (i.e. $\omega+\omega+\omega=3\omega$ and $3\omega+\omega+\omega=5\omega$) are simultaneously phase matched. Although it is not straight forward, such simultaneous phase matching of several processes can be achieved by using a multicomponent gas mixture or appropriate quasi-phased-matched (QPM) structures [2, 11, 12].

To best of current knowledge there is no systematic investigation of the role of focusing on the efficiency of FHG in this very interesting case. This paper presents a detailed study of the optimal conditions for FHG with a focused fundamental beam through the chain of interactions A as shown in Table 1. The system of reduced amplitude equations (1) describing the process of fifth harmonic generation in focused beams in three levels of approximation were solved. The case of weak and arbitrary focusing are considered separately in the condition of the non depleted approximation of the fundamental beam. To account for the deletion of the fundamental beam, a direct numerical approach was used. It was found that optimal phase

	Type of the process	I step	II step	Observed at phase matching
A	cascaded	$\omega+\omega+\omega$ $=3\omega$	$3\omega+\omega+\omega$ $=5\omega$	$k_1+k_1+k_1\sim k_3$ or/and $k_3+k_1+k_1\sim k_5$ or $k_1+k_1+k_1+k_1+k_1\sim k_5$
B	cascaded	$\omega+\omega+\omega$ $=3\omega$	$3\omega+3\omega-\omega$ $=5\omega$	$k_1+k_1+k_1\sim k_3$ or/and $k_3+k_3-k_1\sim k_5$ or $k_1+k_1+k_1+k_1+k_1\sim k_5$
C	direct	$\omega+\omega+\omega+\omega+\omega=5\omega$		$k_1+k_1+k_1+k_1+k_1\sim k_5$

TABLE 1 Some of the interactions and the phase matching conditions for fifth harmonic generation in single nonlinear cenrosymmetric media

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mismatches $\Delta k_{1,\mathrm{opt}}$ and $\Delta k_{2,\mathrm{opt}}$ exist for both steps and that the optimal position of the beam waist is in the center of the nonlinear medium. Any deviation of Δk_1 and Δk_2 from their optimal value results in a shift of the optimal position of the focus.

2 Main equations

The effect of FHG as a result of the simultaneous action of the processes of third harmonic generation, and four wave mixing of the fundamental and third harmonic wave (chain A in Table 1), can be described by the following system of reduced amplitude equations, derived in the assumption of zero absorption of all interacting waves:

$$\begin{aligned}
&\left(\frac{\partial}{\partial z}+\frac{\mathrm{i}}{2k_1}\Delta_\perp\right)A_1 = \\
&-\mathrm{i}\gamma_1 A_3\left(A_1^*\right)^2\exp(-\mathrm{i}\Delta k_1 z)-2\mathrm{i}\gamma_2 A_5 A_3^* A_1^*\exp(-\mathrm{i}\Delta k_2 z),\\
&\left(\frac{\partial}{\partial z}+\frac{\mathrm{i}}{2k_3}\Delta_\perp\right)A_3 = \\
&-\mathrm{i}\gamma_1 A_1^3\exp(\mathrm{i}\Delta k_1 z)-\mathrm{i}3\gamma_2 A_5\left(A_1^*\right)^2\exp(-\mathrm{i}\Delta k_2 z),\\
&\left(\frac{\partial}{\partial z}+\frac{\mathrm{i}}{2k_5}\Delta_\perp\right)A_5 = -\mathrm{i}5\gamma_2\left(A_1\right)^2 A_3\exp(\mathrm{i}\Delta k_2 z). \qquad (1)
\end{aligned}$$

Here A_1, A_2, A_3 denote the complex amplitudes of the fundamental, third and the fifth harmonic waves, $\Delta_\perp$ stands for the operator $\frac{\partial^2}{\partial x^2}+\frac{\partial^2}{\partial y^2}$, and $\gamma_1 = 6\pi\chi^{(3)}_{(\mathrm{eff,THG})}/(8\lambda_1 n_1)$, and $\gamma_2 = 6\pi\chi^{(3)}_{(\mathrm{eff,FWM})}/(8\lambda_1 n_1)$ are the nonlinear coupling coefficients. The wave-vector mismatches are defined as $\Delta k_1 = k_3 - 3k_1$ and $\Delta k_2 = k_5 - k_3 - 2k_1$, where k_j are the wave vectors of the waves involved in the process.

