Fully efficient adiabatic frequency conversion of broadband Ti:sapphire oscillator pulses

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By adiabatic difference-frequency generation in an aperiodically poled nonlinear crystal—a nonlinear optical analog of rapid adiabatic passage in a two-level atomic system—we demonstrate the conversion of a 110 nm band from an octave-spanning Ti:sapphire oscillator to the infrared, spanning 1550 to 2450 nm, with near-100% internal conversion efficiency. The experiment proves the principle of complete Landau-Zener adiabatic transfer in nonlinear optical wave mixing. Our implementation is a practical approach to the seeding of high-energy ultrabroadband optical parametric chirped pulse amplifiers.   © 2012 Optical Society of America

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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). Such systems today include wavelength multiplexing schemes that coherently synthesize few-cycle OPA pulses of several colors to generate high-energy subcycle waveforms [1], and these require a multiple-octave-spanning seed spectrum. Ti:S oscillator pulses, extended to other spectral ranges by sum- or difference-frequency generation (SFG, DFG), are often used. For example, in [1–4], 2 μm optical parametric chirped pulse amplifier (OPCPA) systems amplify a broadband Ti:S pulse converted to the mid-IR via intrapulse DFG. This method has poor efficiency, which is 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 seed energy in these systems is only a few pJ, 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 [5].

In this letter, we report near-100% conversion of a broadband Ti:S oscillator band to the mid-IR using the adiabatic DFG technique [6], thus proving the principle of complete Landau-Zener (LZ) adiabatic transfer in nonlinear optical wave mixing. The complete conversion succeeds for the full temporal and spatial extents of the Ti:S beam. Moreover, it covers a 0.7 octave idler band and is potentially scalable to multiple octaves from a single nonlinear crystal. Here we discuss an implementation ideally suited for the seeding of OPCPAs, but the method could be useful generally for providing nJ energy broadband infrared pulses with up to an MHz repetition rate.

Adiabatic frequency conversion applies the principle of rapid adiabatic passage (RAP) for full transfer of population between states of a 2-level atom to optical frequency conversion, a concept explored in several previous works [6–9]. Notably, in the mixing of frequencies, $\omega_3 = \omega_1 - \omega_2$, where $\omega_1 > \omega_2, \omega_3$, through a quadratic electric susceptibility, the equations of motion are isomorphic to the driven optical Bloch equations for a 2-level atom (neglecting radiative losses) when the wave of frequency ω_2 is strong and undepleted during propagation [6–8]. In the first examples of the technique, adiabatic SFG was demonstrated with a narrowband tunable source seeded and pumped by an optical parametric oscillator [6,7]. Later, the method was used for broadband conversion of an amplified Ti:S pulse with up to 50% internal conversion efficiency, converting an 80 nm pulse to a 40 nm pulse centered at 450 nm via adiabatic SFG and a 30 nm pulse to a 400 nm pulse centered at 3.15 μm via adiabatic DFG [8]. These first experiments showed only partial adiabatic transfer in accordance with the LZ theory.

In our work, the "pump" is the middle-frequency wave, ω_2, whereas the "signal" is the high-frequency wave, ω_1. Our system operates with a strong, undepleted ω_2 pump wave, whereas previously an undepleted-pump condition was often applied to ω_1. This latter condition was shown to be useful; e.g., as a means of pulse shaping of the idler in DFG [10] and as a way to allow arbitrarily broad gain bandwidth in an OPA with undepleted pump [11]. We apply the former condition, making conversion between ω_1 and ω_3 analogous to population transfer in RAP possible and allowing 100% conversion efficiency in DFG or SFG.

In our experiment (see Fig. 1), an octave-spanning 85 MHz Ti:S oscillator (Idesta QE) served as the front end. Its longer wavelengths, reflected from a dichroic
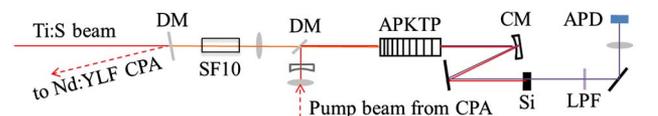

Fig. 1. (Color online) Experimental setup. DM = dichroic mirror, SF10 = 8 cm SF10 prism, CM = collimating mirror, Si = 1 cm Si prism, LPF = long pass filter, APD = PbSe amplified photodiode.

