250-MHz synchronously pumped optical parametric oscillator at 2.25-2.6 μm and 4.1-4.9 μm

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Abstract: A compact and versatile femtosecond mid-IR source is presented, based on an optical parametric oscillator (OPO) synchronously pumped by a commercial 250-MHz Er:fiber laser. The mid-IR spectrum can be tuned in the range 2.25-2.6 μm (signal) and 4.1-4.9 μm (idler), with average power from 20 to 60 mW. At 2.5 μm a minimum pulse duration of 110 fs and a power of 40 mW have been obtained. Active stabilization of the OPO cavity length has been achieved in the whole tuning range.

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1. Introduction

The near-mid infrared spectral region from 2 to 10 \( \mu \text{m} \) is of primary importance in molecular spectroscopy, due to the strong absorption provided by the fundamental roto-vibrational bands and first overtones of many molecules. In this context, the availability of stable and accurate probes in the mid-IR spectral window is greatly desirable for a wide range of applications, including air quality and environment monitoring, industrial processes control, human breath analysis and detection of biologically hazardous or explosive materials. State of the art performances have been reached by exploiting phase-stabilized mid-IR femtosecond comb sources in direct multiplex spectroscopy experiments, where high-sensitivity and parallel detection of multiple molecular species have been simultaneously demonstrated [1]. Different approaches have been used to extend frequency combs in the mid-infrared. Recently, a femtosecond Tm:fiber source has been proposed to cover the range 1-2.5 \( \mu \text{m} \) with a power per mode below 10 nW exploiting nonlinear broadening in a silica fiber [2]. Mode-locked (ML) Cr:ZnSe lasers are good candidates in the region from 2.2 to 2.8 \( \mu \text{m} \), but their emission is presently limited to a 70-nm band around 2.4 \( \mu \text{m} \) [3]. Various systems based either on difference frequency generation [4, 5] or on OPOs [6–10] have been also reported at wavelengths above 3 \( \mu \text{m} \), with power levels as high as 1.5 W [6] and ultrawide spectral coverage over the range 2.6-6.1 \( \mu \text{m} \) [7]. However, no mid-IR fiber-pumped synthesizer has been demonstrated so far at repetition frequencies higher than 250 MHz, which would be very useful for direct comb spectroscopy techniques and, in particular, for dual-comb spectroscopy [11–13].

In this paper, we describe a robust OPO synchronously pumped by a commercial Er:fiber femtosecond laser, operating at a repetition frequency of 250 MHz. The OPO signal and idler have been tuned in the range 2.25-2.6 \( \mu \text{m} \) and 4.1-4.9 \( \mu \text{m} \), respectively, with average powers from 20 to 70 mW; this corresponds to a power per mode above 1 \( \mu \text{W} \) at all wavelengths, i.e. above the minimum power level generally required in gas trace detection experiments, which makes this system especially suitable to cover the lack of frequency comb sources for molecular spectroscopy in the region from 2.2 to 2.6 \( \mu \text{m} \). In a previous work based on a similar OPO system, power levels of \( \sim \)30 mW in the 3.7-4.7 \( \mu \text{m} \) spectral range were obtained at a repetition frequency of 90 MHz [14]. Here, we report on a completely different design of the OPO cavity, exploiting a ring configuration to achieve unidirectional operation and thus to minimize the losses. Particular attention has been devoted to the optimization of the OPO signal at 2.25-2.6 \( \mu \text{m} \), where we were able to obtain pulses with a minimum duration of 110 fs at 2.5 \( \mu \text{m} \) and an average power of \( \sim \)40 mW. In addition, long term intensity stabilization of the signal in the whole tunability range from 2.25 to 2.6 \( \mu \text{m} \) has been achieved by locking the OPO resonator length, which makes this system suitable for direct comb spectroscopy and ultrafast spectroscopy applications in the mid-IR. Once aligned, our OPO system is very robust and guarantees hands-free operation for hours.

