Femtosecond deep-infrared optical parametric oscillator pumped directly by a Ti:sapphire laser

Callum F. O'Donnell*[a, b], S. Chaitanya Kumar,[a, b], Kevin T. Zawilski,[c] Peter G. Schunemann,[c] and Majid Ebrahim-Zadeh[a, b, d]

[a]Radiantis, Polígon Camí Ral, 08850 Gavà, Barcelona, Spain; [b]ICFO-Institut de Ciencies Fotoniques, The Barcelona Institute of Science and Technology, 08860 Castelldefels, Barcelona, Spain; [c]BAE Systems, Incorporated, MER15-1813, P.O. Box 868, Nashua, New Hampshire 03061-0868, USA; [d]Institucio Catalana de Recerca i Estudis Avancats (ICREA), Passeig Lluis Companys 23, Barcelona 08010, Spain.

ABSTRACT

We report a high-repetition-rate femtosecond optical parametric oscillator (OPO) for the deep-infrared (deep-IR) based on the nonlinear optical crystal, $CdSiP_2$ (CSP), pumped directly by a Ti:sapphire laser, for the first time. By pumping CSP at <1 μm, we have achieved practical output powers at the longest wavelengths generated by any Ti:sapphire-pumped OPO. Using a combination of pump wavelength tuning, type-I critical phase-matching, and cavity delay tuning, we have generated continuously tunable radiation across 6654–8373 nm (1194–1503 cm^{-1}) at 80.5 MHz repetition rate, providing up to 20 mW of average power at 7314 nm and >7 mW beyond 8000 nm, with idler spectra exhibiting bandwidths of 140–180 nm across the tuning range. Moreover, the near-IR signal is tunable across 1127–1192 nm, providing up to 37 mW of average power at 1150 nm. Signal pulses, characterised using intensity autocorrelation, have durations of ~260–320 fs, with corresponding time-bandwidth product of $\Delta\upsilon\Delta\tau$~1. The idler and signal output exhibit a TEM_{00} spatial profile with single-peak Gaussian distribution. With an equivalent spectral brightness of ~6.68×10^{20} photons s^{-1} mm^{-2} sr^{-1} 0.1% BW^{-1}, this OPO represents a viable table-top alternative to synchrotron and supercontinuum sources for deep-IR applications in spectroscopy, metrology and medical diagnostics.

Keywords: Optical parametric oscillators, nonlinear optics, ultrafast optics, nonlinear materials, infrared sources.

1. INTRODUCTION

High power, high-repetition-rate femtosecond sources of coherent deep-infrared (deep-IR) radiation are in demand for ever increasing range of applications, from vibrational spectroscopy to medical imaging.[1] In particular, the spectral region of 7-8.5 μm is of great interest due to the presence of strong absorption lines corresponding to the amide III functional group, and various chemical pollutants.[2] The primary diagnostic technique used to analyse such samples is Fourier Transform Infrared Spectroscopy (FTIR), involving the illumination of a sample using a broadband IR source such as a globar. Although the spectral coverage is broad, the diffuse and incoherent nature of the light source places limitations on the achievable spatial resolution and signal-to-noise ratio.[3] In the absence of conventional solid-state lasers at these wavelengths, a variety of different approaches towards the development of high-power coherent mid-IR and deep-IR sources have been explored, including supercontinuum generation, synchrotron sources, and quantum cascade lasers (QCLs).[4-6] However, the use of these technologies is associated with compromises in spectral intensity, available beam time, and tunability, respectively. On the other hand, nonlinear parametric down-conversion techniques can exploit well-established near-IR solid-state and fibre laser technology to generate powerful coherent radiation far into the infrared.[7] Furthermore, the temporal qualities of the pump laser are transferred to the down-converted beams, enabling high-repetition-rate quasi-continuous-wave pulse trains with kW-level peak power. While commercial OPOs are capable of directly producing hundreds of milliwatts of average power up to and above ~4 μm in the mid-IR, efforts to extend their wavelength access into the deep-IR have been hampered by stringent nonlinear material requirements. The critical properties include high nonlinear coefficient, d_{eff}, resistance to damage at high optical intensities, high bulk

