Narrowband carrier-envelope phase stable mid-infrared pulses at wavelengths beyond 10 μm by chirped-pulse difference frequency generation


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We report on the generation of narrowband carrier-envelope phase stable mid-infrared (MIR) pulses between 10 and 15 μm. High pulse energies and narrow bandwidths are required for the selective nonlinear excitation of collective modes of matter that is not possible with current sources. We demonstrate bandwidths of <2% at 12.5 μm wavelength through difference frequency generation between two near-infrared (NIR) pulses, which are linearly chirped. We obtain a reduction in bandwidth by one order of magnitude, compared to schemes that make use of transform-limited NIR pulses. The wavelength of the narrowband MIR pulse can be tuned by changing the optical delay between the two chirped NIR pulses.© 2017 Optical Society of America

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Optical pulses at mid-infrared (MIR) frequencies (6–20 μm wavelength) are a powerful tool to control the functional properties of solids and molecular systems [1,2], for example, by driving lattice vibrations to large amplitudes [3,4]. Typically, tunable MIR pulses between 10 and 20 μm are obtained by difference frequency generation (DFG) between two femtosecond near-infrared (NIR) pulses [5,6]. However, the broad bandwidths of the NIR pulses that are used in these cases translate into MIR pulses with relative bandwidths of 10%–30% \(\Delta \omega /\omega_0\). These are much larger than the vibrational mode linewidths in condensed matter, which are often of the order of a few percent only, resulting in poor spectral selectivity of the excitation. In order to selectively access such modes and, in particular, to separately drive closely spaced modes, it is necessary to reduce the MIR bandwidth by at least one order of magnitude.

The most straightforward method to reduce the bandwidth of either the interacting NIR pulses or of the resulting MIR light consists of linear spectral filtering by bandpass filters (such as Fabry–Perot filters) or by slits placed in the Fourier plane of a zero-dispersion pulse shaper [7]. However, this approach is intrinsically inefficient, since its energy loss is proportional to the achieved spectral narrowing. Narrowband pulses can be efficiently generated by nonlinear interaction between suitably chirped broadband pulses, allowing for frequency components with the same sum or difference to interact and directly transfer their energy into the narrowband output. This approach has been successfully applied in various spectral regions: narrowband visible pulses have been obtained via sum frequency generation of NIR pulses with opposite chirp [8]. Analogously, narrowband MIR pulses with wavelengths shorter than 10 μm have been generated by DFG between NIR pulses having chirp with the same sign [9–11]. However, the spectral region between 10 and 20 μm, of interest for vibrational control in solids, has not been addressed.

Furthermore, the narrowband MIR pulses generated so far were lacking CEP stability, a feature shown only in the THz range, at 300 μm wavelength [12]. However, this CEP stability is a big asset for the investigation of mid-infrared and THz vibrational control in condensed matter as it enables to explore the coherent perturbation of electronic, magnetic, or structural degrees of freedom on the sub-cycle phonon timescale [13], as well as phase-dependent effects [14].

In this Letter, we apply chirped-pulse DFG to generate narrowband, CEP-stable MIR pulses in the 10–15 μm wavelength range. We achieve pulses with relative bandwidths of \(\Delta \omega /\omega_0 = 1.6\%\) at a 12.5 μm wavelength, one order of magnitude narrower than the bandwidth obtained from the unchirped NIR pulses.
The MIR carrier wavelength is easily tuned by changing the delay between the chirped NIR pulses.

Figure 1 summarizes the principle of narrowband MIR pulse generation by DFG among chirped pulses. Panels (a)–(c) show the time-frequency Wigner distributions [15] of the interacting NIR pulses; the MIR light is generated at the difference frequency (DF) between frequency components interacting at the same time in the nonlinear medium. The MIR bandwidth can be estimated at the zero order as \( \Delta \Omega = \Omega_2 - \Omega_1 \) where \( \Omega_{1/2} \) is the frequency difference between the closest/farthest interacting frequency components. If the two NIR pulses are transform limited, all their spectral components interact simultaneously [see Fig. 1(a)], giving rise to the broadest MIR pulse [dotted line in Fig. 1(d)]. If the two NIR pulses are linearly chirped with different group delay dispersion (GDD), as depicted in Fig. 1(b), only a subset of their frequency components can interact, leading to a decrease in the MIR bandwidth [dashed line in Fig. 1(d)]. In this case, the MIR chirped pulses can be described as 
\[
E_{\text{MIR}}(t) = E_{01}(t) E_{02}(t) \exp[i(\omega_1 t + C_1 t^2/2) - i(\omega_2 t + C_2 t^2/2)],
\]
different chirp configurations; the MIR components are generated at the DF between frequency components interacting at the same time delay. This potentially leads to the generation of monochromatic MIR pulses. However, the DF spectrum gets inevitably broadened, since any cut at time \( t' \) of the time-frequency distribution of the NIR pulses has a finite bandwidth. A first-order estimation of the MIR bandwidth \( \Delta \omega \) [solid line in Fig. 1(d)] can be obtained by recalling that the MIR pulse cannot be longer than the interacting stretched NIR pulses. For this reason, fixing \( \Delta \omega \) corresponds to choosing the target MIR duration and, hence, the NIR stretching.