2. OPO experimental setup

The scheme of the OPO cavity, pump system and locking electronics is shown in Fig. 1. The pump source is a 250-MHz ML Er:fiber laser (MenloSystems, FC1500) with output power of
580 mW, pulse duration of 40 fs, and vertical linear polarization on the fundamental mode. The pump beam is first collimated by an anti-reflection (AR) coated lens with focal length of 500 mm and then focused onto a periodically poled 5% doped MgO:LiNbO$_3$ (PPLN) crystal by an AR coated lens with focal length of 75 mm. The calculated pump beam waist inside the PPLN crystal is $24 \mu m$. The OPO cavity is constituted by four mirrors in a ring configuration with an unfolded length of $\sim 1.2$ m: a curved mirror with radius of curvature (ROC) of 50 mm and a dichroic coating (AR at 1.55 $\mu m$, high-reflection (HR)$ > 99.8\%$ at 2-3 $\mu m$), a curved mirror with ROC of 75 mm and a dichroic coating for extraction of the idler (AR at 4-5 $\mu m$, HR at 2-3 $\mu m$, YAG substrate), a plane HR mirror and an output coupler (R = 99%) for the 2-3 $\mu m$ range. The calculated mode radius at the intracavity focus (signal wavelength) in the PPLN crystal is $\sim 27 \mu m$, with good matching to the pump beam. The 5-mm long PPLN crystal, located at the intracavity focus of the signal mode, has six different poling period from 29.5 to 34.5 $\mu m$ with steps of 1 $\mu m$, and is AR coated with R < $4\%$ at the pump, signal, and idler wavelengths.

Fig. 2. (a) Spectra of the OPO signal and idler as acquired with poling period of 29.5 $\mu m$ (violet), 30.5 $\mu m$ (red), 31.5 $\mu m$ (green), 32.5 $\mu m$ (blue). (b) Pulse duration (filled circles) and power level (open circles) of the OPO signal and idler. (c) Autocorrelation of the 110-fs OPO signal pulses for a poling period of 32.5 $\mu m$. 

Fig. 1. Scheme of the OPO cavity and setup for offset frequency locking.
Table 1. OPO performance at room-temperature.

| Poling period (μm) | Threshold pump power (mW) | OPO wavelength signal (μm) | Output power signal (mW) | Idler wavelength (μm) | Idler output power (mW) | Pulsewidth signal (fs) | Pulsewidth idler (fs) |
|-------------------|---------------------------|---------------------------|-------------------------|----------------------|-------------------------|-----------------------|-----------------------|
| 29.5              | 280                       | 2.35                      | 20                      | 20                   | 295                     | 400                   |
| 30.5              | 160                       | 2.35                      | 24                      | 22                   | 163                     | 510                   |
| 31.5              | 165                       | 2.43                      | 60                      | 59                   | 133                     | 492                   |
| 32.5              | 155                       | 2.48                      | 37                      | 50                   | 110                     | 470                   |

The PPLN crystal is mounted inside an oven to allow for a fine tuning of the OPO emission frequency; for brevity temperature tuning curves are not reported in this paper, and experimental data refer to room temperature. The combination of different poling periods with temperature tuning allows for a stepwise coverage of the OPO peak wavelengths (continuous tuning needs a crystal with fan out grating [6]). The signal and idler spectra, acquired using a scanning monochromator with a resolution bandwidth (RBW) of 0.5 nm and a PbSe photodiode, are reported in Fig. 2(a) for the poling periods available from 29.5 to 32.5 μm. The corresponding signal and idler wavelengths cover the range 2.25-2.6 μm and 4.1-4.9 μm, respectively, with a maximum spectral width of 100 nm at 2.48 μm. Due to strong water vapor resonances in the 2.55-2.7 μm range, it was not possible to drive the OPO above threshold with the remaining poling periods. Figure 2(b) shows the average power and pulse duration of the signal and idler as a function of the central wavelength for each spectrum. The power levels are in the range 20-60 mW, with a maximum of 60 mW for both signal and idler when using the 31.5 μm poling period. The pulse durations have been measured by an interferometric autocorrelator based on collinear second harmonic generation in a LiIO3 crystal. Figure 2(c) shows the autocorrelation trace of the signal pulse train with the minimum duration of 110 fs (40-mW power), as retrieved by a fit assuming a sech^2-pulse shape, corresponding to a time-bandwidth product of 0.46. The reduction of the signal bandwidth when changing the poling period is due to the decrease of the phase-matching acceptance bandwidth and to the reduced transmission of the PPLN crystal (cut-off at ~5 μm), which introduces increasing losses on the idler. This is also confirmed by the trend of the power levels, with the exception of the data corresponding to the 32.5 μm poling, where the reported values are affected by the water vapor absorption at 2.55 μm. Table 1 summarizes the OPO performance.

