

Spectral and Linewidth Properties Investigation of Backward-Wave Optical Parametric Oscillators Pumped by Single-Frequency Nanosecond Pulses for IPDA Applications

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ABSTRACT

We report on the experimental investigation of the fine spectral properties of backward-wave optical parametric oscillators (BWOPs) pumped by narrow-linewidth nanosecond pulses. This is enabled by implementing high-frequency heterodyne beat-note measurement techniques between the BWOP output and a narrow-linewidth laser diode. With this technique, instead of the pure Fourier-transform-limited pulses predicted by plane-wave theory, measurements show an asymmetric Airy-type spectral broadening for the parametric waves with multiple side-lobes spanning over 1.5 GHz at the -20 dB level. We interpret this structure as arising from an extremely slight variation in the order of $10\text{ m}^{-1}/\text{mm}$ in the phase matching condition within the crystal itself, which would correspond to a deviation of a few pm/mm in the poling period along propagation direction. This interpretation is supported by different observations and numerical simulations. The origin of the phase matching condition variations is potentially multi-factorial, and its amplitude is extremely small, which makes it difficult to predict or control. We thus propose conditions of implementations for the BWOP, which would enable to circumvent the issue, and to generate narrow-linewidth nanosecond pulses, that are required for numerous applications such as trace-gas sensing in the atmosphere. In particular, using a temporal pump pulse profile close to a top-hat shape is expected to bring narrow-linewidth spectral shapes, even in the case of imperfect phase matching condition. Promising preliminary results have been obtained on this latter topic, by using a hybrid fiber-bulk Ytterbium laser amplifier with active pulse shaping capabilities as the pump laser for the BWOP.

Keywords: Backward Wave Optical Parametric Oscillator, Infrared, Spectroscopy, Lidar, IPDA

1. INTRODUCTION

Recently, the backward-wave Optical Parametric Oscillator (BWOP) approach has been proposed as promising alternative to conventional "co-propagating" optical parametric oscillators (OPOs) for infrared trace-gas spectroscopy and remote sensing applications.¹⁻³ In the particular context of space-borne greenhouse gases monitoring missions with the integrated path differential absorption lidar (IPDA) method such as ACDL,^{4,5} and MERLIN,^{6,7} which rely on OPOs as the master tunable laser source, the BWOP would provide very attractive characteristics including mirrorless operation, and intrinsic narrow linewidth emission without using injection seeding schemes.⁸ Up to now, most of the research work on BWOPs has been carried out in pulsed regime with sub-nanosecond multi-mode pump laser sources,⁹ but space-borne IPDA requires extreme laser spectral purity and narrow linewidth operation that may only be accessible for longer pulse durations in the nanosecond regime^{10,11} with single longitudinal mode pump lasers. On this topic the focus has essentially been on power characteristics and conversion efficiencies,^{12,13} while the efforts to measure and study the fine spectral properties of these sources has been limited in terms of resolution to a few hundreds of MHz.^{2,14} To assess more

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precisely the spectral output in this regime, we have implemented heterodyning techniques to obtain shot by shot time-resolved spectra of the output backward and forward waves produced by a BWOPO. This method has been employed in different set-ups and wavelength ranges of interest for IPDA measurement of CO_2 or CH_4 from space (at 2051 nm and 1645 nm respectively), with either a solid-state diode-pumped injection-seeded Nd:YAG at 1064 nm, or a hybrid fiber-bulk Ytterbium-based master oscillator power amplifier (MOPA) at 1030 nm. In all cases, we found that the BWOPO waves present an unexpected spectral broadening and structuring with an asymmetric Airy-type shape that expands up to a few GHz depending on the intensity of the pump. Based on experimental observations we interpret this spectral broadening as arising from a slight phase-matching (PM) condition inhomogeneity distributed along the propagation direction in the nonlinear crystal. By incorporating a linear PM condition variation into a numerical model of the backward waves interaction, the observed features can be reproduced, thereby providing a better understanding of the potential defects responsible for the PM's inhomogeneity, as well as their amplitude. In the first part of this paper we present the results obtained in the case of a 2 μm BWOPO pumped by a commercial injection seeded 1064 nm Nd:YAG laser that is representative of current pump sources developed for space lidar applications.^{15,16} In this case the backward is emitted at 2051 nm, which could be used to perform CO_2 spectroscopy in a low-pressure gas-cell. In a second part we propose a more advanced set-up employing a hybrid fiber-bulk Ytterbium MOPA at 1030 nm as the pump source for a BWOPO working at the MERLIN wavelength 1645 nm for CH_4 . In this case we show promising preliminary results where the pulse shaping capabilities offered by the MOPA approach can be leveraged to improve the spectral quality, with the use of top-hat shaped pump pulses as proposed by Zheng et al.¹⁷