For low input fundamental intensities, the effects of depletion of the fundamental and third harmonic waves can be neglected and eqn. (1) reduces to

$$\begin{aligned}
&\left(\frac{\partial}{\partial z}+\frac{\mathrm{i}}{2k_1}\Delta_\perp\right)A_1 = 0,\\
&\left(\frac{\partial}{\partial z}+\frac{\mathrm{i}}{2k_3}\Delta_\perp\right)A_3 = -\mathrm{i}\gamma_1 A_1^3\exp(\mathrm{i}\Delta k_1 z),\\
&\left(\frac{\partial}{\partial z}+\frac{\mathrm{i}}{2k_5}\Delta_\perp\right)A_5 = -\mathrm{i}5\gamma_2 A_3 A_1^2\exp(\mathrm{i}\Delta k_2 z). \qquad (2)
\end{aligned}$$

Equation (2) allows some of the results to be obtained in analytical form that will help to gain physical insight of the process of cascaded FHG.

The other possible chain of cascaded interactions leading to FHG (marked as B in Table 1) can be described with the system of differential equations similar to (2) except for the last equation that has the form:

$$\left(\frac{\partial}{\partial z}+\frac{\mathrm{i}}{2k_5}\Delta_\perp\right)A_5 = -\mathrm{i}5\gamma_2\left(A_3\right)^2 A_1^*\exp(\mathrm{i}\Delta k_4 z) \qquad (3)$$

where $\Delta k_4 = k_5 - 2k_3 + k_1$.

The rest of this paper will concentrate on the chain of processes **A**; results for the second cascading possibility can be obtained in a similar way.

Systems (1) and (2) are solved assuming only one input beam – the fundamental that has Gaussian spatial distribution. Its propagation is described by [4, 9]

$$A_1(x,y,z) = A_\mathrm{o} u_1(x,y,\xi_1) \qquad (4)$$

with

$$u_1(x,y,\xi_1) = \frac{1}{1-\mathrm{i}\xi_1}\exp\left[-\frac{x^2+y^2}{w_{\mathrm{o}1}^2(1-\mathrm{i}\xi_1)}\right]$$

where $\xi_1 = 2(z-z_\mathrm{o})/b_1$, with z_o is marked the position of the focal spot, b is the confocal parameter of the fundamental beam, $b_1 = k_1 w_{\mathrm{o}1}^2$, $w_{\mathrm{o}1}$ is the focal spot radius, and A_o is the electric field amplitude at the center of the focal spot $(0, 0, z_\mathrm{o})$. With these notations the distribution of the fundamental field at the entrance of the nonlinear medium ($z = 0$) is

$$A(x,y,0) = A_\mathrm{in}\exp\left[-\left(x^2+y^2)(\frac{1}{w_\mathrm{in}^2}-\frac{\mathrm{i}k_1}{2R_1}\right)\right], \qquad (5)$$

where the amplitude, beam radius, and wave-front curvature at the nonlinear medium entrance are $A_\mathrm{in} = (\frac{w_{\mathrm{o}1}^2}{w_\mathrm{in}^2}-\mathrm{i}\frac{b}{2R_1})A_\mathrm{o}$, $w_\mathrm{in}^2 = w_{\mathrm{o}1}^2(1+4\frac{z_\mathrm{o}^2}{b^2})$, and $R_1 = (z_\mathrm{o}+\frac{b^2}{4z_\mathrm{o}})$ respectively.

3 Non depleted approximation

The starting point of considerations will be the solution for the amplitude of the fifth harmonic beam that is obtained from (2) by applying the procedure of trial solution as described in [13, 14].

