High-resolution indirect pulse shaping by parametric transfer

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The phase and amplitude profile of a shaped pulse in the visible is transferred to a pulse in the near-infrared via an optical parametric amplification (OPA) process. Complex shaped pulses, such as multiple-pulse trains and pulses with high-order phase chirp, are produced at 1.2 μm. Theoretical conditions necessary for high-fidelity parametric shape transfer are discussed. Similar schemes can be implemented for other OPA systems pumped at near-infrared wavelengths to generate high-resolution shaped pulses in the mid-infrared.

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High-resolution pulse-shaping capabilities exist for wavelengths ranging from the visible to the near-infrared (NIR).[1,2] There is much interest in generating mid-infrared (MIR) shaped pulses, which are useful for various applications, such as extending ultrafast coherent control studies to vibrational systems of chemical and biological molecules. Although direct amplitude shaping of MIR pulses has been performed by use of a patterned amplitude mask in the spectral domain,[3] direct phase and amplitude pulse shaping have not been demonstrated. The main difficulty is the lack of appropriate materials for the usual direct methods of pulse shaping. Liquid-crystal modulators absorb in the MIR; acousto-optic materials such as TeO_2 and GaP, although they are transparent, have low diffraction efficiency. One solution is to shape pulses in the visible or NIR wavelengths and transfer the shape to a pulse of another wavelength by a nonlinear optical process. There have been some recent successes with this approach.[4–6] In all these cases, the shaped pulses are the result of nonlinear optical process between two pulses with similar bandwidth. The resolution of the final pulse shape may be limited, as it is a nontrivial convolution between the two input pulses. In this Letter we demonstrate high-fidelity transfer of a broadband pulse shape to another pulse of a different wavelength by an optical parametric amplification (OPA) process pumped by a pulse with a narrower spectral bandwidth. Noncollinear optical parametric amplification[7–9] (NOPA) of shaped pulses in the visible[10] concurrently produces shaped pulses in the NIR. This scheme can be directly applied to an OPA system that is pumped in the NIR (~800 nm) to produce high-resolution shaped pulses in the MIR. Similar concepts have been applied to sum-frequency-generation experiments that produce shaped pulses in the ultraviolet.[11]

In an OPA process, the idler pulse that is generated as the difference-frequency component between a monochromatic pump and a broadband signal pulse acquires a group-delay profile with frequency dependence opposite to that of the signal. In practice, it is more convenient to use an ultrashort pulse stretched to an appropriate length as a pump pulse instead of a quasi-monochromatic source. This stretching can be achieved by, for example, use of a diffraction grating pair. Although this linear frequency chirp does not affect the phase profile of the amplified signal pulse, it does affect the phase profile of the idler pulse. We present a simple discussion to describe the effect of the pump pulse's chirp on the deviation in shape that the idler pulse acquires from the signal. For a NOPA process with broadband phase matching, the generated idler electric field $E_i(\omega)$ can be approximated as[12]

$$E_i(\omega) \propto i \int_{-\infty}^{\infty} \mathrm{d}\omega' {E_s}^*(\omega') E_p(\omega' + \omega) . \qquad (1)$$

We let both the pump- and the signal-pulse electric field bear linear frequency chirps and assume the form $E_x(\omega) = \exp[-(\alpha_x - i\beta_x)(\omega - \omega_x)^2]$, where $x = p$ and $x = s$ denote the pump pulse and the signal pulse, respectively, β is the group-velocity dispersion (GVD), and α is related to the pulse's spectral FWHM Δf_x by $\alpha \equiv 2 \ln 2/(2\pi\Delta f_x)^2$. The acquired GVD of the idler pulse from the resultant expression of relation (1) is

$$\beta_i = -\mathrm{Im}\left[\left(\frac{1}{\alpha_s + i\beta_s} + \frac{1}{\alpha_p - i\beta_p}\right)^{-1}\right] \cdot \qquad (2)$$

In the limit of a monochromatic, zero-chirp pump pulse ($\alpha_p \gg \beta_p$), Eq. (2) gives $\beta_i = -\beta_s$; i.e., the idler pulse obtains the negative GVD of the signal pulse. For a nonmonochromatic pump pulse, if $|\beta_p| \gg |\alpha_p|, |\alpha_s|, |\beta_s|$, we obtain from Eq. (2)

$$\beta_i \approx -\beta_s - \frac{{\beta_s}^2 - {\alpha_s}^2}{\beta_p} \cdot \qquad (3)$$

