Multistep quadratic cascading in broadband optical parametric generation

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We theoretically and experimentally investigate multistep parametric processes in broadband optical parametric generators (OPGs) based on periodically poled 1 mol. \% MgO-doped stoichiometric LiTaO$_3$. We demonstrate that parametric collateral processes may deplete or enhance spectral portions of the OPG output, depending on pump pulse duration. © 2012 Optical Society of America

A wide range of applications, encompassing ultrafast spectroscopy, high-harmonic and attosecond pulse generation, frequency combs, and remote sensing, spur current research on broadband optical sources in the infrared \([1,2]\). Coherent ultra-broadband parametric gain suitable for ultrashort pulse generation and amplification can be afforded by quadratic frequency downconversion in quasi-phase-matched (QPM) materials operated close to their zero group velocity dispersion point. Based on this principle, broadband optical parametric generation (OPG) has been achieved in periodically patterned GaAs \([3]\), KTiOPO$_4$ \([4]\), LiNbO$_3$ \([5]\), and, recently, LiTaO$_3$, with a record gain bandwidth of 180 THz \([6]\).

The spectral flatness of the parametric gain curves is a feature almost as important as the breadth of their spectral coverage. However, multistep $\chi^{(2)}$ processes triggered by broadband OPG, such as sum-frequency generation (SFG), often disrupt the flatness of gain profiles in QPM media \([3-6]\).

In this letter, we investigate theoretically and experimentally the impact of multistep $\chi^{(2)}$ processes on broadband OPG in the pulsed regime. With reference to SFG, we show how quadratic cascading \([7]\) can be used to engineer the OPG spectral profiles. Specifically, we demonstrate that the SFG-induced gain dips commonly observed in broadband OPG \([3-6]\) can be turned into peaks by suitably choosing the pulse duration of the OPG pump. Experiments conducted in periodically poled MgO-doped stoichiometric LiTaO$_3$ (PPLN) \([6]\) provide evidence for theoretical predictions based on a model accounting rigorously for broadband OPG and multistep processes \([8]\).

The multistep quadratic interactions that can affect ultra-broadband parametric generation in a periodically poled medium are schematically illustrated by Fig. 1(a). In the $\chi^{(2)}$ material, the optical pump at $\lambda_P$ excites parametric generation in the infrared region ($\lambda_{\text{OPG}}$) by means of first-order QPM processes. High-order resonances of the QPM grating can simultaneously enable SFG between the pump and specific components ($\lambda_S$ in Fig. 1) within the broadband OPG spectrum.

The SFG process ($1/\lambda_P + 1/\lambda_S \to 1/\lambda_{\text{SFG}}$) subtracts energy from the OPG band, carving a gain dip at $\lambda_S$ as highlighted on the spectrum of Fig. 1(b). This is the situation typically encountered in experiments \([3-6]\).

The unwanted effects of parasitic SFG processes on the parametric gain could in principle be alleviated through more sophisticated QPM grating designs, yet this option becomes ever more challenging as the OPG spectra get wider. Alternatively, instead of trying to remove the QPM resonances responsible for upconversion, one could exploit SFG cascading to reroute power into the OPG band by difference-frequency generation (DFG; cf. Fig. 1(a)).

In principle, according to the theory of quadratic cascading for nondegenerate frequency upconversion \([9]\), the direction of energy flow in the SFG step is reversible (even at phase matching) as long as depletion of one of the waves (either $\lambda_S$ or $\lambda_P$) is locally achieved in the device. In our configuration (Fig. 1(a)), the signal ($\lambda_S$) is generated by the OPG pump, and therefore the control of SFG cascading ultimately resides only in the latter (i.e., the input at $\lambda_P$).
To assess the impact of SFG cascading on broadband parametric generation, we performed a systematic numerical analysis based on a recently developed mathematical model [8], which can account rigorously for all the spectral components and multiple interactions of Fig. 1. We explored the device response in the pulsed regime under experimentally viable conditions [6], referring to OPG in a 1-cm-long PPMgSLT crystal, doped with 1 mol. % MgO, with a QPM grating with a period of 25 μm and a duty cycle of 42.5%, operated at a temperature $T = 80\,^\circ C$ and pumped close to its zero-dispersion point ($\lambda_P = 860\, nm$) by Gaussian pulses at 1 kHz, with peak intensities in the range 1–10 GW/cm² and pulse durations ranging between 0.5 and 30 ps.

