Broadband sum-frequency generation using cascaded processes via chirped quasi-phase-matching

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Broadband sum-frequency generation using cascaded processes via chirped quasi-phase-matching

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An efficient broadband sum frequency generation (SFG) technique using the two cascaded optical parametric processes $\omega_3 = \omega_1 + \omega_2$ and $\omega_4 = \omega_1 + \omega_3$ is proposed. The technique uses chirped quasi-phase-matched gratings, which, in the undepleted pump approximation, make SFG analogous to adiabatic population transfer in three-state systems with crossing energies in quantum physics. If the local modulation period first makes the phase match occur for ω_3 and then for ω_4 SFG processes then the energy is converted adiabatically to the ω_4 field. Efficient SFG of the ω_4 field is also possible by the opposite direction of the local modulation sweep; then transient SFG of the ω_3 field is strongly reduced. Most of these features remain valid in the nonlinear regime of depleted pump.

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I. INTRODUCTION

Recent advances in quasi-phase-matching (QPM) techniques [1, 2] have drawn analogies between optical parametric processes and two- and three-state quantum systems [3–5]. By using an analogy to stimulated Raman adiabatic passage (STIRAP) in atomic physics [6–9] Longhi proposed [3] a scheme in which the fundamental frequency field is directly converted into the third harmonic without a transient generation of the second harmonic. This proposal requires the continuous simultaneous phase matching of second harmonic generation ($\omega + \omega = 2\omega$) and sum frequency generation ($\omega + 2\omega = 3\omega$); because this condition can be fulfilled only for a specific frequency this technique is not broadband. Suchowski *et al.* [4, 5] used an aperiodically poled QPM crystal to achieve both high efficiency and large bandwidth in sum frequency generation (SFG) in the undepleted pump approximation using ideas from the technique of adiabatic passage via a level crossing in quantum physics [8, 10].

In this paper, we make use of the analogy between coherent population transfer in three-state quantum systems and the two simultaneous collinear second-order parametric processes $\omega_3 = \omega_1 + \omega_2$ and $\omega_4 = \omega_1 + \omega_3 = 2\omega_1 + \omega_2$ to design a potentially highly efficient broadband SFG technique. To this end, we use linearly chirped QPM gratings [11–13], which provide the analogy to level crossings in atomic systems [14–16].

In our proposal there is no requirement for continuous phase matching as in the STIRAP-like technique proposed by Longhi [3] but we require phase mismatch varying along the nonlinear crystal. This is far easier to achieve than continuous phase matching, as has been demonstrated already experimentally [4, 5]. We point out here that the broadband generation in Refs. [4, 5] is done in the undepleted pump approximation, i.e. in the linear regime, and for a single SFG. We show here that these ideas still work in the nonlinear regime of a depleted pump (delivering much higher absolute intensities). Moreover, we extend the idea of Refs. [4, 5] from a single SFG to simultaneous SFG processes; this is analogous to the extension of adiabatic passage from a two-state quantum system to a three-state one, with three level crossings and multiple evolution paths [8]. The present paper therefore extends the idea of Refs. [4, 5] for a single SFG linear process in the undepleted pump approximation to simultaneous multiple SFG linear and nonlinear processes in and beyond the undepleted pump approximation.

II. CASCADED SUM FREQUENCY GENERATION

The two simultaneous SFG processes $\omega_3 = \omega_1 + \omega_2$ and $\omega_4 = \omega_1 + \omega_3$, for a QPM crystal with susceptibility $\chi^{(2)}$ and local modulation period $\Lambda(z)$ are described by the set of nonlinear differential equations [1, 2]

$$i\partial_z E_1 = \Omega_1 \left(E_2^* E_3 e^{-i\Delta_1 z} + E_3^* E_4 e^{-i\Delta_2 z} \right), \tag{1a}$$

$$i\partial_z E_2 = \Omega_2 E_1^* E_3 e^{-i\Delta_1 z}, \tag{1b}$$

$$i\partial_z E_3 = \Omega_3 \left(E_1 E_2 e^{i\Delta_1 z} + E_1^* E_4 e^{-i\Delta_2 z} \right), \tag{1c}$$

