Research Article

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High peak-power laser system tuneable from 8 to 10 μ**m**

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Abstract: A high peak-power rapidly tuneable laser system in the long-wave infrared is obtained using an external cavity quantum-cascade laser (EC-QCL) broadly tuneable from 8 to 10 μm and an optical parametric amplifier (OPA) based on quasi phase-matching in orientation-patterned gallium arsenide (OP-GaAs). To provide an efficient amplification, the nonlinear crystal is pumped by a pulsed fiber laser source. With a pump laser source tuneable around 2 μm, quasi phase-matching remains satisfied with a fixed grating period in the OP-GaAs crystal when the EC-QCL wavelength is swept from 8 to 10 μm. The OPA demonstrates parametric amplification from 8 to 10 μm and achieves output peak powers up to 140 W, with spectral linewidths below 3.5 cm $^{-1}$ and a beam profile quality (M²) below 3.4 in both horizontal and vertical directions.

Keywords: fiber optics amplifiers and oscillators; laser; parametric oscillators and amplifiers; parametric processes in nonlinear optics; quantum cascade lasers; tuneable lasers.

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1 Introduction

There is an increasing demand for laser sources in the 8 to 12-μm spectral range for stand-off detection of chemical traces. These applications require a fast and large spectral tuneability with a narrow linewidth to achieve spectral resolution of the components to identify $\left($ < 3 cm⁻¹ for typical chemicals deposited on surfaces) and both a good output beam quality and high peak power to achieve longrange detection. On one hand, narrow linewidth pulsed optical parametric oscillators (OPO) can achieve most of these targets but so far within complex systems with slow tuning means. A widely tuneable pulsed OPO based on orientation-patterned gallium arsenide (OP-GaAs) was demonstrated in Vodopyanov et al. [1] at the expense of a complex pump at 3 μm. An intracavity OPO based on zinc germanium phosphate medium (ZGP) was reported in Robertson et al. [2], but the wide tuneability was provided by angle tuning of the crystal. A narrow linewidth OP-GasAs-based OPO was reported in Clément et al. [3], but the crystal temperature tuning range remains limited and slowly addressed. On the other hand, external cavity quantum cascade lasers (EC-QCL) have demonstrated a very broad tuneability (\sim 300 cm⁻¹) with a good output beam quality and a narrow linewidth around 1 cm⁻¹ in the full tuning range [4]. These sources are well adapted except that at room temperature, the available short pulses of a few hundred-milliwatt peak power limit the stand-off detection range to a few meters [5–7]. We propose here to enhance the peak power of an EC-QCL tuneable from 8 to 10 μm, thanks to optical parametric amplification (OPA).

OPA requires a high peak-power pump source and a nonlinear medium-enabling frequency conversion from the pump to signal and idler waves. GaAs is one of the most interesting mid-IR nonlinear materials with a very large second-order nonlinear optical coefficient $d_{14} \sim 90$ pm/V near 10 μm [8]. An efficient nonlinear conversion can be obtained if the relative phases between interacting waves are controlled and maintained constant all along the propagation in the crystal. This phase-matching

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condition is usually achieved in birefringent nonlinear crystals by rotation, temperature tuning of the crystal, leading to slow and complex systems.

Quasi phase-matching (QPM) is an alternative technique where the sign of the nonlinear coefficient is periodically reversed to reset the phase relationship between the interacting waves. In the case of GaAs, the growth of orientation-patterned crystals (OP-GaAs) was achieved [8], and thanks to QPM, several optical parametric oscillators demonstrated an efficient conversion from the pump to the generated signal and idler waves [9, 10]. OPA based on OP-GaAs as a nonlinear crystal formerly showed high gain at a fixed wavelength of 4.5 μm while preserving the spectral and spatial properties of an amplified DFB-QCL [11]. Clement et al. [12] demonstrated the OPA of a singlefrequency continuous-wave QCL tuneable from 7.8 to 8.4 μm in ZGP, but again, the tuneability was achieved by angle tuning the nonlinear medium.