The simulations employed the following quadratic nonlinear envelope equation [8]:

$$\frac{dA}{dz} + iDA = -\frac{i\chi^{(2)}(\omega_0)}{4\beta_0 c^2}\left(1 - \frac{i}{\omega_0 \sigma \tau}\right)[A^2e^{i\phi} + 2|A|^2e^{-i\phi}],$$

where $A(z,t)$ is the broadband complex electric field envelope at a reference pulse $\omega_0$. $\phi(z,t) = i\omega_0 t - i(\beta_0 - k_0 \omega_0)z$, and $\beta_0 = \text{Re}[k(\omega_0)]$ with $k(\omega) = (\omega/c)\sqrt{1 + \chi(\omega)}$ the propagation constant and $\chi(\omega)$ the linear electric susceptibility. $D = \sum_{m=2}^\infty \frac{1}{m!} km\left(-i\frac{\sigma}{\tau}\right)^m$ is the dispersion operator, $k_m = \frac{2\pi n_m^2}{\lambda_m^2}(\omega_0)$, $\chi^{(2)}$ is the second-order nonlinearity, $z$ is the propagation coordinate, and $\tau = t - k_1 z$ is the coordinate system, moving with the pump group velocity. The model refers to a plane-wave configuration, with no dependence on the transverse coordinates. Material dispersion is accounted for based on [10].

We simulated OPG from a classical Gaussian pump and a semiclassical pulse shot noise of 1 photon per mode with random phase on each spectral discretization bin. The OPG response was statistically evaluated over the ensemble of multiple simulations, performed with different random noise seeds. We typically used an average of 30 realizations, which compares well with the experimental conditions described in what follows.

Figure 2 illustrates key features of the calculated response, by considering OPG in PPMgSLT with a pump at 860 nm. In each panel, the upper plot shows the broadband OPG spectrum (sweeping from 1 to 3.5 μm) at the output, while the contour plot illustrates its evolution in propagation (along $z$) within the PPMgSLT crystal. Figures 2(a) and 2(b) show two different pump pulse durations, namely, 10 and 0.7 ps, and peak intensities, 7 and 15 GW/cm², respectively.

Figure 2(a) refers to the situation generally encountered in the experiments, where SFG induces localized dips in the gain spectra. In this specific case, the dips, located at 1.4, 1.5, 1.95, and 2.2 μm, are the signatures of two distinct SFG processes occurring in the PPMgSLT device, namely,

- SFG between $\lambda_P$ and $\lambda_{S1} = 1.4\, \mu m$, generating a wave at $\lambda_{SF1} = 533\, nm$ by third-order QPM;
- SFG between $\lambda_P$ and $\lambda_{S2} = 1.95\, \mu m$, generating a wave at $\lambda_{SF2} = 600\, nm$ by second-order QPM.

The two SFG processes also affect the OPG gain at the conjugate wavelengths [$\lambda_{S1} = \lambda_S\lambda_P/(\lambda_S - \lambda_P)$], located at $\lambda_{S1s} = 2.2\, \mu m$ and $\lambda_{S2s} = 1.5\, \mu m$, respectively.

![Figure 2](image)

Figure 2(b) shows simulations for the same device as Fig. 2(a), this time pumped with pulses of a much shorter duration (0.7 ps). As is apparent from the output spectrum, a gain enhancement now occurs at the very same spectral locations ($\lambda_{S1}, \lambda_{S2}, \lambda_{S1s}, \lambda_{S2s}$) where SFG was carving dips in the gain curve of Fig. 2(a). This happens as a consequence of the onset of SFG cascading, which brings power back into the OPG band by activating the DFG channel of Fig. 1(a).

The simulation results point out a crucial role played by pulse walk-off in the onset of SFG cascading. As the pump pulse duration is decreased, the walk-off among the spectral components involved in the SFG process ($\lambda_P$, $\lambda_{SF}$, and $\lambda_S$) increases. Accordingly, as these components lose their overlap upon propagation, the SF wave begins to back-convert its power to $\lambda_P$ and $\lambda_S$, yielding an enhancement of the OPG output. This agrees well with what would be expected for a single three-wave mixing interaction, where periodic upconversion and downconversion take place with a spatial period that depends on field intensities and temporal walk-off [11].

