Flexible and rapidly configurable femtosecond pulse generation in the mid-IR

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Received September 17, 2002

We demonstrate a new experimental approach for flexible femtosecond pulse generation in the mid-IR by use of difference-frequency generation from two tightly synchronized Ti:sapphire lasers. The resultant mid-IR pulse train can be easily tuned, with an adjustable repetition frequency up to 100 MHz, a pulse energy of $\sim 1.5 \times 10^{-13}$ J, and an intensity noise similar to that of the Ti:sapphire. Rapid switching of the mid-IR wavelength and programmable amplitude modulation are achieved by precision setting of the time delay between two original pulses. © 2003 Optical Society of America

OCIS codes: 320.0320, 320.7110, 140.3070, 140.7090.

Mid-infrared (MIR) ultrashort light pulses are of primary importance for the study of ultrafast dynamics in chemical reactions, molecular vibrations, and the application of femtosecond IR spectroscopy to problems in solid-state physics. In particular, experiments in coherent control require a flexible, tunable source of MIR femtosecond pulses. New techniques for controlled pulse shaping in the MIR[1,2] in combination with such adaptable sources promise to make possible abundant progress in the science of coherent control.

High-repetition-rate sources are desirable for high-speed acquisition of data and signal averaging. Two methods have been employed for the generation of MIR pulses at high repetition rates, usually by frequency mixing in nonlinear crystals such as $AgGaS_2$, $AgGaSe_2$, or GaSe. In one type of experiment, 800-nm pulses directly from a Ti:sapphire (Ti:s) laser are used. An 82-MHz two-color Ti:s laser mixed in $AgGaS_2$ yielded 5-μW pulses tunable from 7.5 to 12.5 μm.[3] Mixing two ends of the spectrum from an 88-MHz broadband Ti:s laser produced 9–18-μm femtosecond pulses with an average power of 1 μW.[4] A 1-MHz cavity-dumped Ti:s laser employed the same technique to produce 5 μW of average power that was tunable from 7–20 μm.[5] These methods are limited by the fixed amount of power available in the spectral region accepted by the crystal for the mixing process and are tunable only via nonlinear crystals' phase-matching angles. A second type of experiment involves the use of an optical parametric oscillator system, in which the signal and idler waves, centered near 1.6 μm, are used for the difference-frequency generation (DFG) process. An 82-MHz (76-MHz) optical parametric oscillator system generated 500 μW (2 mW) of average power that was tunable from 2.5 to 5.5 μm (5.2–18 μm).[6,7] These systems have the advantage of producing a larger MIR power at the cost of increased complexity and greater initial pump power.

In this Letter we demonstrate a flexible, compact source for the generation of femtosecond MIR pulses by use of two highly synchronized passively mode-locked Ti:s lasers. The individual tunability of the two Ti:s lasers would in principle make possible DFG generation with spectral coverage from 3 μm down to the terahertz region. We recently synchronized pulse trains from two independent femtosecond Ti:s lasers to a timing jitter ~1% of the pulse duration.[8,9] These pulse trains, operating independently at separate wavelengths, are used for DFG in a GaSe crystal. Tuning the crystal's phase-matching angle as well as the wavelength separation of the two input pulses generates MIR pulses from 7.5 to 12.5 μm, with the measured upper (lower) tuning range limited by the MIR monochromator (the nonlinear crystal). A direct measurement of the MIR pulse duration shows <670-fs pulses at a wavelength of 11.5 μm. The average power of the pulses exceeds 15 μW, which to our knowledge represents the highest power in the >7.5-μm spectral range obtained from direct DFG of Ti:s pulses. More importantly, precision setting of the time delay between the two original chirped-pulse trains allows rapid switching of the MIR wavelength as well as programmable amplitude modulation.

A schematic of the setup is depicted in Fig. 1. Two passively mode-locked Ti:s lasers are each pumped at 2.5 W, producing ~100-MHz pulse trains. An average power of 200 mW per laser is generated while the lasers are operated at center wavelengths 20–40 nm on either side of 800 nm, with (uncompressed) pulse durations of ~140 fs. The shorter-wavelength laser is free running (master), whereas the longer-wavelength