2. BWOPO PUMPED BY A SINGLE-FREQUENCY SOLID-STATE Nd:YAG LASER

2.1 Experimental set-up

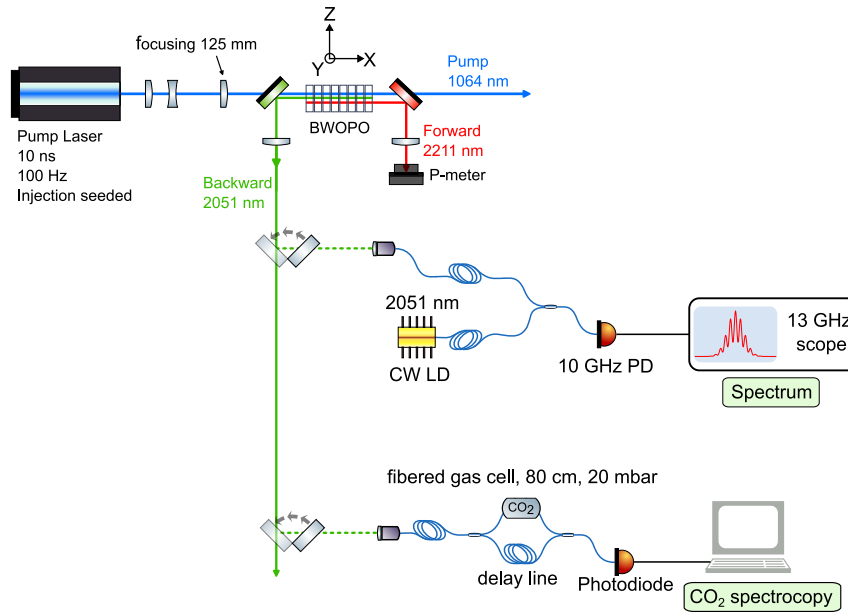


Figure 1. Experimental set-up. The BWOPO is based either on a 15 mm or 23 mm-long rubidium-doped periodically-poled KTP (Rb:PPKTP) with a poling period of 560 nm. Multiple diagnostics are implemented : BWOPO output power, heterodyne spectrum, and low-pressure CO_2 gas cell transmission measurements.

The overall experimental set-up is described in Fig. 1. The BWOPO optical implementation is compact: pump beam focusing, nonlinear crystal, and filtering dichroic mirrors to extract forward and backward waves. The nonlinear crystal consists in a 1 mm-thick, and 15 mm- (crystal #1) or 23 mm- (crystal #2) long Rb:PPKTP, with a poling period of 560 nm enabling the generation of a backward-wave at 2051 nm close to room temperature for a pump wavelength of 1064 nm. The poling period corresponds to the first order quasi-phase matching condition

