

# Quadratic Cascading Effects in Broadband Optical Parametric Generation

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**Abstract:** We investigate theoretically and experimentally multistep processes in broadband optical parametric generation (OPG) and demonstrate the possibility to control fine features in the gain spectra via the OPG pump.

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## 1. Introduction

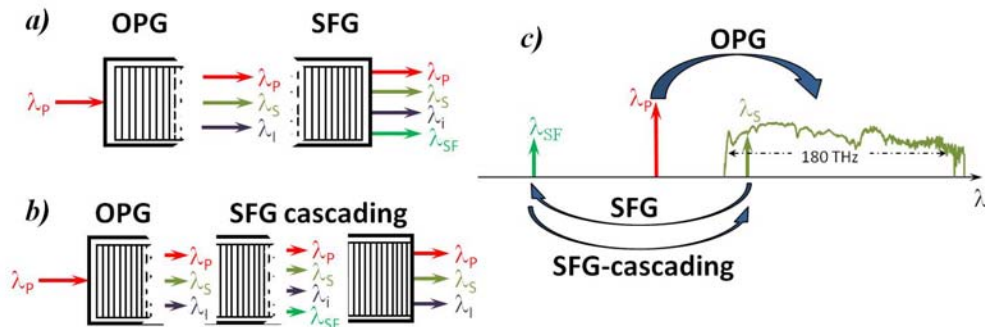
Access to coherent broad-band optical gain in the infrared is of considerable interest for applications ranging from ultrafast spectroscopy to high energy physics and frequency-comb generation. Frequency down-conversion processes in quadratic media have proven particularly well-suited to this aim,<sup>1</sup> especially since the recent developments of quasi-phase-matching (QPM)<sup>2</sup> techniques to engineer parametric resonances so to operate close to zero group-velocity-dispersion.<sup>3</sup> Broadband optical parametric generation (OPG) has been demonstrated based on this principle in several QPM materials, such as GaAs,<sup>4</sup> KTiOPO<sub>4</sub>,<sup>5</sup> LiNbO<sub>3</sub>,<sup>6</sup> and, most recently, in MgO-doped stoichiometric LiTaO<sub>3</sub> (MgO:SLT), where the broadest bandwidth (180 THz) to date have been achieved.<sup>7</sup>

The spectral flatness of the parametric gain curves is a feature almost as important as the breadth of their spectral coverage for practical applications. Concurrent nonlinear processes accompanying broadband OPG often contribute to disrupting the flatness of the gain profiles in QPM materials.<sup>5-7</sup> Collateral processes typically affecting OPG are sum-frequency generation (SFG) between the pump ( $\omega_p$ ) and specific spectral components ( $\omega_s$ ) in the OPG output (selected by higher-order QPM).<sup>6-7</sup> SFG ( $\omega_p + \omega_s \rightarrow \omega_{SF}$ ) subtracts energy from the OPG band, carving dips in the gain spectrum located at  $\omega_s$  and at its conjugate OPG frequency ( $\omega_p - \omega_s$ ).

Here we demonstrate how local spectral features due to SFG can be controlled so to bring energy back to the OPG band, through a suitable choice of pump pulse parameters. We analyse theoretically<sup>8</sup> the interplay of OPG and SFG in the pulse regime for different pump pulse durations and identify the possibility to reverse the direction of energy flow in the SFG step by means of *quadratic cascading*.<sup>9</sup> Furthermore, we provide experimental evidence for the predicted effects and their impact on the OPG spectra in periodically poled MgO:SLT (PP:MgO:SLT), showing how SFG dips in the spectrum of a broadband parametric generator can be turned into peaks by decreasing the OPG pump pulsewidth.