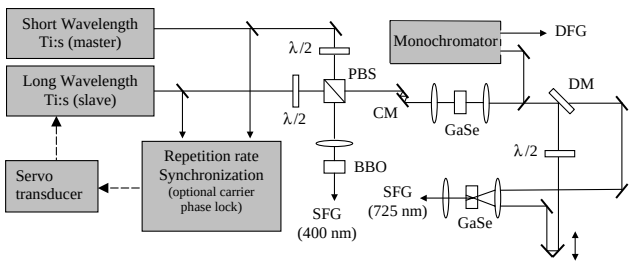

Fig. 1. Experimental system used to generate and characterize femtosecond MIR pulses: Two fs Ti:s laser with separate center wavelengths are tightly synchronized and are mixed in a 1-mm-thick GaSe crystal that is angle tuned for type I difference-frequency mixing. $\lambda/2$, half-wave plates; BBO, β-barium borate; CM, chirped-mirror pair; DM, dichroic beam splitter; PBS, polarizing beam splitter.

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laser (slave) utilizes both a fast piezo-actuated small mirror and a regular mirror mounted on a slow piezo with large dynamic range for active control of its repetition rate. Both the fundamental repetition rate (100 MHz) and its 140th harmonic (14 GHz) of each laser are used in a configuration with two phase locked loops.[8,9]

Two Ti:s beams are combined collinearly with orthogonal polarization into two arms. In one arm of the experiment, the pulse trains are focused into a thin β-barium borate crystal cut for type I sum-frequency generation (SFG), rotated 45° so that polarization components of both pulse trains propagate as ordinary beams in the crystal. The resultant SFG signal is used to characterize the timing jitter of the pulse trains when the timing offset corresponds to half a pulse width. In the other arm of the experiment, the pulses are given a slight negative chirp by use of two negatively chirped mirrors and focused ($f = 12$ cm) onto a GaSe crystal for DFG.

The 1-mm-thick uncoated GaSe crystal is cleaved normal to its z axis, since it is not yet possible to polish under arbitrary angles. This results in external incident angles of 51–74° for type I phase matching ($e - o = o$ for $\omega_{SW} - \omega_{LW} = \omega_{IR}$) of the 800-nm light. Even though such steep external angles entail large (>40%) reflection losses at the surfaces of the crystal, GaSe exhibits a higher nonlinear susceptibility and a wider transparency range than crystals such as $AgGaS_2$ or $AgGaSe_2$. Recent research with indium-doped GaSe also indicates a developing capability of producing arbitrary-cut GaSe crystals as well as antireflection coating.[10]

After the GaSe crystal, the MIR light is separated from the two collinear near-IR beams with a long-pass filter. The MIR beam's intensity is measured by use of a liquid-nitrogen-cooled HgCdTe detector. An IR monochromator can be used before the HgCdTe detector for spectral measurements of the MIR pulses. For temporal measurements of the MIR pulses, the shorter-wavelength Ti:s and the MIR DFG pulses are mixed in a second GaSe crystal by use of type I phase matching ($o + o = e$ for $\omega_{IR} + \omega_{LW} = \omega_{SW}$) for another SFG-based cross-correlation measurement. A photomultiplier tube combined with lock-in detection and suitable interference filters is used to measure the resultant SFG power as a function of the delay between the Ti:s and MIR pulses.

Figure 2 shows the cross-correlation data between the two Ti:s pulse trains, measured simultaneously in the SFG and DFG arms. The cross correlation is measured to be slightly shorter when the DFG signal is used because of the compression offered by the chirped-mirror pair; the differing optics in either arm of the experiment also cause slight differences in the temporal profiles of the pulses. The insets show timing-jitter measurements made while the locking system is activated, demonstrating similar timing jitter of 1.3 and 1.5 fs in a 160-Hz low-pass bandwidth when measured with SFG and DFG, respectively. Flexible DFG repetition rates are achievable by setting of the Ti:s lasers at different commensurable repetition frequencies.[8] Phase locking the carrier frequencies of the two Ti:s lasers would permit generation of optical spectra in three different regions with the desired phase coherence.[11]

The broad tunability in wavelength of our DFG scheme is depicted in Fig. 3(a). The Ti:s lasers are tuned to various separations, and the maximum MIR power obtainable at each separation is recorded as a function of the MIR wavelength. Figure 3(b) shows how this process is executed: At a given wavelength separation of the Ti:s beams, the spectra of the MIR pulses are recorded for various external angles (ranging from 59° to 71°) of the GaSe. Then the external angle that generates the greatest power

