Two photon frequency conversion

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Abstract: We study the case of two simultaneous three-wave-mixing processes, where one frequency is converted to another through an intermediate frequency. The common assumption is that these processes can occur only when the material is transparent at all participating frequencies. Here we show experimentally that, under appropriate conditions, the intermediate frequency remains dark throughout the interaction. This means that even if the material is opaque at the intermediate frequency, the conversion will remain efficient. New possibilities of frequency conversion are therefore available, e.g. through absorptive bands in the ultraviolet or mid-infrared. Moreover, though it was hitherto assumed that the phase mismatch value is governed only by dispersion, we show here that phase matching also depends on light intensity. These findings promise novel all optical switching techniques.

OCIS codes: (190.4223) Nonlinear wave mixing; (190.4410) Nonlinear optics, parametric processes.

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1. Introduction

In the field of nonlinear optics, available lasers are mixed to produce new frequencies, using three-wave-mixing processes in quadratic nonlinear optical crystals. For example, in sum frequency generation (SFG), two frequencies ω_1 and ω_{p_1} are summed to produce a higher frequency $\omega_2 = \omega_1 + \omega_{p_1}$. In order for this process to have significant efficiency, momentum conservation needs to be maintained. For the given example, this would require that $\vec{k}_2 = \vec{k}_1 + \vec{k}_{p_1}$ where $\vec{k}_i$ is the momentum (wave-vector) of the wave with frequency ω_i, a requirement also known as phase-matching. Three-wave-mixing processes can also be cascaded [1], e.g. the generated sum-frequency can be further combined with another frequency in another SFG process, which produces $\omega_3 = \omega_2 + \omega_{p_2} = \omega_1 + \omega_{p_1} + \omega_{p_2}$.

Recently, a wideband and robust frequency conversion method was developed, by using an analogy between the dynamics of sum or difference frequency generation (DFG) process and a two state quantum system induced by electromagnetic (EM) field [2, 3].

Here, a method to perform frequency conversion via simultaneous three-wave-mixing (STWM) is developed and demonstrated experimentally. Most notably, phase-matching is found to be dependent on light intensity. This is in contrast to the hitherto accepted view that phase-matching depends only on the dispersion properties of the nonlinear medium [4]. Moreover, the intermediate frequency remains dark, hence conversion can be preformed through frequencies at which the nonlinear medium is opaque. The method presented here relies on an analogy drawn between an EM-excited three state quantum system and *two* STWM processes. This analogy leads to mapping of atomic phenomena such as dark states and Stark shifts to the field of nonlinear optics.

2. Theoretical analysis

2.1. Dynamical equations and atomic analogy

We consider two simultaneous three wave mixing processes (STWM), where the generated frequency is further combined with another frequency, ω_{p_2}, in an additional process, generating a new frequency, ω_3. For the case of two SFG processes, we obtain $\omega_3 = \omega_2 + \omega_{p_2} = \omega_1 + \omega_{p_1} + \omega_{p_2}$. Generally, two distinct pumps can drive the interaction: the first transfers energy between ω_1 and ω_2, while the second connects ω_2 with ω_3. We consider the case where the pumps are much more intense than the other waves and thereby are negligibly affected by the interaction (undepleted pumps approximation). The procedure of adiabatic elimination [5, 6] is used to reduce the three wave system into an effective two coupled waves system. Notably, a work conducted in parallel to ours applied this procedure to the case termed 'direct third harmonic generation' [7]. Notwithstanding, here we analyze STWM by analogy to a three level quantum system, following the approach of Suchowski et al. [2, 3].

Let us start by writing the coupled equations that govern the evolution of the two STWM processes along a nonlinear crystal:

$$\frac{d}{dz}\begin{bmatrix} A_1 \\ A_2 \\ A_3 \end{bmatrix} = i\begin{bmatrix} 0 & \sigma_{12}e^{-i\Delta k_1 z} & 0 \\ \sigma_{21}e^{i\Delta k_1 z} & 0 & \sigma_{23}e^{-i\Delta k_2 z} \\ 0 & \sigma_{32}e^{i\Delta k_2 z} & 0 \end{bmatrix}\begin{bmatrix} A_1 \\ A_2 \\ A_3 \end{bmatrix} \tag{1}$$

