Generation of broadly tunable sub-30-fs infrared pulses by four-wave optical parametric amplification

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We report on the generation of sub-30-fs near-IR light pulses by means of broadband four-wave parametric amplification in fused silica. This is achieved by frequency downconversion of visible broadband pulses provided by a commercial blue-pumped beta-barium borate crystal-based noncollinear optical parametric amplifier. The proposed method produces the IR idler pulses with energy up to ~20 μJ and tunable in wavelength from 1 to 1.5 μm. The shortest pulse duration is 17.6 fs, measured at 1.2 μm. © 2011 Optical Society of America

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Broadband, few-optical-cycle IR pulses are required for diverse studies in ultrafast IR spectroscopy, biomedicine, and laser–matter interactions. The demand for intense few-optical-cycle IR pulses has grown to urgency, due to the rapidly developing range of applications in high-harmonic generation, in connection with the evolving discipline of attosecond science [1], as well as for fundamental studies of new aspects of light and matter interactions [2]. Blue-pumped beta-barium borate (BBO) crystal-based noncollinear optical parametric amplifiers (NOPAs) serve as an indispensable tool for the generation of ultrashort signal pulses in the visible spectral range [3]. Although the idler wave, which lies in the IR spectral range, is readily produced by the amplification process, its use is almost impractical because of irregular spatial chirp. Moreover, BBO crystal-based NOPAs fail to support broadband phase matching for IR wavelengths using an 800 nm Ti:sapphire laser pump. Therefore, the NOPA principle with 800 nm pumping has been demonstrated using different nonlinear crystals, which provide suitable group velocity matching between the signal and idler wave, and thus broadband amplification: potassium titanyl phosphate (KTP) [4,5], bismuth triborate (BIBO) [6,7], lithium iodate [8], and periodically poled stoichiometric lithium tantalate (PPSLT) [9,10]; also see [11] for a review.

Alternatively, broadband amplification in the IR spectral range could be performed in gases by means of four-wave parametric processes in the filamentation [12] or guided-wave propagation regimes [13]. To this end, four-wave parametric amplifiers based on wide-bandgap solids provide broad gain bandwidth as well, supporting amplification of sub-10-fs pulses in the visible [14,15] and in the UV [16]. Moreover, using cylindrically focused beams, the energy of the pump pulse could be scaled up to millijoule level without the onset of optical damage. An important feature of four-wave interactions is that the wavelength of the signal wave is shorter than that of the pump, so the four-wave interaction allows either frequency upconversion or downconversion amplification, a process that is not possible with conventional $\chi^{(2)}$-based parametric amplifiers. Indeed, by taking advantage of this possibility, frequency upconversion of visible pulses has been recently demonstrated for achieving 10 μJ, 30 fs pulses in the UV [17] range.

In this Letter we present a proof-of-principle demonstration that allows broadband frequency downconversion of a commercial blue-pumped BBO crystal-based NOPA. The results presented here refer to a single-pass four-wave optical parametric amplifier (FWOPA) in fused silica, pumped at 800 nm and seeded by broadband visible pulses. With a NOPA signal tuning range of 550–700 nm, the frequency downconverted near-IR pulses cover a 1–1.5 μm range with sub-30-fs pulse duration and energy up to 20 μJ.

The configuration of the single-pass FWOPA is sketched in Fig. 1(a). The experiment has been performed using an amplified 1 kHz repetition rate, 130 fs Ti:sapphire laser (Spitfire PRO, Newport-Spectra Physics, California, USA) as a pump source. The pump pulse has been split in two: one part (up to 1.8 mJ) is used to

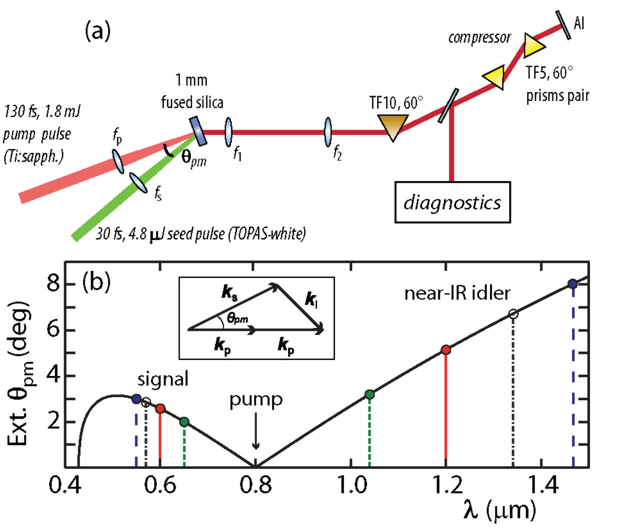

Fig. 1. (Color online) (a) Experimental setup and (b) phase-matching curve for the FWOPA pumped at 800 nm in fused silica. The wavelength of the seed pulse is continuously tunable. The highlighted circles represent four different experimental conditions, from 550 to 650 nm, with corresponding idler pulse wavelength from 1.47 to 1.04 μm. Inset shows the wave vectors diagram.

