Multi-microjoule GaSe-based mid-infrared optical parametric amplifier with an ultra-broad idler spectrum covering 4.2-16 μm

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We report a multi-microjoule, ultra-broadband mid-infrared optical parametric amplifier based on a GaSe nonlinear crystal and a ~2 μm pump laser. The generated idler pulse has a flat spectrum spanning from 4.5 to 13.3 μm at -3 dB and 4.2 to 16 μm in the full spectral range, with a central wavelength of 8.8 μm. To our best knowledge, this is the broadest -3 dB spectrum ever obtained by optical parametric amplifiers in this spectral region. The spectrum supports ~19 fs Fourier-transform-limited pulse width, corresponding to 0.65 cycle, centered at 8.8 μm. ~3.4 μJ pulse energy is obtained with a pump-to-idler conversion efficiency of ~2%.

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High-energy, mid-infrared (mid-IR), ultra-broadband, few-cycle lasers have been attracting extensive attentions for their applications in molecular spectroscopy [1-4] and strong-field physics [5-14]. In the recent years, the research has been extended to >6 μm long-wavelength mid-IR region. For molecular spectroscopic applications, the long-wavelength mid-IR is more important than the short-wavelength mid-IR, because the absorption spectrum in the wavelength range of 6.7-25 μm, i.e. the fingerprint region, is distinctive for each molecule, which allows their unique identification [3, 4]. In the applications of strong-field physics, with long mid-IR laser wavelength, the energy of the re-colliding electron increases substantially. It enables reaching deep into the tunneling ionization regime even at moderate laser intensity, without causing optical damage. The long-wavelength mid-IR few-cycle lasers have triggered a number of researches in the ultrafast electronics in molecules, solid materials as well as nano-structures [7-14]. More interestingly, long-wavelength mid-IR pulses have their photon energies far below the typical electronic interband resonances of bulk semiconductors. So the phase-stable few-cycle long-wavelength mid-IR pulses could serve as a precisely adjustable bias for ultrafast electronics studies [11]. With the demand from the high-fidelity mid-IR spectroscopy and advanced attosecond electronics, high-energy few-cycle long-wavelength mid-IR lasers with the stable carrier-envelope (CE) phase are highly desired.

Due to the lack of broadband gain media in the mid-IR region, nonlinear down conversions such as optical parametric amplifier (OPA), optical parametric chirped-pulse amplifier (OPCPA) and difference-frequency generation (DFG) are commonly used to generate high-energy few-cycle mid-IR pulses. Non-oxide nonlinear crystals such as AgGaS$_2$ [15, 16], CdSiP$_2$ [17], and ZnGeP$_2$ [18-20] are employed to extend the high-energy few-cycle pulses to the wavelength range of 4-11 μm. 8.5 μm, 150 μJ, few-cycle pulses are demonstrated by mixing the 1.8 μm pump and the 2.4 μm signal in an AgGaS$_2$-based DFG system [15], 33 μJ, 0.88-cycle pulses covering 2.5-10 μm spectral range are generated by the coherent synthesis of signal and idler pulses of a CdSiP$_2$ OPA, pumped by a 2 μm OPCPA [17]. The energy of 7 μm few-cycle pulses is boosted to half milijoule in a ZnGeP$_2$-based OPCPA system, with a powerful 2 μm cryogenic-cooled pump [18]. Recently, the wide bandgap nonlinear crystal LiGaS$_2$ is emerging, 7-11 μm few-cycle pulses with nano-joule-level pulse energy have been demonstrated in OPA or intrapulse DFG based on LiGaS$_2$ pumped by near-IR pulses [21-24]. The energy scaling potential is foreseeable for this wide-bandgap crystal in the near future. In a future step, to pursue the longer wavelength up to 20 μm, AgGaSe$_2$ [25] and GaSe [26, 27] nonlinear crystals have been employed in the DFG systems, thanks to their longer transparent wavelength cutoff. In order to achieve broad phase-matching (PM) bandwidth and avoid parasitic two- or three-photon absorption, they should ideally be pumped at ~2 μm wavelength. Compared with AgGaSe$_2$, GaSe has higher damage threshold, larger nonlinear coefficient.
In this letter, we report a GaSe-based mid-IR OPA driven by ~2 μm pump for the first time. The generated ultra-broadband idler pulse from the mid-IR OPA has a central wavelength of 8.8 μm, spanning from 4.5 to 13.3 μm at -3 dB and 4.2 to 16 μm in the full spectral range. To our best knowledge, this is the broadest mid-IR spectrum at -3 dB spectral range, ever obtained from the long-wavelength OPA systems. The measured spectrum supports a Fourier-transform-limited pulse width of ~19 fs, which corresponds to 0.65 cycle, centered at 8.8 μm. ~3.4 μJ idler pulse energy is demonstrated with the conversion efficiency of ~2%. The idler pulse has a passively stable CE phase by principle, due to the DFG nature in the OPA. We believe the generated idler pulse has a passively stable CE phase by principle, due to the DFG nature in the OPA. We believe the generated idler pulse has a passively stable CE phase by principle, due to the DFG nature in the OPA. We believe the generated idler pulse has a passively stable CE phase by principle, due to the DFG nature in the OPA.

