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Efficiency of cascaded third harmonic generation in single quadratic crystal in focused beam

Rosen Ivanov *, Kaloian Koynov, Solomon Saltiel

Quantum Electronics Department, Faculty of Physics, University of Sofia, BG-1164 Sofia, Bulgaria

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Abstract

Generation of third harmonic by cascading of two phase-matched or two near phase-matched second order processes in single quadratic crystal with focused fundamental beam is theoretically investigated. The optimized conditions for maximum conversion into third harmonic are found with the help of analytical and numerical investigations. In general the optimal position of focusing depends on the values of the mismatches Δk_1 and Δk_2 for both "steps" of the second order cascading ($\omega + \omega = 2\omega$; $\omega + 2\omega = 3\omega$). It is shown that this method of third harmonic generation requires specially chosen $\Delta k_{1.\mathrm{opt}}$ and $\Delta k_{2.\mathrm{opt}}$ and focusing into the center of the nonlinear media in order to obtain best efficiency.

Keywords: 42.65.−k; 42.65.Ky; 42.79.Nv

Keywords: Nonlinear optics; Harmonic generation; Frequency conversion; Optical frequency converters

1. Introduction

Third harmonic generation (THG) in single quadratic crystal is one of the most extensively investigated cascading schemes. This interaction is a result of simultaneous action of two phase-matched processes $\omega + \omega = 2\omega$; $\omega + 2\omega = 3\omega$. The first proposals for single crystal THG on the base of second order nonlinearities can be found in the book of Akhmanov and Khokhlov [1]. One can find there the plane wave analysis of the condition for third harmonic generation in single quadratic crystal. A great part of the experimental works on cascaded THG have been done in conditions that only one of the two steps (second harmonic generation or sum frequency mixing) was phase-matched [2–12]. The maximum efficiency achieved with one of the phase-matched steps is 6% [8,9]. Much higher efficiency can be expected if both steps are simultaneously phase-matched. The first attempts [13,14] to fulfill simultaneously two phase-matching conditions were not successful. Nowadays the situation is totally different due to the methods for designing nonlinear media with periodical and quasi-periodical spatial modulation of the quadratic nonlinearity

* Corresponding author. Fax: +359-2-96-25-276.
E-mail address: ris@phys.uni-sofia.bg (R. Ivanov).

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[15,16]. Several methods that allow simultaneous phase-matching of two or more second order processes have been suggested [16–26]. In THG experiment in $LiTaO_3$ crystal with Fibonacci quasi-periodic superlattices with both fulfilled phase-matching conditions, 23% efficiency of the overall process $\omega \to 3\omega$ was achieved [21,22]. Experimental double phase-matched third harmonic generation is reported also in [27]. Recently it is calculated that a simultaneously phase-matching of both steps can be obtained in a photonic band gap structure [23,24,28]. Two-dimensional nonlinear photonic crystals are also promising nonlinear media for simultaneous phase-matching of several processes [18,19].

Very recently several theoretical investigations [29–32] of plane wave interactions in quadratic media demonstrated the possibility of 100% conversion for THG, if the ratio of the nonlinearities responsible for the two processes is optimized. Multiple nonlinear optical interactions including cascaded THG in quasi-periodic superlattices with nonzero vector mismatches have been reported recently in single KTP crystal [25]. In this work the authors used focusing in the center of the crystal. Not exact phase-matching for both steps is the reason for small THG efficiency. However, as we will show the efficiency could be improved by using off-center focusing.

One of the approaches to achieve better conversion with moderate input intensities in the processes for harmonic generation is to use focused beams. The cases of single process of second [33,34], third [34–36] and fifth [34,36,37] harmonic generation in focused beams have been investigated in the past and optimum conditions have been defined. For example, the optimal focusing for the direct processes is in the center of the nonlinear crystal. Concerning single quadratic crystal THG, by now only the cases when only one of the two steps ($\omega + \omega = 2\omega$ or $\omega + 2\omega = 3\omega$) are phase-matched were considered [2,3]. The theoretical and experimental investigations of Rostovtceva et al. [2,3] demonstrate that the optimal position of the fundamental beam focus depends on the fact which step is phase-matched: if the first step is phase-matched, the position of the focus should be at the output face of the quadratic crystal and if the second step is phase-matched, the position of the focus should be at the input face of the quadratic crystal. According to our knowledge no attempts have been done to calculate the optimum conditions for single crystal THG when both steps are phase-matched or near phase-matched.