## 2. Cascaded processes in broadband OPG

Fig.1 depicts the wave-mixing processes we are going to consider. A single PP:MgO:SLT crystal, pumped at  $\lambda_p$  (~860 nm) generates a broad OPG spectrum in the infrared (Fig. 1c, experiments of ref. 7). The presence of an OPG component at  $\lambda_s$  excites a SFG process between the latter and the pump ( $\lambda_p + \lambda_s \rightarrow \lambda_{SF}$ ), phase-matched via high-order QPM in the same PP:MgO:SLT grating (Fig. 1a).



**Fig.1.** Cascaded interactions in an ultra-broadband parametric generator, as in ref. 7. *a)* OPG followed by SFG between the pump ( $\lambda_p$ ) and the OPG component at  $\lambda_s$  results in depletion of the output at  $\lambda_s$ ; *b)* OPG followed by SFG cascading (up-conversion to  $\lambda_{SF}$  followed by down-conversion to  $\lambda_s$  and  $\lambda_p$ ) results in back-transfer of power to  $\lambda_s$ ; *c)* Illustration of the energy transfer mechanisms across the spectrum.

SFG up-converts OPG power from  $\lambda_S$  to  $\lambda_{SF}$ , as shown in Fig. 1c, resulting in the appearance of a spectral dip in the OPG output at  $\lambda_S$  (and at its conjugate wavelength  $\lambda_I^{-1} = \lambda_P^{-1} - \lambda_S^{-1}$ ).

Even for perfectly phase-matched SFG, the theory of quadratic *cascading* predicts the possibility of reversing the energy transfer, from up- ( $\lambda_P + \lambda_S \rightarrow \lambda_{SF}$ ) to down- ( $\lambda_{SF} \rightarrow \lambda_P + \lambda_S$ ) conversion, for sufficiently high conversion rates and imbalanced inputs.<sup>10</sup> If this occurs, SFG *cascading* (Fig. 1b) channels power back into the OPG band, suppressing the dips at  $\lambda_S$  (and  $\lambda_I$ ) in the output spectra. Quadratic *cascading* introduces an *all-optical* control of nonlinear interactions. In our case, it implies the possibility to control the direction of energy transfer ( $\lambda_S \leftrightarrow \lambda_{SF}$ ) in SFG by means of the optical input at  $\lambda_P$  (the OPG pump). In the pulsed regime, one can envisage to control the spectral features of the OPG output at  $\lambda_S$  and  $\lambda_I$  by adjusting the input temporal profile and peak power of the optical pump.

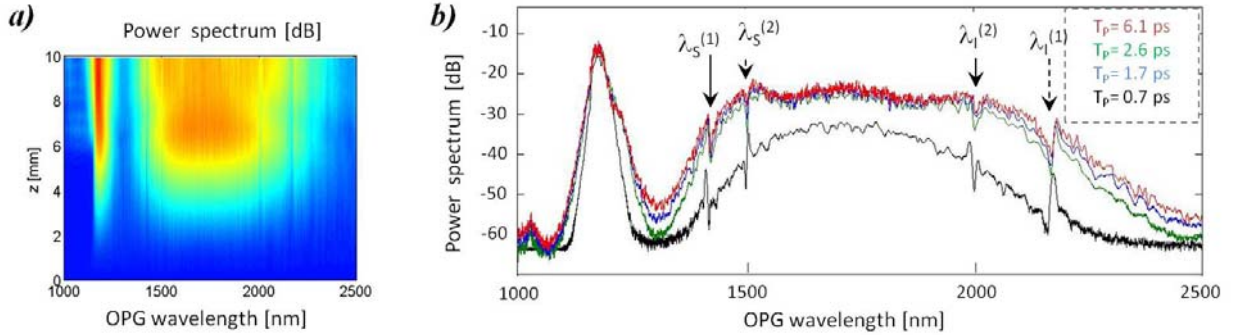
### 3. Simulations

We analysed numerically OPG in the presence of multistep parametric processes (Fig. 1) with a model that can rigorously account for the propagation of ultra-broadband pulses in quadratic media.<sup>8</sup> Fig. 2 illustrates simulation results for ultrabroadband OPG in a PP:MgO:SLT device with realistic conditions,<sup>7</sup> i.e. a 1cm-long PP:MgO:SLT grating with a uniform period and a non-50% duty-cycle, supporting the following SFG resonances:

1. pump+*signal* SFG (third order QPM) for:  $\lambda_S^{(1)} \sim 1.4 \mu\text{m}$  [OPG conjugate =  $\lambda_I^{(1)}$ ]
2. pump+*idler* SFG (second order QPM) for:  $\lambda_I^{(2)} \sim 2.0 \mu\text{m}$  [OPG conjugate =  $\lambda_S^{(2)}$ ].