Relation (3) implies that the larger β_p is, the smaller is the deviation of the imparted shape from that at the monochromatic pump limit, but this needs to be balanced with the necessary intensity threshold for the OPA process to occur, as well as the length and shape of the shaped pulse that one desires. The pump-pulse usage can be maximized by introduction of a significant basic GVD in the signal pulse to match its length with that of the pump pulse. Any desired modulation is then added on top of this GVD. The resultant idler pulse will bear the opposite (negative) of this basic GVD as well as the desired modulation. This basic negative GVD can then be compensated for by normal

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material dispersion, giving a final pulse of the desired modulation. This strategy is not unlike chirped-pulse amplification and subsequent compression of shaped pulses in the NIR near 800 nm.[13,14]

Our setup is shown in Fig. 1. The two-stage NOPA setup is similar to the setup described in Ref. 10. A first-stage NOPA is used to produce broadband visible pulses, which are then shaped by an acousto-optic pulse shaper. This preamplified shaped pulse ($\sim$0.2 μJ) seeds a second-stage NOPA pumped by a pulse centered at 401 nm (30 μJ). This pulse has been stretched from $\sim$150 fs to $\sim$4 ps by a diffraction grating pair. The signal pulse is amplified to $\sim$3 μJ. Because of the phase matching in the optical parametric process, the idler beam ($\sim$80-nm bandwidth centered at 1210 nm) exits the β-barium borate (BBO) crystal vertically 13° from the pump, with an angular dispersion of 1.5°. Since in this case the paraxial approximation still holds, an $f = 120$ mm spherical mirror is used to collimate the beam in both the horizontal and the vertical directions. The idler's pulse-front tilt is not measured but can be calculated from the angular dispersion[15] to be $\sim$19°. A prism pair (fused silica) then compensates for the spatial chirp resulting from the collimation, yielding a shaped pulse energy of $\sim$0.8 μJ (pulse-to-pulse energy fluctuation, $\sim$5%). The energy throughput of the prism pair is $\sim$60% since the polarization of the idler is perpendicular to the direction of the spatial chirp. The exit-beam spatial profile is slightly elliptical because of the astigmatism caused by the vertical tilt of the collimating spherical mirror. Compensation for the angular dispersion can also be achieved by a combination of spherical mirror, diffraction grating, and cylindrical mirrors.[16]

We characterize both the signal and the idler shaped pulses with the cross-correlation spectrally and temporally resolved upconversion technique (STRUT). The setup for the characterization process was described elsewhere.[10] By tilting the crystal for phase matching and using a flipper mirror to select between the signal and the idler pulses, we obtain sum-frequency-generation (type I, 80-μm BBO) spectra as a function of delay between a reference pulse (10-nm bandwidth pulse at 800 nm) and the signal or the idler shaped pulses. From the resultant STRUT trace, the pulse group delay versus frequency plot can be recovered.

To demonstrate the capability of this scheme, we produced a pulse with a fourth-order dispersion (4OD). We used a pulse centered at 600 nm with a bandwidth of $\sim$25 nm ($\alpha_s = 80$ fs^2) to seed the NOPA stage. We programmed the acousto-optic pulse shaper (together with the setup's material dispersion) to impose a phase profile of GVD of 5400 fs^2 and a 4OD of 2,080,000 fs^4 on this signal pulse. The resulting idler pulse centers at $\sim$1215 nm. The experimental STRUT traces of the amplified signal and idler shaped pulses are depicted in Figs. 2(c) and 2(d), respectively. As the arguments above predict, the frequency dependence of the idler pulse's group delay bears the mirror image of that of the signal pulse. Theoretical STRUT traces in Figs. 2(a) and 2(b) are provided for comparison. Figures 2(e) and 2(f) depict the experimental spectrum and the recovered group delay of the signal and idler shaped pulse, respectively. The recovered phase information for the idler pulse has a GVD of -5350 fs^2 and 4OD of $-2,170,000$ fs^4. The pump pulse for the second-stage NOPA has a spectral bandwidth of 4.4 THz ($\alpha_p = 1800$ fs^2) and a GVD of $\beta_p = 135,000$ fs^2. Because of the 4OD, the GVD of the signal pulse, β_s, ranges from 5000 fs^2 at the center wavelength to $\sim$10,000 fs^2 at the wings of its spectrum. The conditions leading to relation (3)