The purpose of the present work is to investigate the process of THG by cascading two quadratic processes that are simultaneously near phase-matched in condition of the focused fundamental beam. We consider separately the case of weak and arbitrary focusing in condition of nondepleted approximation of the fundamental beam. To account for the depletion of the fundamental beam we used direct numerical approach.

2. Main equations

The effect of THG, as a result of simultaneous action of the processes of second harmonic generation and sum frequency mixing of the fundamental and the second harmonic wave, is described by the following system of differential equations [2,38,39], derived in assumption of zero absorption of all interacting waves:

$$\begin{aligned}
\left(\frac{\partial}{\partial z} + \frac{i}{2k_1}\Delta_\perp\right)A &= -\mathrm{i}\sigma_1 SA^* \exp(-\mathrm{i}\Delta k_1 z) \\
&\quad - \mathrm{i}\sigma_2 TS^* \exp(-\mathrm{i}\Delta k_2 z), \\
\left(\frac{\partial}{\partial z} + \frac{i}{2k_2}\Delta_\perp\right)S &= -\mathrm{i}\sigma_1 A^2 \exp(\mathrm{i}\Delta k_1 z) \\
&\quad - \mathrm{i}2\sigma_2 TA^* \exp(-\mathrm{i}\Delta k_2 z), \\
\left(\frac{\partial}{\partial z} + \frac{i}{2k_3}\Delta_\perp\right)T &= -\mathrm{i}3\sigma_2 SA \exp(\mathrm{i}\Delta k_2 z),
\end{aligned} \tag{1}$$

where A, S, T denotes the complex amplitudes of the fundamental, second and the third harmonic waves. $\Delta_\perp$ stands for the operator $\frac{\partial^2}{\partial x^2} + \frac{\partial^2}{\partial y^2}$. Nonlinear coupling coefficients are calculated as $\sigma_1 = 2\pi d_{\mathrm{eff,SHG}}/(\lambda_1 n_1)$ and $\sigma_2 = 2\pi d_{\mathrm{eff,SFM}}/(\lambda_1 n_1)$, where the magnitude of the $d_{\mathrm{eff,SHG}}$ and $d_{\mathrm{eff,SFM}}$ depends on the phase-matching method and type of the nonlinear media that are used. The wave vector mismatches are defined as $\Delta k_1 = k_2 - 2k_1$

and $\Delta k_2 = k_3 - k_2 - k_1$, where k_j are the wave vectors of the waves involved in the process. Note that the sum of the two-phase mismatch parameters is exactly the phase mismatch parameter Δk_3 for the direct process for THG ($\omega + \omega + \omega = 3\omega$; $\Delta k_3 = \Delta k_1 + \Delta k_2 = k_3 - 3k_1$). In general third harmonic wave will be also generated as a result of a direct process governed by the cubic nonlinearly of the media. This contribution to the TH signal is in the same order of magnitude as the TH signal, due to the second order cascading when only one of the cascading steps is phase-matched. But if both Δk_1 or Δk_2 are small, the TH signal generated due to inherent cubic nonlinearly can be neglected.

For not very high input fundamental intensities the effects of depletion of the fundamental and second harmonic waves can be neglected and the system (1) is reduced to

$$\begin{aligned}
&\left(\frac{\partial}{\partial z} + \frac{i}{2k_1}\Delta_\perp\right)A = 0,\\
&\left(\frac{\partial}{\partial z} + \frac{i}{2k_2}\Delta_\perp\right)S = -\mathrm{i}\sigma_1 A^2 \exp(\mathrm{i}\Delta k_1 z), \qquad (2)\\
&\left(\frac{\partial}{\partial z} + \frac{i}{2k_3}\Delta_\perp\right)T = -\mathrm{i}3\sigma_2 SA \exp(\mathrm{i}\Delta k_2 z).
\end{aligned}$$

System (2) allows some of the results to be obtained in analytical form that will help to obtain physical insight of the cascaded THG process.