*Corresponding author: callum.odonnell@radiantis.com

transparency, and ability to be phase-matched at the desired pump, signal and idler wavelengths. The breakthrough finally came by the successful development of cadmium silicon phosphide, $CdSiP_2$ (CSP), a chalgocenide crystal which was first grown to sufficient optical quality in 2010.[8] With a nominal transparency across 1–7 μm, thermal conductivity of 13.6 W/m·K, and d_{eff} ~84 pm/V, it enables single-stage conversion from 1.06 μm to the mid-IR and deep-IR.[9] Moreover, its nonlinear figure-of-merit ($FOM=d_{eff}^2/n^3$) ranks among the highest of all existing nonlinear crystals. To date, ultrafast OPOs based on CSP have been operated at a fixed angle under noncritical phase-matching (NCPM), with spectral tuning achieved via cavity delay, or by varying the pump wavelength.[10-13] A summary of previous demonstrations of femtosecond OPOs based on CSP is presented in Table 1. In this work, we add another important milestone to this list, by demonstrating a deep-IR femtosecond OPO based on CSP, pumped directly at by a Kerr-lens-mode-locked (KLM) Ti:sapphire laser, for the first time. In doing so, we show that tunable deep-IR radiation can be generated in a single conversion stage using well-established ultrafast pump laser technology.

Table 1: Summary of previous demonstrations of femtosecond OPOs based on CSP.

Pump	Rep. rate (MHz)	Tuning method[a]	Idler (μm)[b]	Pump power (**W**) (wavelength (nm))	Idler power (**mW**) (wavelength (μm)[c]	Idler pulse duration (fs)	Ref.
Yb:KYW	100	-	6.2	**1.6** (1053)	-	-	10
Yb:KGW	43	C	6.8–7.0	**5** (1029)	**110** (7.0)	~500 (est.)	11
Intracavity MgO:PPLN OPO	76	C	5.9–8.1	**0.9** (1024)	**0.064** (6.8)	-	12
Commercial OPO	80	P, C	6.3–7.0	**1** (1015–1074)	**32** (6.8)	~200 (est.)	13
Ti:sapphire	80	P, A, C	6.7–8.4	**0.83** (993–1011)	**20** (7.3), **7** (8.1)	~300 (est.)	This work

[a]Tuning methods: C: Cavity delay tuning, P: Pump wavelength tuning, A: Angle tuning.
[b]Idler range defined relative to weighted centre of most extreme recorded spectra, not 10 dB limit.
[c]Peak idler power and power at selected wavelengths of interest.

2. RESULTS AND DISCUSSION

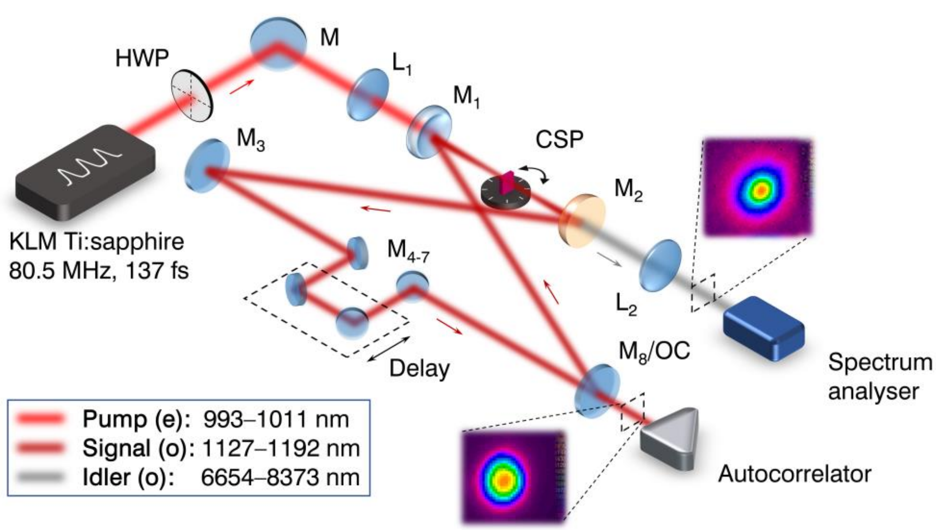

Figure 1. Schematic of the experimental setup for the Ti:sapphire-pumped CSP femtosecond OPO. HWP: Half wave plate, M: Mirrors, OC: Output coupler, L: Lenses, F: Filters.