It must be emphasized that in the non depletion approximation the fundamental beam will preserve its Gaussian form. Moreover, the generated third and fifth harmonics will be Gaussian beams as well, but with different focal spot radii:

$$A_3(x,y,z) = T(z)u_3(x,y,\xi_3) \qquad (6a)$$

with $u_3(x,y,\xi_3) = \frac{1}{1-\mathrm{i}\xi_3}\exp\left[-\frac{x^2+y^2}{w_{\mathrm{o}3}^2(1-\mathrm{i}\xi_3)}\right]$

$$A_5(x,y,z) = F(z)u_5(x,y,\xi_5), \qquad (6b)$$

with $u_5(x,y,\xi_5) = \frac{1}{1-\mathrm{i}\xi_5}\exp\left[-\frac{x^2+y^2}{w_{\mathrm{o}5}^2(1-\mathrm{i}\xi_5)}\right]$

In (6a) and (6b), $\xi_3 = 2(z-z_\mathrm{o})/b_3$, and $\xi_5 = 2(z-z_\mathrm{o})/b_5$. The amplitudes A_o, T and F that appear in (4) and (6) are the electric field amplitudes that correspond to the center of the focal spot $(0, 0, z_\mathrm{o})$. Importantly, they depend on the length of the nonlinear media. The spot radii are connected by the relation $w_{\mathrm{o}j} = \frac{w_{\mathrm{o}j}}{\sqrt{j}}$ [13–15]. As a result all interacting waves have practically the same confocal parameters $b_5 \cong b_3 \cong b_1 = b$ (neglecting a slight difference in the index of refraction of the three waves) and this means that $\xi_5 \cong \xi_3 \cong \xi_1 = \xi$.

Substituting (4) and (6) into (2) and noting that

$$\begin{aligned}
&(u_1)^3 = \frac{u_3}{(1-\mathrm{i}\xi)^2}\\
&(u_1)^2 u_3 = \frac{u_5}{(1-\mathrm{i}\xi)^2} \qquad (7)
\end{aligned}$$

gives the following system of first order differential equations, that are equivalent to (2) but easier to solve.

$$\frac{\partial A_o}{\partial z} = 0,$$
$$\frac{\partial T}{\partial z} = -\frac{\mathrm{i}\gamma_1 A_o^3}{(1-\mathrm{i}\xi)^2}\exp(\mathrm{i}\Delta k_1 z),$$
$$\frac{\partial F}{\partial z} = -\frac{\mathrm{i}5\gamma_2 T A_o^2}{(1-\mathrm{i}\xi)^2}\exp(\mathrm{i}\Delta k_2 z). \tag{8}$$

The straightforward solution of (8) gives

$$T(\xi) = -\frac{\mathrm{i}\gamma_1 A_o^3 b}{2}\exp(\mathrm{i}\Delta k_1 z_o)\int_{\beta_1}^{\xi}\frac{\exp(\mathrm{i}\Delta k_1 b\tau/2)}{(1-\mathrm{i}\tau)^2}\mathrm{d}\tau \tag{9}$$

$$F = G\int_{\beta_1}^{\beta_2}\frac{\exp(\mathrm{i}\Delta k_2 b\xi/2)}{(1-\mathrm{i}\xi)^2}\int_{\beta_1}^{\xi}\frac{\exp(\mathrm{i}\Delta k_1 b\tau/2)}{(1-\mathrm{i}\tau)^2}\mathrm{d}\tau\mathrm{d}\xi \tag{10}$$

where the limits of integrations are $\beta_1 = -m(1+2l)$, $\beta_2 = m(1-2l)$; with m denoting the strength of focussing $m = L/b$, (L is the length of the nonlinear medium), and l is the dimensionless parameter indicating the position of the focus, $l = (2z_o - L)/2L$. The coefficient G in (10) is

$$G = -\frac{5\gamma_1\gamma_2 A_o^5 b^2}{4}\exp(\mathrm{i}z_o(\Delta k_1 + \Delta k_2)). \tag{11}$$

The efficiency of the fifth harmonic generation is defined as

$$\eta_{5\omega} = \frac{\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} A_5 A_5{}^*\mathrm{d}x\mathrm{d}y}{\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} A_1 A_1^*\mathrm{d}x\mathrm{d}y}. \tag{12}$$

After performing the integration, the following is obtained

$$\eta_{5\omega} = \frac{1}{5}\frac{|F|^2}{|A_o|^2}.$$

Equation (10) can be solved analytically in the weak focussing approximation. For arbitrary focusing (10) has to be solved numerically.