We solve systems (1) and (2) with only one input beam – the fundamental that has Gaussian spatial distribution. Its propagation is described by [36]

$$A(x,y,z) = \frac{F_0}{1 - \mathrm{i}v} \exp\left[-\frac{x^2 + y^2}{w_0^2(1 - \mathrm{i}v)}\right], \qquad (3)$$

where $v = 2(z - z_0)/b$, z_0 marks the position of the focal spot, b is the confocal parameter of the fundamental beam, $b = k_1 w_0^2$, w_0 is the focal spot radius, F_0 is the electric field amplitude at the center of the focal spot $(0, 0, z_0)$. With these notations the distribution of the fundamental field at the entrance of the nonlinear media $(z = 0)$ is

$$A(x,y,0) = A_0 \exp\left[-\left(x^2 + y^2\right)\left(\frac{1}{w_1^2} - \frac{\mathrm{i}k_1}{2R_1}\right)\right], \qquad (4)$$

where the maximum amplitude, beam radius and wave-front curvature at the crystal entrance are

$$A_0 = \left(\frac{w_0^2}{w_1^2} - \mathrm{i}\frac{b}{2R_1}\right)F_0,$$

$$w_1^2 = w_0^2\left(1 + 4\frac{z_0^2}{b^2}\right),$$

$$R_1 = \left(z_0 + \frac{b^2}{4z_0}\right),$$

respectively.

3. Nondepleted approximation

The starting point of our consideration will be the solution for the amplitude of the third harmonic beam that is obtained from system (2) by applying Green's functions technique

$$T(x,y,z) = G\int_{-m(1+2l)}^{m(1-2l)} \frac{\exp(\mathrm{i}b\Delta k_2\eta/2)}{1 - \mathrm{i}\eta} \times \int_{-m(1+2l)}^{\eta} \frac{\exp(\mathrm{i}b\Delta k_1\tau/2)}{1 - \mathrm{i}\tau}\, \mathrm{d}\tau\, \mathrm{d}\eta, \qquad (5)$$

where m denotes the strength of focusing $m = L/b$, (L, the length of the nonlinear media) and l is the dimensionless parameter indicating the position of the focus, $l = (2z_0 - L)/2L$. The coefficient in Eq. (5) is

$$G = -\frac{3\sigma_1\sigma_2 F_0^3 b^2}{4} \frac{\exp(-3(x^2 + y^2)/w_0^2(1 - \mathrm{i}v))}{(1 - \mathrm{i}v)} \times \exp(\mathrm{i}z_0(\Delta k_1 + \Delta k_2)).$$

3.1. Weak focusing limit

Let us first consider weak focusing, $m \ll 1$. Then the limits in (5) will be much smaller than 1 and as a result we have

$$T(x,y,z) = \frac{4G\mathrm{e}^{(-\mathrm{i}ma_3 l)}}{a_3}\left[\frac{a_3}{a_1 a_2}\exp\left(\frac{\mathrm{i}m(a_2 - a_1)}{2}\right) - \frac{\exp(\mathrm{i}ma_3/2)}{a_1} - \frac{\exp(-\mathrm{i}ma_3/2)}{a_2}\right], \qquad (6)$$

where $a_1 = b\Delta k_1 + 2$, $a_2 = b\Delta k_2 + 2$, $a_3 = b\Delta k_3 + 4$.

Analyzing expression (6) we find out four possibilities for phase-matched generation of cascaded third harmonic in quadratic crystals in condition of weak focusing:

(a) phase-matching for the first step $\omega + \omega = 2\omega$. The maximum TH efficiency is obtained when $a_1 = b\Delta k_1 + 2 = 0$ and the required deviation from exact phase-matching is: $\Delta k_1 L = -2m$. The conversion efficiency into the third harmonic in condition of $a_1 \ll a_2$ is

$$\eta_{3\omega}(a_1 \ll a_2) = \frac{12\sigma_1^2\sigma_2^2|F_0|^4 b^4}{a_3^2} \frac{\sin^2(ma_1/2)}{a_1^2}; \tag{7}$$

(b) phase-matching for the second step. The maximum TH efficiency is obtained when $a_2 = b\Delta k_2 + 2 = 0$ that corresponds to deviation $\Delta k_2 L = -2m$. The magnitude of the third harmonic field in condition of $a_2 \ll a_1$ is

$$\eta_{3\omega}(a_2 \ll a_1) = \frac{12\sigma_1^2\sigma_2^2|F_0|^4 b^4}{a_3^2} \frac{\sin^2(ma_2/2)}{a_2^2}; \tag{8}$$

