Two Photon Frequency Conversion

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Abstract: We experimentally demonstrate mid-IR to visible frequency conversion via simultaneous two photon excitation, while the intermediate frequency remains dark throughout the process. The phase mismatch in such mechanism is shown to be intensity dependent.

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We study two simultaneous three wave mixing (STWM) processes, where one frequency is converted to another through an intermediate frequency. We demonstrate experimentally that, under appropriate conditions, the intermediate frequency remains dark throughout the interaction. Efficient conversion can therefore be obtained despite significant absorption of the intermediate frequency, enabling conversion through absorptive bands in the ultraviolet or mid-infrared. Furthermore, we show that phase matching depends on light intensity, in addition to its well known dependence on material dispersion. These phenomena open new possibilities of all optical switching techniques.

In the general case of STWM studied here, each process can be either a sum or difference frequency generation (SFG/DFG) process driven by a different pump. For the case of two SFGs we obtain \( \omega + \omega_{p1} = \omega_2 \) and \( \omega + \omega_{p2} = \omega_3 \), where \( \omega_{p1} \) and \( \omega_{p2} \) are the pumps frequencies. The phase mismatches of the first and second process are \( \Delta k_1 = k_1 + k_{p1} - k_3 \) and \( \Delta k_2 = k_2 + k_{p2} - k_3 \), whereas the sum of the phase mismatches, \( \Delta k_{TP} = \Delta k_1 + \Delta k_2 \), is the two-photon phase mismatch parameter. We assume undepleted pumps, i.e. light at frequencies \( \omega_{p1} \) and \( \omega_{p2} \) is negligibly affected by the interaction, leaving three interacting frequencies.

The dynamics of this interaction is analogous to the dynamics of a three level atom excited by two electromagnetic (EM) fields [1]: the interacting fields amplitudes, pumps and phase mismatches correspond to the population probability amplitudes of the atomic levels, exciting EM fields and the detuning between the atomic resonances and the EM fields frequencies, respectively.

Inspired by this analogy, we use the procedure of adiabatic elimination, previously applied in the atomic context (and, in parallel to our work, for direct third harmonic generation [3]), in the case of STWM. This is done by assuming that the phase mismatch of each of the two processes is very large, while their sum is kept small. Under these conditions, the amplitude at the intermediate frequency will oscillate much faster than those at the other two interacting frequencies. Consequently, it will never build up to a significant value, and will remain always low. It is thus said to be adiabatically eliminated. However, significant energy transfer between the two remaining interacting waves is still possible. This would require that the generation rate of the intermediate wave, by the first process, is matched to the rate of its consumption, by the second process. In this manner, a net exchange of energy between \( \omega_1 \) and \( \omega_3 \) is obtained, as in a four wave mixing process where \( \omega_1 = \omega_2 + \omega_{p1} + \omega_{p2} \) for the case of two SFG processes. We emphasize that the conversion efficiency will not change even if the material is opaque at the intermediate frequency.

In the analysis, we found that the effective phase mismatch in such process has two contributions - the first is the two photon phase mismatch (defined above), and the second, which corresponds to the atomic Stark effect [2], is \( \delta k_S = \sigma_1 I_{p1}/\Delta k_1 + \sigma_2 I_{p2}/\Delta k_2 \), \( I_{p1} \) and \( I_{p2} \) are the pumps intensities, \( \sigma_1 \) and \( \sigma_2 \) are coupling coefficients that depend on the interacting frequencies and the material’s nonlinear coefficient. The overall phase mismatch is thus

\[
\Delta k_{eff} = \Delta k_{TP} + \delta k_S(I_{p1}, I_{p2})
\]  

(1)

Since the effective phase matching condition is \( \Delta k_{eff} = 0 \), it depends on pump intensity. This is in contrast to the generally accepted view that phase matching depends only on material dispersion. The reason for this dependence is that the rate of each of the two STWM processes depends on its pump intensity, and we require these two rates to match, as explained above.

The effective phase-matching condition can be satisfied using the quasi-phase-matching (QPM) technique. We performed a numerical simulation where a single pump at \( \lambda_{p1} = \lambda_{p2} = 1064nm \) with intensity of 10GW/cm² was used to
drive two SFG processes in a QPM KT$_2$OPO$_4$ crystal. The results are displayed in fig. 1a, showing energy oscillations between the $\lambda_1 = 3010\text{nm}$ input and the $\lambda_3 = 452\text{nm}$ output. The peak output intensity at $\lambda_3$ is higher than that at $\lambda_1$ due to contribution from the pump. The inset shows the fast oscillations of the low intensity at the adiabatically eliminated intermediate wavelength $\lambda_2 = 786\text{nm}$, which are 3 orders of magnitude lower than the peak output intensity.

Fig. 1b illustrates the Stark effect in frequency conversion. It shows analytically calculated photon conversion efficiency as a function of the two-photon phase-mismatch, using the same parameters as the numerical simulation. For low pump intensity of $1\text{GW/cm}^2$ the Stark effect is negligible, so the maximum efficiency is obtained near $\Delta k_{TP} = 0$. However, for high pump intensity of $12.4\text{GW/cm}^2$ the Stark effect is significant, and the efficiency peak shifts by $\delta k_S$ to $\Delta k_{TP} = -\delta k_S$.

![Graphs showing energy oscillations and Stark effect](image)

Fig. 1. (a) Numerically simulated intensities of the interacting waves. Introducing a $10\text{cm}^{-1}$ absorption at 786nm decreases the 452nm intensity by just 0.5%. Inset: close-up view of the intermediate wave. 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.

We proceeded to perform STWM with adiabatic elimination experimentally. We observed the theoretically predicted effective phase matching by examining the conversion efficiency as a function of crystal temperature, as depicted in figure 2a. The power at the intermediate wavelength $\lambda_2 = 786\text{nm}$ 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 452nm output power, verifying the theoretical prediction of adiabatic elimination. Fig. 2b displays the experimentally measured 452nm output power obtained with various values of pump power. An excellent correspondence with theory is obtained.

![Graphs showing experimental results](image)

Fig. 2. Experimental results. Blue dots are experimentally measured 452nm output power vs. (a) crystal temperature (b) pump power. The green line was calculated analytically.

To conclude, frequency conversion through a dark intermediate frequency was demonstrated experimentally, and was shown to have light intensity dependent phase matching. These new effects enable conversion through frequencies at which the nonlinear medium is absorptive, as well as new methods of all optical switching.

References