Synchronously pumped at 1064 nm OPO based on $CdSiP_2$ for generation of high power picosecond pulses in the mid-infrared near 6.4 µm

Andre Peremans,[a)] Dan Lis,[a)] Francesca Cecchet,[a)] Peter G. Schunemann,[b)] Kevin T. Zawilski,[b)] Valentin Petrov[c)]

[a]Centre de Recherche en Physique de la Matiere et du Rayonnement (PMR), University of Namur, 61 rue de Bruxelles, B-5000, Namur, Belgium;
[b]BAE Systems, Inc., MER15-1813, P.O. Box 868, Nashua, NH 03061-0868, USA;
[c]Max-Born-Institute for Nonlinear Optics and Ultrafast Spectroscopy, 2A Max-Born-Str., D-12489 Berlin, Germany

ABSTRACT

The recently developed chalcopyrite $CdSiP_2$ is employed in a picosecond, 90°-phase-matched synchronously pumped optical parametric oscillator pumped at 1064 nm, to produce quasi-steady-state idler pulses near 6.4 µm with an energy as high as 2.8 µJ at 100 MHz. The train of 2 µs long macropulses, each consisting of 200 (picosecond) pulses, follows at a repetition rate of 25 Hz. This corresponds to an average power of 14 mW. The pump depletion (conversion efficiency) exceeds 40%. Without intracavity etalon, the 12.6 ps long mid-IR micropulses have a spectral width of 240 GHz.

Key words: synchronously pumped optical parametric oscillators; mid-infrared; cadmium silicon phosphide; picosecond pulses

1. INTRODUCTION

Synchronously pumped optical parametric oscillators (SPOPOs) represent a potentially efficient source of high repetition rate (~100 MHz) ultrashort pulses at wavelengths not available from conventional mode-locked lasers. Difference-frequency generation (DFG) can also be used to this aim but yields very low quantum efficiency and requires two synchronized wavelengths. Depending on the pump source, e.g. mode-locked Ti:sapphire and Nd-lasers or their second harmonics, SPOPOs normally generate 100 fs to 100 ps long pulses. Being down-conversion devices, their coverage in the mid-IR is mostly limited to 4-5 µm, the absorption edge of oxide based nonlinear crystals. Normally, the signal wavelength is only resonated and although in some special cases (high parametric gain in the exit layer of periodically poled $LiNbO_3$, PPLN) the generated idler extended up to 7.25 µm,[1] the single pulse energy at such wavelengths is extremely low (~0.3 pJ).

The use of chalcogenide crystals, transparent in the mid-IR, has been reported only in a few cases. Preserving the high repetition rate was possible only by cascaded operation of two SPOPOs. Thus, a non-critically phase-matched CdSe based SPOPO was pumped and tuned by the signal wave of a PPLN based SPOPO at 120 MHz, and generated picosecond idler pulses but the wavelength range was limited to 9.1-9.7 µm and the maximum single pulse energy was 90 pJ.[2] Another SPOPO based on $AgGaSe_2$ was pumped at 1.55 µm by the signal wave of an 82 MHz femtosecond $CsTiOPO_4$ based SPOPO and generated 0.4-0.5 ps long pulses up to 7.9 µm, with single pulse energy of 270 pJ at 5.25 µm.[3]

In general the cavity length stabilization and synchronization of 3 short-pulse devices (mode-locked pump laser and 2 SPOPOs) is a serious problem which is detrimental for the long term stability and potential applications. Thus, for

Nonlinear Frequency Generation and Conversion: Materials, Devices, and Applications IX,
edited by Peter E. Powers, Proc. of SPIE Vol. 7582, 75820G · © 2010 SPIE
CCC code: 0277-786X/10/$18 · doi: 10.1117/12.840215

attainment of devices with improved practicability, compact design and utility, it is of great interest to develop mid-IR SPOPOs based on chalcogenide materials that can be directly pumped by conventional mode-locked lasers operating in the near-IR, especially near 1 µm. However, nonlinear crystals like the commercially available $AgGaS_2$ (AGS), that can be used at 1064 nm without the onset of two-photon absorption (TPA), have poor thermo-mechanical characteristics like thermal conductivity, anisotropy of the thermal expansion and damage threshold. Three concepts to reduce the load to the nonlinear material in terms of average power have been implemented so far: a 32-fold mechanical chopper was used at 40 Hz to reduce the average power of the 20 W, 100 ps Nd:YAG pump laser operating at 76 MHz,[4] a similar mode-locked laser was additionally Q-switched at 2 kHz to produce an average power of up to 8.5 W at a repetition rate of 2 kHz for the non-stationary macropulses consisting of about 20 micropulses of 145 ps duration,[5] and a pulsed Nd:YAG oscillator-amplifier system, generating about 100 micropulses of 12.5 ps duration with an average power of up to 1.2 W, at 25 Hz for the macropulses, was employed in Ref. 6. Note that these approaches lead to substantial increase of the single pulse energy which reached the nanojoule[4] and microjoule[5,6] level in the 4-5.5 µm wavelength range for the idler.