mirror, seeded a 1047 nm Nd:YLF CPA system [3], while its shorter wavelengths, transmitted through the mirror, served as the DFG signal. The CPA delivered 4 mJ, 12 ps Gaussian pulses at a 1 kHz repetition rate. Up to 9% of the energy was used as the DFG pump, recombined collinearly with the signal by means of a second dichroic mirror and sent to a 20 mm aperiodically poled potassium titanyl phosphate (APKTP) crystal. Prior to combination, the signal pulses were stretched in 80 mm of SF10 glass, which provided 7.3 ps of group delay difference between the 630 and 740 nm wavelengths to avoid signal-idler, group-velocity walk-off. The pump beam was Gaussian, elliptical, and collimated, with 590 and 990 μm e^{-2} beam diameters. The signal beam was Gaussian and slightly expanding, with mean beam diameters of 250 and 290 μm.

Using the quasi-phase matching (QPM) technique, we designed an adiabatic 2 cm APKTP grating that satisfies the constraints imposed by the adiabatic inequality [7] for conversion of a signal range of 600 to 760 nm to an idler range of 1405 to 2800 nm with a 1047 nm pump. In order to induce an adiabatic longitudinal change of the effective total phase mismatch parameter, defined as $\Delta k(z) = k_2 + k_3 - k_1 - G(z)$, we chose the function $G(z) = -90z^2 - 560z - 3350$ [1/cm], where z (in cm) is measured from the center of the crystal. This design optimized the uniformity of the adiabatic conversion rate across signal frequencies. The QPM period, $\Lambda(z) = 2\pi/G(z)$, ranged from 15.6 to 21.9 μm. Numerical finite difference simulations for a pump intensity of 2.5 GW/cm^2 predicted an ~100% conversion to an idler range of 1.5 to 2.5 μm in saturation (see Fig. 4, inset).

Initially, conversion of the signal to idler was verified by monitoring the transmitted signal power with a Si photodiode, filtered by an 11.5 nm wide bandpass filter centered at the 680 nm peak of the signal spectrum. The 1 kHz clock signal from the pump CPA was used to trigger the photodiode signal on an oscilloscope for monitoring each signal pulse synchronized to a pump pulse. Once overlap of signal and pump in the APKTP crystal was achieved, a roughly 90% depletion of the signal in the range of the bandpass filter was measured (Fig. 2).

Figure 3 shows the idler spectrum, measured with an extended-InGaAs CCD-based grating spectrometer. The power spectrum covers 1550 to 2450 nm at −20 dB relative to peak, with the lower than expected measured conversion above 2400 nm attributed to diminishing sensitivity of the CCD. The observed transfer of the signal

spectral shape to the idler is inherent to a saturated adiabatic DFG process, and illustrates an equal conversion rate for all idler wavelengths.

In order to verify the conversion dynamics were those of a complete adiabatic transfer, we measured the response of a PbSe photodiode to the generated idler power while varying the pump intensity. We filtered the idler beam with a 1 cm Si plate and a 1430 nm long-pass filter to fully block the collinear pump beam and any residual signal, ensuring collection of idler wavelengths only. The transmitted idler beam was collected by a CaF$_2$ lens and fully focused onto a PbSe-amplified photodetector (Thorlabs PDA20H). Figure 4 shows the PbSe photodiode response to the incident idler power plotted against peak pump intensity. In the adiabatic limit, the LZ theory for adiabatic passage predicts a conversion efficiency [7],