(QPM) enabling counter-propagating signal and idler waves. The nonlinear crystal is mounted on a copper plate thermo-regulated at 23°C. The pump beam is extracted from an injection seeded Spitlight DPSS 100 from Innolas GmbH emitting single-frequency 10 ns pulses, that might be used for future BWOPO-OPA (optical parametric amplification) experiments. Only a very small fraction of the available pump energy is extracted (325 μJ out of 100 mJ). The beam is focused with a 105 μm diameter inside the BWOPO crystal, in order to maintain the pump fluence well below the light induced damage threshold (LIDT) in KTP being around 10 J/cm². The pump beam spatial profile is not perfect and contains a right-shaped pedestal surrounding a central gaussian distribution with a respective energy ratio of 28 % and 72 %. Peak fluence estimations presented in the following figures thus take into consideration this 72 % ratio from the total pump energy, and the 10 ns laser pulse duration. The beam diameter size is chosen much smaller than the crystal physical thickness (1 mm) as the aperture where the QPM poling is well-developed is reduced to an effective area of about 300 μm . Indeed, despite constant research efforts to improve the fabrication process of the Rb:PPKTP samples, the quality control is still a challenging point,¹⁸ and is not systematically reproducible from one batch to another yet. The backward wave beam is then sent in different diagnostic paths for power and spectral properties characterizations. For spectral measurements, the BWOPO is mixed with a CW-DFB laser diode (linewidth better than 5 MHz) at 2051 nm on a fast photodiode (electronic bandwidth > 10 GHz) to produce single-shot beat-note spectra, from which the BWOPO pulses spectrum may be extracted. Additionally, the backward wave beam can be sent into a low-pressure gas cell containing pure CO₂ at 15 Torr in order to carry out a transmission measurement experiment and illustrate the potential of BWOPO for spectroscopy applications.

2.2 Power characteristic

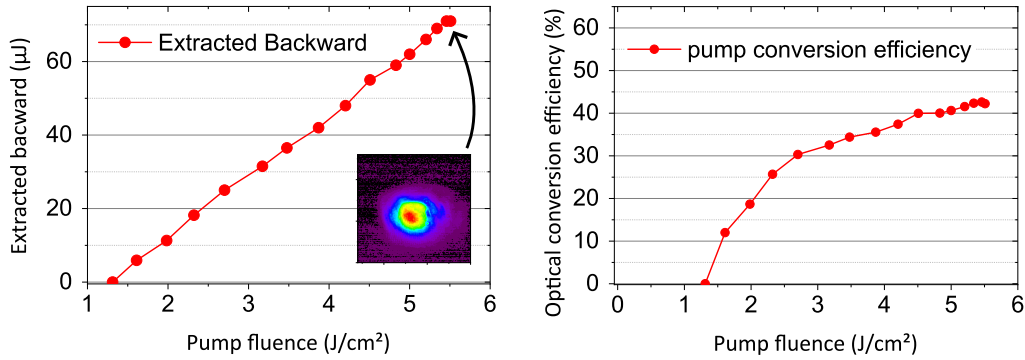


Figure 2. Backward-wave output energy at 2051 nm and optical conversion efficiency (pump toward signal + idler) extracted from crystal #2 (23 mm-long). The values are not corrected from Fresnel losses from the unpolished crystal facets and filtering optics. In inset is the beam profile at maximum power viewed with a Pyrocam IV.

With this set-up, both crystals exhibit similar output performances characteristics. Fig. 2 illustrates crystal #2 characteristics. The BWOPO threshold is around 100 μJ , and for the maximal pumping rate (3.4 times the oscillation threshold), the extracted backward wave output energy was typically 70 μJ corresponding to a total pump conversion efficiency superior to 40 %. These power characteristics are slightly lower than those reported by Mølster *et al*² and Liljestrand *et al*,¹³ which may be attributed to imperfect duty cycle in the crystal z-axis direction. As mentioned earlier, imperfect duty cycle is commonly noticeable for ultra-short poling periods (see the paper from Mutter *et al*¹⁸) for example.

2.3 Heterodyne single shot spectra and plane-wave modeling

Using the optical heterodyne wave mixing detection system described in Fig. 1, we can measure on a pulse-to-pulse basis the spectrum of the backward wave. Although the plane-wave theory in perfect phase-matching conditions predicts that the BWOPO should produce Fourier-transform-limited backward and forward pulses when pumped by a single-frequency pulse,⁸ we found that the spectra present an Airy-type shape that expands further for higher pumping rates up to 1.5 GHz, with side-lobes separated by a couple of hundred MHz. As