The experimental setup for the CSP-based femtosecond OPO is depicted in Fig. 1. The OPO is synchronously pumped by $\Delta\tau$~137 fs input pulses from a KLM Ti:sapphire laser (Spectra-Physics, *Mai Tai HP*) at a repetition rate of 80.5 MHz. For this experiment, to conform with the chosen optical coatings, we operated at pump wavelengths across 993–1011 nm, where the measured FWHM spectral bandwidth was $\Delta\lambda$~10.5 nm, corresponding to a time-bandwidth product of $\Delta\upsilon\Delta\tau$~0.44, close to the transform limit for $sech^2$ pulses. After transmission through L_1 and M_1, the average pump power incident on the crystal is ~430–690 mW. The CSP sample is 1-mm-long with a 4 mm × 5 mm aperture, and cut at θ=90° (ϕ=45°) for type-I ($e\rightarrow oo$) NCPM at normal incidence. The crystal is maintained at room temperature and mounted on a precision rotation stage, with both end faces antireflection (AR)-coated for high transmission at the pump (T>99% over 950–1080 nm), signal (T>99% across 1100–1200 nm), and idler (T>93% across 7000–8500 nm). The pump polarisation is controlled using a half-wave plate (λ/2) and the beam is focused to a waist radius of w_0 ~28 µm at the centre of the crystal using a lens (L_1) of focal length, f=100 mm. The OPO cavity comprises two plano-concave mirrors ($M_{1\text{-}2}$) with radius of curvature, r=100 mm, and six plane mirrors ($M_{3\text{-}8}$). M_1 transmits the pump (T~83% at 993 nm to T~83% at 1011 nm) and is highly reflecting (HR) for the signal (R>99% across 1100–1300 nm). M_2 is a 3-mm-thick ZnSe substrate, AR-coated for the deep-IR idler (T>94% across 6500–9000 nm), and HR for the signal (R>97% across 1100–1300 nm). M_8 is a 5% signal output coupler (OC) (R=95% across 1100–1300 nm). All remaining mirrors ($M_{3\text{-}7}$) are fused silica substrates, HR for the signal (R>99.8% across 1100–1300 nm), and negatively chirped for dispersion compensation (GDD~-100 – -400 fs^2 across 1100–1160 nm). The OPO is thus singly-resonant for the near-IR signal wave, with the undepleted pump exiting the cavity through M_8, while the single-pass deep-IR idler is transmitted through M_2. Finally, after appropriate filtering, the idler is collimated by a CaF_2 lens with good deep-IR transmission (T>60% over 6000–8500 nm).