The contour plot of Fig. 2a shows the evolution of the OPG spectrum upon propagation ( $z$ ) in the PP:MgO:SLT device, starting from broadband noise in the OPG band and a 2.6 ps-long (FWHM) Gaussian pump pulse at  $\lambda_P = 860\text{nm}$ , with a bandwidth of 2nm.

Fig. 2b illustrates the impact of different pump durations on the OPG response. Each curve is the statistical average over several (>30) simulations of the kind of Fig. 2a, with different noise seeds.



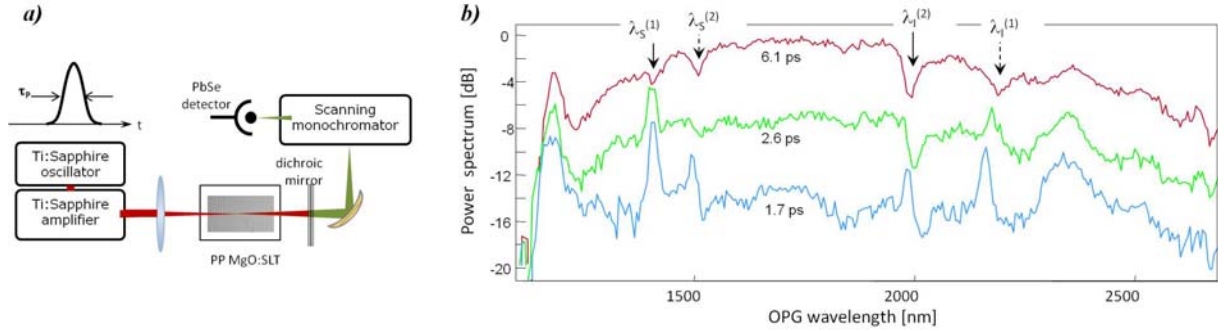
**Fig. 2.** Simulations of ultra-broadband OPG with cascaded SFG. **a)** Evolution of the OPG spectrum in propagation ( $z$ ). Inputs: Gaussian pump at 860 nm, with a pulsewidth (FWHM)  $T_p = 2.6$  ps and peak intensity  $I_0 = 5 \text{ GW/cm}^2$  and broadband noise in the OPG band. **b)** OPG outputs calculated for  $T_p = 6.1, 2.6, 1.7$  and  $0.7$  ps (red, green, blue and black curve, respectively), with the same peak intensity:  $I_0 = 5 \text{ GW/cm}^2$ .