3.1 *Weak focussing limit*

When considering very weak focusing, $m \ll 1$ the limits of integration in (10) will be much smaller than 1 and as a result yields

$$F(L) = \frac{4G\exp(-\mathrm{i}ma_3 l)}{a_3}\left[\frac{a_3}{a_1 a_2}\exp\left(\frac{\mathrm{i}m(a_2-a_1)}{2}\right) - \frac{\exp(\mathrm{i}ma_3/2)}{a_1} - \frac{\exp(-\mathrm{i}ma_3/2)}{a_2}\right], \tag{13}$$

where, $a_1 = b\Delta k_1 + 4$, $a_2 = b\Delta k_2 + 4$, $a_3 = a_1 + a_2$. Analyzing expression (13) gives four possibilities for the phase-matched generation of the cascaded fifth harmonic in a cubic media in a condition of weak focussing:

i) Phase-matching for the first step $\omega+\omega+\omega = 3\omega$. The maximum FH efficiency is obtained when $a_1 = b\Delta k_1 + 4 = 0$ and the required deviation from exact phase matching is: $\Delta k_1 L = -4m$. The conversion efficiency into the fifth harmonic with the condition $a_1 \ll a_2$ is

$$\eta_{5\omega}(a_1 \ll a_2) = \frac{20\gamma_1^2\gamma_2^2|A_o|^8 b^4}{a_3^2}\frac{\sin^2(ma_1/2)}{a_1^2}; \tag{14}$$

ii) Phase-matching for the second step. The maximum FH efficiency is obtained when $a_2 = b\Delta k_2 + 4 = 0$ that corresponds to the deviation $\Delta k_2 L = -4m$. Then for $a_2 \ll a_1$ the following is obtained

$$\eta_{5\omega}(a_2 \ll a_1) = \frac{20\gamma_1^2\gamma_2^2|A_o|^8 b^4}{a_3^2}\frac{\sin^2(ma_2/2)}{a_2^2}; \tag{15}$$

iii) Phase-matching when both steps $\omega+\omega+\omega = 3\omega$ and $\omega+\omega+3\omega = 5\omega$ are non-phase-matched, but $a_3 = a_2 + a_1 \approx 0$. The optimal phase mismatch for this case is $(\Delta k_2 + \Delta k_1)L = \Delta k_3 L = -8m$. The generated fifth harmonic when $a_3 \ll a_1, a_2$ is governed by

$$\eta_{5\omega}(a_3 \ll a_1, a_2) = \frac{20\gamma_1^2\gamma_2^2|A_o|^8 b^4}{a_1^2}\frac{\sin^2(ma_3/2)}{a_3^2}; \tag{16}$$

iv) The last and most interesting possibility that is characterized with the highest FH efficiency is the situation when both steps are simultaneously phase-matched, $a_1 \approx 0$, $a_2 \approx 0$. The expression for FH conversion efficiency in these conditions takes the form:

$$\eta_{5\omega}(a_1, a_2 \le 1) \approx 20\gamma_1^2\gamma_2^2|A_o|^8 b^4 \times \frac{\sin^2(ma_2/2)}{a_2^2}\frac{\sin^2(ma_1/2)}{a_1^2}. \tag{17}$$

The optimal deviations from the exact phase-matching condition for the both steps are equal: $\Delta k_2 L = \Delta k_1 L = -4m$. Equation (17), when double phase-matching conditions are fulfilled, exceeds (14)–(16) with the magnitude of the square of the normalized phase mismatch $|a_1|^2$ or $|a_2|^2$of the "step" that is not phase-matched.

The main conclusion at this level of consideration is that even at weak focussing a deviation from the exact phase matching conditions is required in order to optimize the process of cascaded FHG. The required "shift" from the exact phase matching is $-4m$. It was verified by direct numerical integration of (10) that the analytical formulas (13)–(17) can be used until $m < 1$.

3.2 *Arbitrary focussing*

This section considers the arbitrary value of the strength of focusing m, while still keeping the nondepletion approximation for the fundamental and third harmonic beam. The efficiency conversion in FH is calculated from (10) and is found to be:

$$\eta_{5\omega} = \frac{5S^4}{16}\left|\int_{\beta_1}^{\beta_2}\frac{\exp(\mathrm{i}\Delta k_2 b\xi/2)}{(1-i\xi)^2}\int_{\beta_1}^{\xi}\frac{\exp(\mathrm{i}\Delta k_1 b\tau/2)}{(1-i\tau)^2}\mathrm{d}\tau\mathrm{d}\xi\right|^2 \tag{18}$$

