This is the published version of a paper published in *Optics Express*.

Citation for the original published paper (version of record):


Access to the published version may require subscription.

N.B. When citing this work, cite the original published paper.

Permanent link to this version:
http://urn.kb.se/resolve?urn=urn:nbn:se:kth:diva-249969
Narrowband, tunable, infrared radiation by parametric amplification of a chirped backward-wave OPO signal

ANNE-LISE VIOTTI,* ANDRIUS ZUKAUSKAS, CARLOTA CANALIAS, FREDRIK LAURELL, AND VALDAS PASISKEVICIUS

Department of Applied Physics, Royal Institute of Technology, Roslagstullsbacken 21, 106 91 Stockholm, Sweden
*alviotti@kth.se

Abstract: The strict momentum conservation constraints for backward-wave optical parametric oscillators (BWOPOs) gives an inherently narrowband backward-generated wave, even with broadband pumping. Unfortunately, the limited tuning range of this wave restricts potential applications. Here we demonstrate a method to circumvent this restriction and increase the tuning range by more than one order of magnitude. A linearly chirped pump modulation is transferred to the forward-generated BWOPO wave, which is then mixed with an identically chirped pump in a conventional optical parametric amplifier to obtain narrowband (38 GHz), broadly tunable, infrared radiation around 1.86 µm, with an output energy of 19 µJ.

© 2019 Optical Society of America under the terms of the OSA Open Access Publishing Agreement

1. Introduction

Methods for generating two infrared narrowband pulses with tunable frequency separation over a few THz with energy scaling potential would be of interest for stimulated Raman scattering spectroscopy [1,2] studies of phonon-polariton modes in ferroelectrics [3,4] and other linear and nonlinear materials. Narrowband tunable sub-nanosecond pulses in the near-to mid-IR can be employed in imaging techniques such as coherent anti-Stokes Raman scattering (CARS) where longer excitation wavelengths would help to reduce the Rayleigh scattering efficiency, avoid damage on biological samples and potentially provide higher measurement sensitivity [5–7].

Narrowband signal and idler waves can be generated in over-constrained doubly-resonant optical parametric oscillator (OPO) cavities where the signal and idler phases are fixed by the boundary conditions of the cavity [8]. However, stable, steady-state operation in the sub-nanosecond regime with such a device would be difficult to obtain. A method for bandwidth tailoring using a fiber-based four-wave mixing (FWM) OPO cavity has been suggested in [9], where the FWM has to operate at degeneracy and at the zero GVD point of the photonic crystal fiber, harnessing the large dispersive propagation length in the rest of the cavity in order to achieve substantial stretching. However, such a device is inherently sensitive to pump frequency fluctuations and environmental factors. Additionally it would be very challenging to generate picosecond, narrowband and tunable pulses with energies in the µJ range with this technique. Synchronously-pumped OPOs with intra-cavity spectral filtering have been used to generate dual narrowband, tunable picosecond pulses [10,11], but with low output powers. Reaching µJ energies would require employing a separate synchronized laser to pump an additional optical parametric amplifier (OPA) and a pulse-picker to reduce the repetition rate appropriate for the pump laser.

Bandwidth tailoring of infrared idler waves in OPA can be done by monitoring the chirps of the pump and the seed through the concept of double-chirp optical parametric amplification [12,13]. Backward-wave optical parametric oscillator (BWOPO) is the only
device which can generate a properly chirped OPA seed directly from the pump, i.e. without invoking any additional OPA stage that in turn has to be seeded with an external, narrowband and synchronized laser source [14]. Other techniques, including OPA seeded with supercontinuum [15], or exploiting optical parametric generation (OPG) [16,17] would require additional spectral phase tailoring steps to properly adjust the chirp of the seed. Moreover, in the OPG case, the temporal coherence is limited by the amplified quantum noise which, by definition, has a random phase with respect to the pump.