illustrated in Fig. 3, these features are observed for both crystals #1, and #2, but at varying degrees, depending on the crystal and the pumping rate. This asymmetric spectral broadening and structuring can be interpreted as stemming from a varying phase-matching condition distributed in the propagation direction of the nonlinear crystal, inducing phase variations during the BWOPO pulse that are directly dependent on the pump pulse intensity variations over time.^{17,19} For instance, numerical simulations incorporating a linear phase-mismatch with a linear chirp of $\delta k = 46 \text{ m}^{-1}/\text{mm}$, and a gaussian-shaped pump pulse, qualitatively reproduces the experimental asymmetric broadening and side lobes structuring observed for crystal #1. Such a chirped PM condition would correspond to a relative refractive index gradient of $7.9 \cdot 10^{-6}/\text{mm}$ for the pump wave in the nonlinear crystal, or 35 pm deviation in the poling period over the 15 mm length of the crystal. As illustrated in Fig. 3, crystal #2 presents an even smaller spectral broadening. In this case simulations suggest that the phase-mismatch chirp would be in the order of $\delta k = 9 \text{ m}^{-1}/\text{mm}$. These values are extremely small compared to either the refractive index of KTP or to the QPM poling period, and may be due to a combination of numerous reasons such as for example internal stress, rubidium doping inhomogeneity, or imperfect alignment of the crystal during the UV-photolithography process to produce the QPM poling. Sorting out, and ultimately controlling these different potential origins, thus appear extremely challenging. In part 3 we propose an optical solution based on pump pulse shaping that is expected to circumvent the issue for narrow-linewidth BWOPO operation.

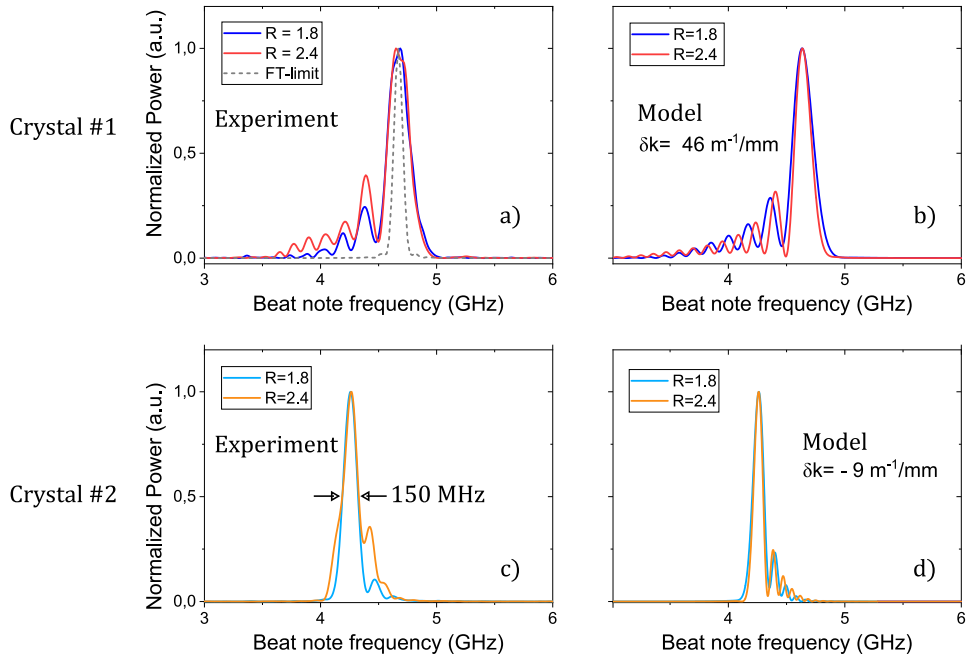


Figure 3. a) Normalized experimental beat-note spectra for BWOPO #1, the gray dashed line gives the Fourier-Transform-limit (FT-limit) of the BWOPO pulse in the case of a pumping rate of 2.4. b) Plane-wave simulation of BWOPO #1 with a linear phase-mismatch coefficient $\delta k = 46 \text{ m}^{-1}/\text{mm}$, and a gaussian pulse shape (10 ns at FWHM). c) Normalized experimental beat-note spectra for BWOPO #2, and d) plane-wave simulation with a linear phase-mismatch coefficient $\delta k = 9 \text{ m}^{-1}/\text{mm}$. In all cases, and for visualization purposes, the local oscillator frequency (or the zero-frequency relative position in the simulation) are chosen so that the peaks of the different spectra overlap at the same position.