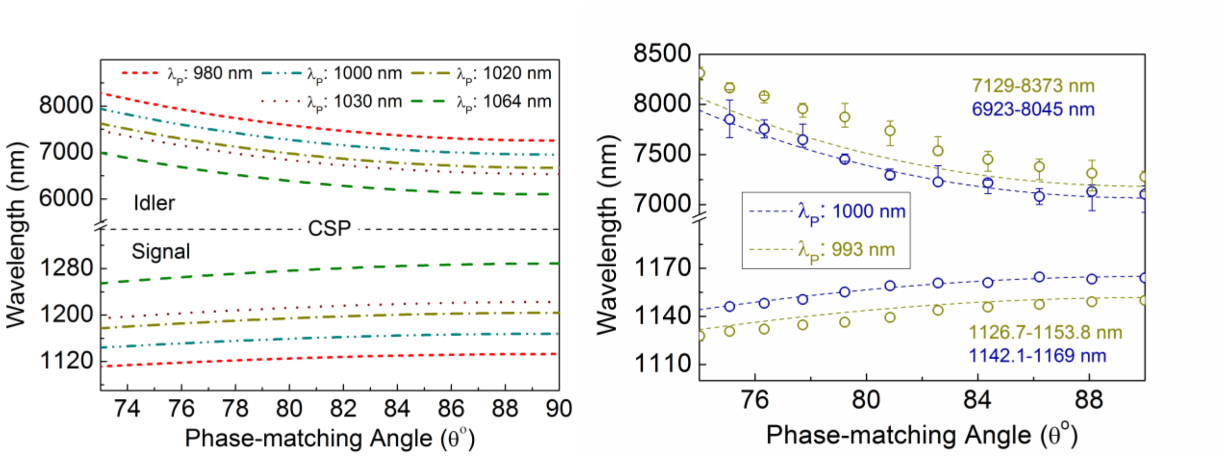

Figure 2. a) Signal and idler tuning curves for type-I ($e\rightarrow oo$) critical phase-matching as a function of the CSP internal angle for various pump wavelengths in the range so far used to pump CSP OPOs, b) Idler and signal tuning as a function of the internal phase-matching angle in the CSP crystal, for pump wavelengths of 993 nm and 1000 nm, compared to the wavelength tuning curves predicted using the Sellmeier equations[8]. Error bars represent additional cavity delay tuning.

Operation of the OPO was achieved by assembling the cavity as shown in Fig. 1, and synchronisation was obtained by fine adjustment of the delay line. Deployment of the KLM Ti:sapphire laser offers the important advantage of pump wavelengths <1 µm, which enables the generation of longer idler wavelengths in the deep-IR. This is illustrated in Fig. 2(a), where the signal and idler wavelength tuning as function of the CSP phase-matching angle under critical type-I ($e\rightarrow oo$) phase-matching for various discrete pump wavelengths of interest, from Yb-based solid-state and fibre lasers down to the Ti:sapphire laser, is presented. As can be clearly seen, for all phase-matching angles (including θ=90° at NCPM), longer deep-IR idler wavelengths can be generated by deploying shorter pump wavelengths in the Ti:sapphire laser wavelength range. The results of angle tuning from θ=90° to 74° are shown in Fig. 2(b), where it can be seen that the near-IR signal wavelength could be tuned across 1126.7–1153.8 nm, with the idler covering a spectral range of 6923–8373 nm in the deep-IR, for two pump wavelengths at 993 nm and 1000 nm. The measured values are also compared to the Sellmeier predictions,[8] with the observed offset accredited to strong self-phase modulation (SPM) redistributing the spectral density towards shorter wavelengths.

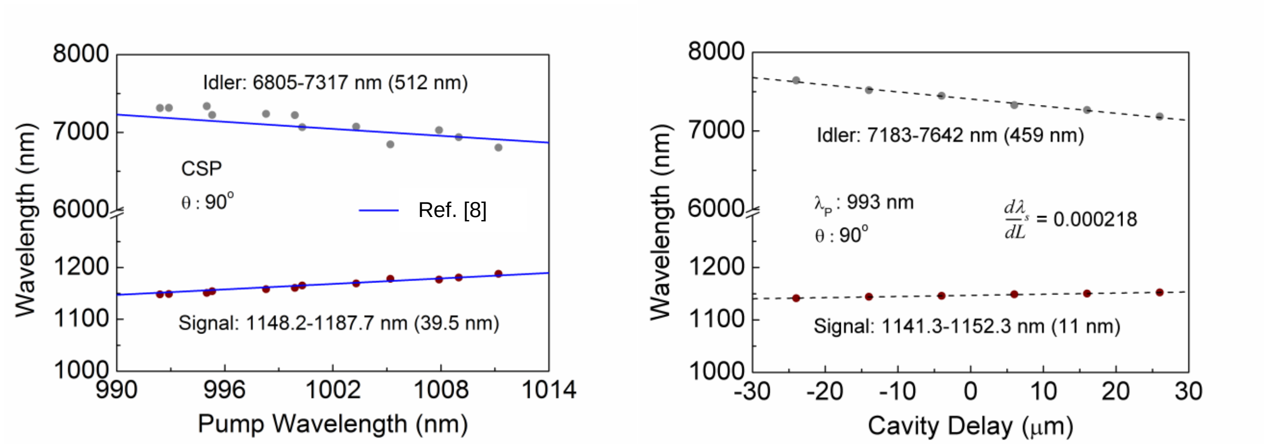

Figure 3. a) Pump tuning of the OPO compared to the Sellmeier predictions,[8] b) Cavity delay tuning at normal incidence for a pump wavelength of 993 nm.

