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Origin and suppression of back conversion in a phase-matched nonlinear frequency down-conversion process

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Back conversion is an intrinsic phenomenon in nonlinear frequency down-conversion processes. However, the physical reason for its occurrence is not well understood. Here, we theoretically reveal that back conversion is the result of a π-phase jump associated with the depletion of one interacting wave. By suppressing the idler phase jump through a deliberate crystal absorption, the back conversion can be inhibited, thus enhancing the conversion efficiency from the pump to the signal. The results presented in this Letter will further the understanding of nonlinear parametric processes and pave the way toward the design of highly efficient down-conversion systems.

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Optical quadratic nonlinear processes, such as sum-frequency generation (SFG), difference-frequency generation (DFG), and optical parametric amplification (OPA), provide a powerful and flexible tool for generating coherent light in the spectral regions inaccessible to laser emissions. One important merit of such quadratic three-wave nonlinear interactions lies in their outstanding designability. On the one hand, various phase-matching schemes can be applied, including quasi-phase matching (QPM), adiabatic phase matching, and multistep cascade phase matching. On the other hand, nonlinear crystals can also be fabricated with distinct structures, including periodically poled grating patterns and sectional tiling, and can be doped with rare-earth ions.

One inherent characteristic of quadratic nonlinear processes is that they allow both forward and backward energy transfers among the interacting waves in the crystal. Such back conversion will be detrimental when a unidirectional energy transfer is desired. It is well known that phase mismatch among the interacting waves can cause back conversion, which can be prevented using QPM or sectional tiling. Under the perfect phase-matching conditions, however, back conversion can still occur when one interacting wave is depleted. This back conversion will set an ultimate limit on the application performance of a quadratic nonlinear process. As of now, the physical reason for the occurrence of this back conversion is still unclear, and there is still no effective method to inhibit it.

One can usually answer the most basic and important questions about optics through the phase of a wave. A phase jump of π widely exists in linear optics, such as the half-wave loss associated with an interface reflection and the Gouy phase-shift associated with beam focusing. In this Letter, we numerically find an analog of this phase jump in nonlinear optics, which occurs when one wave is completely depleted in the process of three-wave interactions. This nonlinear phase jump is indeed the origin of back conversion in a phase-matched DFG process. From this new perspective, we can explore a method to inhibit such a back conversion, which has long been regarded as impossible. For a parametric down-conversion process, such as DFG or OPA, the back conversion can be inhibited by removing the phase jump of the idler wave through deliberate crystal absorption. Our studies would be beneficial for a deep understanding of optical quadratic nonlinear processes and pave the way toward the design of highly efficient DFG systems.

We start with the standard nonlinear coupled-wave equations that govern three-wave interactions (represented by $W_1$, $W_2$, and $W_3$), with the angular frequencies $\omega_1 < \omega_2 < \omega_3$, $\omega_1 + \omega_2 = \omega_3$,

\[
d\frac{A_1}{dz} = -i \frac{\omega_1 d_{\text{eff}}}{n_1 c} A_2 A_3 e^{-i\Delta k z},
\]

\[
d\frac{A_2}{dz} = -i \frac{\omega_2 d_{\text{eff}}}{n_2 c} A_1^* A_3 e^{-i\Delta k z},
\]

\[
d\frac{A_3}{dz} = -i \frac{\omega_3 d_{\text{eff}}}{n_3 c} A_1 A_2 e^{i\Delta k z},
\]