Phase-matching can also be realized with wavelength tuning of both pump and signal beams, avoiding any action on the nonlinear crystal [13–15]. In particular, Klein et al. [13] demonstrated a PPLN-based OPO whose wavelength is tuned from 3.16 to 3.5 μm within 330 μs with a fiber laser all-electronically tuneable through an acoustooptic tuneable filter (AOTF).

We present, here, a pulsed tuneable laser system in the long-wave infrared using an EC-QCL broadly tuneable from 8 to 10 μm and OPA without any moveable part. The OPA is based on OP-GaAs of a fixed period and at a fixed temperature, and a thulium-doped fiber pump system acousto-optically tuneable around 2 μm. In the OPA, QPM is satisfied over the whole tuning range of the EC-QCL by simultaneous wavelength tuning of the pump, thus, avoiding any action on the nonlinear crystal.

2 Experimental setups

2.1 Tuneable pulsed fiber pump system

The architecture of the tuneable pulsed fiber pump system is described in Figure 1. It is based on the fiber pump system previously described in Ref. [16]. The system was improved in this work using only polarization-maintaining fibers so that the output pulses have a linear polarization, independent of the emitted wavelength. The architecture is a tuneable Q-switched thulium-doped silica fiber laser oscillator followed by a fiber amplification stage. The laser and the amplifier are based on short lengths of highly-doped core-pumped active fibers to prevent nonlinear effects in the fibers.

The laser is based on a 44-cm long thulium-doped silica fiber with high-core dopant concentration (2.5% wt. T m_2O_3), with a core of diameter 6 μm and NA 0.22 that is single-mode above 1.75 μm and

Figure 1: Setup of the tuneable pulsed fiber pump system. PM, polarization-maintaining component; O.C., output coupler.

slightly multimode at the 1.54 μm pump wavelength. The Tm-doped fiber is core pumped at 1.54 μm through a WDM coupler fibered with a NUFERN PM1950 fiber to match the active fiber mode diameter. A home-made CW Er/Yb fiber laser up to 6 W was used as pump source. The other port of the WDM coupler is spliced to an equivalent length of passive fiber with a FC/APC connector before coupling and collimation to a free-space path. The free-space path allows an external cavity feedback comprising a lens of focal length 5 mm and NA ~ 0.4, an acousto-optic modulator (AOM) for cavity Q switching, an electronically controlled AOTF to tune the emitted wavelength with a line-width of 2 nm (FWHM) and a broadband highly reflective mirror to close the cavity on the AOTF first diffraction order. At the output of the active fiber, a simple FC/PC connector provides 4% back reflection to close the cavity and forms the output coupler. This connectorized fiber was chosen with a core diameter of 10 μm and NA 0.10 to limit spectral broadening at the laser output and to match the input fiber of the amplifier. The optical cavity in the external cavity feedback is aligned on the first diffraction order of the AOM to reduce amplified spontaneous emission (ASE) generated at low repetition rates between laser pulses and to have shorter pulse widths [16]. In this work, the WDM coupler is placed opposite the output coupler to reduce ASE at the laser output. Fiber lengths were reduced as much as possible between components, and the total cavity length is 4.6 m. The laser output is coupled to the amplifier input fiber by means of two collimating lenses of identical focal lengths (5 mm).

The acousto-optic tunable filter (AOTF) enables a wavelength selection with a delay ultimately limited by the establishment of the acoustic wave over the diameter of the beam to diffract. The beam diameter of 1 mm in the free-space path let us expect a response time of 1 μs. Tuneability is provided by tuning the RF frequency in the range of tens of MHz from a PC-controlled driver. Such AOTFs demonstrated rapid tuning with a programmable digital synthesizer [13].