GaSe has broad band bandwidth, pumped at ~2 μm wavelength. Fig. 1(a) shows the PM function \(|\sin(\Delta kL/2)|\) with respect to the PM angle and PM wavelength, in a GaSe crystal with a length (L) of 1 mm, for the type-I phase match, at 2.15 μm pump wavelength. (b) The measured optical parametric generation spectrum in a 1 mm GaSe crystal pumped by the 2.15 μm, 51 fs laser.

The mid-IR OPA has a single amplification stage as shown in the schematic in Fig. 2. It starts with a commercial OPA system (TOPAS from Light Conversion, which is driven by a 5 μJ, 30 fs, 800 nm Ti: Sapphire laser system) with a 2.15 μm central wavelength, 420 μJ pulse energy, 51 fs pulse width, and 1 kHz repetition rate. A CaF₂ wedge placed at 22° with respect to the pump beam functioning as a beam splitter is employed to reflect ~10 μJ, 2.15 μm, p-polarized pump for the supercontinuum (SC) generation. It is rotated to s-polarization by a half-wave plate, and focused into a 6-mm thick BaF₂ by L₁ to generate signal pulses via SC. The generated SC is collimated by L₂ and resized to ~2.5 mm/1/e² diameter by a telescope comprised of L₃ and L₄. The transmitted 2.15 μm beam enters the pump line, which includes a delay line and a telescope (L₅ and L₆), to form a collimated pump beam with a 1/e² diameter of ~2.5 mm. The collimated beam is then employed to pump a 1 mm-thick uncoated GaSe crystal with a type-I phase match. As the GaSe crystal could be cleaved only along the (001) plane (z-cut, \(\theta = 0°\)), an internal PM angle of ~11.1° corresponding to an external angle of ~32° is introduced, which causes ~16% loss for the p-polarized pump. Note that the pump beam is routed to the crystal using multiple silver mirrors with ~96% reflection each. Taking into account all the losses, the maximum available pulse energy of the 2.15 μm pump is ~300 μJ, giving an estimated peak intensity of ~224 GW/cm². Two 200-μm thick silicon windows placed at Brewster angle of 73.8° with respect to the pump beam are used as the beam combiner and beam splitter, respectively, with ~71% reflection for the s-polarized signal and idler beams. The generated idler pulse is separated from the amplified signal using a long-pass filter (LPF) with a cut-off wavelength at 45 μm. It is then characterized by a thermal sensitive power meter and a mid-IR monochrometer with a liquid-nitrogen-cooled MCT detector. For the spectral measurement, an uncoated ZnSe lens with a near-flat transmission over 1-15 μm and a dielectric-coated mid-IR hollow-core fiber (from OptoKnowledge) with a near-flat transmission in the range of 3-16 μm are employed to couple the idler pulses into the mid-IR monochrometer.