The recently discovered cadmium silicon phosphide, $CdSiP_2$ (CSP),[7] is a negative uniaxial II-IV-V_2 chalcopyrite compound (space group 42m) that allows 1064 nm pumping without TPA and possesses a useful transparency up to 6.5 µm, limited by intrinsic multi-phonon peaks. As shown in Ref. 8, it outperforms all other mid-IR nonlinear materials that can be pumped near 1 µm in almost every aspect with the main problem yet to be solved being the residual absorption close to the band-gap which is not intrinsic. In addition, it is the only material which, without being a solid solution, still allows non-critical phase-matching when pumped near 1 µm, with a maximum effective nonlinearity of $d_{eff}=d_{36}=84.5$ pm/V.[8,9] We realized now the first SPOPO based on 90°-cut CSP, pumped at 1064 nm.

2. EXPERIMENTAL SETUP AND CRYSTAL SAMPLE

The sample used in the present study (Fig. 1) was grown by directional solidification in a modified, high-temperature transparent furnace using the horizontal gradient freeze (HGF) technique. It was cut at $\theta=90°$, $\varphi=45°$ and had a length of 9.5 mm. Its aperture was 6 mm (along the ***c***-axis) × 6.75 mm. The residual losses measured for the relevant polarizations (e for the pump and o for the signal and idler) are 0.185 cm^{-1} at 1064 nm, 0.114 cm^{-1} at 1.3 µm, and 0.014 cm^{-1} at 6.4 µm, see Fig. 2. Both faces were AR-coated for the three wavelengths (pump, signal, and idler) and the 8-layer coating (TwinStar) resulted in averaged reflectivity per surface of ~0.35% at 1064 nm, ~0.4% at 1275 nm and ~0.8% at 6400 nm.

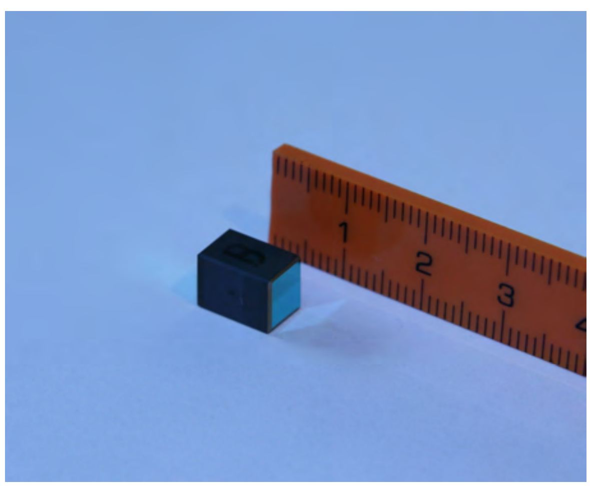

Fig. 1. Photograph of the AR-coated sample CSP 17B used in the present study.

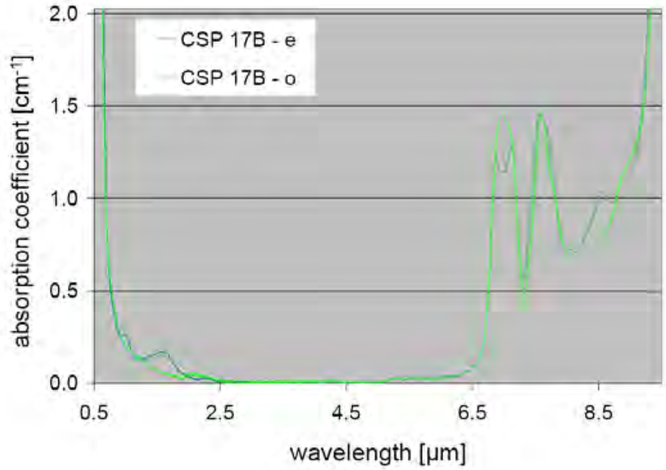

Fig. 2. Polarized transmission of sample CSP 17B measured prior to coating (b).