Backward-wave optical parametric generation is the second-order nonlinear process in which the signal and idler waves are forced by momentum conservation to propagate in opposite directions [18,19]. The counter-propagating parametric waves cross-seed each other with the energy from the pump, thereby establishing distributed feedback and fulfilling the condition for oscillation without a need for external mirrors [20]. With the introduction of quasi-phase matching [21], additional freedom in momentum conservation through the grating wave-vector was obtained, making it possible to demonstrate BWOPO, or as it is also called, mirrorless OPO [22]. The periodically poled crystals necessary for BWOPOs require a dense domain structure with a sub-μm periodicity. For bulk interactions, this has only been observed in crystals from the KTiOPO4 family [23]. In contrast to conventional OPOs, BWOPOs demonstrate unique spectral properties arising from the strong momentum conservation constraints imposed by the counter-propagating parametric interaction. The consequence is that the (idler) wave generated in the direction opposite to the pump will be inherently narrowband. For broadband pumping, the bandwidth of the backward-generated wave typically comprises 1% of the bandwidth of the pump [24]. As required by energy conservation, the spectral information from the pump is then translated into the forward-generated parametric wave. Therefore, a BWOPO can be viewed as a frequency translator which, for example, can find application in quantum state manipulation protocols [25]. The narrowband backward-generated wave, which is typically in the mid-IR, could be utilized for spectroscopic applications, e.g. for seeding mid-IR optical parametric amplifiers used in MIR LIDARs [26]. Interestingly, for BWOPOs, the wavelength of the backward-generated wave is quite insensitive to the pump wavelength, which eventually limits its tuning range. For instance, tuning of the backward idler wave by 1 THz would require pump tuning over 100 THz, comparable to the entire Ti:Sapphire gain bandwidth. Correspondingly, angular and temperature tuning ranges are limited to approximately one hundred GHz [27].

In this work, we present a method which allows circumvention of this tuning limitation, exploiting the frequency-translation properties of the forward-generated wave (the signal in this case). While pumping the BWOPO with broadband chirped pulses, the chirp is transferred to the forward-propagating signal wave at a longer central wavelength. The BWOPO signal is then used to seed a conventional co-propagating OPA pumped by the same chirped pump pulses, which acts as a double-chirp OPA [12]. The idler generated in the OPA retains all the narrowband properties of the BWOPO idler, but it is widely tunable by simply adjusting the temporal delay between the OPA pump and the BWOPO signal. Using stretched pulses derived from a Ti:Sapphire regenerative amplifier as the pump allowed us to reach a tuning range of 2.7 THz for the narrowband idler. Such a tuning range would be impossible to achieve for the idler directly from the BWOPO by tuning the pump wavelength using any known laser technology.

2. Experimental setup and results

The experimental setup, illustrated in Fig. 1, consists of a BWOPO followed by an OPA, both realized in periodically poled Rb-doped KTiOPO4 crystals (PPRKT) [23,28]. The BWOPO crystal had a poling period of 500 nm and a grating length of 7.3 mm. The uncoated sample was pumped by 240 ps-long (full width at half maximum (FWHM)), stretched pulses generated by a Ti:Sapphire regenerative amplifier operating at 1 kHz. The central wavelength of the pump pulses was 800 nm with a FWHM bandwidth of about 5.5 nm. The
The uncompressed output of the regenerative amplifier was used, leading to chirped pump pulses. A small fraction of the pump was used for the BWOPO pumping, while the rest of the pump was employed in the OPA after an appropriate delay.

![Illustration of the experimental setup with the BWOPO and the OPA crystals.](image)

The pump pulses were loosely focused into the BWOPO crystal to a beam waist radius of 105 µm with an f = 200 mm lens, and the pump power was controlled by a wave plate-polarizer arrangement. The optical pulses were polarized along the z-axis of the crystal and propagated along its x-axis. The pump generated a co-propagating signal centered at 1.4 µm and a backward-propagating idler at 1.867 µm. The intensity at the threshold of the BWOPO process was 720 MW/cm². The effective nonlinearity for the PPRKTP crystal was estimated to be 7.4 pm/V [20], which is reasonably close to the maximum expected nonlinearity of 10.7 pm/V for a perfectly poled crystal [29,30].