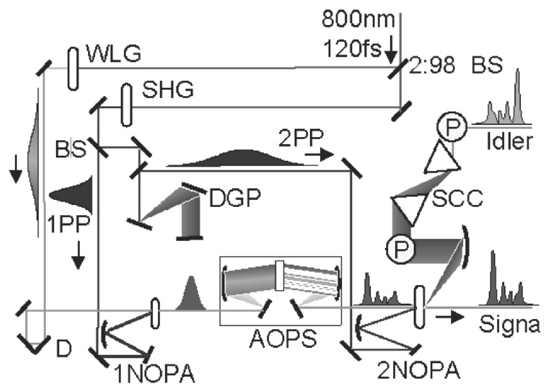

Fig. 1. Experimental setup of the two-stage NOPA system to produce shaped pulses in both the visible and the infrared. BSs, beam splitters; WLG, white-light generation (sapphire); SHG, second-harmonic generation (type I, BBO). D, delay. DGP, diffraction grating pair; AOPS, acousto-optic pulse shaper; 1PP and 2PP, first- and second-stage pump pulses, respectively. 1NOPA and 2NOPA, first- and second-stage noncollinear parametric amplification processes (type I, BBO), respectively; P's, periscopes; SCC, prism-pair spatial chirp compensator.

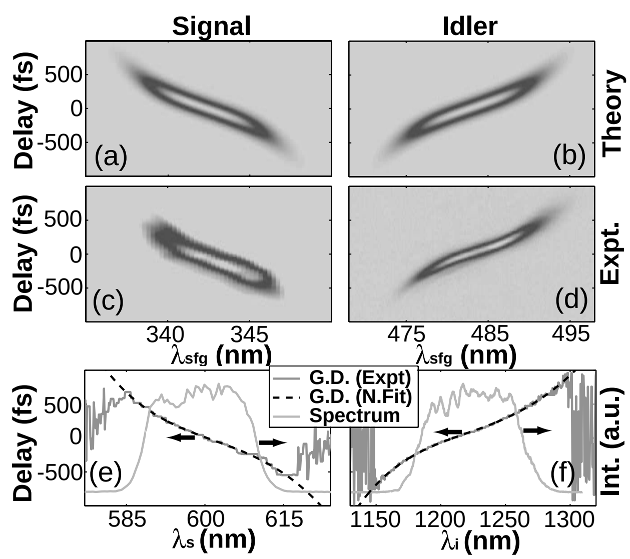

Fig. 2. (a), (b) Theoretical STRUT trace of the shaped signal and idler pulses, respectively. (c), (d) Corresponding experimental STRUT traces of the shaped signal and idler pulse, respectively. (e), (f) Spectra and the recovered group delay (G.D.) of the signal and idler pulses, respectively. Numerical fits (N.Fit) give a GVD of 5400 fs^2, a 4OD of 2,080,000 fs^4 for the signal pulse, a GVD of -5350 fs^2, and a 4OD of $-2,170,000$ fs^4 for the idler pulse. sfg, sum-frequency generation.

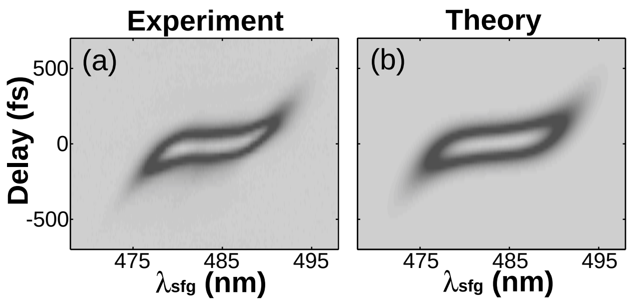

Fig. 3. (a) Experimental STRUT trace of the shaped idler described in Fig. 2 after the trace is passed through 4 mm of GaP crystal to compensate for the GVD, giving a shaped pulse with only 4OD. (b) Theoretical STRUT trace for comparison.

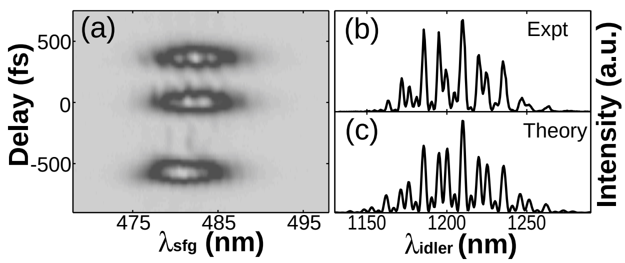

Fig. 4. (a) Experimental STRUT trace of a three-pulse train with delays of 600 and 400 fs between them. (b) Experimental and (c) theoretical spectra of the shaped idler pulse.

are hence satisfied. Relation (3) predicts that the deviation of the group delay of the idler shaped pulse will not deviate beyond that of the monochromatic pump limit by more than 7.5%. We compensate for the shaped idler pulse's basic negative GVD of $-5350\ \mathrm{fs}^2$ by passing the beam through 4 mm of GaP crystal. The fine tuning of the basic GVD to match the dispersion compensation can be done easily by programming of the acousto-optic pulse shaper. The STRUT trace of the resultant pulse shaped with a 4OD dispersion in Fig. 3(a) compares favorably with the corresponding theoretical STRUT trace in Fig. 3(b).