2.4 Low-pressure CO₂ gas-cell transmission experiment

As illustrated in Fig. 3 the backward wave out of crystal #2 presents a slightly narrower spectrum despite some residual broadening and side-lobes modulation, with a full linewidth at half-maximum of 150 MHz. To push further the demonstration of the BWOPO potential for spectroscopy applications we thus used this backward

wave to measure the transmission of a fibered 15 Torr CO₂ gas-cell. The experimental set-up is schematically described in Fig. 1. The backward wave radiation is injected in a single mode fiber coupler separating two paths: one through the gas cell, and the other one through a delay line for normalization. The two paths are then recombined on the same photo-diode. The wavelength is tuned by slightly shifting the crystal temperature over a couple of °C in order to sweep through a few GHz, with a temperature tuning coefficient of -2.28 GHz/°C. Fig. 4 shows the transmission spectrum of one of the CO₂ absorption lines present in the 2051 nm cluster with an expected linewidth of 330 MHz. The points are single shot measurements. A slight broadening of the transmission line is observed due to the residual spectral broadening of the backward wave itself. This result is nonetheless an improvement with respect to previous spectroscopy experiment carried out with sub-ns BWOPO pumped by multi-mode laser sources,³ and bring promising perspectives for IPDA applications.

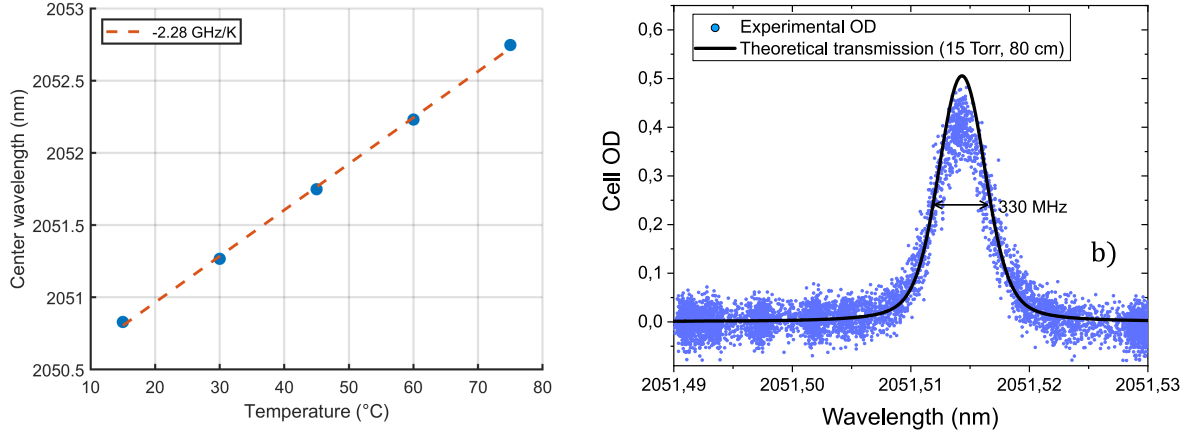


Figure 4. a) Temperature tuning characterization of the backward wave. tuning slope is -2.28 GHz/°C. b) Low-pressure (15 Torr) CO₂ gas cell transmission measurement experiment with BWOPO radiation from crystal #2 pumped at 2.4 times the oscillation threshold. Blue points are raw single shot measurements. The wavelength is tuned by sweeping the temperature over ~2°C. The black line is the theoretical expected transmission.