The spectral and temporal profiles of the 2.15 μm pump from TOPAS are firstly characterized. The measured spectrum as shown in Fig. 3(a) has a full width at half maximum of ~220 nm which supports ~31 fs Fourier-transform-limited pulse width. The intensity autocorrelation shown in Fig. 3(b) reveals the pulse width of ~51 fs, assuming a Gaussian temporal profile. This agrees with the residual dispersion from the TOPAS optics, including...
\[ \sim -400 \text{ fs}^2 \text{ dispersion from the 3 mm fused silica beam splitter} \]

(reflecting 800 nm, and transmitting 2.15 \( \mu \text{m} \)). Fig. 3(c) shows the long-wavelength side of the SC spectrum (measured through a InF fiber and a 2.4 \( \mu \text{m} \) LPF) which serves as the signal of the mid-IR OPA. It extends to \(~4.3 \mu \text{m}\). Here the influence of the nonlinear effects from the InF fiber on the spectral measurement has been excluded. With the knowledge of the long-wavelength edge of the signal, the calculated spectral edge on the short-wavelength side of the generated idler is \(~4.3 \mu \text{m}\), using 2.15 \( \mu \text{m} \) as the pump wavelength. Thus a LPF with a 4.5 \( \mu \text{m} \) cut-off wavelength is used to separate the generated idler from the amplified signal.

The spectrum of the amplified signal is shown in Fig. 3(d), measured through the mid-IR hollow-core fiber and the 2.4 \( \mu \text{m} \) LPF. It is to be noticed that the short wavelength edge is limited by the mid-IR hollow-core fiber with the transmission range of 3-16 \( \mu \text{m} \), and the non-zero intensity at \(~4.3 \mu \text{m}\) is attributed to the emergence of idler pulse at the degenerate wavelength.

The spectrum of the generated idler pulse is measured through the mid-IR hollow-core fiber and LPFs with different cut-off wavelengths, with 20 nm spectral resolution, as shown in Fig. 4(a). The responsivity of the MCT detector, the diffraction efficiency of the grating in the monochrometer, and the transmission of LPFs, ZnSe lens and the mid-IR hollow-core fiber are calibrated over the broadband mid-IR spectral range. The possibility of the spectral shaping is carefully excluded to ensure the accuracy of the spectral measurement. The measured spectrum of the idler pulse spans from 4.2 to 16 \( \mu \text{m} \), with the -3 dB bandwidth ranging from 4.5 to 13.3 \( \mu \text{m} \). This agrees with the predicted bandwidth by the PM function shown by the maroon curve in Fig.4 (a). It should be noted that the 4.5 \( \mu \text{m} \) LPF is responsible for the steep edge at \(~4.6 \mu \text{m}\). The water absorption contributes to the broad dip centered at \(~5.5 \mu \text{m}\). The peaks appearing at\(~7 \mu \text{m}\) and\(~12 \mu \text{m}\) are attributed to the best PM condition. It is clear that the PM function has its maximal value at these two wavelengths. The dip at \(~9.6 \mu \text{m}\) is also predicted by the PM function. While the PM condition is still satisfied over 13-15 \( \mu \text{m}\), the increasing transmission loss in GaSe beyond 13 \( \mu \text{m}\) prevents the efficient idler generation, as shown in the measured spectrum.

\[ \text{Intensity (a.u.)} \]

\[ \text{Wavelength (\mu m)} \]

\[ \text{Delay (fs))} \]

\[ \text{LPF with ~3.4 \mu J output energy (black curve). The PM function \[\text{sinc}(\alpha dL/L/2)\] (maroon curve) at a PM angle of ~11.05° is compared. The type-I phase match in the 1 mm GaSe, pumped at 2.15 \( \mu \text{m} \) wavelength is used in the calculation. (b) The calculated temporal profile and electrical field of the idler pulse using the measured idler spectrum, assuming a Fourier-limited pulse, and the zero CE phase.} \]