$$i\partial_z E_4 = \Omega_4 E_1 E_3 e^{i\Delta_2 z}, \tag{1d}$$

where z is the position along the propagation axis, c is the speed of light in vacuum, and E_j is the electric field of the j-th laser beam. Here $\Omega_j = \chi^{(2)}\omega_j/4cn_j$ ($j = 1, 2, 3, 4$) are the coupling coefficients, while $\Delta_1 = n_1\omega_1/c + n_2\omega_2/c - n_3\omega_3/c + 2\pi/\Lambda$ and $\Delta_2 = n_1\omega_1/c + n_3\omega_3/c - n_4\omega_4/c + 2\pi/\Lambda$ are the phase mismatches for the ω_3 and ω_4 SFG processes, where ω_j and n_j are the frequency and the refractive index of the j-th laser beam.

A. Undepleted pump approximation

The coupled nonlinear equations (1) are often linearized assuming that the incident pump field E_1 is much stronger than the other fields and therefore its amplitude is nearly constant (undepleted) during the evolution. Then Eqs. (1) are reduced to a system of three linear equations,

$$i\partial_z \mathbf{A}(z) = \mathbf{M}(z)\mathbf{A}(z), \tag{2a}$$

$$\mathbf{M} = \begin{bmatrix} -\Delta_1 & \Omega_p^* & 0 \\ \Omega_p & 0 & \Omega_s^* \\ 0 & \Omega_s & \Delta_2 \end{bmatrix}, \tag{2b}$$

with $\mathbf{A}(z) = [A_2(z), A_3(z), A_4(z)]^T$, where

$$\Omega_p = E_1\sqrt{\Omega_2\Omega_3}, \tag{3a}$$

$$\Omega_s = E_1\sqrt{\Omega_3\Omega_4}, \tag{3b}$$

$$A_2 = E_1 E_2 e^{i\Delta_1 z}\sqrt{\Omega_3\Omega_4/2}, \tag{3c}$$

$$A_3 = E_1 E_3\sqrt{\Omega_2\Omega_4/2}, \tag{3d}$$

$$A_4 = E_1 E_4 e^{-i\Delta_2 z}\sqrt{\Omega_2\Omega_3/2}. \tag{3e}$$

Upon the substitution $z \to t$, Eq. (2) becomes identical to the time-dependent Schrödinger equation for a three-state quantum system in the rotating-wave approximation, which is studied in great detail [8]; the vector $\mathbf{A}(z)$ and the driving matrix $\mathbf{M}$ correspond to the quantum state vector and the Hamiltonian, respectively. We note that the quantity $|\mathbf{A}(z)|^2 = |A_2(z)|^2 + |A_3(z)|^2 + |A_4(z)|^2$ is conserved, like the total population in a coherently driven quantum system. By definition, in the adiabatic regime the system stays in an eigenvector of the "Hamiltonian" $\mathbf{M}$. We assume that $\Delta_1(z)$ and $\Delta_2(z)$ change linearly along z, which can be achieved, for example, by varying the local modulation period $\Lambda(z)$. Explicitly, we assume that either $\Delta_1 = \delta_1 - \alpha^2 z$ and $\Delta_2 = \delta_2 - \alpha^2 z$, which is called "intuitive sweep" (for reasons that will becomes clear shortly) or $\Delta_1 = \delta_1 + \alpha^2 z$ and $\Delta_2 = \delta_2 + \alpha^2 z$ which is called "counterintuitive sweep". For the sake of generality, we take hereafter α as the unit of coupling and $1/\alpha$ as the unit of length. Then the three eigenvalues of $\mathbf{M}$ will cross each other at three different distances z_m ($m = 1, 2, 3$), thereby creating a triangle crossing pattern [14–16]. These crossings allow us to design recipes for efficient broadband SFG, in analogy to adiabatic passage techniques in quantum physics [8–10, 14–16]. Because of the analogy to the Schrödinger equation the condition for adiabatic evolution can be derived using the Landau-Zener-Stückelberg-Majorana model [17–20] and reads (for linear chirping and constant couplings): $|\Omega_x| \gtrsim \alpha$, where Ω_x is the relevant coupling at the respective crossing.