The experimental setup for the generation of narrowband MIR pulses is illustrated in Fig. 2. The NIR pulses were obtained from two-stage optical parametric amplifiers (OPAs), pumped with 100 fs, 800 nm pulses from a commercial Ti:sapphire regenerative amplifier at a 1 kHz repetition rate. The two OPAs were seeded by the same white light continuum; hence, they possess the same CEP fluctuations in their signal output pulses [16]. The MIR pulses generated by DFG are thus CEP stable [16–18], making it possible to measure their electric fields by EOS [5]. To this end, we used 20 fs gate pulses at a 900 nm wavelength, derived from a synchronized non-collinear OPA [19].

The NIR pulses were tuned to 1.31 (~229 THz) and 1.46 \( \mu \)m (~205 THz), with pulse energies of 330 and 310 \mu\)j, respectively. These were made to interact in a 600 \mu\)m thick GaSe crystal to generate MIR pulses at 12.5 \mu\)m (24 THz). At this wavelength, \( \Delta \omega/\omega_0 \approx 2\% \) corresponds to a pulse duration of about 1 ps, setting a lower limit for the duration of the chirped NIR pulses. The NIR wavelengths were tuned to above 1.2 \mu\)m to prevent two-photon absorption in GaSe.

The NIR pulses were chirped by linear propagation in highly dispersive transparent materials, which are far simpler to handle than gratings [8,9] or prism pairs [11], and introduce an easily reproducible dispersion. They are also easily inserted or removed from the optical setup to switch between the broadband and narrowband generation schemes, without affecting the footprint or the alignment. These materials should be transparent in the NIR, with a high enough dispersion to stretch the NIR pulses in a contained space. They should introduce suitable GDD and third-order dispersion (TOD), allowing for the matching of the two NIR chirps, and have a high bandwidth to prevent two-photon absorption. In Table 1, we show the calculated GDD and TOD for three readily available materials commonly used in the NIR: zinc selenide (ZnSe), cadmium telluride (CdTe), and silicon (Si). For any of these materials, rods of different thicknesses \( L_I \) and \( L_{II} \) are required to obtain the same GDD for the two different NIR frequencies, respectively. Once \( L_I \) and \( L_{II} \) are set, the material with the optimum dispersion properties is the one for which TOD\( I \) and TOD\( II \) are closest to each other, making TOD\( I/TOD_{II} \) a good figure of merit.
Table 1. Dispersion at the Pump (λ₁) and Signal (λ₂) Wavelengths Introduced by Propagation in 1 cm of Highly Dispersive Materials

| Material | \( \text{GDD/} \lambda [\text{fs}^2/\text{cm}] \) | \( \text{TOD/} \lambda [\text{fs}^3/\text{cm}] \) | \( \text{GDD/} \lambda [\text{fs}^2/\text{cm}] \) | \( \text{TOD/} \lambda [\text{fs}^3/\text{cm}] \) | \( \text{TOD/} \text{TOD} \)
<table>
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<tbody>
<tr>
<td>ZnSe</td>
<td>4741</td>
<td>4277</td>
<td>4131</td>
<td>3992</td>
<td>0.93</td>
</tr>
<tr>
<td>CdTe</td>
<td>10530</td>
<td>11500</td>
<td>8938</td>
<td>10080</td>
<td>0.96</td>
</tr>
<tr>
<td>Si</td>
<td>16865</td>
<td>60530</td>
<td>12330</td>
<td>17320</td>
<td>2.55</td>
</tr>
</tbody>
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*The last column is the ratio between the TOD and TOD/\( \lambda \) at plate thicknesses where GDD/\( \lambda \) = GDD/\( \lambda \). The dispersion was calculated with Sellmeier coefficients from [20–22].