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Here $A_j(z)$ is the complex amplitude of the wave with frequency ω_j, $j = 1,2,3$. $\sigma_{12} = \left(\chi^{(2)}\omega_1^2/k_1c^2\right)A_{p_1}^*$ and $\sigma_{23} = \left(\chi^{(2)}\omega_2^2/k_2c^2\right)A_{p_2}$, where $\sigma_{ij} = \frac{\omega_i^2}{\omega_j^2}\frac{k_j}{k_i}\sigma_{ji}^*$, are the effective coupling coefficients between the fields. A_{p_1} and A_{p_2} are the complex amplitude of each pump, $\chi^{(2)}$is the second-order nonlinear coefficient of the material (considered to be frequency independent) and c is the velocity of light. $\Delta k_1 = k_1 + k_{p_1} - k_2$ and $\Delta k_2 = k_2 + k_{p_2} - k_3$ are the phase mismatches of the two nonlinear processes, where each wave vector is defined as $k_i = n_i(\omega_i)\omega_i/c$. The undepleted pumps approximation $dA_{p_1}/dz = 0$ and $dA_{p_2}/dz = 0$ is implicit in these equations. These dynamical equations could describe four possible frequency conversion processes: two DFG processes, two SFGs, SFG followed by DFG and DFG followed by SFG. The difference between these processes is hidden in the direction of the phase mismatch parameters, Δk_1 and Δk_2, where in our notation, positive is in the +z direction.

The same set of equations describes the dynamics of a three level atom interacting with two EM fields [5, 8, 9]. In the context of atomic physics, the complex amplitude of each field $A_i(z)$ corresponds to $a_i(t)$, the probability amplitude of population in each state. Each effective coupling coefficient σ_{ij} represents the strength of the dipole interaction between levels i and j, usually described by the Rabi frequency $\Omega_{ij} = d_{ij}\cdot\varepsilon(t)/\hbar$, where d_{ij} is the dipole moment between states i and j, $\varepsilon(t)$ is the induced EM field, and $\hbar$ is Planck's constant. Respectively, the terms Δk_1 and Δk_2 correspond to the detunings, Δ_1 and Δ_2, with the two atomic transitions.

We note that, for the optical case, the notation used here assumes quasi-monochromatic laser beams, which in the visible and near infrared means pulses longer than about 1ps [2]. However, since the interaction is coherent, femtosecond pulses can also be used. Transform limited femtosecond pulses can be stretched before entering the crystal, and the generated frequency pulse can be compressed to a transform limited duration [10].

2.2. Adiabatic elimination

Utilizing the adiabatic elimination procedure is done by assuming that each of the SFG or DFG processes exhibits a very large phase mismatch, but their sum is rather small, i.e. $|\Delta k_1|L \gg 1$, $|\Delta k_2|L \gg 1$ while $|\Delta k_1 + \Delta k_2|L \ll 1$, where L is the length of the nonlinear crystal. The sum of the phase-mismatches, which is a central notion here, will be defined as the *two-photon phase mismatch*, and denoted $\Delta k_{TP} = \Delta k_1 + \Delta k_2$. Under these conditions, it is reasonable to assume that the oscillating terms in Eq. (1) vary much faster than the fields amplitudes at ω_1 and ω_3, allowing us to write an approximation for the intermediate frequency complex amplitude:

$$\begin{aligned} A_2 &= \int [i\sigma_{21}A_1 exp(i\Delta k_1 z) + i\sigma_{23}A_3 exp(-i\Delta k_2 z)]\,dz \approx \\ &\approx \tfrac{\sigma_{21}}{\Delta k_1} exp(i\Delta k_1 z)A_1 - \tfrac{\sigma_{23}}{\Delta k_2} exp(-i\Delta k_2 z)A_3 \end{aligned} \tag{2}$$

As a consequence of this approximation, the amplitude of the intermediate frequency, $|A_2|$, will remain always low along the propagation in the nonlinear crystal.