The pump source was an upgraded version of the one described in Ref. 6, see Fig. 3, more details will be provided in a separate publication. Briefly Part I shows the mode-locked Nd:YAG laser oscillator built around one Nd:YAG rod (110 mm long and 6 mm in diameter) that is pumped with a flashlamp at a repetition rate of 25 Hz. The oscillator cavity length of about 1.5 m is adjusted to the 100 MHz frequency of the acousto-optic mode-locker (AOML). Mode-locking is achieved using frequency doubling nonlinear mirror (FDNLM) constituted by a SHG crystal (BBO) and a dichroic mirror (DM) which provides a positive feedback on the peak intensity of the pulse circulating in the cavity. The FDNLM effect is balanced by the negative feedback provided by the nonlinear absorption in a GaAs semiconductor platelet. These two antagonist actions result in the stabilization of the pulse energy and duration. The pulse duration at the oscillator output, as inferred from background-free second-order autocorrelation, is between 15 and 20 ps.

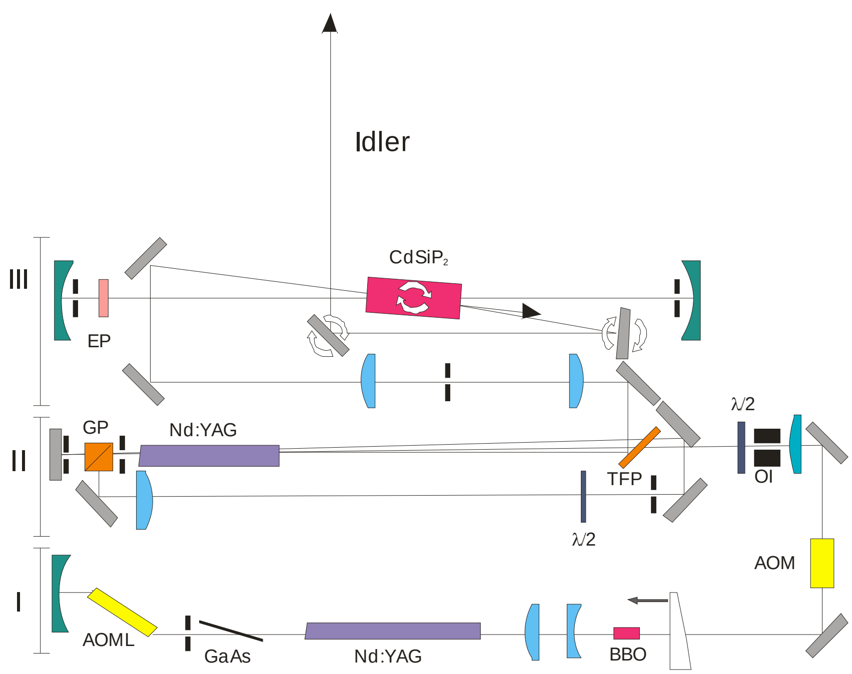

Fig. 3. Schematic drawing of the laser system. Part I: laser oscillator mode-locked using FDNLM. Part II: 3-pass amplification stage in Nd:YAG. Part III: SPOPO for the mid-IR. The etalon EP was not used in the present study.

The extracavity AOM suppresses the initial, non-stationary part of the train, and the resulting, almost steady-state macro-pulse consists of about 200 micropulses with 10 ns separation. The oscillator is protected by an optical isolator (OI).

A 3 pass amplification scheme is adopted as illustrated in Part II of Fig. 3. The amplification stage is built around an identical Nd:YAG laser rod (110 mm long and 6 mm in diameter) which is pumped by two flashlamps. After the Glan polarizer, GP, the beam is horizontally polarized and amplified by a double pass through the Nd:YAG rod. The second pass trajectory is slightly tilted off from the first, so that it is directed towards the half-wave plate ($\lambda/2$) that rotates the polarization to vertical. The beam is then re-injected through the Nd:YAG rod by the polarizer GP for the third pass amplification. This amplified beam is separated from the first and the second pass by a thin film polarizer (TFP). The lenses set the beam diameter in the Nd:YAG rod to 3 mm for the first and second passes, and to 5 mm for the third pass amplification, in order to optimize the energy extraction from the laser rod. Maximum output pump power of about 3 W, e.g. 3/25=0.12 J per bunch (macro-pulse) is achieved. The micropulse duration is 2 µs. The maximum picosecond micropulse energy is thus 120/200=0.6 mJ.