(c) phase-matching when both steps $\omega + \omega = 2\omega$ and $\omega + 2\omega = 3\omega$ are nonphase-matched, but $a_3 = a_2 + a_1 \approx 0$. This is the case of THG in the conditions for direct third harmonic generation by the process $\omega + \omega + \omega = 3\omega$. The optimal phase mismatch for this case is $(\Delta k_2 + \Delta k_1)L = \Delta k_3 L = -4m$. This result coincides with the optimal Δk shift for the THG based on direct $\chi^{(3)}$ process [34–36]. The amplitude of the third harmonic field in condition of $a_3 \ll a_1, a_2$ is

$$\eta_{3\omega}(a_3 \ll a_1, a_2) = \frac{12\sigma_1^2\sigma_2^2|F_0|^4 b^4}{a_1^2} \frac{\sin^2(ma_3/2)}{a_3^2}; \tag{9}$$

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

$$\eta_{3\omega}(a_1, a_2 \leqslant 1) \approx 12\sigma_1^2\sigma_2^2|F_0|^4 b^4 \frac{\sin^2(ma_2/2)}{a_2^2} \times \frac{\sin^2(ma_1/2)}{a_1^2}. \tag{10}$$

The optimal deviations from the exact phase-matching condition for the both steps are equal: $\Delta k_2 L = \Delta k_1 L = -2m$. The expression (10) when double phase-matching conditions are fulfilled exceeds the expressions (7)–(9) 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 focusing it is required deviation of exact phase-matching conditions in order to optimize the process of cascaded THG. The required "shift" from the exact phase-matching is $-2m$. We verified by direct numerical integration of Eq. (5) that the analytical formulas (7)–(10) can be used until $m \leqslant 1$.

3.2. Arbitrary focusing

In this section we consider arbitrary value of the strength of focusing m and nondepletion approximation for the fundamental and second harmonic beam.

The efficiency conversion in TH is calculated from (5) and is found to be

$$\eta_{3\omega} = \frac{3S^4}{16}\left| \int_{-m(1+2l)}^{m(1-2l)} \frac{\exp(\mathrm{i}b\Delta k_2\eta/2)}{1-\mathrm{i}\eta} \times \int_{-m(1+2l)}^{\eta} \frac{\exp(\mathrm{i}b\Delta k_1\tau/2)}{1-\mathrm{i}\tau}\,\mathrm{d}\tau\,\mathrm{d}\eta \right|^2, \tag{11}$$

where $S = \sqrt{\sigma_1\sigma_2}|F_0|b$.

The dependence of $\eta_{3\omega}$ on the four parameters, describing the system: strength of focusing m, position of focusing l and conditions for phase-matching of both steps $\Delta k_1 b$, $\Delta k_2 b$ was investigated. For maximum THG 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 on Figs. 1(a) and (b) calculated for three different strength of focusing. Fig. 1(a) illustrates the two-dimensional phase-matching curves when focusing is in the center of the nonlinear crystal, $l = 0$ while Fig. 1(b) is calculated for focusing position $l = 0.4$, close to the output face of the crystal. All amplitude distributions are normalized to their maximum.