where $A_j(z) = \rho_j(z) \exp[i\varphi_j(z)]$ is the electric field envelope, and $\rho_j$ and $\varphi_j$ refer to the amplitude and phase, respectively. $n_j$ is the refractive index of the crystal, $d_{\text{eff}}$ is the effective nonlinear coefficient, and $c$ is the light speed in a vacuum. $\Delta k = k_3 - k_2 - k_1$ is the wave-vector mismatch among the three interacting waves. After defining a total phase $\theta = \Delta k z - \varphi_2(z) + \varphi_3(z) + \varphi_1(z)$, employing the transformation $e^{i\theta} = \cos \theta + i \sin \theta$, and equating the real and imaginary parts, Eqs. (1)–(3) become
From Eqs. (4)–(6), we can see that the direction of the energy flow among the three interacting waves is determined by \( \sin \theta \). When \( \sin \theta < 0 \), \( \frac{d\rho_1}{dz} > 0 \) and \( \frac{d\rho_3}{dz} < 0 \), the energy flow is from wave \( W_3 \) to waves \( W_1 \) and \( W_2 \) (i.e., \( W_3 \rightarrow W_1 + W_2 \)). When \( \sin \theta > 0 \), \( \frac{d\rho_2}{dz} > 0 \) and \( \frac{d\rho_3}{dz} < 0 \), the energy flow is inverse, i.e., \( W_1 + W_2 \rightarrow W_3 \). From the definition of \( \theta \), a phase accumulation due to \( \Delta k \) can induce the symbol change of \( \sin \theta \), so a phase mismatch can cause the back conversion. However, here we only consider the perfect phase-matching condition (\( \Delta k = 0 \)). In this case, \( \sin \theta = \sin[\varphi_1(z) + \varphi_2(z) - \varphi_3(z)] \), that is, the direction of the energy flow depends on the phase changes of the three interacting waves.

Let us consider a DFG process, in which only \( W_2 \) and \( W_3 \) are seeded onto the crystal. The initial phases of these two waves are assumed to be equal to zero, i.e., \( \varphi_2(0) = 0 \) and \( \varphi_3(0) = 0 \). In this case, the newly generated wave \( W_1 \) accommodates its phase to \( \varphi_1(0) = -\pi/2 \) in order to maximize the conversion rate from \( W_3 \) to \( W_1 \) and \( W_2 \) (\( \sin \theta = -1 \)). As there is no phase mismatch, \( \sin \theta \) remains at a value of \(-1\) until the pump \( W_3 \) is completely depleted at a position \( z = L_0 \). From Eq. (9), when \( \rho_3 = 0 \), \( \frac{d\rho_3}{dz} \rightarrow \infty \), so there exists a phase jump on \( \varphi_3 \). This phase jump activates the back conversion in the SFG. From Eqs. (4)–(6), to maximize the rate of such a back conversion, it requires \( \sin \theta = 1 \), i.e., \( \varphi_1(L_0) + \varphi_2(L_0) - \varphi_3(L_0) = \pi/2 \). As \( \varphi_1(L_0) = -\pi/2 \) and \( \varphi_2(L_0) = 0 \), \( \varphi_3 \) has to jump from 0 to \(-\pi\), i.e., \( \varphi_3(L_0) = -\pi \). Such a phase jump is the underlying reason for the back conversion. At the position \( z = 2L_0 \), the idler will be completely depleted. Similar to the above analysis, \( \varphi_1(2L_0) \) will jump from \(-\pi/2\) to \(\pi/2\) to change \( \sin \theta \) from 1 to \(-1\) to activate the forward DFG process again. In such a way, the forward and backward conversions happen periodically. Note that the phase of \( W_2 \) does not change with the distance during the whole process, because it cannot be depleted at any time.

To verify the above theoretical analysis, we numerically solved Eqs. (1)–(3) by the symmetrized split-step Fourier method. A DFG process with \( \lambda_3 = 532 \) and \( \lambda_2 = 800 \) nm in a collinear phase-matched \( \beta \)-BBO crystal orientated at 22.05° was simulated. Pump wave \( W_3 \) is a Gaussian pulse with a full width at half-maximum (FWHM) duration of 100 ps, and its peak intensity is 5 GW/cm², while the incident signal wave \( W_2 \) is a super-Gaussian pulse with an FWHM duration of 240 ps and a peak intensity of 1 GW/cm² (Fig. 1). Dispersions, walk-off, and diffraction effects are all neglected in our calculations.

Figure 2 summarizes the calculated intensity and phase evolutions of the three interacting waves in a 10-mm-long crystal.