By substituting the outcome of Eq. (2) into Eq. (1) and moving to the rotating frame using $A_1 = \tilde{A}_1(z)\,exp\left[-i\left(\Delta k_{eff}/2 - \sigma_{12}\sigma_{21}/\Delta k_1\right)z\right]$ and $A_3 = \tilde{A}_3(z)\,exp\left[i\left(\Delta k_{eff}/2 - \sigma_{23}\sigma_{32}/\Delta k_2\right)z\right]$, a reduced two coupled wave equations set for ω_1 and ω_3 is obtained,

$$\frac{d}{dz}\begin{bmatrix}\tilde{A}_1\\ \tilde{A}_3\end{bmatrix} = i\begin{bmatrix}\Delta k_{eff}/2 & -\sigma_{12}\sigma_{23}/\Delta k_2\\ \sigma_{21}\sigma_{32}/\Delta k_1 & -\Delta k_{eff}/2\end{bmatrix}\begin{bmatrix}\tilde{A}_1\\ \tilde{A}_3\end{bmatrix} \tag{3}$$

where Δk_{eff} is the phase-mismatch of the effective process connecting ω_1 and ω_3, defined as

$$\Delta k_{eff} = \Delta k_1 + \Delta k_2 + \sigma_{12}\sigma_{21}/\Delta k_1 + \sigma_{23}\sigma_{32}/\Delta k_2 \tag{4}$$

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The physical interpretation of Eqs. (2) and (3) is as follows: since each of the two STWM processes is greatly phase-mismatched, the amplitude of the intermediate frequency, $|A_2|$, will oscillate very rapidly in comparison to variation in the amplitudes of ω_1 and ω_3. As a result, $|A_2|$ will never build up to a significant value and thus remain low throughout the interaction. Nevertheless, energy can still be transferred between ω_1 and ω_3 via ω_2, effectively imitating a four-wave-mixing process with undepleted pumps, where $\omega_3 = \omega_1 + \omega_{p_1} + \omega_{p_2}$ in the case of two simultaneous SFG processes. We emphasize that the efficiency of conversion from ω_1 to ω_3 does not change even if the material is opaque at ω_2.

The rate of energy transfer and the amount of energy participating in this process are determined by the effective phase-mismatch Δk_{eff}. In this mechanism, two additional contributions to the conventional phase mismatch parameters appear. These contributions, which depend on the coupling coefficients (and thus on the intensities of the pumps, I_{p_1} and I_{p_2}), are the equivalent of the atomic Stark shift [5]. In the nonlinear optics context, we choose to define it as $\delta k_S = \sigma_{12}\sigma_{21}/\Delta k_1 + \sigma_{23}\sigma_{32}/\Delta k_2$.

This term is critical where significant energy transfer between ω_1 and ω_3 is desired, which occurs when the effective phase-matching parameter is equal to zero, i.e. when $\Delta k_{eff} = \Delta k_1 + \Delta k_2 + \delta k_S = 0$. Since $\delta k_S \propto I_p$ it introduces a pump intensity dependence into the phase-matching condition. This is contrary to the conventional wave-mixing phase mismatch, which is considered to depend solely on the dispersion properties of the nonlinear material. Such dependence exists since the rate of each of the two STWM processes depends on pump intensity. To obtain a net flow of energy from one frequency to another, these rates must be compatible over a significant interaction length. The mechanism here is thus fundamentally different from phase-modulation in four-wave-mixing processes, which stems from intensity dependent refractive index.

The analytical solution of Eq. (3), for the initial condition $A_3(0) = 0$, is

$$\tilde{A}_3(z) = (K_3/\kappa)\tilde{A}_1(0)\,sin(\kappa z)\,exp\left(i\Delta k_{eff} z/2\right) \tag{5}$$

where $\kappa = \sqrt{-K_1K_3 + \Delta k_{eff}^2/4}$ is the effective Rabi frequency, $K_1 = -i\sigma_{12}\sigma_{32}/\Delta k_2$ and $K_3 = i\sigma_{21}\sigma_{32}/\Delta k_1$. It is interesting to note that having reduced the system to an effective two-states system, it can now be represented geometrically using the method introduced by Bloch [11] and Feynman et al. [12].