$$\eta_{LZ} = 1 - \exp(-4\kappa^2/\pi|d\Delta k/dz|), \tag{1}$$

where $\kappa^2 = 2.232 d_{eff}^2 I_2/n_1 n_2 n_3 \lambda_1 \lambda_3 c$; d_{eff} is the effective second-order susceptibility in pm/V; I_2 is the pump intensity in MW/cm^2; n_1, n_2, n_3, are signal, pump, and idler indices of refraction; λ_1 and λ_3 are signal and idler wavelengths in cm; $c = 3 \times 10^{10}$ cm/s; and the sweep rate $|d\Delta k/dz|$ is in cm^{-2}. The exponent has a linear dependence on I_2 and on d_{eff}^2, and is inversely proportional to $|d\Delta k/dz|$. An exponential fit, $\eta(I_2) = A(1 - \exp(BI_2))$, of the data demonstrates outstanding confidence of fit with the LZ theory and shows the photon number conversion of signal to idler power asymptotically approached 100%. The best fit exponential parameter ($B = -2.72 \pm 0.05$ cm^2/GW) is within 20% of the experimental value for the peak idler wavelength of 1.9 μm (−2.3 cm^2/GW), which is remarkable considering both the variation in $|d\Delta k/dz|$ over the full bandwidth range and the ~40% variation in local pump intensity across the full spatial and temporal extent of the overlapped signal. The observed insensitivity of efficiency to pump intensity in saturation is an essential practical advantage of adiabatic compared to conventional DFG, as it allows full
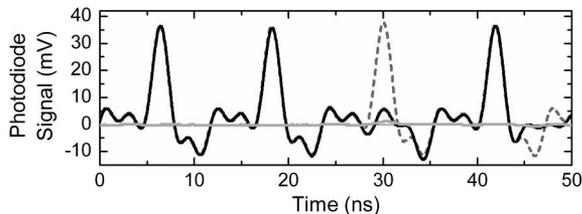
Fig. 2. Oscilloscope traces of Si photodiode response to the 674 nm to 686 nm band of the Ti:S signal pulses transmitted through the APKTP crystal with (dashed line) >20 ps delay or (dark solid line) 0 delay between the pump and signal pulses. The depletion of the synchronized Ti:S pulse at 30 ns is ~90%. Note, blocking the Ti:S pulse reveals a slight leakage of the strong pump pulse into the photodiode (light solid line).
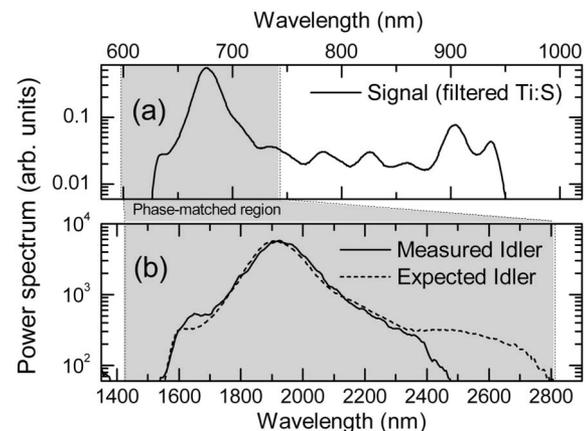
Fig. 3. (a) Ti:S oscillator spectrum transmitted through the dichroic mirror. (b) Measured DFG idler spectrum (solid) and expected spectrum (dashed) based on simulated conversion of the signal spectrum in (a) to idler with a pump wavelength of 1047 nm and 2.5 GW/cm^2 intensity. The shaded regions indicate the signal and idler bands chosen for conversion in the APKTP crystal.
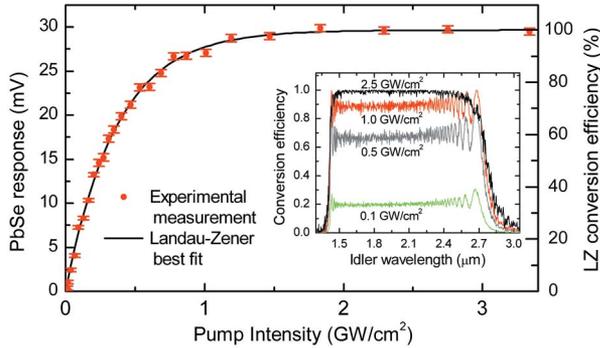
Fig. 4. (Color online) Measured PbSe photodiode response plotted versus peak pump intensity. Error bars indicate the relative measurement error. Solid line: exponential fit $A(1 - \exp[BI_2])$ of the experimental data. The adjusted $R^2 = 0.998$. (Inset: Predicted conversion efficiency versus idler wavelength for several pump intensities based on numerical simulations.)

conversion even with bell-shaped pump beam and pulse profiles. With $|d\Delta k/dz| = 561$ cm^{-2} for $\lambda_3 = 1.9\,\mu$m in our experiment, the condition for adiabatic transfer [7], $|d\Delta k/dz| \ll \kappa^2$, reduces to $I_2 \gg 580$ MW/cm^2. This corroborates the apparent requirement for full conversion of $I_2 \gtrsim \sim 2$GW/cm^2 that can be inferred from Fig. 4.

This measurement shows we reached the *maximum* conversion that can be achieved from any nonlinear crystal in a difference frequency conversion process, as compared to $<50\%$ achieved previously in adiabatic frequency conversion [8]. The only practical losses of the conversion apparatus were few-percent Fresnel reflections from the APKTP and SF10 prism faces, which could be mostly eliminated by means of antireflection coatings.