The SPOPO is schematized in part III of Fig. 3. It consists of two RC=-3 m curved mirrors for the signal only, at a separation of 1.5 m giving a Gaussian mode diameter of 3.2 mm in the middle where the nonlinear crystal was placed. A telescope adapts the pump beam spot diameter (~2 mm) to the cavity mode. The pumping of the nonlinear crystal is slightly non-collinear (the pump beam makes an angle of about 2° with respect to the SPOPO cavity axis) meaning that the signal beam oscillates in the cavity while the idler beam is emitted out of the SPOPO cavity axis. Synchronous pumping requires the optical length of the cavity to be adjusted to that of the Nd:YAG oscillator (1.5 m).

3. RESULTS AND DISCUSSION

We studied the input/output characteristics of the OPO, including the oscillation threshold, at normal incidence and minimum possible cavity length (18.5 mm). The tuning curve required tilting of the crystal and the corresponding cavity length was slightly increased (20.5 mm). We estimated the idler energy both from the pump depletion and using a calibrated pyroelectric detector. The threshold corresponded to an average pump power of 15 mW or a single (micro)pulse energy of 3 µJ. The depletion together with the simultaneously measured transmission of the crystal is shown in Fig. 4.

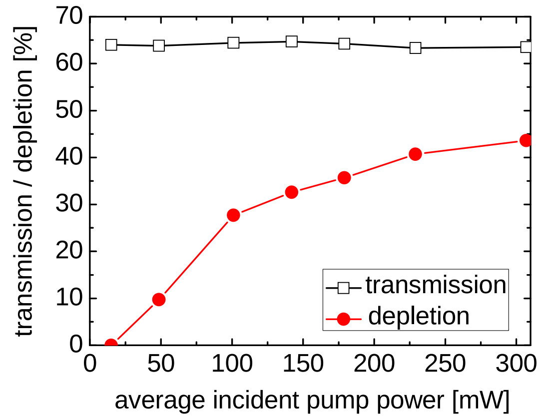

Fig. 4. Pump depletion and crystal transmission at 1064 nm vs. pump power.

The incident pump power was limited to slightly above 300 mW because strong pump depletion, reaching typically >40%, was observed starting from about 100 mW of pump power. The actual crystal transmission at the pump wavelength, when the OPO cavity was blocked, was lower than expected but this was confirmed in additional extracavity measurements, using also a CW source with comparable beam size and can be explained by inhomogeneous crystal quality (defects inside). However, no signs of TPA are seen in Fig. 4.

Figure 5 shows the input-output characteristics. The directly measured idler power was in good agreement with estimations from the pump depletion. The maximum idler power of 14 mW corresponds to a single (micro)pulse energy of 2.8 μJ.

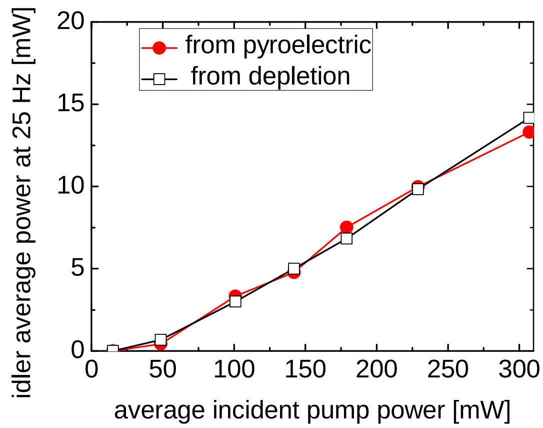

Fig. 5. Idler power vs. pump power.

Only normal incidence was studied but the slightly noncollinear interaction resulted in ~200 nm longer idler wavelengths. From the measured signal wavelength of 1276.55 nm (Fig. 6), one arrives at an idler wavelength of 6397.5 nm, This was confirmed by calculations using the refined Sellmeier equations,[9] which predicted the same idler wavelength for an internal angle of 0.666° between the signal and the pump waves, see Fig. 6.