Fig. 3(a) shows the experimental data for pump tuning, compared to the theoretical predictions in blue, calculated using the Sellmeier equations.[8] As seen in Fig. 2(b), the relatively wide parametric gain bandwidth of CSP enables wavelength tuning by variation of cavity length. This is a convenient method for rapid tuning across a large idler bandwidth. An example of cavity delay tuning from this OPO at a pump wavelength of 993 nm and phase-matching angle of θ=90° is shown in Fig. 3b). Using these methods in tandem, we are able to rapidly tune across 6805–7642 nm in the deep-IR.

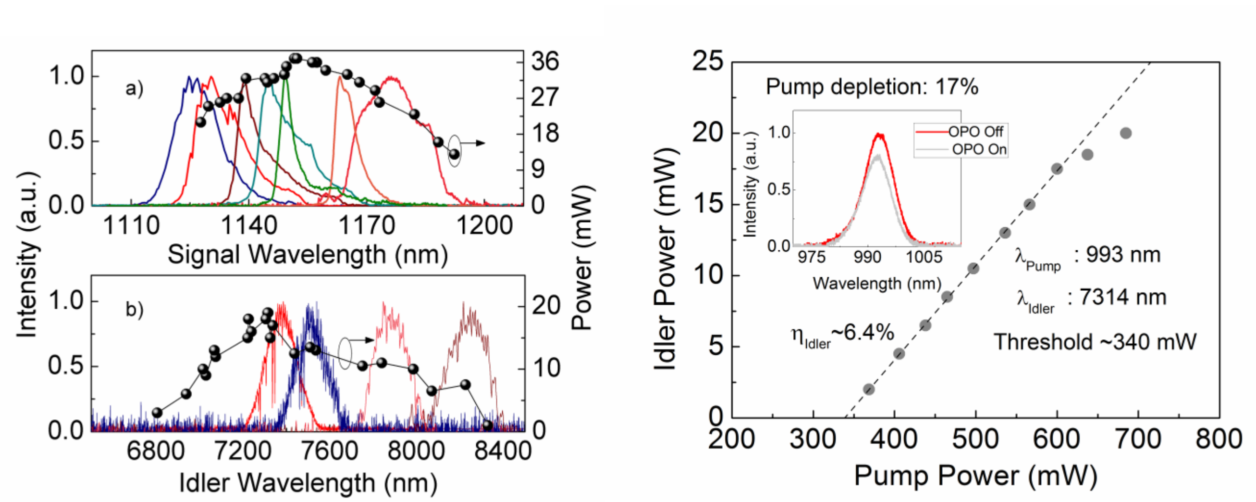

Figure 4. a) Signal, and b) idler power and spectra across the OPO tuning range, c) Power scaling of the idler extracted from the CSP femtosecond OPO. Inset: Input pump spectrum and depleted pump spectrum while the OPO is operating at maximum power.

As shown in Fig. 4(a), we extracted deep-IR idler average powers of >10 mW across >65% of the tuning range, with a maximum of 20 mW at 7314 nm and >6 mW available up to 8227 nm. This is equivalent to a peak idler quantum conversion efficiency of 21.3%, and when the beam divergence and spectral bandwidth are considered, is equal to a spectral brightness of ~6.68×10^{20} photons s^{-1} mm^{-2} sr^{-1} 0.1% BW^{-1}. Furthermore, with a 5% output coupler in place, we measured signal powers up to 37 mW at 1152 nm. Also plotted are the detailed spectral qualities of the signal and idler, characterised using two spectrum analysers for the near-IR and deep-IR, with resolutions of 0.7 nm and ~4 nm respectively. The signal spectra were found to be highly asymmetric, containing an SPM-induced long-wavelength tail, as expected for a cavity with a large net positive GVD. In contrast, idler spectra were measured to be almost symmetric with evidence of atmospheric water absorption at shorter wavelengths in the form of deep modulations. We investigated idler power scaling of the OPO, using a high reflector in place of M_8 to maximise the deep-IR power, with the results shown in Fig. 4(b). The pump power is corrected for a ~83% transmission loss through M_1. At a pump wavelength of 993 nm, the OPO threshold was measured to be ~340 mW, and the idler slope conversion efficiency is ~6.4 %, with a pump depletion of ~17%.

