Abstract: We demonstrate adiabatic downconversion of a 110-nm band from a Ti:sapphire oscillator to the infrared, spanning 1550-2450 nm, with near-100% efficiency. The technique can potentially deliver multi-octave-spanning pulses for parametric amplification.

OCIS codes: (320.7110) Ultrafast nonlinear optics; (190.4975) Parametric processes

Today’s demand for octave-spanning-bandwidth sources of coherent optical pulses—at wavelengths other than the Ti:sapphire (Ti:S) oscillator’s 800-nm centered band—is widened by the need for seed pulses for ultrabroadband optical parametric amplifiers (OPAs). Today’s broadband OPAs include wavelength multiplexing schemes that coherently synthesize few-cycle OPA pulses of several colors to generate high-energy sub-cycle waveforms [1], thus requiring a multiple-octave-spanning seed spectrum. In such systems, seed pulses are generated from Ti:S oscillator pulses extended to other spectral domains by sum- or difference-frequency generation (DFG). For example, in [1–3], 2-μm optical parametric chirped-pulse amplifier (OPCPA) systems employ the downconversion of a broadband Ti:S pulse to the mid-IR via intrapulse DFG. This method has poor efficiency, a result of the tight focusing and transform-limited duration needed to reach an intensity high enough for nonlinear interaction, resulting in a short interaction length. The DFG pulse in these designs is only a few pJ in energy, less than 1% of the Ti:S power. This has practical consequences, since the low seed energy in these high-gain amplifiers is the root of severe superfluorescence noise contamination, effectively limiting the overall amplifier gain [4].

In this report, we have used instead the adiabatic DFG technique [5] to achieve near-100% conversion efficiency of a broadband Ti:S oscillator band to the infrared. Our experiment proves the principle of complete Landau-Zener (LZ) adiabatic transfer in nonlinear optical wave mixing for the first time, and does so for a broad 0.7-octave idler band. We predict that this is a modest result for the technique, which could potentially produce multi-octave-spanning mid-IR pulses with a suitable aperiodically poled nonlinear crystal. This report discusses an implementation ideally suited for the seeding of OPCPA systems, but the technique could be useful generally to provide nJ-energy broadband infrared pulses with up to MHz repetition rate.

The experiment employed a common setup for an OPCPA with octave-spanning Ti:S oscillator front end. The 1047-nm component of the oscillator injection seeded a 12-ps Nd:YLF CPA system [3], while the sub-1-μm wavelengths were available for frequency conversion and amplification. Up to 360 μJ of the available CPA pulse energy was used as the adiabatic DFG pump. This was recombined collinearly with the 630-940 nm portion of the Ti:S pulses (spectrum shown in Fig. 1a) in a dichroic mirror. These short-wavelength components of the Ti:S laser serve as the DFG “signal”, and were spatially and temporally overlapped with the 1047-nm pump in a 20-mm aperiodically poled potassium titanyl phosphate (APKTP) crystal, with domain periodicity varied from 15.6 to 21.9 μm along a crystal length of 20 mm to induce a third-order polynomial change of the phase mismatch parameter. This design satisfies the constraints imposed by the adiabatic inequality [5] for conversion of a signal range of 600–760 nm to an idler range of 1405–2800 nm with a 1047-nm pump. Prior to combination, the signal pulses were stretched in 80 mm of SF10, which provided 7.3 ps of group delay difference between the 630 and 740 nm wavelengths to avoid the effect of signal-idler group-velocity walkoff. The pump beam and slightly larger signal beam were Gaussian and roughly collimated. 360 μJ of pump energy produced a 3.5-GW/cm² intensity.