The effective phase-matching condition can be satisfied using the quasi-phase-matching (QPM) technique [13]. For a periodically poled crystal with period Λ, the phase-mismatch of each of the two STWM processes should be modified with the addition of the periodicity pattern, i.e. in the first order Fourier approximation, $\Delta\tilde{k}_1 = \Delta k_1 + 2\pi/\Lambda$ and $\Delta\tilde{k}_2 = \Delta k_2 + 2\pi/\Lambda$. Substituting these expressions into the phase-matching condition produces a cubic equation with one solution that adheres to the conditions of adiabatic elimination.

3. Numerical simulation

A QPM solution as described above was used to numerically simulate the interaction. A single pump at $\lambda_{p_1} = \lambda_{p_2} = 1064nm$ was used to drive both processes. We have used a periodically poled $KTiOPO_4$ (PPKTP) crystal with a poling period of $\Lambda = 8.6\mu m$, which was calculated to phase-match conversion from $\lambda_1 = 3010nm$ to $\lambda_3 = 452nm$, via the two implicit SFG processes $3010nm + 1064nm \rightarrow 786nm$ and $786nm + 1064nm \rightarrow 452nm$, when the crystal temperature is $125^{\circ}C$. The simulation does not assume undepleted pump or adiabatic elimination.

The resulting evolution of the intensities of the interacting waves, with pump intensity of $I_p = 10GW/cm^2$, is plotted in Fig. 1(a). Energy oscillations between the $\lambda_1 = 3010nm$ input and the $\lambda_3 = 452nm$ output are clearly seen. Since each photon converted from λ_1 to λ_3 is paired

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with two pump photons, the peak output intensity at λ_3 is higher than that at λ_1. The inset shows the fast oscillations of the low intensity at the adiabatically eliminated intermediate wavelength $\lambda_2 = 786nm$, which are over 3 orders of magnitude lower than the peak output intensity.

Using the same parameters, the Stark effect in frequency conversion was analyzed. In Fig. 1(b) the photon conversion efficiency from λ_1 to λ_3 is calculated for two cases: low pump intensity of $1GW/cm^2$ and high pump intensity of $12.4GW/cm^2$. The results are plotted as a function of the two-photon phase-mismatch $\Delta k_{TP} = \Delta k_1 + \Delta k_2$. For low pump intensity the Stark shift is negligible, so the peak efficiency is obtained near the two-photon resonance condition $\Delta k_{TP} = 0$. However, when the pump intensity is high, the Stark shift can no longer be neglected and the efficiency peak shifts by δk_S to $\Delta k_{TP} = -\delta k_S$, which is the exact shift amount predicted analytically using Eq. (4).

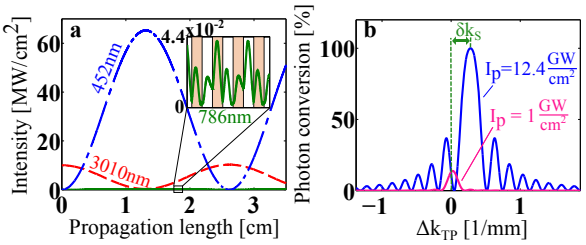

Fig. 1. (a) Numerically simulated intensities of the 3010nm input wave (red dashed line), 452nm output wave (blue dash-dotted line) and 786nm intermediate wave (green solid line). Introducing a $10cm^{-1}$ absorption at 786nm decreases the 452nm intensity by just 0.5%. Inset: close-up view of the intermediate wave over $34.4\mu m$. Shading indicates poling. (b) Analytically calculated photon conversion efficiency vs. the two-photon phase-mismatch. The difference between the peak efficiencies phase-mismatches is the Stark shift.

4. Experiment

Following the numerical simulation, STWM with adiabatic elimination had been experimentally realized. The experimental system is depicted in Fig. 2. A 4.4nsec Nd:YAG laser beam with $\lambda_p = 1064nm$, which served as the strong pumps, was passed through a temperature controlled PPKTP crystal with a $\Lambda = 8.6\mu m$ period. A laser with $\lambda_1 = 3010nm$ and an estimated $10nm$ FWHM bandwidth, which was produced from the same pump through an optical parametric oscillator, served as our input. The emerging power at the $\lambda_3 = 452nm$ output and the intermediate wavelength $\lambda_2 = 786nm$ were independently measured, using power detectors or a spectrometer, and peaks of both wavelengths were observed, corresponding to the expected output and intermediate wavelengths. When either the pump or the input were prevented from entering the crystal, both peaks would vanish.