In the OPG output spectra of Fig. 2b one can clearly recognize features at  $\lambda_S^{(1)}$  and  $\lambda_I^{(1)}$  (due to SFG between  $\lambda_S^{(1)}$  and  $\lambda_P$ ) and features at  $\lambda_I^{(2)}$  and  $\lambda_S^{(2)}$  (due to SFG between  $\lambda_I^{(2)}$  and  $\lambda_P$ ). Such features take the form of dips in the gain curve when OPG is pumped with relatively long pump pulses (e.g. top curve,  $T_p = 6.1\text{ps}$ ), i.e. when walk-off and cascading effects are not significant in SFG, so that power from  $\lambda_S^{(1)}$  and  $\lambda_I^{(2)}$  is simply up-converted to the visible band (Fig. 1a). On the other hand, as the OPG pump pulse duration is decreased (e.g. bottom curve,  $T_p = 0.7\text{ps}$ ), the OPG spectral dips turn into peaks, i.e. the output at the OPG wavelengths involved in the SFG process is actually locally enhanced. This is a signature of SFG *cascading* (fig.1b) feeding back power into the OPG band. The dependence of *cascading* on the pump pulse duration is a consequence of the nonlinear interplay of the interacting pulses in propagation, which is rigorously accounted for by the numerical model.<sup>8</sup> Intuitively the behaviour can be justified by considering that, as the pump pulse duration is reduced, the walk-off between the pulses involved in the SFG process increases (for a fixed device length). The SF will continue to extract power from the infrared wavelengths as long as it is travelling with them. As the pump pulse duration is decreased below a critical value, the SF walks off from the infrared components, which triggers its down-conversion, feeding power back into  $\lambda_S^{(1)}$  and  $\lambda_I^{(2)}$ . As a result, the OPG output can be locally enhanced, as seen in Fig. 2b for the shorter pulse case.

### 4. Experiments

Seeking experimental evidence for the above SFG cascading phenomena we investigated the response of a PP:MgO:SLT optical parametric generator with different pump durations. For details on the samples and

experimental setup (Fig. 3a) we refer the reader to ref. 7. Here we simply illustrate, in Fig. 3b, key experimental results which confirm the theoretical predictions, showing a clear signature of SFG cascading on broadband OPG.



**Fig. 3. a)** Experimental setup to investigate SFG effects in broadband OPG with PP:MgO:SLT. **b)** OPG spectra measured with a pulsed OPG pump at 860 nm for pulsewidths of 6.1, 2.6 and 1.7 ps (top, middle and bottom curves, respectively) with the same pump peak intensity.

In the experiments (Fig. 3a), a PP:MgO:SLT sample with a poling period of  $25\mu\text{m}$  was pumped by a chirped pulse amplified Ti:sapphire system delivering 1kHz ps-pulses of 2 nm bandwidth, focused to a beam radius of  $100\mu\text{m}$  by a 300 mm lens. To obtain the data shown in Fig. 3b, we varied the pump pulse duration by adjusting the group velocity dispersion introduced by the final compressor stage and recorded the OPG spectra at the device output, with a scanning monochromator (Horiba Jobin Yvon iHR550, PbSe detector) averaging over 900 pulses. The pump intensity was adjusted to maintain the peak intensity constant at  $6\text{ GW/cm}^2$ . On the experimental OPG gain curves of Fig. 3b, measured for three different pump pulse durations (1.5, 2.6 and 6.1ps), we have highlighted the spectral locations of the SFG resonances, as per previous discussion.

From Fig. 3b it is apparent that, in accordance with the previous argumentations, with long pump pulses (top curve, in red), the SFG processes simply deplete the OPG output and carve dips into the gain curve at  $\lambda_S^{(1)}$  and  $\lambda_I^{(2)}$  [and their conjugates:  $\lambda_I^{(1)}$  and  $\lambda_S^{(2)}$ ]. On the other hand, with shorter pump pulses (bottom curve, in blue), SFG *cascading*, triggered by the SF walk-off, enhances the OPG output at the specific signal and idler pairs involved in the SFG process, transforming the OPG dips into peaks.

#### 4. Conclusions

We have reported on the experimental observation, corroborated by numerical modeling, of quadratic *cascading* effects arising during ultra-broadband parametric generation in periodically poled MgO:SLT. In a pulsed OPG regime, *cascading* triggered by temporal walk-off can reverse the direction of energy flow in the SFG processes which normally accompany broadband OPG. As a result, narrow-band spectral features in the OPG output, arising from SFG, can be controlled *all-optically* by varying the OPG pump input. In the experiments we could turn on and off the OPG gain at specific wavelengths by adjusting the temporal duration of the OPG pump pulses. These results point out to new potential routes to ‘dynamically’ engineer the spectral response of broadband parametric generators and amplifiers.

#### 5. Acknowledgements

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