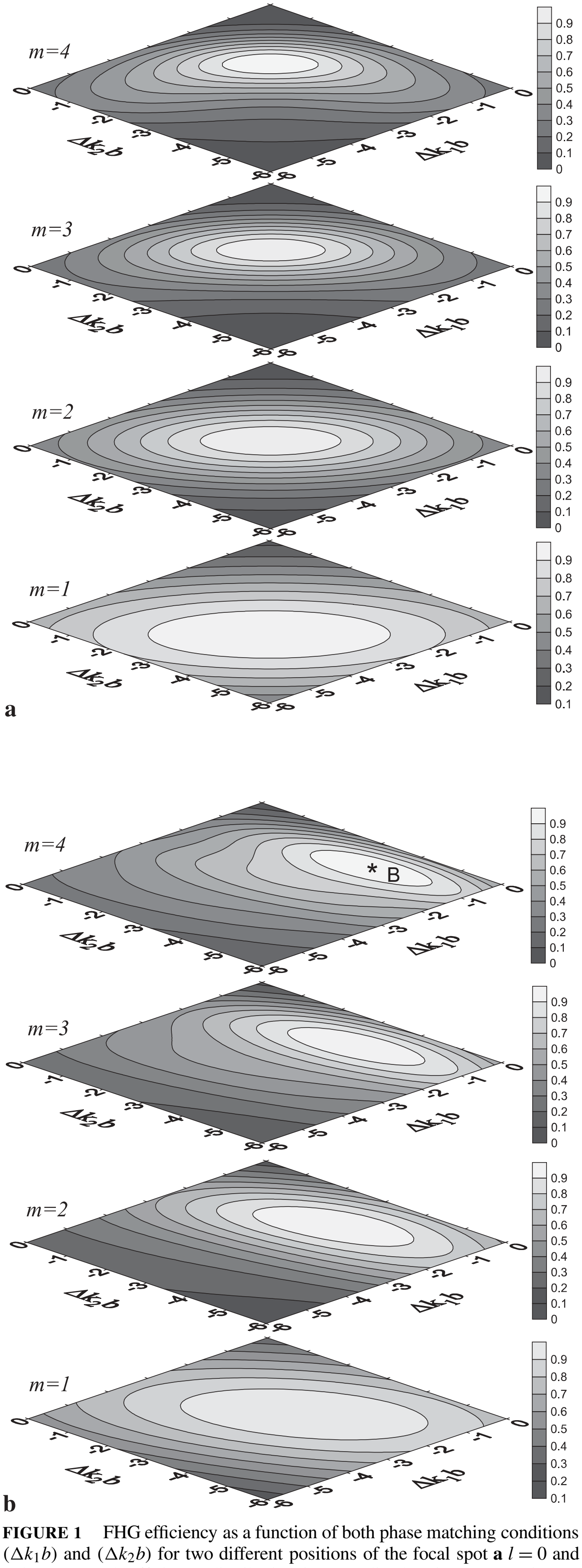

FIGURE 1 FHG efficiency as a function of both phase matching conditions $(\Delta k_1 b)$ and $(\Delta k_2 b)$ for two different positions of the focal spot **a** $l = 0$ and **b** $l = 0.4$. Each efficiency distribution is normalized to its own maximum. Normalized input intensity $S = 0.3$

In (18) $S = \sqrt{\gamma_1\gamma_2}|A_{\mathrm{o}}|^2 b = \sqrt{\gamma_1\gamma_2}\dfrac{8P_1}{\varepsilon_{\mathrm{o}} c\lambda_1}$ (where P_1 – input power)

The dependence of $\eta_{5\omega}$ on the four parameters, describing the system: strength of focusing m, position of focusing l, and conditions for phase matching of both steps $\Delta k_1 b$, $\Delta k_2 b$ were investigated. For maximum FHG efficiency it is necessary to tune both mismatches to their optimal values $(\Delta k_1, \Delta k_2) = (\Delta k_{1,\mathrm{opt}}, \Delta k_{2,\mathrm{opt}})$. This can be seen on the contour plots shown in Fig. 1a and b as calculated for four different strengths of focusing. Figure 1 a illustrates the 2D phase-matching curves when focusing is in the center of the nonlinear medium, $l = 0$ while Fig. 1b is calculated for the focusing position $l = 0.4$, close to the output face of the nonlinear medium. Each efficiency distribution is normalized to its own maximum. Figure 2a and b allow the obtaining of the