![Fig. 2. Intensity and phase evolutions of the three interacting waves.](image-url)
As is clearly shown in Fig. 1, respectively. $\omega_3$ shows the intensity evolutions of flat-top profile, or the pump and signal pulses can be converted effects, the pump pulses can be shaped to a spatiotemporally Gaussian laser. To mitigate the back effect of back conversion on the conversion efficiency is more serious in the OPA, which is typically pumped by $A$ and $B$. To illustrate this, we assume that the absorption coefficient of $W_1$ is $a$ and rewrite Eq. (1) as

$$\frac{dA_1}{dz} = -i a n_1 c A_1^* A_3 e^{-i A_3^* A_3} - \frac{1}{2} A_1. \quad (10)$$

By numerically solving Eqs. (1)–(3) and (10), the intensity and phase evolutions of the three waves under two absorption levels of $A_{L_{\text{abs}}}$ are shown in Fig. 4, where the nonlinear length $L_{\text{nl}}$ is fixed at 1.5 mm. By inducing an absorption of $a L_{\text{abs}} = 8$ on $W_1$, the back conversion indeed no longer happens as expected, as shown in Fig. 4(b). At each position of the crystal, it can be viewed as a DFG process without the interference of the idler. Therefore, phase $\phi_1$ keeps at $-\pi/2$ across the entire crystal, while $\phi_2$ and $\phi_3$ remain at a constant value of zero [Fig. 4(d)]. This in the case of the conversion from $W_3$ to $W_2$ becomes unidirectional without back conversion [Fig. 4(b)]. The inhibition of the back conversion process ensures a complete depletion of $W_3$ and a maximum amplification of $W_2$.

In the case of $a L_{\text{abs}} = 2$, although the back conversion can still occur initially, it will diminish gradually, as shown in Fig. 4(a). Compared to the case of no absorption in $W_1$, the depletion of $W_1$ is enhanced by the absorption, so its phase jump takes place earlier than usual [Fig. 4(c)]. This earlier phase jump activates the forward conversion process before the end of a complete back conversion. In such a way, $W_2$ still obtains a net gain in one forward–backward conversion process. As a result, $W_2$ keeps rising with the decreasing oscillation amplitude [Fig. 4(a)]. Finally, $W_2$

![Fig. 3. Intensity evolutions of $W_3$ at three different temporal points. The black, red, and blue curves correspond to points A, B, and C in Fig. 1, respectively.](image1)

![Fig. 4. Intensity and phase evolutions of $W_1$ (blue curves), $W_2$ (red curves), and $W_3$ (black curves) under the conditions of $a L_{\text{abs}} = 2$ [(a) and (c)] and 8 [(b) and (d)]. All calculations correspond to point A in Fig. 1.](image2)
will approach to the quantum-limited intensity with negligible back conversion, and $W_3$ is almost completely depleted. In practice, the choice of $\alpha$ depends on the initial pump and signal intensities and also on the crystal length. On the one hand, to effectively suppress the back conversion, the value of $\alpha$ should be larger than a certain value, $\alpha L_{\text{nl}} > m$ ($m = 0.3$ in the case of Ref. [9]). On the other hand, to ensure a maximum conversion efficiency within a limited crystal length $L$, the value of $\alpha$ should be smaller than a certain value, $\alpha L < n$.

Due to the suppression of the back conversion, each temporal coordinate on $W_2$ can obtain a maximum gain at the output end of the crystal (Fig. 5), which ensures the maximum conversion efficiency regardless of the profile of $W_3$. Figure 5 also implies that such a DFG process with absorption in $W_1$ is robust against the variation of the pump intensity. A more stable output can be expected from such a DFG process without back conversion.

In conclusion, we reveal that the phase jump, accompanied by one depleted interacting wave, is responsible for the intrinsic back conversion in quadratic nonlinear processes. Based on this understanding, we find an effective method to inhibit the back conversion in a parametric down-conversion process. By using crystal absorption to enhance the depletion of the undesirable wave, the back conversion can be terminated, and the conversion efficiency from the pump to the desirable wave could reach the quantum limit. This method for suppressing the back conversion by absorbing the idler wave is compatible with the technique of noncollinear phase matching, so it applies to broadband nonlinear amplification. In the case of broadband amplification, absorption on a specific wavelength (often corresponding to the magic wavelength) can be obtained by doping specific rare-earth ions in an yttrium calcium oxyborate crystal, as we did in Ref. [9]. In addition to the linear absorption method, one could also use a nonlinear method to deplete the undesirable wave, e.g., designing a second-harmonic generation process for this wave in the same crystal[12], which, however, is hard to realize in conventional bulk crystals. The results presented in this study further our understanding of quadratic nonlinear processes and pave the way toward the design of highly efficient parametric down-converters.

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References