The above intuitive explanation for the results of Fig. 2 is also consistent with the evolution of the interacting fields observed in the time domain, illustrated by Fig. 3. As seen in Fig. 3(a), with the longer input pulses (10 ps), because the pump and the generated visible and infrared waves maintain their overlap, the temporal distribution of the total pulse exhibits no significant offset with respect to the input pulse ($t = 0$). This leads to efficient SFG and, hence, depletion of the OPG outputs at $\lambda_{S1}, \lambda_{S2}$ and $\lambda_{S1s}, \lambda_{S2s}$ (Fig. 2(a)). On the other hand, with shorter pulses (0.7 ps) the visible and infrared components
generated in the device get away faster from the pump. This effect can be recognized in the time-domain propagation plot of Fig. 3(b), where a skewed trace (asssociated with components walking off from the pump) clearly emerges for $z \geq 6$ mm. The progressive reduction of the temporal overlap among the spectral components involved in SFG locally alters the frequency-conversion dynamics, starting to feed power back in the infrared region through DFG (Fig. 2(b)).

To confirm the above predictions, we then set to investigate the response of ultra-broadband OPG in PPMgSLT [6] under varying pumping conditions, seeking experimental evidence for SFG cascading. The samples used for these experiments were 500 $\mu$m-thick, 1 cm-long, $z$-cut substrates poled with a constant period of 25 $\mu$m.

The pump source was a tunable picosecond Ti:sapphire amplifier system, consisting of a Nd:YVO$_4$-pumped femtosecond source that seeded a regenerative chirped-pulse amplifier, delivering microjoule pulses at 1 kHz with bandwidths of 2 nm, tunable from 820 to 900 nm. We varied the pump pulse durations by adjusting the group-delay dispersion introduced by the final compressor stage of the amplifier. We used a 300 mm lens to loosely focus the pump beam to a radius of 100 $\mu$m at the center of the PPMgSLT grating. At the sample ends, the radius increased only by 10%, justifying the plane-wave approximation used in the model. The crystal was kept at 80 °C. At its output, after rejecting the residual pump with a dichroic filter, we recorded the signal spectra with a 0.55 m spectrometer (Horiba Jobin Yvon iHR550) sensitive up to 4.8 $\mu$m, averaging over 900 pulses.

Within the limits of our experimental setup, we systematically investigated the impact of input pulse durations and powers on the output OPG spectra. Figure 4 summarizes the key results by comparing two measurements taken for the same pump peak intensity (6 GW/cm$^2$) but with two different input pulse durations, namely, 6.1 and 1.7 ps.

A comparison with the spectral responses expected with long and short pump pulses, respectively (Figs. 2(a), 2(b)), confirms the good agreement between our experimental observations and theory. The spectrum recorded with long pump pulses (6.1 ps; Fig. 4, red curve) bears the characteristic signatures of SFG, exhibiting a marked depletion of the OPG output at four signal wavelengths (1.4, 1.5, 1.95, and 2.2 $\mu$m; highlighted by arrows), which match well the theoretical predictions (Fig. 2(a)). On the other hand, when pulse durations are reduced (1.7 ps; Fig. 4, black curve), the OPG output is locally enhanced at the same spectral locations where the gain reduction was apparent with long pulses, as predicted by theory (Fig. 2(b)), as a result of SFG cascading.

In conclusion, we have studied the interplay of multistep quadratic processes with broadband OPG in the pulsed regime. We demonstrated the possibility of countering the effect of SFG on the parametric gain by triggering SFG cascading in order to channel energy back into the OPG band. Simulations and experiments in PPMgSLT show how the energy flow in the SFG step can be reversed by reducing the pump pulse duration so as to drive walk-off-initiated downconversion of the sum-frequency wave, turning the gain dips typically observed during broadband OPG in QPM media into peaks. Simulations also predict the possibility of accessing an intermediate regime, without dips or peaks, with significant improvements in the flatness of the OPG spectra, by fine-tuning the input pulse duration in relation to the sample length. Ultimately, these results point to new possibilities to engineer entirely optically the response of broadband parametric sources and amplifiers in QPM materials.

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### References