The amplifier comprises a WDM coupler, a 30-cm-long Tmdoped fiber (estimated doping \sim 1.4% wt. Tm₂O₃) with a 25-µm core diameter and 0.09 NA as an active fiber and a polarization-sensitive isolator inserted before the WDM to avoid emission from the amplification stage to be coupled back into the laser. The WDM coupler and the isolator are fibered with the 10-μm core diameter fiber chosen for the output coupler of the laser. A mode field Adaptor is inserted

between the output of the WDM coupler and this second Tm-doped fiber to preserve single-mode propagation of the laser signal when passing from the passive fiber of the 10-μm core diameter to the active fiber of 25-μm core diameter. This larger core active fiber is pumped by a second home-made CW Er/Yb fiber laser source delivering up to 12 W. The active fiber output is spliced to an equivalent length of corresponding passive fiber with an angled FC/APC output connector to avoid back reflection and to prevent the amplifier from lasing.

The laser oscillator provides stable Q-switch operation for any AOM repetition rates between 1 kHz and 50 kHz. Characterizations were performed at the optimum repetition rate of 2 kHz that was found for the previous fiber pump system [16]. Pumping conditions (i.e. launched power) can be found so that a stable Q-switch operation is obtained over a 100-nm tuning range. Increasing the pump power makes ASE develop so that it finally clamps the gain available for the tuneable Q-switch pulse and reduces the tuning range. A tradeoff is chosen between pump rate (pulse energy) and tuning range. Pulse energies were measured with an energy meter. Their temporal profile was observed on a 1-GHz bandwidth oscilloscope with a fast-response photodetector of spectral range 830–2100 nm and rise time/fall time below 50 ps (eotech ET-5010F). The exact temporal profiles of the pulses were exploited to estimate the emitted peak power from the signal amplitude and area of the fast-response photodetector and independently of the pulse shape and width. For a launched power of 1.2 W, the laser is tuneable from 1880 to 1980 nm with pulse energy in the 8–17 μJ range. Pulse widths are measured in the 23 to 37-ns range, and output peak powers range between 300 and 700 W (see Figure 2A). Spectral measurements were performed with a resolution of 0.05 nm. The spectra of the output pulses have a linewidth below 0.7 nm (FWHM) over the whole tuning range after further propagation in 1 m of passive fiber. ASE building up between output pulses could be evaluated at a level 40 dB below the laser one (Figure 2B). The output polarization is linear with an extinction ratio larger than 10 dB.

Further amplification is required as the output peak power achieved is not large enough to provide an efficient parametric gain in nonlinear crystals. With the laser operated at 2 kHz repetition rate, the amplifier delivers pulses tuneable from 1880 to 1980 nm with pulse widths between 23 and 34 ns. Output energy scales up to 200 μJ when operated with a pump power up to 9 W in the amplifier.

This maximum launched pump power was limited by the damage threshold of the antireflection coating on the nonlinear crystal. At the amplifier output, 15% of the output energy appears to come from the cladding, thus, it cannot be properly focused and contributes to losses. Nevertheless, after filtering these losses with a diaphragm, the amplifier still provides peak powers between 3 and 6.7 kW depending on the wavelength (see Figure 3A). The amplified pulses have a spectral linewidth of 0.4 to 0.9 nm at -3 dB (FWHM) and below 2.4 nm at − 10 dB over the whole tuning range (see example in Figure 3B). The amplifier output beam is linearly polarized (ellipticity 0.7–0.8), with a small \pm ^{5°} variation of the polarization axis depending on the wavelength.

The beam quality is measured at the output of the amplifier for 1.2-W launched power in the oscillator operated at 2-kHz repetition rate and 9-W pump power in the amplifier. Transverse profile characterizations were performed with a pyro-electric detector behind a rotating slit (NanoScan) after collimation with an achromatic reflective mirror of focal length 7 mm and focusing with a lens of focal length 60 mm. The beam remains nearly diffraction limited with M^2 <1.4 both in the horizontal and vertical directions. An important figure for the application is the fraction of pulse peak power comprised in the 2-nm pump acceptance bandwidth targeted. A factor of comparison is estimated as follows: the average pulse power integrated over a 2-nm span centered on the peak is compared to the overall integral across the entire range. We thus estimate that 70% to 90% of amplified output power stands in a 2-nm bandwidth (corresponding to the spectral acceptance bandwidth of the OPA), making this tuneable fiber pump suited to pump an OPA based on OP-GaAs.