Considering applications, the scheme used here is immediately suited for the seeding of a degenerate OPCPA with a narrowband $1\,\mu$m pump and chirped $2\,\mu$m signal, as in [2–4], 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, ps pulses at a 100 kHz to few-MHz repetition rate (e.g., [12,13]) operating at 10 to 100 W of average power. Used as the DFG pump, they could provide tens of mW of DFG power and many Watts of few-cycle pulses when further amplified by the same pump in an OPA. We note that the DFG idler spectral phase contains two polynomial components, one related to the poling periods of the quasi-phase-matched crystals [14] and the other due to material dispersion. Both can be removed using conventional linear compression methods.

The adiabatic DFG technique could be used potentially to generate multiple-octave-spanning spectra. While KTP begins absorbing at $2.8\,\mu$m, lithium niobate could be used to extend the idler spectrum beyond $4\,\mu$m, while further extension into the mid-IR would require identification of suitable quadratic materials for poling. A Ti:S oscillator

with a flatter spectral profile (employing, for example, an inverse-gain output coupler [15]) would result in a more even spread of spectral power density. Finally, since the signal and pump pulses are derived from the same oscillator pulse, we expect a vanishing carrier envelope phase of the idler up to the relative phase error of signal and pump caused by slow environmental fluctuations in the Nd:YLF CPA. An active low-frequency feedback loop could be implemented for stabilization if necessary.

In conclusion, broadband conversion of Ti:S oscillator pulses 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 can be used for transferring an octave-spanning Ti:S spectrum throughout the infrared and is well suited for seeding ultrabroadband OPA systems.

References

1. S. W. Huang, G. Cirmi, J. Moses, K. H. Hong, S. Bhardwaj, J. R. Birge, L. J. Chen, E. Li, B. J. Eggleton, G. Cerullo, and F. X. Kärtner, Nat. Photon. **5**, 475 (2011).
2. T. Fuji, N. Ishii, C. Teisset, X. Gu, T. Metzger, A. Baltuska, N. Forget, D. Kaplan, A. Galvanauskas, and F. Krausz, Opt. Lett. **31**, 1103 (2006).
3. J. Moses, S. W. Huang, K. H. Hong, O. D. Mücke, E. L. Falcao-Filho, A. Benedick, F. Ö. Ilday, A. Dergachev, J. A. Bolger, B. J. Eggleton, and F. X. Kärtner, Opt. Lett. **34**, 1639 (2009).
4. K. H. Hong, S. W. Huang, J. Moses, X. Fu, C. J. Lai, G. Cirmi, A. Sell, E. Granados, P. Keathley, and F. X. Kärtner, Opt. Express **19**, 15538 (2011).
5. C. Manzoni, J. Moses, F. X. Kärtner, and G. Cerullo, Opt. Express **19**, 8357 (2011).
6. H. Suchowski, D. Oron, A. Arie, and Y. Silberberg, Phys. Rev. A **78**, 063821 (2008).
7. H. Suchowski, V. Prabhudesai, D. Oron, A. Arie, and Y. Silberberg, Opt. Express **17**, 12731 (2009).
8. H. Suchowski, B. D. Bruner, A. Ganany-Padowicz, I. Juwiler, A. Arie, and Y. Silberberg, Appl. Phys. B **105**, 697 (2011).
9. N. B. Baranova, M. A. Bolshtyanskii, and B. Y. Zel'dovich, Quantum Electronics **25**, 638 (1995).
10. G. Imeshev, M. M. Fejer, A. Galvanauskas, and D. Harter, J. Opt. Soc. Am. B **18**, 534 (2001).
11. M. Charbonneau-Lefort, B. Afeyan, and M. M. Fejer, J. Opt. Soc. Am. B **25**, 463 (2008).
12. J. Neuhaus, D. Bauer, J. Zhang, A. Killi, J. Kleinbauer, M. Kumkar, S. Weiler, M. Guina, D. H. Sutter, and T. Dekorsy, Opt. Express **16**, 20530 (2008).
13. J. Limpert, F. Roser, D. N. Schimpf, E. Seise, T. Eidam, S. Hadrich, J. Rothhardt, C. J. Misas, and A. Tunnermann, IEEE J. Sel. Top. Quantum Electron. **15**, 159 (2009).
14. M. A. Arbore, A. Galvanauskas, D. Harter, M. H. Chou, and M. M. Fejer, Opt. Lett. **22**, 1341 (1997).
15. L.-J. Chen, M. Y. Sander, and F. X. Kärtner, Opt. Lett. **35**, 2916 (2010).