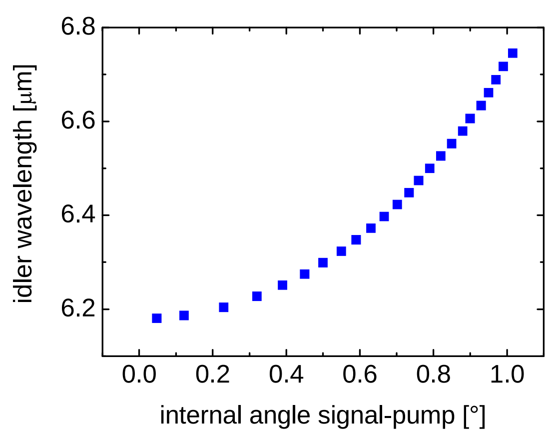

Fig. 6. Idler wavelength versus internal angle between the signal and pump waves for noncollinear type-I interaction in CSP. The angle between the pump wave and the optical axis is fixed at 90° corresponding to the noncritical configuration. The corresponding internal angle between the idler and the pump weaves varies between 0 and 5.6°.

The signal spectrum was measured from a cavity leakage at a pump level of 200 mW, with 0.4 nm spectral resolution, Fig. 7. The signal bandwidth corresponds to 8 cm^{-1} or 240 GHz which is comparable, though slightly larger than the spectral acceptance of CSP for DFG.

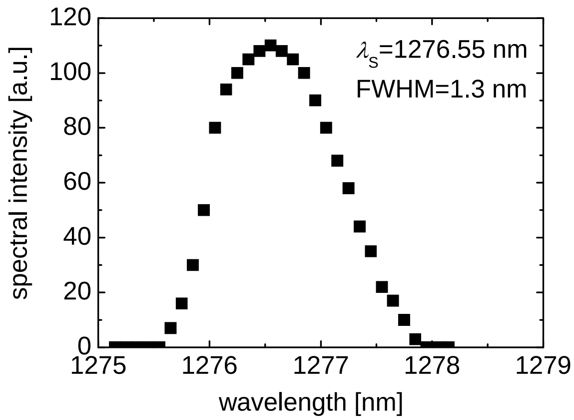

Fig. 7. Signal spectrum.

We measured the duration of the idler pulses using non-collinear SHG in a 2-mm-thick $HgGa_2S_4$ crystal cut at φ=45°, θ=40° for type-I interaction. A ZnSe plate served as a beam splitter and the second harmonic was detected by a PbS resistor connected to a lock-in amplifier. Figure 8 shows the result of averaging 4 traces.

The Lorentzian fit is better than the Gaussian one, and gives a FWHM ~25 ps for the trace, which means a pulse FWHM of 12.6 ps. Assuming the same spectral bandwidth for the idler, one arrives at a time-bandwidth product of 3, more than an order of magnitude above the Fourier limit.

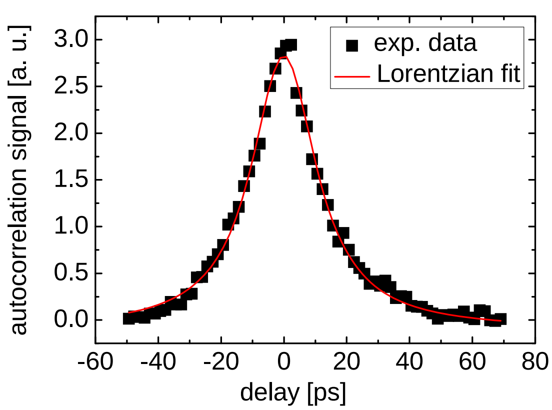

Fig. 8. Experimental data (symbols) and Lorentzian fit (curve) of a background-free autocorrelation trace for the idler pulses.

Finally, by using a hair dryer, we confirmed that slight heating of the crystal results in shorter (about 2 nm) signal wavelength which translates into an idler wavelength in the 6450 nm range, important for medical (surgical) applications.[10] This is an indication of the possibility for practical tuning by temperature variation in the non-critical configuration which potentially can cover the transparency range up to the long-wave limit.

When CSP was replaced by AGS (10×10×10 mm^3) cut for type-I phase-matching, the parametric oscillation threshold increased to ~100 mW, i.e. about an order of magnitude. Moreover, the use of AGS in this SPOPO was accompanied by gradual surface blackening. Surface lifetime could be kept above ~400 h if only the pump average power did not exceed ~200 mW.