3. PUMP PULSE SHAPING FOR NARROW-LINEWIDTH BWOPO OPERATION

3.1 BWOPO pumped by top-hat shaped pulses modeling

To further improve the spectral linewidth from nanosecond BWOPO in the prospect of space-borne applications, we have suggested to use pump pulses with a square shape in order to limit the effect of a distributed phase-mismatch inside the nonlinear crystal,^{17,19} by systematically addressing the same local phase-matching condition for the backward parametric process. To illustrate this effect, numerical simulation with the same parameters used in Fig. 3b) are shown in Fig. 5. These simulation show that the BWOPO pulse phase is such that the instantaneous frequency tends to follow the BWOPO pulse shape in the case of a chirped phase matching condition. Consequently, for a gaussian pump pulse, identical frequencies are swept during the rising and falling edges of the pulses, which results in the characteristic side-lobes structure, which can be interpreted as the spectral beating of identical optical frequencies shifted in time by a duration equivalent to the pulse duration. Conversely, with a top-hat shape, most of the time is spent at a constant intensity, and thus at constant frequency, which results in a much narrower pulse spectrum close to its Fourier-transform limit.

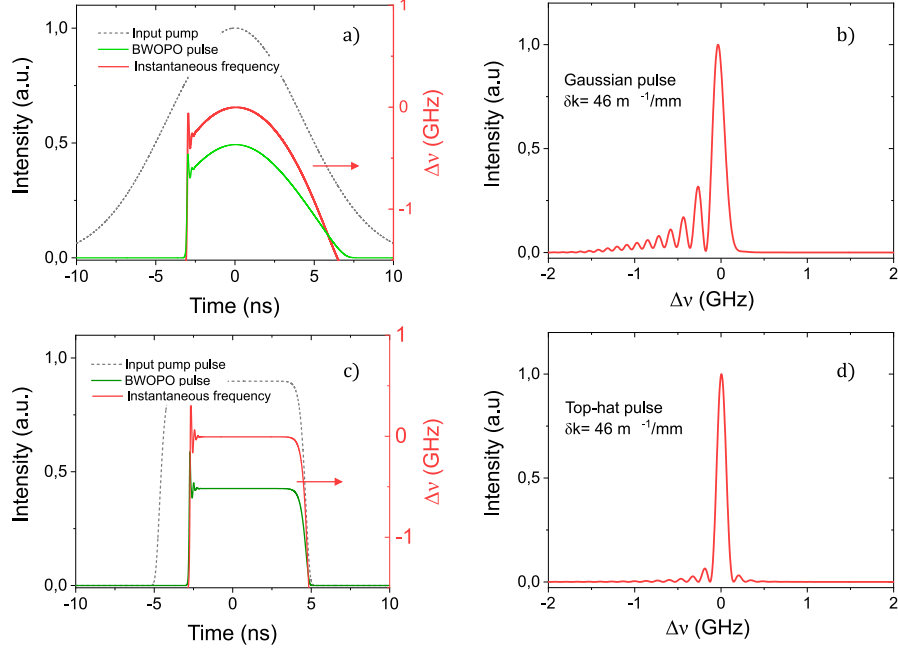


Figure 5. a) Simulated BWOP temporal pulse shape and instantaneous frequency shift defined as $\nu_{\text{instant}} = \frac{1}{2\pi} \frac{d\phi(t)}{dt}$, where $\phi(t)$ is the local phase shift of the backward wave, for an initial gaussian pump pulse shape. b) Corresponding spectrum. c) Simulated BWOP temporal pulse shape and instantaneous frequency shift in the case of a top-hat shaped pump pulse. d) Corresponding spectrum. In all cases a linear phase mismatch distribution $\delta k = 46 \text{ m}^{-1}/\text{mm}$ is modeled. The pump pulse shapes are chosen with 10 ns full width at half-maximum, and the peak powers are chosen so that the pumping rates are identically equal to 2.4. The top-hat profile is simulated with a high order super-gaussian shape. The frequency scales are arbitrary chosen so that the zero frequency is located at the peak of the spectrum