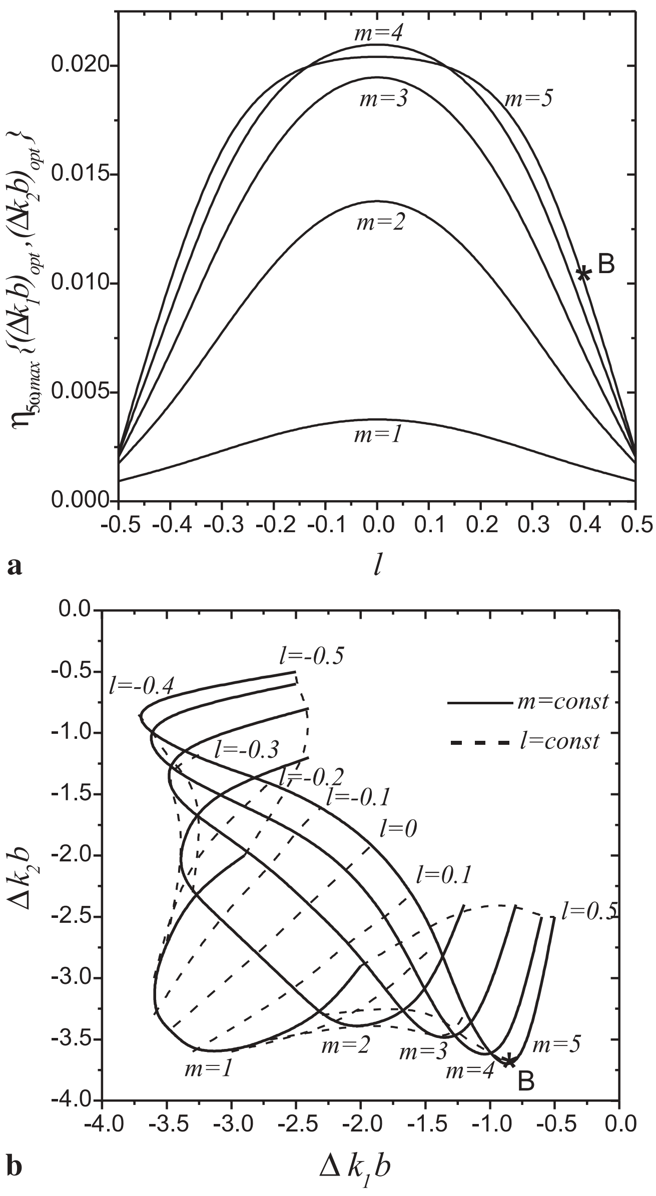

FIGURE 2 **a** FHG efficiency $\eta_{5\omega}$ calculated at optimal phase matching conditions as a function of position of focusing l and **b** optimal phase matching for both steps for several values of level of focusing m and the position of focusing l. Normalized input intensity $S = 0.3$

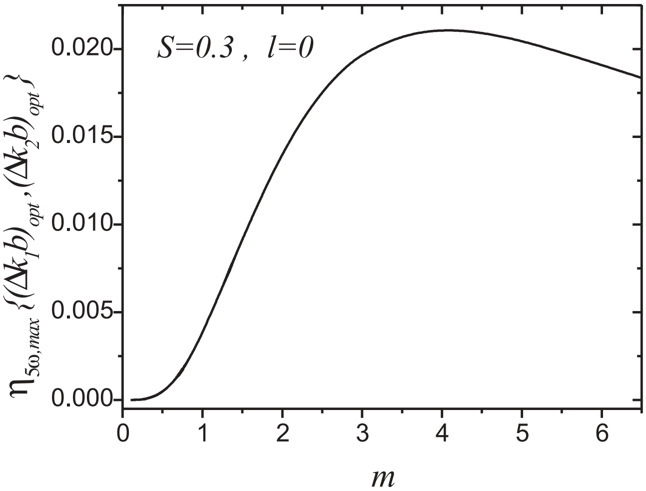

FIGURE 3 FHG efficiency as a function of the strength of focussing m for focusing in center of the nonlinear cenrosymmetric media