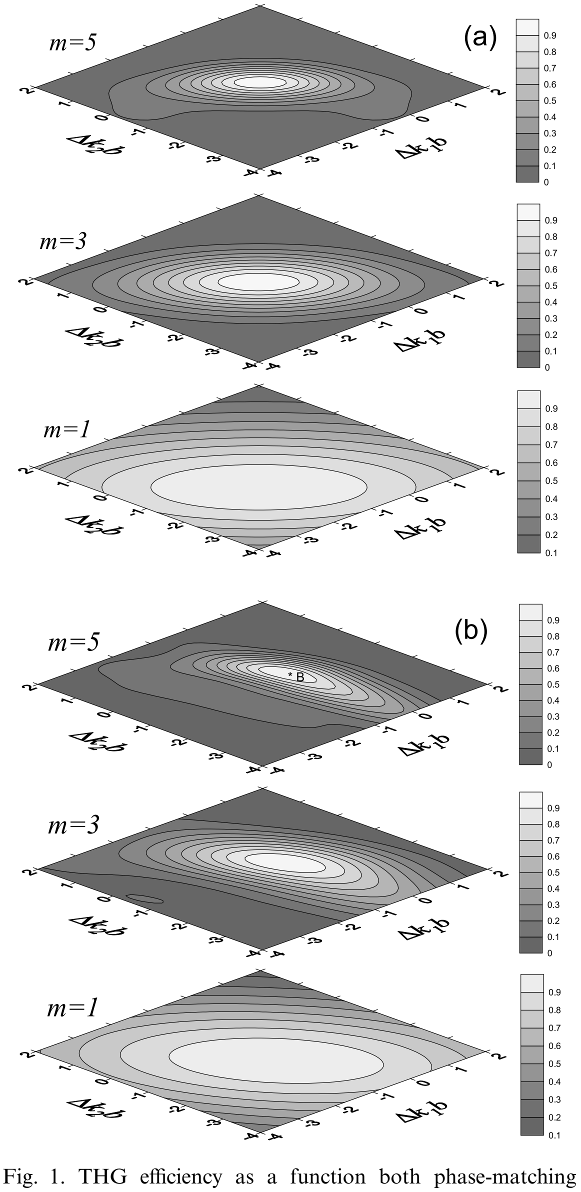

Fig. 1. THG efficiency as a function both phase-matching conditions ($\Delta k_1 b$) and ($\Delta k_2 b$) for two different positions of the focal spot (a) $l = 0$ and (b) $l = 0.4$. All amplitude distributions are normalized to their maximum. Normalized input amplitude $S = 0.3$.

Figs. 2(a) and (b) allow one to obtain the maximum $\eta_{3\omega}$ for arbitrary values of these four parameters. Fig. 2(a) shows the dependence of the efficiency $\eta_{3\omega}$ 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. 2(a) can be found from Fig. 2(b). For example, as seen from

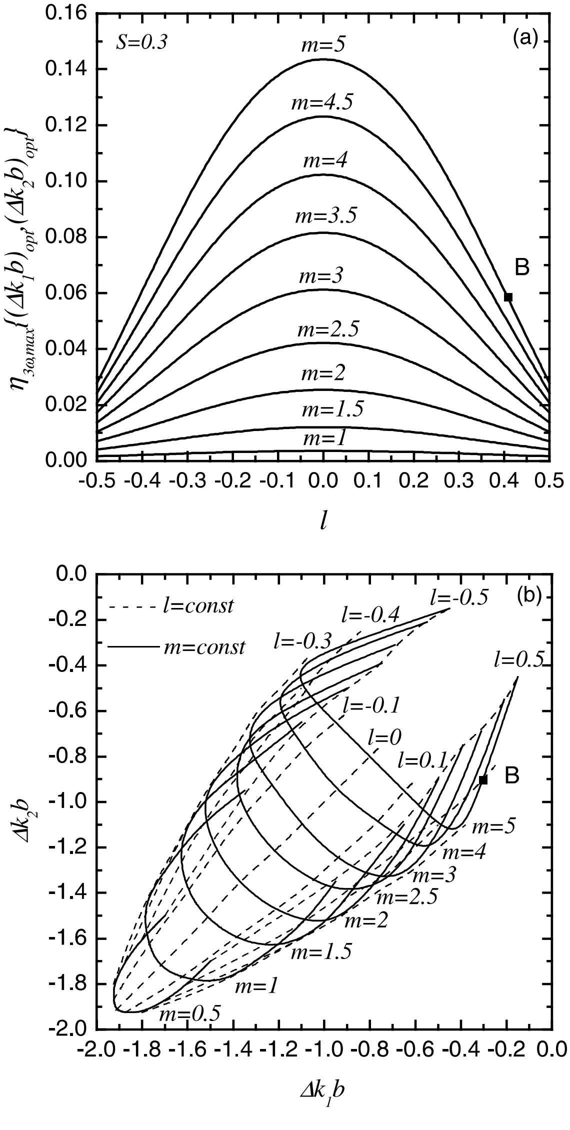

Fig. 2. (a) THG efficiency $\eta_{3\omega}$ calculated at optimal phase-matching conditions and (b) optimal phase-matching for both steps for several values of level of focusing m and the position of focusing l. Normalized input amplitude $S = 0.3$.

Fig. 2(a) point B ($m=5$, $l=0.4$, $S=0.3$ with efficiency $\eta_{3\omega}=6.1\%$) corresponds to the following optimal phase mismatches $(\Delta k_1 b)_{\mathrm{opt}}=-0.3$, $(\Delta k_2 b)_{\mathrm{opt}}=-0.9$ as can be found from Fig. 2(b). We found out that in process of cascaded third harmonic generation in single quadratic crystal with 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.