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directly pump the FWOPA, while the second part (0.5 mJ) is frequency doubled and serves to pump a commercial NOPA device (TOPAS-White, Light Conversion Ltd., Lithuania), which generates 30 fs pulses with a tunable wavelength between 550 and 700 nm. These visible light pulses are used as a seed for the FWOPA. The seed signal is then noncollinearly overlapped with the pump beam inside the 1-mm-long fused-silica sample. The precise value of the crossing angle, $\theta_{\rm pm}$, is dictated by the phase-matching conditions plotted in Fig. 1(b). The black solid line represents the phase-matching curve (external angles) for the four-wave mixing process, $k_p + k_p = k_s + k_i$, where $k(\omega) = \omega n(\omega)/c$ is the wave number and indexes p, s, and i stand for pump, signal, and idler wave, respectively. In order to achieve broadband phase-matching along the tuning range, we varied the external angle, $\theta_{\rm pm}$, from ~2 to 3 deg for four different input seed central wavelengths, namely, 650, 600, 570, and 550 nm [colored circles in Fig. 1(b)]. The corresponding amplified near-IR idler pulses are centered at 1.04, 1.2, 1.34, and 1.47 μm, respectively, and are highlighted in Fig. 1(b). We note that, due to the slope of the phase-matching curve, the output idler pulses are tilted with a pulse front tilt angle, $\delta = \arctan(\lambda d\theta/d\lambda)$, [18,19]. For example, around 1.34 μm, the pulse front tilt is $\delta = 9.86°$ (open black circle in Fig. 1(b)].

The pump and seed signal beams are focused with lenses, $f_p = +800$ mm and $f_s = +500$ mm, respectively, so as to ensure mode matching at the input facet of the fused silica sample and good spatial overlap along its entire length. The FWHM dimensions of the input beams are 795 μm and 325 μm for pump and seed signal, respectively. Finally, the output near-IR idler pulse has been imaged with a $4f$ system [$f_1 = +125$ mm, and $f_2 = +300$ mm in Fig. 1(a)] onto a TF10 glass prism with a 60° apex angle in order to remove the pulse tilt. After tilt removal, the pulse is compressed in a double-pass prism-pair compressor that uses 60° apex angle TF5 glass prisms at Brewster's angle. The IR pulse spectrum is monitored by a fiber spectrometer (QE65000, Ocean Optics); its spatial profile is recorded by an InGaAs complementary metal-oxide semiconductor (CMOS) camera (Xenics, Xeva 202); and its temporal profile is measured by a scanning autocorrelator.

Figure 2 shows the experimental results. When pump and seed pulses are temporally overlapped, the FWOPA supports broadband signal amplification and conversion into the corresponding near-IR idler pulse, as shown by the spectra plotted in Figs. 2(a) and 2(b), respectively, and according to the phase-matching relations of Fig. 1(b). The wavelength tunability is clearly visible, and the spectral width of the near-IR pulses at FWHM, ranging from 70 to 140 nm, supports sub-30-fs pulse duration along the whole tuning range and even sub-20-fs duration for the idler pulse centered at 1.2 μm [red solid line in Fig. 2(b)]. After the pulse front tilt removal and compression stage, we indeed measured almost transform-limited pulse durations for all the near-IR pulses. In Fig. 2(c) we show the autocorrelation of the compressed pulse at 1.2 μm. The retrieved pulse duration is 17.6 fs, which is fairly close to the transform-limited pulse duration supported by the spectrum, 15 fs FWHM. We note that the broad bandwidth at 1.2 μm and the resulting shorter pulse duration at this particular wavelength are the result of the broader bandwidth seed pulse at 600 nm generated by the commercial NOPA. This therefore shows that the solid-state FWOPA may support very large bandwidths. The largest bandwidth would be obtained for a nearly constant-angle interaction, i.e., by seeding at 510 nm (idler generated at 1.85 μm), while seeding at 443 nm would lead to an idler pulse at 4.14 μm with zero tilt, thus removing the need for the prism post-compensation stage; however, these wavelengths are beyond the tuning range of Topas-White. Finally, in Fig. 2(d) we show the spatial profile of the collimated near-IR beam after tilt removal, which has an FWHM width of 416 μm.