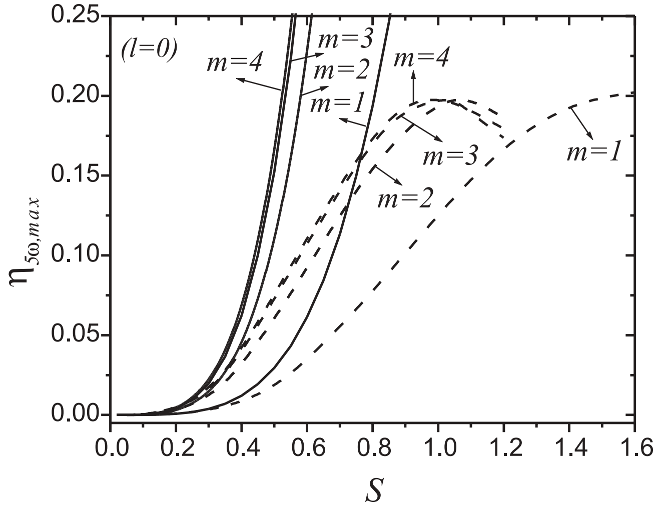

FIGURE 4 Dependence of FHG efficiency on normalized input intensity. *Solid line* – calculations with the semi analytical approach given in Sect. 3.2. *Dash line* – direct numerical integration of system (1) for the case $\gamma_1 = \gamma_2$

maximum $\eta_{5\omega}$ for arbitrary values of these four parameters. Figure 2a shows the dependence of the conversion efficiency $\eta_{5\omega}$ on the strength of focusing m, and the position of focusing l. The optimal $(\Delta k_1 b)_{\rm opt}$ and $(\Delta k_2 b)_{\rm opt}$ for each point of Fig. 2a can be found from Fig. 2b. For example, as seen in Fig. 2a point B ($m = 5$, $l = 0.4$, $S = 0.3$ with efficiency $\eta_{5\omega} = 1.04\%$), this corresponds to the following optimal phase mismatches: $(\Delta k_1 b)_{\rm opt} = -0.85$, $(\Delta k_2 b)_{\rm opt} = -3.75$, as can be found from Fig. 2b.

It was found that in the process of cascaded fifth harmonic generation in a single cubic nonlinear medium, with a focussed beam, that for maximum efficiency the phase matching $\Delta k_1 b$ and $\Delta k_2 b$ have optimal values, different from 0, and that these values are different for both m and l. However, for best conversion the optimal position for focusing is in the center of the nonlinear medium. Figure 3 shows the dependence of the FH efficiency on the focusing parameter m. The optimal strength of focusing for a small conversion efficiency is $m = 4.02$. Recall that the direct process of FHG at the phase matching condition has no optimum focusing parameter [8].

4 Accounting for depletion

For calculating the process of the single crystal cascade FHG, without neglecting the depletion of the fundamental and third harmonic beams, and evaluating the applicability of the nondepleting approach used in the proceeding sections, (1) was solved by direct numerical integration. For this purpose FORTRAN code was written based on the Split-Step Fourier Method. The base principle of this method is the assumption that in propagating the nonlinear media over a small distance h, the diffraction and nonlinear effects act independently. The Fast Fourier Transformation (FFT) algorithm was used to calculate the diffraction effects, and the Runge–Kuta method was used for the nonlinear effects.

The investigation, carried out at low input fundamental intensities, confirms the results in paragraph 3.2. – the optimal position of focusing is in the center of the nonlinear medium with $(\Delta k_1 b)_{\rm opt}$ and $(\Delta k_2 b)_{\rm opt}$, and the optimal strength of focusing has the same value. Figure 4 shows calculations for $m = 1$; 2; 3 and 4, and allows the establishment of the maximum normalized intensity of the semi analytical approach described in Sect. 3.2. It can be concluded that this approach can be used for the normalized fundamental beam intensity $S \leq 0.3$. In addition, it is seen that with the depletion effects taken into account, the maximum conversion practically does not depend on the strengths of focusing.

5 Conclusion

In conclusion, this work presented both analytical and numerical investigations of the process of cascaded fifth harmonic generation in single centrosymmetric nonlinear media, in a condition of simultaneous phase matching of both steps and a focused fundamental beam. If the design of the nonlinear media allows tuning of the phase matching conditions to its optimal values, then optimal focussing is in the center of the crystal. If the phase matching parameters can not be changed and they deviate from the optimal values, then the optimal position of the focus spot should be calculated according to the analysis presented here.

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