3.2 Preliminary experimental results

In the following section we show preliminary results obtained on a nanosecond BWOP pumped by an Ytterbium hybrid laser source with pulse-shaping capabilities, that we previously used to demonstrate BWOP pump tuning.^{1,2} The experimental set-up used is very similar to the one illustrated in Fig. 1 except that the pump beam is at 1030 nm, with a diameter of 190 μm in the Rb:PPKTP crystal, and a pulse duration of 21 ns. The BWOP crystal is 22 mm long, and the QPM polig period is 755 nm, which brings BWOP wavelengths at 1645.5 nm for the forward wave (useful for CH_4 detection from space), and 2754 nm for the backward wave. In the Yb-doped fiber amplifier stage, the pulses shapes are actively carved with acousto-optic modulators (AOM) that may be electronically controlled to pre-compensate gain saturation and thus flatten the pump pulse shape. The experimental results obtained with this technique are shown in Fig. 6, where the BWOP properties are characterized for two different pump pulse shapes. The power characteristics are rather similar for either shapes, and at maximal pumping rate the extracted energy at 1645 nm is $> 100 \mu\text{J}$. We see from the recorded pulse shapes that the rising and falling edges of the pump are mostly unchanged, the main effect of AOM pre-compensation being homogenization and flattening of the peak. The effect on the heterodyne spectrum is nevertheless quite significant, with a large reduction of the relative intensity of the side-lobes as well as narrowing of the central peak from approximately 180 MHz down to 80 MHz. These results are confirming the interest of pump pulse shaping for BWOP spectral narrowing purposes, and future improvement may be envisioned by including electro-optic modulation (EOM) with high bandwidth waveform generation in the fiber lase amplifier, in order to steepen the rising and falling edges of the pulse shape, and thus further reduce the amplitude of the side-lobes.

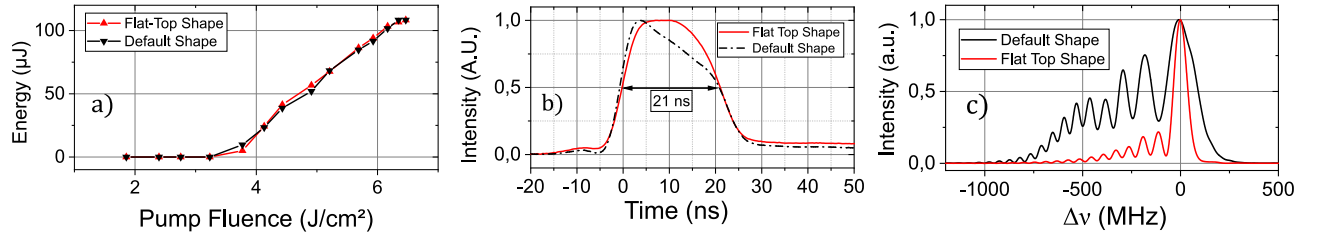


Figure 6. a) Power characteristic of the BWOPO at 1645 nm pumped by a hybrid Yb-based laser amplifier. The fluence is estimated for 21 ns pulse duration and a beam diameter of 190 μm in the nonlinear crystal. b) Pump pulse time shapes with (red) and without (black) AOM pre-compensation. And c) BWOPO heterodyne spectra of the forward wave at 1645 nm with and without AOM pre-compensation. Pump pulses and BWOPO spectra are recorded at maximal pump fluence, corresponding to a pumping rate of 2.

4. CONCLUSION

In summary, BWOPOs are extremely promising parametric sources that may be used in straightforward laser architectures to efficiently generate a wide range of useful wavelengths in the mid-infrared for IPDA applications. However, the challenging nature of the fabrication process and the sensitivity to small inhomogeneities in Rb:PPKTP crystals with sub-micrometric QPM period, might result in the production of a broadened and structured spectrum spanning up to a few GHz when pumped with single-frequency gaussian nanosecond pulses. This broadening might be mitigated, e.g. by reducing the pumping rate, and we could demonstrate the potential of the source for high resolution spectroscopy with a low-pressure gas-cell transmission measurement experiment. To further improve the BWOPO spectrum in the nanosecond regime, it is proposed to used top-hat pump pulse shapes in order to increase the time spent at a given frequency. Preliminary experiments based on AOM modulation and gain pre-compensation in the fiber stages of a hybrid laser amplifier suggest that the spectral purity can indeed be improved with this method. Future work will consist in incorporating electro-optic modulation in order to further sharpen the rising and falling edges of the pump pulses with faster modulation, and thus further enhance the spectral purity.

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