Figure 1 plots the eigenvalues of $\mathbf{M}$ of Eq. (2) vs z. Initially only the ω_2 field is present, hence the vector $\mathbf{A} = [A_2, 0, 0]$. If the evolution is adiabatic then there are two possible paths that the system can follow (marked by arrows). If the phase match for the ω_3 generation process occurs first (left frames of Fig. 1), then the energy is converted first to the ω_3 field and then to the ω_4 field. This "intuitive" two-step scheme extends the single-step adiabatic passage scenario for SFG [4, 5]. Interestingly, we find that efficient energy transfer directly to the ω_4 field is also possible through the "counterintuitive" direction of the local modulation period sweep when the phase match for the ω_4 generation process occurs first (right frames of Fig. 1). Then the energy flows from the ω_2 field to the ω_4 field with almost no energy transferred to the intermediate ω_3 field.

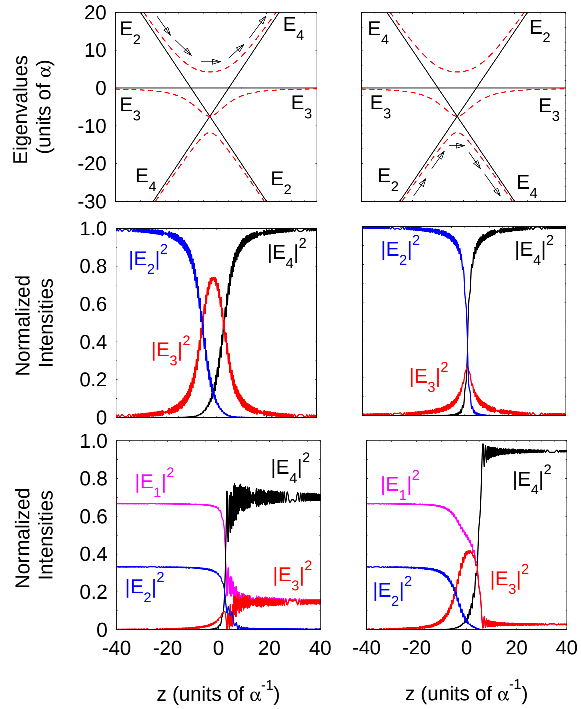

FIG. 1: (Color online) Sequential SFG of ω_4 field. Top frames: Diagonal elements (solid lines) and eigenvalues (dashed lines) of the driving matrix $\mathbf{M}$ of Eq. (2) for the "intuitive" (left frames) and "counterintuitive" (right frames) phase mismatch sweep. The field intensities are calculated numerically from Eqs. (1)for $\delta_1 = 10\alpha$, $\delta_2 = 5\alpha$, $\Omega_1 = \Omega_2 = \Omega_3 = \Omega_4 = 5\alpha$. Middle frames: undepleted pump, $|E_1(z_i)|^2 = 100|E_2(z_i)|^2$, with $z_i = -40\alpha^{-1}$; bottom frames: depleted pump, $|E_1(z_i)|^2 = 2|E_2(z_i)|^2$.

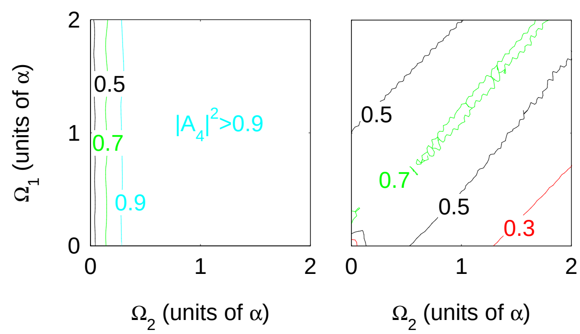

FIG. 2: (Color online) Efficiency of SFG of ω_4 field vs the couplings Ω_1 and Ω_2 obtained by numerical integration of Eqs. (1) for "counterintuitive sweep" with $\delta_1 = 10\alpha$, $\delta_2 = 5\alpha$ and $\Omega_3 = \Omega_4 = 5\alpha$. Left frame: undepleted pump, $|E_1(z_i)|^2 = 100|E_2(z_i)|^2$, with $z_i = -40\alpha^{-1}$; right frame: depleted pump, $|E_1(z_i)|^2 = 2|E_2(z_i)|^2$.