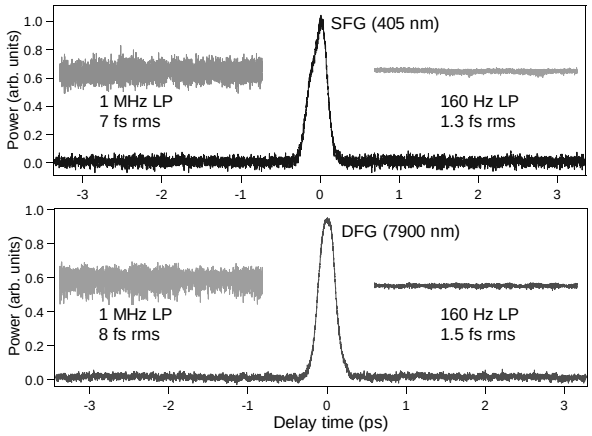

Fig. 2. Cross-correlation measurement of simultaneous SFG (top) and DFG (bottom). The insets show records of timing jitter (of a few second duration) determined from intensity fluctuations of the respective signals, observed through 1-MHz (left) and 160-Hz (right) low-pass filters when the two pulse trains are offset by half a pulse width.

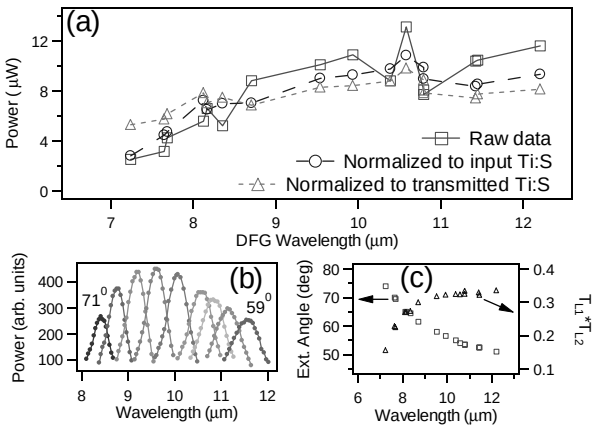

Fig. 3. (a) MIR average power, as a function of wavelength: squares, raw data corrected only for detector response; circles, normalized to the incident power of each Ti:s laser; triangles, normalized to exiting power of each Ti:s laser. (b) Spectra for various external phase-matching angles, all taken for the same separation of the Ti:s center wavelengths. Here, the peak point corresponds to one data point in (a). (c) Squares, external phase-matching angles versus MIR wavelength; triangles, product of the two laser transmissions through GaSe as a function of MIR wavelength.

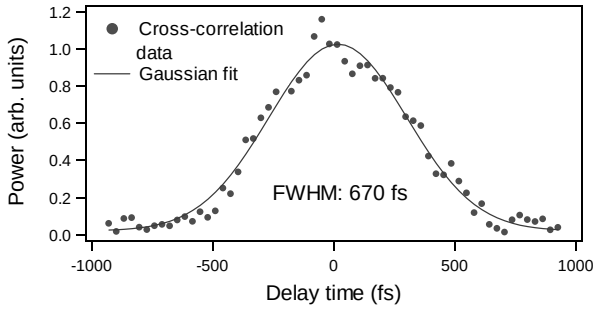

Fig. 4. Cross-correlation measurement of the 11.5-μm MIR pulse versus the shorter-wavelength Ti:s laser, showing a FWHM of 670 fs.

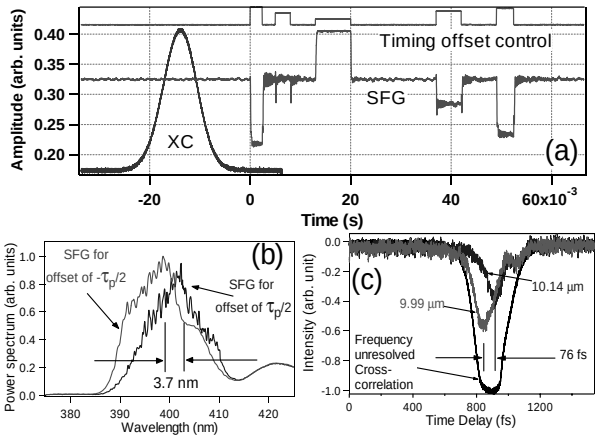

Fig. 5. (a) Arbitrary and programmable amplitude modulation of the SFG or DFG light. (b) Spectra of the SFG light for two different timing offsets by the differently chirped Ti:s pulse trains, illustrating a fast and programmable wavelength jump. (c) Frequency-resolved cross-correlations by use of the DFG signal for two different monochromator settings, while triggering the frequency-unresolved signal.

is used to record a point in Fig. 3(a). In Fig. 3(a), the squares represent the raw data, corrected only for the long-pass filter's response and the IR detector's response. The circles are normalized to the average power of each Ti:s beam just before the GaSe crystal, and the triangles are normalized to the average powers just after the GaSe crystal.