Figure 1 shows the collected idler spectrum (Fig. 1b) and the internal conversion efficiency as a function of pump intensity (Fig.1c). The spectrum, measured with an extended-InGaAs CCD-based grating spectrometer, covers 1550–2450 nm at -20 dB relative to peak, with lower than expected measured conversion above 2400 nm attributed to the diminishing sensitivity of the CCD. The observed preservation of the signal spectral shape seen in the idler is inherent to a saturated adiabatic DFG process. The idler power was measured by a PbSe amplified photodetector (Thorlabs PDA20H). The idler was filtered by 1 cm of Si and a long-pass filter at 1430 nm to fully block the collinear pump beam and any residual signal, thus ensuring collection of idler wavelengths only. This was verified by a null photodiode response in the case of a large delay between pump and signal pulses. The response of the
photodetector and all collection and filtering optics in the path of the idler beam were independently calibrated at 1.9 µm (the peak of the idler radiation) with pulses from a 1-kHz OPCA system [3]. Accounting for an additional 68% of measured losses from the remaining optics in the idler beam path (CaF_2 lens, Si and long-pass filters, 6 silver mirrors), the photodiode response indicated that 214 nW of DFG power was transmitted through the AKTP crystal, or 214 pJ/pulse. The transmitted pulse energy of the Ti:S oscillator in the phase-matched 600-760 nm band was 224 pJ. Thus, accounting for all measurement uncertainties in the conversion of photodiode voltage to idler power, the signal to idler conversion was 96 ± 7%. Fig. 1c shows that the signal to idler conversion efficiency as a function of pump intensity fits extremely well the exponential form of the LZ model of adiabatic population transfer. In this experiment, the conversion saturates at 95%, well within the 7% experimental error of the theoretical 100%. The Fig. 1c inset shows the results of finite-difference simulations of the adiabatic DFG conversion as a function of wavelength. The predicted efficiency as a function of intensity also matches the experimental data extremely well. Note, the insensitivity of conversion efficiency to pump intensity in saturation allows full conversion even with bell-shaped pump beam and pulse profiles, an important practical advantage of the adiabatic downconversion technique.

![Diagram](CTh3B.3.pdf)

Fig. 1. (a) Ti:S oscillator spectrum transmitted through the dichroic mirror. (b) Measured DFG idler spectrum (solid) and expected spectrum (dashed) based on simulated conversion of the signal spectrum in (a) to idler with a pump wavelength of 1047 nm and 2.5 GW/cm² intensity (see c, inset). Shaded regions indicate the signal and idler bands chosen for conversion in the AKTP crystal. (c) Measured DFG conversion efficiency plotted versus peak pump intensity. Error bars indicate relative measurement error. Shaded curve indicates a 7% uncertainty in photodiode voltage to idler power conversion. Solid line: exponential fit A(1-exp[B/l]), accounting for all measurement uncertainties in the conversion of photodiode voltage to idler power, the signal to idler conversion was 96 ± 7%.

Considering applications, the scheme used here is immediately suited for seeding a degenerate OPCA with narrowband 1-µm pump and chirped 2-µm signal as in [3], and the amplified pulses can be subsequently compressed to provide a high-energy, few-cycle source. Alternatively, the idler pulses could be compressed immediately. In such a scheme, a higher pulse-rate pump laser would be desirable. For example, bulk or thin-disk amplifiers can provide ~0.01-1 mJ, sub-ps pulses at 100-kHz to few-MHz repetition rate (e.g., [6,7]). Used as the DFG pump, they could provide 10s of mW of DFG power and many Watts of few-cycle pulses when further amplified by the same pump in an OPA. We note, while KTP begins absorbing at 2.8 µm, lithium niobate could potentially be used to extend the idler spectrum beyond 4 µm (requiring the additional phase-matching of the 760-830-nm Ti:S range), and further extension into the mid-IR range would require identification of suitable quadratic materials for poling. Thus, the adiabatic downconversion technique used here can potentially be used to generate multiple-octave spanning spectra.

In summary, broadband downconversion of Ti:S oscillator pulses to a 0.7-octave idler band with near-100% efficiency has been achieved by an adiabatic DFG process, proving for the first time the principle of complete adiabatic transfer in nonlinear optics. The method is well suited for the seeding of ultrabroadband OPA systems.