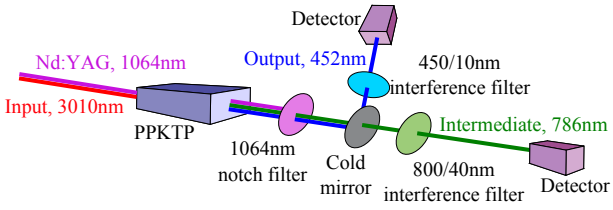

Fig. 2. Experimental apparatus.

Next, we have chosen to examine the conversion efficiency of the process as a function of

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the two-photon phase mismatch. This was done by tuning the crystal temperature from 90^oC to 175^oC, affecting the refractive index [14], and thus the phase-mismatch parameter. The input and pump intensities were kept constant at $71kW/cm^2$ and $44.6MW/cm^2$, respectively. Figure 3(a) shows the experimentally measured 452*nm* output power vs. the PPKTP crystal temperature, after decrementing the background noise, and accounting for the transmission and reflection of the optical components in the apparatus. The detected noise, defined as the power measured when only the pump beam was incident on the PPKTP crystal, was found to be around 152nW and thus negligible compared to the measured output power which reaches $38\mu W$. As seen, optimum two-photon phase matching was obtained at crystal temperature of 133^oC. The small deviation from the expected value of 125^oC can be accounted for by the inaccuracy in the Sellemeier equations that were used to calculate the wavelength and temperature dependence of the crystal's refractive index [14, 15]. The power at λ_2 was below the noise level and therefore could not be measured. Its upper bound was found to be 51.8nW, which is 733 times lower than the peak 452*nm* output power, agreeing with the theoretical prediction of adiabatic elimination.

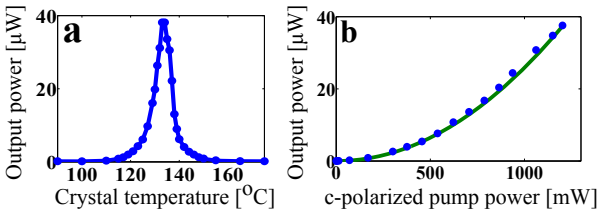

Fig. 3. Blue dots are experimentally measured 452*nm* output power vs. (a) crystal temperature (b) c-polarized pump power. The green line was analytically calculated by Eq. (5).

Finally, the predicted dependence of the 452*nm* output power on pump power was experimentally checked. A half wave plate was placed in the path of the pump beam before it entered the PPKTP crystal, and gradually rotated through 45 degrees. Due to the fact that $KTiOPO_4$ is a biaxial birefringent crystal, its phase-matching condition is polarization-dependent: only the pump power polarized along the PPKTP c axis contributes to the STWM processes. Figure 3(b) shows the experimentally measured 452*nm* output power obtained with various values of c-polarized pump power. In this experiment the noise (defined above) was measured at each wave plate angle and decremented from the detected power. The experimental measurements fit well on the analytically calculated output power (using Eq. (5)), showing a quadratic nature, where the nonlinear coefficient $\chi^{(2)}$ was taken to be 9% higher than the value reported for other processes and wavelengths [16]. This is consistent with the low conversion efficiency limit of Eq. (5), $\left|\tilde{A}_3(z)\right|^2 \approx \left|K_3\tilde{A}_1(0)z\right|^2 \propto I_p^2$. This experimental observation is a clear indication that STWM is in action, since a single TWM process would result in a linear rather than quadratic dependence, while direct third harmonic generation would yield a cubic dependence.

5. Conclusion

We have experimentally demonstrated frequency conversion through an intermediate frequency which remains dark throughout the interaction, enabling conversion through frequencies at which the nonlinear medium is absorptive. Additionally, phase-matching was shown to depend not only on dispersion, but also on pump intensity. This phenomenon enables new methods of all optical switching. We presented an analogy with EM-excited three state quantum systems which lead to these new effects. The same mathematical treatment can also be applied to cases involving three or more STWM processes, analogous to multi-state quantum systems [17].

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Other atomic physics schemes, involving temporal variation of the induced EM fields [5, 18], can be transferred to frequency conversion using aperiodically poled structures.

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