Table 1 shows that ZnSe and CdTe are very similar from this point of view, while Si is significantly worse. We chose ZnSe as a dispersive element because of its smaller nonlinear refractive index, and because of its higher bandgap (2.8 versus 1.5 eV in CdTe) which minimizes two-photon absorption [23]. ZnSe rods of \( L_1 = 4.7 \) cm and \( L_{\text{opt}} = 5.2 \) cm thickness were used to obtain GDD of about 22,500 \( \text{fs}^2 \), which chirps the 60 fs NIR pulses to \( \approx 1 \) ps. The amount of dispersion could be easily doubled to GDD = 45,000 \( \text{fs}^2 \) by introducing two ZnSe rods into each NIR beam.

Spectral broadening due to self-phase modulation was minimized by decreasing the intensities of the two NIR beams with one ZnSe plate in each optical path. Dashed line: the target spectral bandwidth, 4.2 \( \text{μm} \) with 4.2 \( \text{μJ} \) of energy and around 50 \( \text{μm} \) m, obtained by measuring the transmission efficiency, without keeping into account further losses introduced by the optical elements. In our case, the energy efficiency was 35% and 15%, and can potentially be increased by the use of a thicker GaSe crystal or focused NIR beams.

The MIR beam radius in the focus was estimated to be around 50 \( \text{μm} \), obtained by measuring the transmission

Fig. 3. Experimental time-frequency Wigner maps of the NIR OPA pulses, retrieved from the measured SHG-FROG. (a) Pulses as generated (close to the transform limit). (b) Pulses chirped by one ZnSe plate in each optical path. Dashed line: the target spectral chirp with GDD = 22, 500 \( \text{fs}^2 \) and TOD = 20, 000 \( \text{fs}^3 \).

Fig. 4. Normalized EOS (a) traces and (b) spectra of the MIR pulses for different amounts of the NIR chirp. The data in panel (a) are offset for clarity, and the inset shows the EOS traces around 0 ps. The percentage in the legend of panel (b) indicates the relative bandwidth \( \Delta \omega/\omega_0 \).
through a calibrated 75 μm pinhole. This value was constant for all the measurements, indicating that the beam quality is not affected by the NIR stretching setup. For the 0.6 μJ, 1.65 ps pulses discussed above, this corresponds to a 1.9 MV/cm peak electric field.

Delay-dependent frequency tuning of the MIR output is reported in Fig. 5(c), where the red and blue solid lines depict the narrow spectra obtained for different NIR pulse delays (with one ZnSe rod pair), rescaled according to the corresponding measured energy and normalized to the maximum pulse energy (1.4 μJ). Indeed, if the delay between the incoming pulses is changed, the subset of frequencies that can interact at any time t in the DFG is shifted and, hence, their difference frequency changes [11]. In addition, the pulse energy of the narrowband MIR light scales with the shape of the broadband spectrum (black line) because changing the NIR delay affects the amount of interacting frequency components.

An illustrative example is shown in Fig. 5(a), where the delay between the NIR pulses is such that only the frequencies close to each other (red arrow) interact. Since the chirp of the NIR pulses is still the same, the narrow MIR bandwidth remains, but the central frequency is ΩMIR,1 on the low-frequency region of the corresponding broadband pulse. Similarly, Fig. 5(b) shows the situation where the pulse delay only allows for the interaction of the NIR frequency components farther from each other (blue arrow), resulting in the ΩMIR,2 central wavelength, on the high-frequency wing of the corresponding broadband pulse.

Tunability of the MIR source can be also be obtained by tuning the OPA wavelengths, allowing us to optimize the broadband pulse.

In summary, we reported on the generation of narrowband CEP-stable MIR pulses in the mid-infrared wavelength range between 10 and 15 μm (20–30 THz) by DFG between chirped NIR pulses. The technique allowed us to easily reduce the MIR bandwidth by inserting highly dispersive ZnSe rod pairs in the MIR beam paths. We generated MIR pulses with bandwidths down to 1.6% Δω/ω0, a MV/cm peak electric field, and a good time-bandwidth product. These pulses were tunable by delaying the chirped NIR pulses with respect to each other. We expect these pulses to find important applications in the coherent vibrational control of solids.

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**REFERENCES**