Tunable pulses from below 300 to 970 nm with durations down to 14 fs based on a 2 MHz ytterbium-doped fiber system

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A noncollinearly phase-matched optical parametric amplifier pumped by a commercial 2 MHz fiber laser is presented and discussed. The pump system allows the direct generation of a seed continuum from a sapphire plate. Clean pulses with up to 860 nJ energy and down to 14 fs pulse length can be obtained over a fundamental tuning range from 620 to 970 nm. Conversion by second- and third-harmonic generation as well as sum frequency mixing results in an extended tuning range down to well below 300 nm. © 2008 Optical Society of America

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For laser imaging and machining, ultrashort pulses can render the necessary high intensities and fields. The investigation of ultrafast molecular dynamics requires tunable pulses in the sub-20 fs regime. To avoid sample damage in spectroscopy, for good signal-to-noise ratios, and for efficient machining, high repetition rates are needed. Megahertz, fiber-based femtosecond laser systems with pulse energies in the multimicrojoule regime promise a breakthrough for these applications. Recently, a fiber-laser-pumped parametric amplifier was reported that was seeded by a photonic crystal fiber and delivered tunable 250 fs pulses [1]. Noncollinear optical parametric amplifiers (NOPAs) pumped by Ti:sapphire amplifiers efficiently generate tunable visible sub-20 fs pulses at kHz repetition rates and pump energies above 100 μJ [2,3]. They were also demonstrated at 100 kHz and pump energies below 10 μJ [4], and lately even for a cavity-dumped Yb:KYW oscillator at 1 MHz [5]. In this Letter we present a novel NOPA design for a 2 MHz fiber laser system, with a white-light seed generated in a bulk medium. Our new NOPA provides the desired tunability in the visible and near-infrared (NIR) and accesses the 10 fs regime. In addition, we transfer these pulses into the blue and ultraviolet (UV) spectral region, which is essential for spectroscopic applications.

Figure 1(a) shows the NOPA, which is pumped by a commercial Yb-doped fiber-oscillator/amplifier system (IMPULSE; Clark-MXR, Inc.) delivering 10 μJ pulses at 1035 nm (frequency ω) with sub-250 fs length. To efficiently generate green pump light for the parametric process, the entire 1035 nm light is focused into a 800 μm thick type I BBO crystal. Doubling efficiencies up to 30% are achieved, while the remaining NIR pulse is still suitable for continuum generation. A dichroic mirror transmits the NIR part used for white-light generation in a 4 mm thick sapphire crystal with highly parallel faces. Out of several focal lengths tested, we find for the pump beam diameter of ~5 mm a length of 75 mm optimal for low threshold and stability, corresponding to a numerical aperture of 0.035. Due to the comparatively long pump, pulse energies of at least 2.5 μJ have to be used. In a static setup this energy in combination with the high repetition rate damages the sapphire crystal. By rotating the crystal [6] stable operation is achieved. A short-pass filter (Model 102927, LAYERTEC GmbH; 6.35 mm fused silica) blocks the fundamental and transmits the continuum part from 480 to 1000 nm [see Fig. 2(a)]. A 30 mm fused silica lens recollimates and focuses the continuum into a 5 mm type I BBO crystal cut at 26.5° for the para-

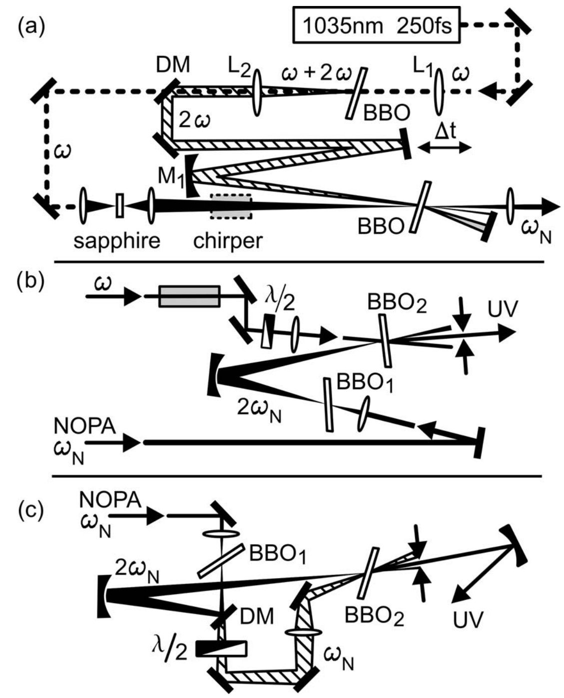

Fig. 1. (a) 2 MHz NOPA. (b) SFM of the doubled NOPA output with the fundamental of the pump. (c) THG of the NOPA. DM, dichroic mirror; L, lens; M, mirror.