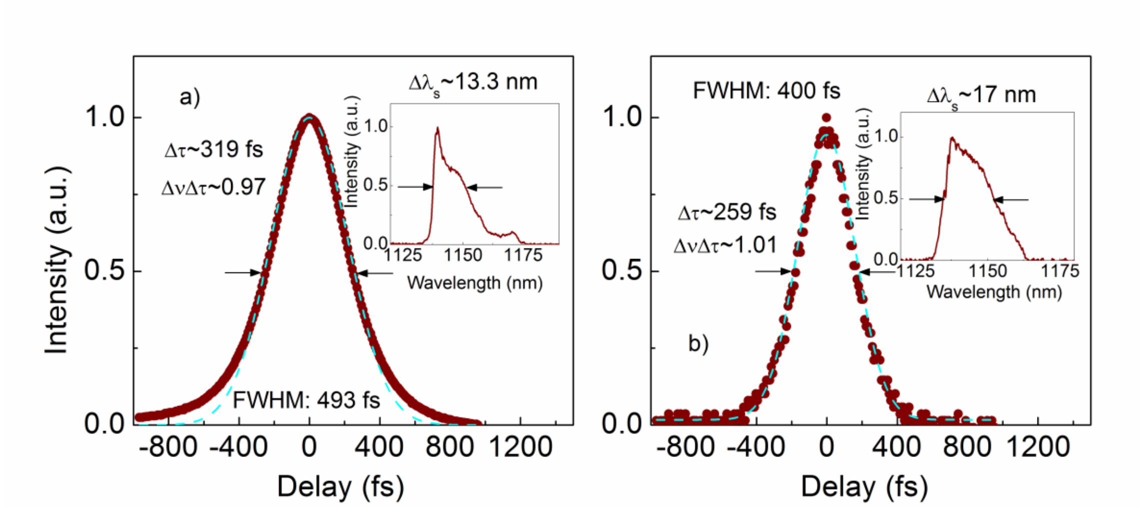

Figure 5. Signal intensity autocorrelations measured (a) at perfect cavity synchronisation, and (b) with the cavity detuned. Inset: simultaneously measured signal spectra.

The pulse duration was measured experimentally using an intensity autocorrelator, confirming strongly chirped pulses. Fig. 5 shows two signal autocorrelation profiles at (a) perfect synchronisation, and (b) when the cavity is slightly detuned, confirming durations of 319 and 259 fs, respectively. Also shown in the insets are the two corresponding spectra, with approximate FWHM bandwidths of 13.3 nm and 17 nm, respectively. When the signal and pump pulses are not perfectly temporally overlapped, the reduced intracavity power induces a smaller nonlinear phase shift and results in a weaker pulse chirp.

3. CONCLUSIONS

In conclusion, we have demonstrated a high-spectral-brilliance, high-repetition-rate femtosecond source of coherent radiation for the deep-IR using a single frequency conversion stage pumped by a KLM Ti:sapphire laser. By exploiting the wide transparency window of CSP, we have been able to achieve successful operation of a deep-IR femtosecond OPO directly using a KLM Ti.sapphire laser, for the first time. In doing so, we have produced up to 20 mW of average power at 7314 nm, equivalent to a spectral brightness of 6.68×10^{20} photons s^{-1} mm^{-2} sr^{-1} 0.1% BW^{-1}, with up to 37 mW generated in the near-IR signal. We have used three separate tuning methods; type-I critical phase-matching, pump wavelength tuning, and cavity delay tuning, to generate coherent radiation across 1127–1192 nm and 6654–8373 nm (1194–1503 cm^{-1}) in the signal and idler, respectively, with typical FWHM spectral bandwidths of 140–180 nm in the deep-IR. In particular, exploiting the tunable properties of the Ti:sapphire laser has permitted rapid tuning of the deep-IR idler wavelength over key spectral regions, including the amide III band, of great interest for medical imaging. The signal and idler beams exhibit excellent TEM_{00} spatial quality and passive long-term power stability of 2.2% and 3.2% rms over 1 hour, respectively. Spectral and temporal characterisation of the signal has revealed chirped pulses with a time-bandwidth product, $\Delta\upsilon\Delta\tau$~1. Both the pulse durations and 10 dB spectral bandwidths were observed to be broader at higher intracavity powers, indicating strong intracavity self-phase-modulation, which could be reduced using appropriate intracavity dispersion management. With high average power at a high repetition rate, this OPO represents an attractive source for low-noise spectroscopy, frequency comb generation and medical diagnostics in the 6.5–8.5 μm spectral region, using a simplified pumping method based on established ultrafast KLM Ti:sapphire laser technology.