3. OPO intensity stabilization

The long-term stabilization of the signal intensity has been achieved by exploiting the wide spectrum of nonlinear mixing signals generated by non phase-matched interactions inside the PPLN crystal with a technique similar to that described in [6]. Figures 3(a) and 3(b) show the nonlinear tones extracted from the HR plane mirror of the OPO cavity in the visible and near infrared spectral regions, respectively. Each tone has been labelled according to the specific nonlinear process involved (e.g. p + i represents the sum frequency between the pump and idler comb). The beam is dispersed using a fused silica prism and focused onto a 125-MHz InGaAs photodiode after proper spectral filtering by a slit. The spectral overlap around 1200 nm between the components p + i and 2s gives rise to a beatnote at \( f_0^p + f_0^s - 2f_0^0 = 2f_0^p - 3f_0^0 \), where \( f_0^p \), \( f_0^s \) and \( f_0^0 \) are the offset frequency of the pump, signal and idler, respectively. The same beat frequency has been observed around 600 and 800 nm, originating from the overlap of \( 2p + s \) with \( 4s \), and \( 2p \) with \( 3s \), respectively. A phase-lock circuit was implemented to stabilize the beatnote against a 15-MHz reference derived from a frequency synthesizer. The signal at the
output of the InGaAs photodiode has been suitably amplified (∼30 dB) to match the input dynamic of the phase detector, and then sent to a proportional-integral servo acting on a piezo mounted cavity mirror. The signal radiation is constituted by frequencies \( \nu = k v/L = k f_r + f_0 \), where \( k \) is the number of wavelengths within the cavity, \( v \) is the mean phase velocity, \( L \) is the cavity length, and \( f_r \) is the repetition frequency. Since the OPO signal is inherently synchronous with the pump, a change in the cavity length at constant \( f_r \) induces only a change of \( f_0 \) [15]. Therefore, phase-locking of this beat note translates into an OPO cavity length control, providing long-term stabilization of the OPO signal intensity.

Figure 4 shows the locked beatnote as measured with an RF spectrum analyzer. The observed FWHM of 38 kHz is consistent with the free-running width of the pump offset frequency \( f_0 \), which is not controlled in this experiment. The beatnote in Fig. 4 has been acquired using the 32.5 μm poling period. Similar results in terms of FWHM with slightly reduced peak values
Fig. 5. (a) RIN (left) and cumulative standard deviation (right) of the locked OPO and Er-fiber pump laser. (b) Signal intensity versus observation time for unlocked (0–30 min) and locked OPO (30–60 min).

have been obtained for all remaining periods, due to the good spectral overlap between the $p + i$ and $2s$ nonlinear tones. Figure 5(a) shows the relative intensity noise (RIN) of the signal, as recorded by an extended-InGaAs photodiode with 10-MHz bandwidth, when the OPO cavity length is locked. For comparison, the RIN of the Er-fiber pump oscillator is also shown. For Fourier frequencies lower than 20 Hz and larger than 1 kHz an excess noise in the OPO RIN is observed, due to conversion of the pump offset frequency phase-noise into OPO amplitude noise [10, 16] and cavity vibrations. The cumulative standard deviation of the OPO intensity is as low as 0.2%, i.e. a factor of $\sim 2$ larger than the pump laser cumulative standard deviation of 0.1%. Long-term intensity stability of the OPO signal is also shown in Fig. 5(b) for 1 h observation time. The free running standard deviation of the OPO intensity is effectively reduced by a factor $\sim 5$ when the active stabilization loop of the OPO cavity length is closed.

4. Conclusion

In conclusion, a compact OPO source synchronously pumped by a commercial 250-MHz Er:fiber laser has been presented. The OPO signal and idler cover the spectral range 2.25-2.6 $\mu$m and 4.1-4.9 $\mu$m, respectively, with average powers from 20 to 60 mW. The minimum observed pulse duration is 110 fs at 2.5 $\mu$m, with a power level of 40 mW. In addition, long-term active stabilization of the signal intensity in the whole tunability range from 2.25 to 2.6 $\mu$m allows for the reduction of the intensity noise down to the 0.2% level. Further stabilization of the pump repetition and offset frequencies will lead to a full mid-IR comb source, which could be used for absolute referencing of solid-state Cr$^{2+}$ lasers and quantum cascade lasers, or for direct comb spectroscopy in the mid-IR spectral region.

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