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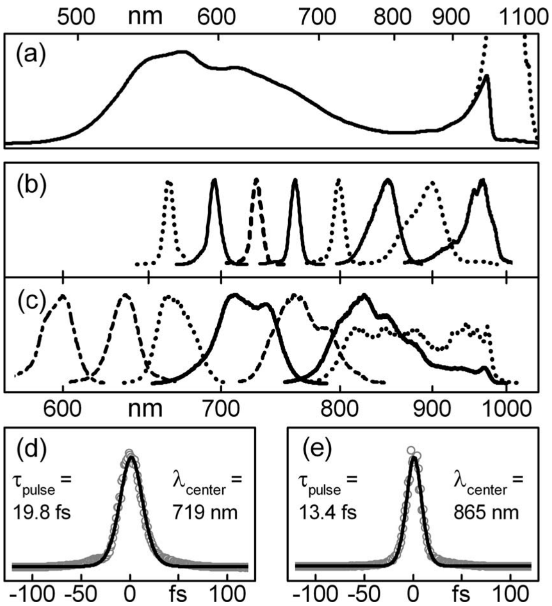

Fig. 2. (a) Spectrum of the seed continuum. (b) NOPA spectra with bandwidth reduced by 10 mm SF57 chirper. (c) NOPA spectra with full bandwidth [spectra of (d) and (e): solid curves]. (d), (e) Autocorrelation (with Gauss fit) at 719 and 865 nm, respectively. The vertical scale is linear in all panels.

metric amplification. The resulting chirp on the continuum corresponds to a group delay between the 650 and 900 nm spectral components of ~0.66 ps. The pulses are characterized with a fiber-coupled spectrometer (not intensity calibrated) and a dispersion-free autocorrelator [7].

The distance between the lenses L_1 and L_2 (both $f = 250$ mm) is adjusted such that the green beam has a diameter of ~2 mm on the focusing mirror M_1 ($f = 300$ mm). A focal diameter of 125 μm and a Rayleigh range of 25 mm are achieved allowing us to place the amplifier crystal within the Rayleigh range [4] and match the spot sizes of pump and seed. For broadband amplification a noncollinear geometry with an external angle of ~3.7° between the pump and the seed beam is employed [1,2]. The precise value is adjusted together with the phase-matching angle for the desired output wavelength. Due to the chirp of the seed only part of its spectrum overlaps temporally with the pump pulse in the crystal and is amplified. To tune the NOPA, the arrival time of the pump pulse is changed by translating a plane mirror. The NOPA output is collimated with an antireflection-coated lens ($f = 500$ mm), and the chirp of the pulses is corrected with a prism sequence. To reduce the bandwidth of the NOPA output the chirp of the white light can be increased by inserting LF7 or SF57 glass (chirper) into the white light (10 mm of SF57 increase the 650/900 nm group delay to ~2.6 ps). Most of the measurements were performed at 500 kHz; experiments at 2 MHz proved that the pulse bandwidth, duration, and spatial shape remain the same at higher repetition rates and the output energies are nearly equal for equal pump energies.

The smooth spectrum of the seed [see Fig. 2(a)] and the compressibility of the NOPA pulses are a direct consequence of the seed generation in bulk sapphire. The latter is only possible for the sub-250 fs pump duration, for longer pulses a photonic crystal fiber has to be used [1], resulting in a low degree of coherence. Figure 2(c) shows typical spectra of NOPA pulses at different center wavelengths. The tuning range extends from below 620 nm to beyond 970 nm. It is limited on the blue side by the absorption of the idler beam in the BBO crystal starting at about 2.7 μm and on the red side by the short-pass filter in the seed light.

Spectral widths of over 44 THz can easily be amplified, providing sufficient bandwidth for pulses with a transform limit around 10 fs [Figs. 2(c) and 3]. Pulse durations (assuming Gaussian pulse shape) of 13.4 fs at 865 nm and 19.8 fs at 719 nm are demonstrated with compression in fused silica prisms [Figs. 2(d) and 2(e)]. They correspond to time–bandwidth products of 0.51 and 0.57, close to the theoretical limit of 0.441. Due to the smooth spectra and the high coherence of the seed, the pulses show nearly no wings. If the seed is chirped with LF7 or SF57 glass, the spectral width is reduced by a factor of about 1.5 or 2.5 [Fig. 2(b) and Fig. 3]. Figure 3 gives an overview of output energies and Fourier limited pulse lengths for the various configurations. The possible durations decrease toward longer wavelengths due to the decreasing seed chirp and due to the increasing group velocity mismatch between the seed and the green pump in the 5 mm BBO crystal that allows the pump to sweep over the seed [group delay 650(o)/517(e) nm, ~0 ps; 900(o)/517(e) nm, ~0.35 ps]. Throughout the fundamental tuning range, energies well above 400 nJ are obtained, with a maximum of 860 nJ at 685 nm. At 2 MHz an average output power of 1.65 W has been observed.