Other complex, high-resolution shaped pulses can also be produced. For example, we produce a three-pulse train with pulse delays of 600 and 400 fs and a controllable phase relationship between them. The STRUT trace is depicted in Fig. 4(a). Figure 4(b) shows the highly structured details of the experimental spectrum versus the theoretical spectrum of such a pulse train centered at 1210 nm with a spectral bandwidth of ~70 nm.

In conclusion, we have demonstrated a novel scheme to transfer a shaped pulse to another wavelength by a NOPA process. This can be achieved with high resolution. Our approach uses a narrow-band pump pulse with large linear chirp, which causes high-fidelity parametric broadband shape transfer in an OPA or difference frequency mixing process. We have also outlined general theoretical considerations regarding the relation between the pump pulse and the shaped idler pulse. Tunability of the NOPA system to 500 nm allows idler shaped pulses with wavelengths of up to 2 μm to be generated. This scheme can likely be extended to produce high-resolution shaped pulses in the MIR wavelengths. Other broadband phase-matched OPA systems (whether noncollinear or collinear), pumped at NIR (~800-nm) wavelengths,[17,18] can amplify shaped signal pulses in the NIR (0.95–1.10 μm). The corresponding shaped idler pulses will be in the spectroscopically interesting MIR 3–5-μm regime.

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References

1. A. M. Weiner, Rev. Sci. Instrum. **71,** 1929 (2000).
2. J. X. Tull, M. A. Dugan, and W. S. Warren, Adv. Magn. Opt. Res. **20,** 1 (1997).
3. R. A. Kaindl, M. Wurm, K. Reimann, P. Hamm, A. M. Weiner, and M. Woerner, J. Opt. Soc. Am. B **17,** 2086 (2000).
4. F. Eickemeyer, R. A. Kaindl, M. Woerner, T. Elsaesser, and A. M. Weiner, Opt. Lett. **25,** 1472 (2000).
5. N. Belabas, J.-P. Likforman, L. Canioni, B. Bousquet, and M. Joffre, Opt. Lett. **26,** 743 (2001).
6. T. Witte, D. Zeidler, D. Prach, K. L. Kompa, and M. Motzkus, Opt. Lett. **27,** 131 (2002).
7. T. Wilhelm, J. Piel, and E. Riedle, Opt. Lett. **22,** 1494 (1997).
8. G. Cerullo, M. Nisoli, and S. De Silvestri, Appl. Phys. Lett. **71,** 3616 (1997).
9. A. Shirakawa and T. Kobayashi, Appl. Phys. Lett. **72,** 147 (1998).
10. H.-S. Tan, W. S. Warren, and E. Schreiber, Opt. Lett. **26,** 1812 (2001).
11. M. Hacker, T. Feurer, R. Sauerbrey, T. Lucza, and G. Szabo, J. Opt. Soc. Am. B **18,** 866 (2001).
12. S. A. Akhmanov, V. A. Vysloukh, and A. S. Chirkin, *Optics of Femtosecond Laser Pulses* (American Institute of Physics, New York, 1992).
13. M. R. Fetterman, D. Goswami, D. Keusters, W. Yang, J.-K. Rhee, and W. S. Warren, Opt. Express **3,** 366 (1998), http://www.opticsexpress.org.
14. T. C. Weinacht, J. L. White, and P. H. Bucksbaum, J. Chem. Phys. B **103,** 10166 (1999).
15. J. Hebling, Opt. Quantum Electron. **28,** 1759 (1996).
16. A. Shirakawa, I. Sakane, and T. Kobayashi, Opt. Lett. **23,** 1292 (1998).
17. F. Rotermund, V. Petrov, and F. Noack, Opt. Commun. **169,** 183 (1999).
18. S. Cussat-Blanc, A. Ivanov, D. Lupinski, and E. Freysz, Appl. Phys. B **70S,** S247 (2000).