In contrast to the process of direct THG where there is an optimal strength of focusing [34], in the case of cascaded *double phase-matched* THG there is no optimal strength of focusing. As illustrated in Fig. 2(a) the higher the level of focusing is the higher the TH efficiency is. The optimal position of the focal spot is more critical for stronger focusing. When the optimal position of the focal spot is in the middle of the crystal ($l=0$) then $\Delta k_{1,\mathrm{opt}}=\Delta k_{2,\mathrm{opt}}=\Delta_{\mathrm{f}}(m)$ and this magnitude depends on the strength of the focusing. As illustrated in Fig. 2(b) for deviations of $(\Delta k_{1,2}-\Delta_{\mathrm{f}})$ the optimal focusing is in the first part of the crystal when $|\Delta k_1/\Delta k_2|>1$ and respectively in the second part of the crystal when $|\Delta k_1/\Delta k_2|<1$.

4. Account for depletion

For calculating the process of single crystal cascade THG without neglecting the depletion of the fundamental and second harmonic beam and evaluating the area of the applicability of the nondepleting approach used in the proceeding sections, we have solved system of Eq. (1) 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 Runge–Kutta method for the nonlinear effects.

The investigation, carried out in not very high input fundamental intensities, confirms the results in Section 3.2 – the optimal position of focusing is in the center of nonlinear medium with $(\Delta k_1 b)_{\mathrm{opt}}$ and $(\Delta k_2 b)_{\mathrm{opt}}$ and there is no optimal strength of focusing.

Fig. 3 calculated for $m=1$, $m=3$ and $m=5$ allows establishing maximum normalized intensity of the semi-analytical approach described in Section 3.2. We may conclude that this approach can be used for normalized fundamental beam amplitudes $S\leqslant 0.3$. Also it is seen when $S>0.3$ and the depletion effects are taken into account, the maximum conversion practically does not depend on the strengths of focusing. As we already note in [29–32] is proved in case of plane wave approximation that the maximum efficiency depends on the ratio of the two second order nonlinearities responsible for each of the steps of this double phase-matched cascaded interaction for THG and one has to use the optimal value $\sigma_1/\sigma_2=1.53$ for 100% efficiency. Our numerical calculations of THG with focused fundamental beam shown on Fig. 3 compare two different cases (i) nonoptimized ratio of nonlinearities for $\sigma_1=\sigma_2$ and (ii) optimized "plane wave" value $\sigma_1=1.53\sigma_2$. We see that the choice of the optimal "plane wave" value gives efficiencies close to 90%.

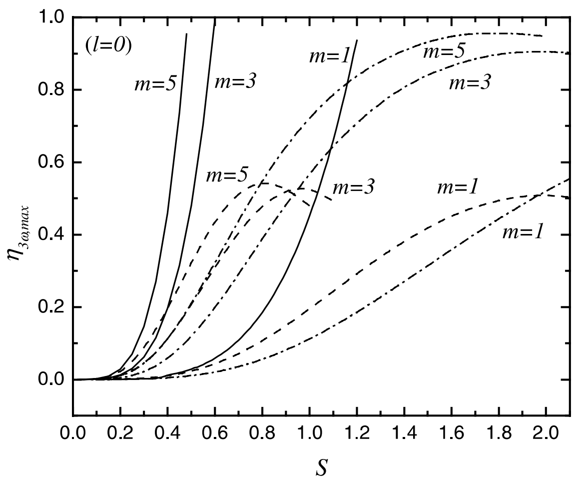

Fig. 3. THG efficiency as a function of normalized input amplitude. Solid line, calculations with the semi analytical approach given in Section 3.2. Dashed line, direct numerical integration of system (1) for the case $\sigma_1=\sigma_2$. Dot-dashed line, direct numerical integration of system (1) for the case $\sigma_1=1.53\sigma_2$.

5. Conclusion

In conclusion, we present both analytical and numerical investigation of the process of cascaded THG in single quadratic crystal in condition of simultaneous phase-matching of both steps and focused fundamental beam. If the design of the nonlinear media allow tuning the phase-matching conditions to its optimal values, then the optimal focusing is in the center of the crystal. If the phase-matching parameters are fixed and they deviate from the optimal values, then the optimal position of the focus spot should be calculated according the analysis presented here.

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