\[ \text{The temporal profile and the electric field of the generated idler pulse are calculated using the measured spectrum, assuming the idler pulse is Fourier-transform limited, and the CE phase is zero. As shown in Fig. 4(b), ~19 fs transform-limited pulse width is supported, which corresponds to 0.65-cycle electric field of an 88 \( \mu \text{m} \) center wavelength. The actual pulse width could be broadened to few cycles by the dispersion from the 1 mm thick 4.5 \( \mu \text{m} \) LPF (Ge substrate) and the intrinsic dispersion from the OPA, for example, the dispersion from the 1 mm thick GaSe. Nevertheless, it could be compressed back to a near-Fourier-limited pulse using bulk materials with proper dispersions. It is worth noting that as the signal and the pump are from the same TOPAS pump system, the CE phase fluctuation in the idler pulse is self-canceled through the DFG nature of the OPA [28, 29]. Recently, the stable CE phase of the idler pulse from a passively CE phase stable mid-IR OPA has been experimentally confirmed [17]. Thus, we, to the large extent, believe that the CE phase for our system is stable. However, its CE phase stability could be degraded by some factors such as the intensity-to-phase noise of the pump, and the temporal jitters between the pump and the signal [28, 29].} \]

\[ \text{Time (ps)} \]

\[ \text{Amplitude (a.u.)} \]

\[ \text{Wavelength (\mu m)} \]

\[ \text{intensity-to-phase noise of the pump, and the temporal jitters between the pump and the signal [28, 29].} \]

\[ \text{The pulse energy of the generated idler is measured as ~3.4 \mu J behind a 4.5 \mu m LPF, with ~300 \mu J pump energy. Considering the 85% transmission of the LPF and ~71% reflection from the silicon beam splitter for idler pulses, ~2% conversion efficiency from pump to idler is achieved. It is to be mentioned that for our OPA system, the measured energy of the optical parametric generation at the maximum pump energy is ~0.12 \mu J, and the energy fluctuation of the generated idler pulses is <0.1 \mu J, within 30 minutes measurement time. In addition, on the long-wavelength side of the generated idler, ~1.9 \mu J is measured using a 7.3 \mu m LPF.} \]
The evenly distributed pulse energy in the short and long wavelength bands of the generated idler spectrum further confirms the flat spectral span as measured in Fig. 4(a). The idler pulse energies with different LPFs as a function of the pump energy are measured. As shown in Fig. 5, no saturation occurs at the maximum pump energy, which explains the obtained moderate conversion efficiency. The beam profile behind the 4.5 μm LPF are also measured exhibiting a good Gaussian profile shown in the inset of Fig. 5. The good beam profile of the idler beam also indicates no back-conversion or crystal damage occurred. It is worth mentioning that with the rapid progress of ~2μm Ho:YLF or Ho:YAG OPCPA systems [18-20,30], the energy scaling up of the ultra-broadband mid-IR pulses is expected through the development of the GaSe-based OPCPA system.

Fig. 5. The dependence of the idler pulse energy on the pump energy behind the 4.5 and 7.3 μm LPFs. The inset is the beam profile of the idler pulse behind the 4.5 μm LPF at the output energy of ~3.4 μJ.

In conclusions, we report a GaSe-based mid-IR OPA system driven by ~2 pump pulses, with the passively stable CE phase by principle. The idler pulse has a flat spectrum spanning from 4.5 to 13.3 μm (at -3 dB) and 4.2 to 16 μm (full spectrum range). To the best of our knowledge, this is the broadest -3 dB spectrum from the OPA systems at this spectral region. The measured spectrum of the idler pulse supports a pulse width of ~19 fs, corresponding to 0.65-cycle electric field, centered at 8.8 μm. The energy of the idler pulse is measured as ~3.4 μJ with a conversion efficiency of ~2%. The developed multi-microjoule, few-cycle, mid-IR laser with a pulse is measured as ~3.4 μJ with a conversion efficiency of ~2%.
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