Tunable Few-Cycle Mid-IR Pulses towards Single-Cycle Duration by Adiabatic Frequency Conversion

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Abstract: Using adiabatic difference frequency generation, we generate Fourier-limited, few-cycle, tunable 2-4-μm mid-IR pulses at μJ-level, with controllable amplitude and phase by shaping before conversion from the near-IR, paving the way for arbitrary single-cycle mid-IR waveforms.
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The fields of nonlinear infrared spectroscopy and strong-field laser-matter interaction science call for intense and ever-shorter sources of mid-IR light [1,2], from few-cycle down to single-cycle duration. In addition, experiments in these fields often demand spectral amplitude and phase control. Recently [3], we demonstrated the generation of an energetic, coherent, and octave-spanning mid-IR spectrum covering 2-5 μm by downconverting a broadband, chirped near-IR pulse in a quasi-phase-matching structure with an aperiodic poling period satisfying the conditions for adiabatic frequency conversion [4]. We achieved >80% photon number conversion and one-to-one spectral amplitude transfer from the near-IR to the mid-IR, highlighting the capability of adiabatic frequency conversion to overcome the usual efficiency-bandwidth trade-off in wave mixing. However, as of yet there has been no experimental verification of the spectral phase transfer also expected in a chirped-pulse adiabatic conversion process, or compression of the coherent mid-IR spectrum to a near transform-limited pulse.

In this proof-of-principle study, we demonstrate compression of the mid-IR idler generated via adiabatic difference frequency generation (ADFG) to transform-limited duration by exploiting the near-IR to mid-IR spectral phase transfer inherent to the process with a narrowband pump. This allowed construction of an amplitude- and phase-shaped mid-IR source capable of generating few-cycle pulses tunable from 2.2-3.7 μm with ~1-μJ energy. Our pulse characterization device has so far limited us to confirmation of 4-cycle pulses, though our measurements indicate a flat spectral phase supporting a 2-cycle pulse and the possibility of single-cycle pulse generation. This result was made possible by adding only an ADFG stage and a bulk Si pulse compressor to the chirped output of a near-IR optical parametric chirped pulse amplifier (OPCPA). Since the spectral phase of the mid-IR idler is simply the combination of the phase on the input near-IR signal, the material dispersion in the ADFG crystal, and a constant conversion phase, control of the mid-IR spectral phase was achieved using a near-IR pulse shaper in the OPCPA.

Our experimental setup (Fig. 1) is the same modified OPCPA used in [3], with the addition of a bulk Si compressor. A Nd:YLF chirped pulse amplifier pumped both a 2-stage noncollinear optical parametric amplifier (NOPA) and the ADFG crystal, with a Ti:sapphire oscillator front end. Initially, numerical propagation simulations of the DFG process were performed to characterize the phase imparted by the ADFG crystal, from which it was concluded that dispersion is dominated by the material dispersion in $LiNbO_3$. This is complicated by the fact that in ADFG different wavelengths are converted at different longitudinal locations in the crystal, resulting in a significant 'effective dispersion' that depends sensitively on the longitudinal variation of the poling period, Λ(z). Our dispersion management scheme was designed based on the numerical simulations, which showed that the spectral phase could be well approximated by $\phi(\lambda) = L_{\mathrm{NIR}}(\lambda)k_{\mathrm{NIR}}(\lambda) + L_{\mathrm{MIR}}(\lambda)k_{\mathrm{MIR}}(\lambda)$, a combination of the material dispersion accumulated before and after frequency conversion, and approximating instantaneous conversion for each wavelength where it is exactly phase matched. For the generated mid-IR pulses in this experiment, this spectral phase was a smooth function that imparted ~1 ps group delay difference across the spectrum, which could be nearly

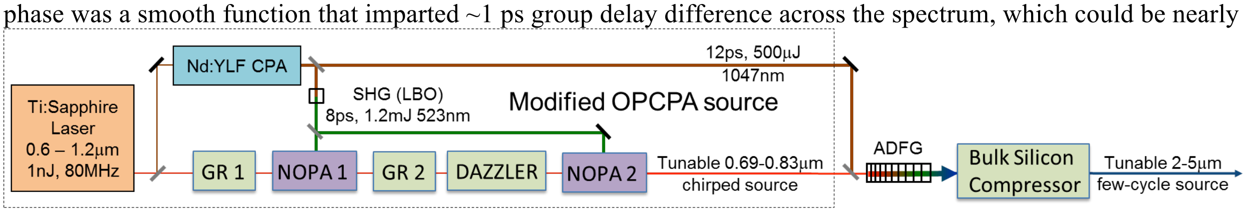

Fig. 1. Experimental setup. Nd:YLF CPA: 12-ps, 1-kHz, 4-mJ chirped pulse amplifier; GR1: grism pair GVD = -6300fs^2; NOPA1: 5-mm BBO; GR2: grism pair GVD = -15000fs^2; Dazzler (Fastlite): GVD = ~12000fs^2; NOPA2: 3-mm BBO; ADFG: aperiodically poled, 20-mm MgO-doped congruent $LiNbO_3$ grating; Bulk Silicon Compressor: 25-mm, single pass.

fully removed by pre-compensation with traditional near-IR dispersion management techniques.