B. Depleted pump

We have found by numerical integration of the nonlinear system (1) that the described scheme is also applicable beyond the undepleted pump approximation, when the ω_1 and ω_2 fields have comparable energies; this is demonstrated in the bottom frames of Fig. 1. Unfortunately, many optical parametric processes such as $|\omega_1 - \omega_2|$, $2\omega_1$, $2\omega_2$, $\omega_1 + 2\omega_2$ become possible in this case and it is not easy to find the conditions for broadband SFG of the ω_4 field.

The contour plot in Fig. 2 demonstrates the robustness of SFG of the ω_4 field against parameter variations. SFG for an undepleted pump (left frame) is remarkably robust in confirmation of the simple analytic theory described above. SFG for a depleted pump (right) is less robust although relatively high SFG efficiency is still possible; because then the simple eigenvalue arguments cannot be used the interpretation is more difficult.

III. THIRD HARMONIC GENERATION

Third harmonic generation (THG) is an important special case of cascaded SFG, which is readily treated in the adiabatic regime. The respective equations are derived from Eqs. (1),

$$i\partial_z A_\omega = \Omega_\omega A_\omega^* A_{2\omega} e^{-i\Delta_1 z} + \Omega_\omega A_{2\omega}^* A_{3\omega} e^{-i\Delta_2 z}, \quad (4a)$$

$$i\partial_z A_{2\omega} = \Omega_{2\omega} A_\omega^2 e^{i\Delta_1 z} + \Omega_{2\omega} A_\omega^* A_{3\omega} e^{-i\Delta_2 z}, \quad (4b)$$

$$i\partial_z A_{3\omega} = \Omega_{3\omega} A_\omega A_{2\omega} e^{i\Delta_2 z}, \quad (4c)$$

where $(z_i = 0)$

$$\Omega_\omega = E_\omega(0)\,\chi^{(2)}\omega/4cn_\omega, \quad (5a)$$

$$\Omega_{2\omega} = E_\omega(0)\,\chi^{(2)}\omega/2cn_{2\omega}, \quad (5b)$$

$$\Omega_{3\omega} = 3E_\omega(0)\,\chi^{(2)}\omega/4cn_{3\omega}, \quad (5c)$$

$$A_\omega(z) = E_\omega(z)/E_\omega(0), \quad (5d)$$

$$A_{2\omega}(z) = E_{2\omega}(z)/E_\omega(0), \quad (5e)$$

$$A_{3\omega}(z) = E_{3\omega}(z)/E_\omega(0). \quad (5f)$$

Figure 3 shows simulations of THG obtained by numerical integration of Eqs. (4) for the nonlinear optical crystal Lithium Tantalate ($LiTaO_3$) [21]. We use typical wavelengths of Nd:YAG laser: 1050 nm (top frame), 1060 nm (middle frame) and 1070 nm (bottom frame). To maintain adiabatic conditions we choose the local poling period to vary slowly from 6.00 μm to 6.59 μm along a crystal length of 120 mm. Figure 3 shows that high THG efficiency is achieved for a relatively broad wavelength window. This window can be enlarged further if one increases the length of the crystal or simultaneously increases the final poling period and the intensity of the initial fundamental field.

As it is seen from Fig. 3, the second harmonic is absent during the evolution and nearly complete transfer of energy to the third harmonic takes place. Because there is no second harmonic then the problem of overlapping fundamental with the second harmonic is not present and the proposed THG technique can be used for nanosecond as well as picosecond pulses. Another advantage that is even more interesting is that the conversion to the third harmonic will remain efficient even when the crystal is not transparent for the second harmonic frequency, in agreement with recent experimental results [22].