The total conversion efficiency for the BWOPO reached 42% at a pump energy of 196 µJ (Fig. 2(a)). The BWOPO signal and idler energies were 47 µJ and 35 µJ, respectively, at the maximum pump energy. The signal bandwidth was measured to be 2.15 THz at a pump energy of 120 µJ, while the measured FWHM bandwidth of the corresponding idler was 0.34 nm (Fig. 2(b)). Correcting for the spectrometer resolution of 0.1 nm (Yokogawa AQ6376), gives the spectral bandwidth of 0.325 nm, or about 28 GHz. As shown in Fig. 2(b), at larger pump energies the BWOPO-generated signal broadens. This is a direct consequence of increasing depletion of the chirped pump pulses and the frequency translation into the BWOPO signal. As can be seen from the pump spectra in Fig. 2(c), the depletion starts on the higher frequency side which corresponds to the trailing part of the pulse. This is due to the time required to establish the distributed feedback and oscillation in the BWOPO. As the pump energy increases, this delay is reduced, and the lower frequency components of the pump spectrum are also depleted and converted to BWOPO signal and idler. Thus, the signal spectrum gradually broadens on the longer wavelength side in conjunction with the pump depletion (Fig. 2(c)). The spectrum of the BWOPO idler remains stable and fixed regardless of the pump depletion.
The expected bandwidth of the backward-generated BWOPO idler, $\delta\omega_i$, can be estimated from the bandwidth of the depleted pump, $\delta\omega_p$, and the momentum conservation condition. Expanding $\delta\omega_i$ in power series with respect to $\delta\omega_p$ to the second-order gives $\delta\omega_i = \omega_i' \delta\omega_p + 0.5 \omega_i'' (\delta\omega_p)^2$, where the first- and second-order derivatives $\omega_i'$ and $\omega_i''$ of the idler frequency with respect to the pump frequency are:

$$\omega_i' = \beta_{i,s} - \beta_{i,p} \bigg/ \beta_{i,s} + \beta_{i,i}$$

$$\omega_i'' = \beta_{i,s} - \beta_{i,p} - \omega_i' \beta_{i,s} - \omega_i' \beta_{i,i} - 2 \omega_i' \beta_{i,i} \bigg/ \beta_{i,s} + \beta_{i,i} \tag{1}$$

In Eq. (1), $\beta_{i,j} = \partial k_j / \partial \omega$ and $\beta_{i,i} = \partial^2 k_j / \partial \omega^2$ are the wave-vector derivatives over the frequency for the pump, the signal and the idler, with the corresponding indices $j = p, s, i$, respectively. From the energy conservation, the derivative for the BWOPO forward signal wave is then $\omega_i' = 1 - \omega_i^{*}$. According to the wavelengths used in this work and employing the Sellmeier expansion from [31], the derivatives of the idler frequencies are: $\omega_i^{*} = -1.53 \times 10^{-3}$
and $\omega'' = -1.5 \times 10^{-5}$. The second-order corrections are three orders of magnitude smaller and can be safely neglected. From the measured FWHM spectral width of the depleted pump of 1.88 THz, at the incident energy of 100.4 µJ, the expected bandwidth of the backward idler wave is 28.8 GHz, in good agreement with the experimentally measured data.