4. CONCLUSION

In conclusion, we demonstrated for the first time to our knowledge SPOPO operation of CSP pumped at 1064 nm. Under 90°-phase-matching the SPOPO produced quasi-steady-state idler pulses near 6.4 µm with an energy as high as 2.8 µJ at 100 MHz. The train of 2 µs long macropulses, each consisting of 200 (picosecond) pulses, follows at a repetition rate of 25 Hz. This corresponds to an average power of 14 mW. The pump depletion (conversion efficiency) exceeds 40%. Without intracavity etalon, the 12.6 ps long mid-IR micropulses have a spectral width of 240 GHz. Future work will be focused on reduction of the residual loss, which alone is expected to lead to 2-fold improvement of the output powers, temperature tuning maintaining the non-critical configuration, and power scaling using crystals of larger aperture.

ACKNOWLEDGMENTS

The research leading to these results has received funding from the European Community's Seventh Framework Programme FP7/2007-2011 under grant agreement n°224042 and from the U.S. Air Force Research Laboratory Materials and Manufacturing Directorate (AFRL/RXPSO) under contract number FA8650-05-C-5425. D. L. acknowledges the Belgian Fund for Agricultural and Industrial Research (FRIA). F. C. and A. P. are respectively Postdoctoral Researcher and Research Director of the Belgian Fund for Scientific Research (FNRS-F.R.S.).

REFERENCES

[1] Watson, M. A., O'Connor, M. V., Lloyd, P. S., Sheperd, D. P., Hanna, D. C., Gawith, C. B. E., Ming, L., Smith, P. G. R., and Balachninaite, O., "Extended operation of synchronously pumped optical parametric oscillators to longer idler wavelengths," Opt. Lett. **27**, 2106-2108 (2002).

[2] Watson, M. A., O'Connor, M. V., Shepherd, D. P., and Hanna, D. C., "Synchronously pumped CdSe optical parametric oscillator in the 9-10 µm region" Opt. Lett. **28**, 1957-1959 (2003).

[3] Marzenell, S., Beigang, R., and Wallenstein, R., "Synchronously pumped femtosecond optical parametric oscillator based on $AgGaSe_2$ tunable from 2 µm to 8 µm," Appl. Phys. B **69**, 423-428 (1999).

[4] Cheung, E. C., Koch, K., and Moore, G. T., "Silver thiogallate, singly resonant optical parametric oscillator pumped by a continuous-wave mode-locked Nd:YAG laser," Opt. Lett. **19**, 631-633 (1994).

[5] McEwan, K. J., "High-power synchronously pumped $AgGaS_2$ optical parametric oscillator," Opt. Lett. **23**, 667-669 (1998).

[6] Mani, A., A., Dreesen, L., Hollander, Ph., Humbert, C., Caudano, Y., Thiry, P. A., and Peremans, A., "Pumping picosecond optical parametric oscillators by a pulsed Nd:YAG laser mode-locked using a nonlinear mirror," Appl. Phys. Lett. **79**, 1945-1947 (2001).

[7] Schunemann, P. G., Zawilski, K. T., Pollak, T. M., Zelmon, D. E., Fernelius, N. C., and Kenneth Hopkins, F., "New nonlinear optical crystal for mid-IR OPOs: $CdSiP_2$," Advanced Solid-State Photonics, Nara, Japan, Jan. 27-30, 2008, Conference Program and Technical Digest, Post-Deadline Paper MG6.

[8] Petrov, V., Noack, F., Tunchev, I., Schunemann, P., and Zawilski, K., "The nonlinear coefficient d_{36} of $CdSiP_2$," Proc. SPIE **7197**, 7197-21/1-8 (2009).

[9] Schunemann, P. G., Zawilski, K. T., Pollak, T. M., Petrov, V., and Zelmon, D. E., "$CdSiP_2$: a new nonlinear optical crystal for 1- and 1.5-micron-pumped mid-IR generation," Advanced Solid-State Photonics, Denver (CO), USA, Feb. 1-4, 2009, Conference Program and Technical Digest, Paper TuC6.

[10] Edwards, G. S., Austin, R. H., Carroll, F. E., Copeland, M. L., Couprie, M. E., Gabella, W. E., Haglund, R. F., Hooper, B. A., Hutson, M. S., Jansen, E. D., Joos, K. M., Kiehart, D. P., Lindau, I., Miao, J., Pratisto, H. S., Shen, J. H., Tokutake, Y., van der Meer, A. F. G., and Xie, A., "Free-electron-laser-based biophysical and biomedical instrumentation," Rev. Sci. Instrum. **74**, 3207-3245 (2003).