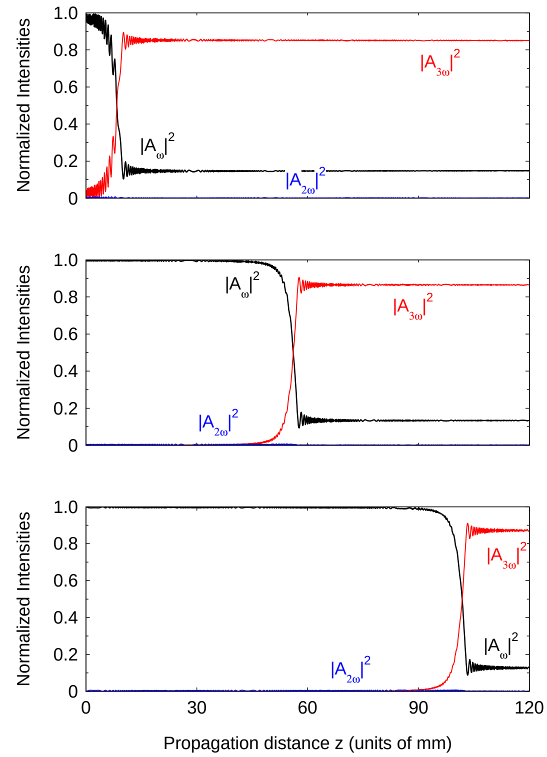

FIG. 3: (Color online) Fundamental, second-harmonic and third-harmonic fields vs. propagation distance z for input intensity of 25 GW/cm^2. Equations (4) are solved numerically for $LiTaO_3$ crystal with local poling period that vary slowly from 6.00 μm to 6.59 μm for three different wavelengths: (Top frame) 1050nm; (Middle frame) 1060nm; (Bottom frame) 1070nm.

Figure 4 demonstrates the efficiency of THG as a function of the input intensity of the fundamental. A typical adiabatic increase [8, 9] of the efficiency toward unity is observed as the initial fundamental input intensity increases.

IV. CONCLUSION

We have used the analogy between the time-dependent Schrödinger equation and the SFG equations in the undepleted pump approximation to propose an efficient broadband SFG technique. A local modulation period sweep along the light propagation direction creates crossings in the phase matching between different parametric pro-

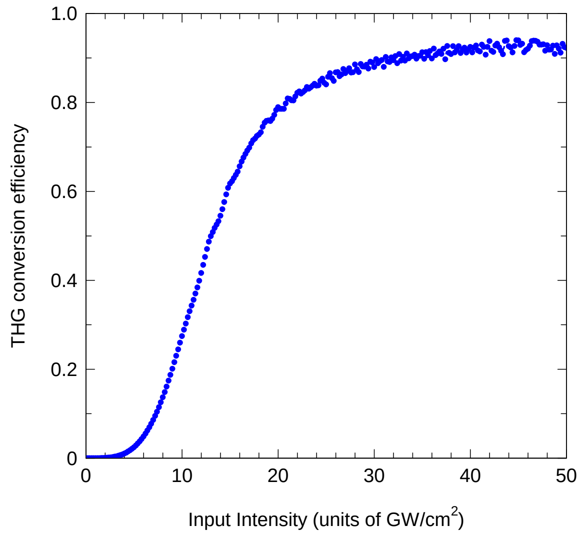

FIG. 4: (Color online) Efficiency of third harmonic generation for 1060nm wavelength vs the input intensity of the fundamental. The other parameters are as in Fig. 3.

cesses, which in combination with adiabatic evolution conditions allow efficient and robust SFG of the desired frequency $\omega_4 = 2\omega_1 + \omega_2$. While the physical picture is transparent in the undepleted pump approximation, the basic feature of the SFG process remain largely intact in the general regime of depleted pump. Chirped QPM gratings offer robustness against variations of the parameters of both the crystal and the electric fields, which include the crystal temperature, the wavelengths of the input electric fields, the crystal length and the angle of incidence.

The present work can be viewed as a generalization of the idea of Suchowski *et al.* [4] from a single SFG to simultaneous SFG processes in and beyond the undepleted pump approximation. This work is also a broadband alternative to the (narrowband) STIRAP-based third harmonic generation proposal of Longhi [3].

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