As shown in Fig. 1, the BWOPO signal was subsequently used as a seed in the co-propagating OPA, after filtering out the remaining BWOPO pump. The OPA sample was 8 mm long and had a poling period of 26.3 µm in order to phase-match the amplification of the BWOPO signal for a pump wavelength of 800 nm. Pump and seed beams were focused to beam waists of 130 µm and superimposed collinearly in the OPA crystal to take advantage of the largest OPG gain in PPRKTP. The parametric gain bandwidth is about 50 THz in the 8 mm-long PPRKTP crystal for the spectral range investigated. As the parametric gain scales with the inverse square root of the crystal length [32], in order to reduce this gain bandwidth to 28 GHz (which corresponds to the BWOPO idler bandwidth) and directly generate a narrowband OPA idler, one would need to increase the sample length 3×10^6 times, which cannot be done in practice. The OPG without the seed reached a threshold of 120 µJ. The OPG spectrum is shown in Fig. 3(a). For an OPA pump energy of 85 µJ, and a seed of 1 µJ, the OPA pump depletion was 23.5% and the OPA gain was 25 dB for the peak of the signal spectrum. A 25 µJ output amplified signal energy was measured, corresponding to an OPA idler energy of 19 µJ. The signal spectrum after the OPA is shown by the purple curve in Fig. 3(b). The apparent wavelength shift of the OPA signal with respect to the BWOPO signal (i.e. the OPA seed) is due to the larger gain available at longer wavelengths, as seen in Fig. 3(a).

![Fig. 3. (a) OPG spectrum; (b) Spectrum of the OPA seed (black curve) and the OPA signal (purple curve) showing the amplification process.](image)

Chirp measurements for both pump and signal were performed using a cross-correlation setup. A small fraction of the Ti:Sapphire regenerative amplifier output was compressed and served as a reference pulse with a pulse duration of approximately 70 fs. From the cross-correlation measurements, we can compare the chirp of the pump, the BWOPO signal and the OPA signal (Fig. 4).
Both the pump and the BWOPO signal have positive linear chirps of similar magnitude, approximately 59 mrad/ps$^2$. This was expected because in a BWOPO configuration, the phase modulation is coherently transferred to the forward wave. We note that the BWOPO signal pulse duration is 120 ps (FWHM), which is half as long as the pump pulse, when pumped at the maximum energy of 195.6 µJ. These chirp measurements indicate that adjusting the delay, $\Delta t$, between the pump and the seed (i.e. the BWOPO signal), before the broadband amplifier, will allow tuning of the OPA idler central wavelength, while preserving the narrowband spectrum for the idler. The width would still be defined by the difference in chirp rates between the seed and the pump [12]:

$$\omega_t(t, \Delta t) = \omega_p - \omega_s + \alpha_p t - \alpha_s (t + \Delta t)$$

$$= \omega_p - \omega_s - \alpha_s \Delta t,$$

where $\alpha_p$ and $\alpha_s$ are the chirp rates of the pump and the seed, respectively. The BWOPO frequency translation properties ensure that the linear chirp rates of the OPA seed and pump are essentially equal (see Fig. 4), which leads to the approximation made in Eq. (2). To verify that the OPA pumped with the chirped pump pulses does not impart additional chirp, we also measured the chirp rate of the amplified signal. As presented in Fig. 4, the measured chirp rate of the OPA signal is found to be 59 mrad/ps$^2$, i.e. the same as that of the seed before amplification. The broadband OPA hence conserves the chirp of the seed and therefore allows tuning of the idler according to Eq. (2) while preserving the narrow bandwidth.

In fact, the chirp rates of the pump and the seed, generated by the BWOPO would be slightly different. Specifically, a chirp rate ratio $\alpha_s / \alpha_p = 1.0153$ is estimated to the first order from equations in [22,24]. This difference in the chirp rates determines the OPA idler bandwidth. First, the OPA idler bandwidth will have a constant component $\delta \nu_I = (\alpha_s - \alpha_p) \tau_s$, given the variation in the frequency difference between the pump and the seed. Here $\tau_s$ is the seed pulse length. Second, this idler frequency variation will be modified during the OPA process due to the group velocity mismatch between the pump and the seed. As both the pump and the seed are located in the normal dispersion region of PPRKTP, the group velocity mismatch will result in an OPA idler bandwidth increase. The total idler bandwidth can then be estimated by:
\[ \delta \nu_{i,\text{OPA}} = \frac{1}{2\pi} \left( \alpha_s \tau - \alpha_p \left( \tau - L \left( \beta_{s,p} - \beta_{s,i} \right) \right) \right), \quad (3) \]

where \( L \) is the OPA crystal length. For the parameters of our experiment, this bandwidth will be about 32 GHz, i.e. only somewhat larger than the bandwidth of the backward wave in the BWOPO.