With seed signal energy of 4.8 μJ, we were able to generate up to 20 μJ sub-30-fs IR pulses across the whole 1–1.5 μm range. In particular, as shown in Fig. 3(a), in a single-pass FWOPA we can achieve near-IR signals of nearly 20 μJ with 1.8 mJ input pump energy and 1% pump-to-idler conversion efficiency. The results of fused-silica-based FWOPA are very attractive compared to those achieved in gaseous media in the guided-wave regime [12,13]. Based on previous studies, e.g., [16,17], we expect to be able to increase this conversion efficiency up to 10 to 15% by increasing the input seed energy; moreover, energy scaling could be achieved by simply increasing the beam size of the pump and signal waves. Note also that an increase in the input pump energy leads to a spectral broadening of the idler pulse [Fig. 3(b)], due to cross-phase modulation (XPM) between the pump and seed pulses inside the fused silica sample. We find that the XPM spectrally broadened pulses are still fully compressible, and indeed the measured pulse durations, after compressor optimization,

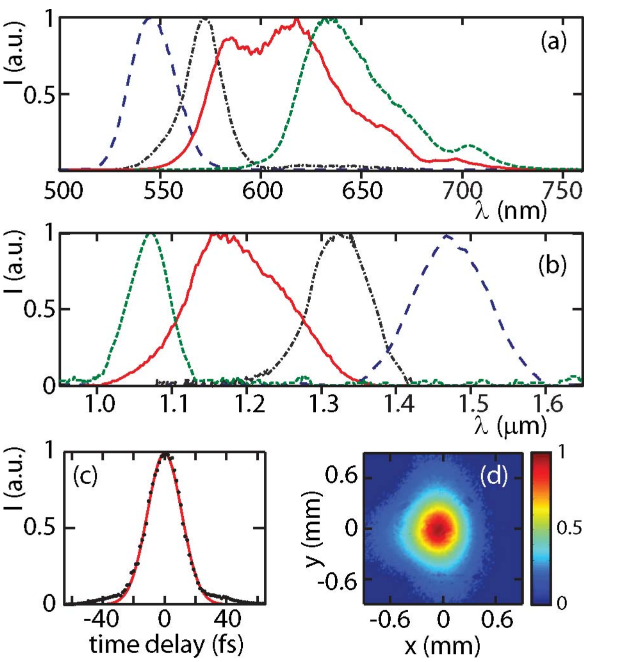

Fig. 2. (Color online) (a) Amplified signal and (b) corresponding near-IR idler spectra, along the tuning range from 1.04 to 1.47 μm. Broadband amplification is evident around 1.2 μm (red solid curve). (c) Autocorrelation trace of the compressed near-IR pulse centered at 1.2 μm. The measured pulse duration is 17.6 fs. (d) Spatial beam profile.

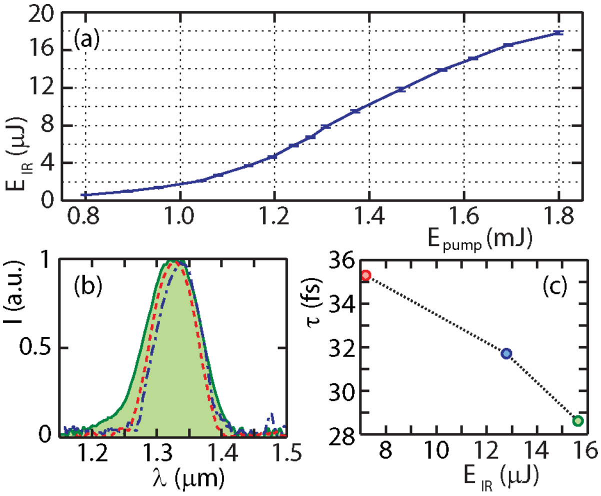

Fig. 3. (Color online) Near-IR pulse energy evolution (a) for a varying pump energy, from 0.8 to 1.8 mJ, and a fixed seed energy at 4.8 μJ. Near-IR spectra (b) and durations (c) at 1.34 μm, for three different output idler energies, namely, 7.15 (red dashed), 12.78 (blue dash-dotted), and 15.62 μJ (green solid).

are shortening with increasing pump energy from 35 fs to 28.6 fs, as shown in Fig. 3(c).

In conclusion, we have demonstrated a proof-of-principle method for broadband frequency down-conversion with a standard Ti:sapphire pumped visible NOPA, which can achieve up to 20 μJ, sub-30-fs pulses tunable in the 1 to 1.5 μm range. To the best of our knowledge, this is the first scheme based on a solid-state Kerr medium (fused silica) that allows visible-to-IR conversion and provides both very large tunability and simultaneous broadband amplification, supporting few-optical-cycle pulses. Moreover, it promises scaling to very high pump energy levels due to the availability of large aperture Kerr media and suitable conditions for frequency downconversion up to 10 μm, using materials with extremely broadband transparency range, such as CaF_2. Despite lower energy conversion efficiency, FWOPA provides very similar output characteristics to conventional three-wave parametric amplifiers based on birefringent crystals. The developed IR source might readily serve for diverse applications and might be considered as an energetic seed pulse with high spatial and temporal quality for front-end high-power IR laser systems [20].

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