The data normalized to the input powers of the Ti:s beams (circles) represent the true capability of our system. More than 15 μW of MIR power has been generated above 9 μm, although less power is available at shorter wavelengths because of higher reflection losses at the GaSe surfaces at the steeper external angles necessary for phase matching. This effect is shown in Fig. 3(c), where the triangles (right-hand axis) represent the product of transmissions of both Ti:s beams through the GaSe crystal. The development of AR coating[10] for GaSe would allow for at least three times enhancement of the conversion efficiency of this system. The squares (left-hand axis) of Fig. 3(c) show the external phase-matching angle as a function of the desired MIR wavelength.

To illustrate the duration of the DFG pulse, in Fig. 4 we show the SFG-based cross-correlation signal (typically near 720 nm) between the DFG (11.5 μm) and the shorter-wavelength Ti:s (770-nm) pulses, with a measured FWHM of 670 fs. Taking into account the Ti:s pulse width before the second GaSe crystal, the group-velocity dispersion at 11.5 μm, and the group-velocity mismatch between the Ti:s and DFG pulses inside the GaSe, we note that the DFG pulse width is estimated to be less than 500 fs.

Using rapid switching of the delay time between the two original pulse trains, the dual-laser approach makes possible significant and unique flexibilities in the control of the DFG and SFG signals. When the Ti:s pulses are well compensated for with minimal chirps, changing the delay offset between these pulses will lead to controlled variations in the nonlinear signal amplitudes, while leaving the associated wavelength intact. This situation is illustrated in Fig. 5(a), showing the possibility of fast and precise switching of the nonlinear signal amplitudes. However, when the two lasers are chirped, we can use this to our advantage by precisely setting and (or) switching the DFG and SFG wavelengths, again with control of the timing offset. When the two lasers have similar chirp sizes and signs, a maximum amount of wavelength switching can be produced in the DFG signal. For SFG, chirps of opposite signs are preferred. Figures 5(b) and 5(c) show the wavelength-switching results in SFG and DFG, respectively. In this case, both lasers are positively chirped. We note that the wavelength-switching capability is limited chiefly by the phase-matching restrictions in the GaSe crystal.

We acknowledge experimental assistance and equipment loans from H. Kapteyn, M. Murnane, and L. Hollberg. The work was funded by the Office of Naval Research, NASA, the National Institute of Standards and Technology, and the National Science Foundation. J. Ye's e-mail address is ye@jila.colorado.edu.

References

1. F. Eickemeyer, R. A. Kaindl, M. Woerner, T. Elsaesser, and A. M. Weiner, Opt. Lett. **25,** 1472 (2000).
2. N. Belabas, J. P. Likforman, L. Canioni, B. Bousquet, and M. Joffre, Opt. Lett. **26,** 743 (2001).
3. M. R. X. de Barros, R. S. Miranda, T. M. Jedju, and P. C. Becker, Opt. Lett. **20,** 480 (1995).
4. R. A. Kaindl, D. C. Smith, M. Joschko, M. P. Hasselbeck, M. Woerner, and T. Elsaesser, Opt. Lett. **23,** 861 (1998).
5. R. A. Kaindl, F. Eickemeyer, M. Woerner, and T. Elsaesser, Appl. Phys. Lett. **75,** 1060 (1999).
6. A. Lohner, P. Kruck, and W. W. Rühle, Appl. Phy. B **59,** 211 (1994).
7. S. Ehret and H. Schneider, Appl. Phys. B **66,** 27 (1998).
8. L.-S. Ma, R. K. Shelton, H. C. Kapteyn, M. M. Murnane, and J. Ye, Phys. Rev. A **64,** 021802 (2001).
9. R. K. Shelton, S. M. Foreman, J. L. Hall, H. C. Kapteyn, M. M. Murnane, M. Notcutt, and J. Ye, Opt. Lett. **27,** 312 (2002).
10. D. R. Suhre, N. B. Singh, V. Balakrishna, N. C. Fernelius, and F. K. Hopkings, Opt. Lett. **22,** 775 (1997).
11. R. K. Shelton, L.-S. Ma, H. C. Kapteyn, M. M. Murnane, J. L. Hall, and J. Ye, Science **293,** 1286 (2001).