As illustrated on the setup scheme (Fig. 1), the OPA pump pulses go through a variable delay line before entering the crystal. Adjusting the temporal delay between the pump and the seed pulses with this delay line allows tuning of the central wavelength of the OPA idler. This method provides a much larger tuning range than the one achievable by tuning the central wavelength of the BWOPO pump. For instance, here, we show a tuning range of 2.7 THz (about 28 nm) obtained by changing the delay by 137 ps, as seen in Fig. 5. In the BWOPO-OPA configuration, the limit of the tuning range comes from the bandwidth of the pump itself. In order to achieve the same tuning range in BWOPO, one would need to tune the BWOPO pump by approximately 177 THz, as given by the relation:

\[ \Delta \omega_p = \Delta \omega_{\nu_p} = \frac{\nu_{\text{g}_p} + \nu_{\text{g}_s}}{\nu_{\text{g}_i} \left( \nu_{\text{g}_i} - \nu_{\text{g}_p} \right)}, \quad (4) \]

where \( \nu_{\text{g}_j} \) are the group velocities for \( j = p, s, i \), respectively.

![Fig. 5. (a) Tuning of the idler wavelength for different delays of the OPA pump. The BWOPO idler spectrum is shown with the dashed red line. (b) Frequency of the tunable OPA idler and the BWOPO idler for different pump delays. The 0 delay corresponds to the middle of the tuning range.](image)

The OPA idler peaks in Fig. 5 were measured with a Jobin-Yvon iHR550 spectrometer with a resolution of 0.25 nm, except for the peak at largest delay of 117 ps (leftmost peak in Fig. 5(a)) where the resolution was decreased to 0.5 nm owing to the lower average power. This was caused by the decreasing temporal overlap between the pump and the seed. The median value of the OPA idler bandwidth throughout the tuning range was 38 GHz, in line with the expectations of Eq. (3). The measured tunability of the idler peak position is shown in Fig. 5(b) with red data points.

A small fraction of the BWOPO backward-propagating idler was sent to the OPA crystal and amplified so that the BWOPO idler frequency could also be measured after the OPA sample at the same time (black squares in Fig. 5(b)). In this way, two narrowband near-IR pulses are generated, with a frequency separation that can be tuned over 2.7 THz.
3. Conclusion

A novel method to generate narrowband infrared radiation, tunable over 2.7 THz limited by the bandwidth of the pump, exploiting counter- and co-propagating parametric processes was presented. The wide tunability range, could for example be of interest e.g. for THz generation. An output energy of 19 µJ for the OPA idler was estimated but the method is energy scalable thanks to the presence of the OPA in the setup. Furthermore, the bandwidth of the tunable OPA idler can be varied down to the transform limit by adjusting the chirp of the OPA seed. This can be done by inserting a dispersive medium on the seed beam path if needed. Such narrow spectral radiation could be used for infrared spectroscopy. Generating narrowband radiation even further in the IR could be envisioned with orientation-patterned semiconductors, in order to target a broader range of IR-active molecules. Another interesting feature of this setup is the efficient phase transfer from the pump to the OPA signal, via the BWOPO counter-propagating interaction. As the OPA signal and the pump show the very same chirp, one should be able to compress the signal in order to get an easily power-scalable femtosecond broadband source in the near infrared region.

Funding

Knut and Alice Wallenberg Foundation (2016-0104); Swedish Research Council (VR) through the Linnaeus Center ADOPT.

Acknowledgments

The authors thank Prof. Ursula Gibson for her help proofreading the manuscript.

References