REFERENCES

[1] Bakker, J. M., Mac Aleese, L., Meijer, G. and von Helden, G., "Fingerprint IR spectroscopy to probe amino acid conformations in the gas phase," Phys. Rev. Lett. 91(20), 203003 (2003).

[2] Bellamy, L. J., [The Infra-red Spectra of Complex Molecules], Springer Netherlands, 231-262 (1975).

[3] Mille, L. M., and Smith, R. J., "Synchrotrons versus globars, point-detectors versus focal plane arrays: Selecting the best source and detector for specific infrared microspectroscopy and imaging applications," Vib. Spectrosc. 38(1), 237-240, (2005).

[4] Petersen, C. R., Møller, U., Kunbat, I., Zhou, B., Dupont, S., Ramsay, J., Benson, T., Sujecki, S., Abdel-Moneim, N., Tang, Z., and Furniss, D., "Mid-infrared supercontinuum covering the 1.4–13.3 μm molecular finger-print region using ultra-high NA chalcogenide step-index fibre," Nat. Photonics 8(11), 830–834, (2014).

[5] Dumas, P., Polack, F., Lagarde, B., Chubar, O., Giorgetta, J. L., and Lefrançois, S., "Synchrotron infrared microscopy at the French Synchrotron Facility SOLEIL," Infrared Phys. Techn. 49(1), 152–160, (2006).

[6] Faist, J., [Quantum cascade lasers], UK: OUP Oxford, (2013).

[7] Ebrahim-Zadeh, M., and Sorokina, I. T., [Mid-infrared Coherent Sources and Applications], Springer, (2008).

[8] Zawilski, K. T., Schunemann, P. G., Pollak, T. M., Zelmon, D. E., Fernelius, N. C., and Hopkins, F. K., "Growth and characterization of large CdSiP2 single crystals," J. Cryst. Growth 312(8), 1127–1132, (2010).

[9] Chaitanya Kumar, S., Schunemann, P. G., Zawilski, K. T., and Ebrahim-Zadeh, M., "Advances in ultrafast optical parametric sources for the mid-infrared based on CdSiP2," J. Opt. Soc. Am. B 33(11), D44–D56, (2016).

[10] Zhang, Z., Reid, D. T., Chaitanya Kumar., S., Ebrahim-Zadeh, M., Schunemann, P. G., Zawilski, K. T. and Howle, C. R., "Femtosecond-laser pumped $CdSiP_2$ optical parametric oscillator producing 100 MHz pulses centered at 6.2 μm," Opt. Lett. 38(23), 5110–5113, (2013).

[11] Chaitanya Kumar, S., Krauth, J., Steinmann A., Zawilski, K. T., Schunemann, P. G., Giessen, H. and Ebrahim-Zadeh, M., "High-power femtosecond mid-infrared optical parametric oscillator at 7 μm based on $CdSiP_2$," Opt. Lett. 40(7), 1398–1401, (2015).

[12] Ramaiah-Badarla, V., Chaitanya Kumar, S., Esteban-Martin, A., Devi, K., Zawilski, K. T., Schunemann, P. G., and Ebrahim-Zadeh, M., "Ti:sapphire-pumped deep-infrared femtosecond optical parametric oscillator based on CdSiP2," Opt. Lett. 41(8), 1708–1711, (2016).

[13] Chaitanya Kumar, S., Esteban-Martin, A., Santana, A., Zawilski, K. T., Schunemann, P. G., and Ebrahim-Zadeh, M., "Pump-tuned deep-infrared femtosecond optical parametric oscillator across 6–7 μm based on $CdSiP_2$," Opt. Lett. 41(14), 3355–3358, (2016).