2.2 QCL OPA setup and results

The tuneable fiber laser system detailed above is employed to pump our OPA setup described in Figure 4.

The OPA is seeded with an EC-QCL [1] based on a two-stack heterocascading-design chip. The EC-QCL emission is tuneable from 8 to 10.2 μm within 1 ms by means of an intra-cavity grating in a Littrow configuration mounted on a fast rotary stage. It delivers pulses of 200-ns pulse width and peak power from 100 to 350 mW

Figure 2: Output characteristics of the laser oscillator at 2 kHz repetition rate with 1.2 W launched power. (A) Laser peak power and pulse width vs. emitted wavelength. (B) Output spectrum for laser tuned at 1933 nm.

Figure 3: Output characteristics of the amplifier with laser pumped with 1.2 W, 2 kHz repetition rate, and amplifier pumped with 9 W. (A) Peak power and pulse width vs. emitted wavelength. (B) Spectrum for laser tuned at 1933 nm.

Figure 4: Description of the EC-QCL and OPA setup. λ/2, zero-order half-wave plate; D, diaphragm; DM, dichroic mirror; LP, long-wave pass filter above 6600 nm; blue arrow; pump beam; red arrow, EC-QCL beam; green arrow, complementary signal generated in the OPA of wavelength between 2.3 and 2.6 μm.

at a maximum repetition rate of 200 kHz. At the output of the QCL chip, a high NA lens provides a collimated output beam with a quasidiffraction-limited profile (M^2 <1.4). Spectral linewidths are below 1 cm − 1 (FWHM) on the whole tuning range.

The OPA is based on an OP-GaAs nonlinear crystal of 66 μm period grating, 500 μm thickness (along the [001] crystallographic axis), 2 mm wide (along $[-110]$) and 32 mm length (along [110], the beam-propagation direction). The crystal temperature was adjusted to 97°C to provide quasi phase-matching from 8 to 10 μm with a pump tuneable from 1880 to 1980 nm. Its facets were antireflection coated at the pump, EC-QCL, and complementary signal wavelength that is generated between 2.3 and 2.6 μm through parametric interaction in the crystal. The EC-QCL was linearly polarized along the [− 110] direction.

At the fiber amplifier output, the pump beam is collimated with an achromatic reflective mirror of focal length 7 mm and NA = 0.4. A zero-order half-wave plate enables to align the polarization axis of the pump at all wavelengths along the [111] crystallographic axis of the OP-GaAs crystal and, thus, maximize the parametric gain in the OPA.

Both the fiber pump beam and the EC-QCL beam are superposed with a dichroic mirror (DM) specifically designed to provide a high transmission on the EC-QCL bandwidth $(T > 80\%$ from 7.5 to 10 μ m) and a high reflexion over a bandwidth covering the pump tuning range (R > 99% at 1850–2000 nm). The beams are focused to overlap

with respective diameters at $1/e^2$ of 190 and 200 μ m at the center of the OP-GaAs crystal. The crystal is tilted in the horizontal plane compared with the beam's direction. A fraction of the EC-QCL emission is reflected by the dichroic mirror and provides a port to control the EC-QCL pulse. At the OPA output, an identical dichroic mirror (DM) reflects the residual pump but partially transmits the complementary signal generated in the OPA of wavelength between 2.3 and 2.6 μm. A long-wavelength pass filter (Spectrogon LP6600) with a transmission of 85% above 6600 nm further discards the complementary signal generated in the OPA of a wavelength between 2.3 and 2.6 μm.

Both the EC-QCL and the fiber pump system are operated at a repetition rate of 2 kHz and at their maximum set points. With proper pulse synchronization, tuning the pump wavelength from 1980 to 1887 nm allows parametric amplification of EC-QCL pulses from 8 to 10 μm. Taking into account the tuning properties of the EC-QCL and the still faster characteristics of the AOTF-based fiber pump, the 2-kHz repetition rate of the system is compatible with a wavelength change between each pulse, anywhere in its tuning range. Temporal profiles of the incident EC-QCL pulse, the pump laser pulse, and the amplified EC-QCL pulse are shown in Figure 5.