Our mid-IR source is a modification of a near-IR OPCPA, with the compressor replaced by the combination of an ADFG crystal and a bulk Si compressor. We adjusted the dispersive properties of the Dazzler to pre-chirp the near-IR pulse such that after conversion to the mid-IR and propagation through the Si compressor the pulse would be fully compressed. Care was taken while choosing compressor length and Dazzler settings to maintain a good temporal overlap between interacting pulses in both the ADFG and second NOPA stages. The resulting compressed pulses were characterized in a homebuilt second-order interferometric autocorrelator, which uses either a 0.1-mm thick BBO crystal (for 2-μm operation) or a 0.4-mm thick AGS crystal (for 2.5-4.5-μm operation) for second harmonic generation (SHG), and an ext-InGaAs detector. The generated mid-IR spectrum can span from nearly 2-5 μm at -10 dB from peak, which supports a single-cycle pulse. However, the phase-matching bandwidth of the AGS autocorrelator crystal limited accurate autocorrelation measurements to ≥4-cycle pulses. Thus, we selected three narrower mid-IR spectral bands using the Dazzler to amplitude-shape the near-IR spectrum before conversion (Fig. 2(a,b)). The measured autocorrelation traces are plotted in Fig. 2(c-e), along with the expected autocorrelations assuming perfect SHG phase matching and a perfectly compressed (Fourier-limited) pulse. The transform limits of the measured spectra are 31 fs at 2.2 μm, 46 fs at 3.0 μm, and 49 fs at 3.7 μm, each ~4 optical cycles. The good match between the measured and expected autocorrelation traces shows there is an insignificant amount of spectral phase variation on the compressed mid-IR pulses. Thus, the pre-chirp imparted on the near-IR signal prior to conversion (experimentally optimized using the Dazzler by applying a quartic polynomial phase) compensated the spectral phase on the mid-IR pulse imparted by the adiabatic converter crystal and Si compressor. Note, an arbitrary phase could be added to the mid-IR pulse, if desired, by tuning the Dazzler. Finally, an autocorrelation trace of the pulses with the full 2-5-μm mid-IR spectrum lasts less than 4 interference fringes (Fig. 2(f)), indicating that a flat spectral phase exists at least over a bandwidth corresponding to a ~2-cycle pulse. Upon the soon arrival of a thinner autocorrelator crystal, we expect verification of a near-single-cycle compressed pulse.

In conclusion, we have demonstrated a few-cycle mid-IR source that delivers ~μJ energy 4-cycle pulses tunable from 2-4 μm, with spectral phase and amplitude profiles that can be electronically tuned using a Dazzler. This first demonstration of compression of an ADFG source also confirms that the spectral phase of the near-IR signal is transferred to the mid-IR idler, and the additional phase imparted by the adiabatic conversion process is smooth and easily removed using traditional dispersion management techniques. After improvement of our characterization device, we anticipate the demonstration of an energetic, amplitude- and phase-tunable, single-cycle, mid-IR source.

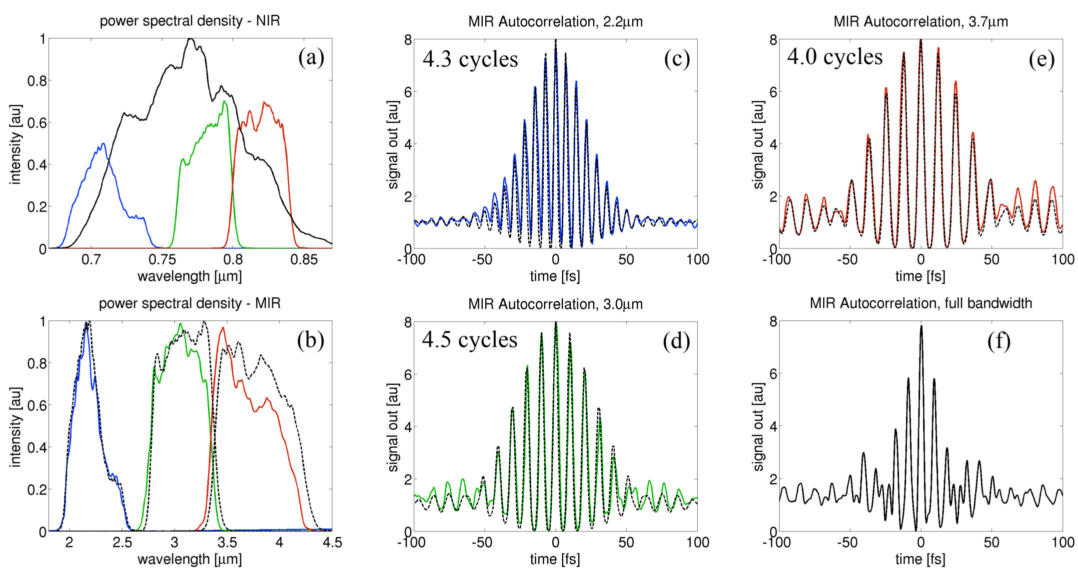

Fig. 2. Measured near-IR spectrum (a), expected mid-IR spectrum (b, dashed), measured mid-IR spectrum (b, solid curves), and autocorrelation traces (c-f), measured (solid) and expected as computed from the measured mid-IR spectrum, assuming perfect compression of the mid-IR pulse (dashed), for 4 different input spectra (blue: 2.2 μm, green: 3.0 μm, red: 3.7 μm, black: full bandwidth).

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