The amplified pulse energy remains too limited to be measured with an energy meter and is, thus, estimated with the output average power and the known 2-kHz repetition rate. The exact temporal profile of the pulses is exploited to estimate the emitted peak power from the signal amplitude and area of a photodetector of bandwidth 250 MHz in the range 8 to 12 μm (VIGO) and independently of the pulse shape and width. The OPA delivers amplified pulses of 10 ns, limited by the pump pulse width and peak power gain between 20 and 26 dB leading to peak powers from 20 to 140 W (see Figure 6A). Spectral measurements are performed with a monochromator (Chromex sm250) with a resolution of 4 nm and the fast-response photodetector whose signal was measured with a lock-in amplifier EG&G model 5210 (see example Figure 6B). Spectral broadening occurs as spectra present a linewidth between 2.5 and 3.5 cm − 1 (FWHM) over the full tuning range. This spectral broadening could come from the coupling observed between the OPA and EC-QCL as the EC-QCL pulse shape observed on the control port changes when the OPA is pumped with a wavelength satisfying QPM.

Beam profile measurements of the amplified EC-QCL were performed with a micro-bolometer camera of 17 μ m × 17 μ m pixel dimensions and 480×640 frame size. An image of the beam profile at the output of the crystal is given in the inset of Figure 6B. The output

Figure 5: Temporal pulse profiles. (A) EC-QCL pulse at 1060 cm − 1 recorded with a photodetector of spectral range 8–12 μm and bandwidth 1 GHz. (B) Pump pulse at 1933 nm recorded with a photodetector of spectral range 830 to 2100 nm and rise time/fall time below 50 ps. (C) Amplified EC-QCL pulse at 1200 cm − 1 recorded with a photodetector of spectral range 8–12 μm and of bandwidth 250 MHz.

Figure 6: Output characteristics of OPA with EC-QCL at maximum output peak power, laser pumped with 1.2 W, amplifier pumped with 9 W, and EC-QCL and pump synchronized at 2 kHz. (A) Peak power and OPA gain vs. EC-QCL wavelength. (B) Spectrum of amplified pulses with EC-QCL tuned ~1144 cm⁻¹ (spectral linewidth FWHM of 20 nm~2.7 cm⁻¹). Inset shows an image of the beam profile at the output of the crystal.

beam presents a M^2 of 3 in the horizontal plane and an M^2 of 3.4 in the vertical plane.

Finally, starting with a QCL delivering pulses of 350 mW peak power, the OPA provides an optical gain from 20 to 26 dB. In the same conditions as reported in Hugger et al. [4] and given that the detection sensitivity varies with the inverse of the square of the distance, the standoff detection range could be increased by a factor of 10–20 to reach 15–30 m.

3 Conclusions

We demonstrated, for the first time in our knowledge, amplification of a tuneable EC-QCL in an OP-GaAs nonlinear crystal of fixed period with a tuneable fiber pump system. The fiber pump system comprises a tuneable Q-switched thulium-doped silica fiber laser followed by a fiber amplification stage delivering pulses tuneable from 1880 to 1980 nm with pulse widths between 23 and 34 ns and peak powers between 3 and 6.7 kW in a quasi-diffraction-limited beam of M^2 < 1.4 and spectral linewidth < 0.9 nm (FWHM).

Starting from an EC-QCL delivering pulses tuneable from 8 to 10 μm with peak power between 100 and 350 mW, an OP-GaAs crystal of 32-mm length, with a fixed 66-μm grating period, demonstrates parametric amplification over the full tuning range and achieves output peak powers up to 140 W. The output beam presents a M^2 of 3 in the horizontal plane and of 3.4 in the vertical plane. Spectral broadening occurs but output spectral linewidths remain below 3.5 cm⁻¹ (FWHM) making this source suitable for remote standoff detection of chemicals deposited on surfaces.

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