Three schemes were applied to convert the NOPA output (frequency ω_N) to the blue and near-UV spectral region: second-harmonic generation (SHG, $2\omega_N$), sum frequency mixing (SFM, $2\omega_N + \omega$) and third-harmonic generation (THG, $3\omega_N$). For SHG, the NOPA output is focused ($f = 75$ mm) into a 200 μm type I BBO crystal [BBO_1, cut at 27.5°; see Fig. 1(b)] placed slightly behind the focus. For SFM [see Fig.

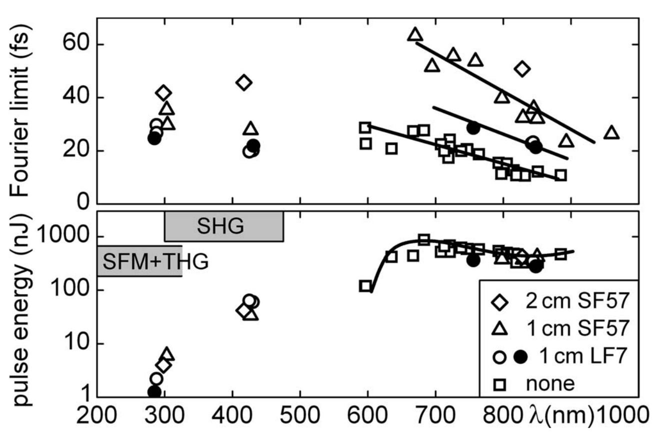

Fig. 3. Fourier limits and energies of the NOPA, SHG, SFM, and THG pulses with and without additional chirpers in the seed at 500 kHz (◦) and 2 MHz (•). Solid lines are guides to the eye.

1(b)] 400 nJ of the 1035 nm pump pulse are stretched by passing through 210 mm of SF57 glass [8]. The polarization is rotated and the beam is focused ($f = 150$ mm) into a 200 μm type-I BBO crystal (BBO_2, cut at 36°). The second harmonic (SH) of the NOPA is refocused by an aluminum mirror ($f = 150$ mm) and overlapped with the NIR pulse under a small angle to spatially separate the SFM pulse. For THG [see Fig. 1(c)] the SH part is reflected off a dichroic mirror and the polarization of the transmitted fundamental is rotated with an achromatic half-wave plate. The SH pulses are refocused by an aluminum mirror ($f = 150$ mm) and overlapped slightly noncollinearly with the focused ($f = 100$ mm) fundamental in a 200 μm type I BBO crystal (BBO_2, cut at 36°). For all conversion processes the seed was chirped with 10 mm LF7 to reduce the NOPA bandwidth and match the acceptance bandwidths of the crystals. Without sacrificing efficiency, 20% of the NOPA pulses can be split off prior to SHG to allow two-color experiments.

Tuning of the NOPA allows SHG from 310 to 485 nm, SFM from 238 to 330 nm, and THG from 206 to 323 nm. The conversion schemes were tested for selected wavelengths (see Figs. 3 and 4). A SHG conversion efficiency up to 21% is observed. For SFM an energy efficiency of 18% (SH to SFM) is achieved, i.e., a pulse energy of 6 nJ at 304 nm. The spectral bandwidth supports pulse lengths of less than 40 fs. Reducing the crystal length to 100 μm does not change the bandwidth but decreases the energy by a factor of ~10, because the NIR mixing pulse is too short to convert the whole SH pulse. Compression of the SH pulse should overcome this problem, but the compressor length has to be readjusted with the NOPA wavelength. For THG, an efficiency (SHG to THG) of 5% is achieved, i.e., energies up to 2.5 nJ. The spectral width of 12 THz at 288 nm supports a pulse duration of 36 fs. The bandwidth was slightly increased by using a 100 μm crystal, leading to a spectral width of 15.7 THz, but again at nearly tenfold lower energy.

The newly developed NOPA is to our knowledge the first parametric amplifier that can efficiently generate tunable sub-20 fs pulses from a purely fiber-based pump system. The minimum pulse length of 13.4 fs represents a ~20-fold shortening of the pump duration. The primary tuning range covering the red and

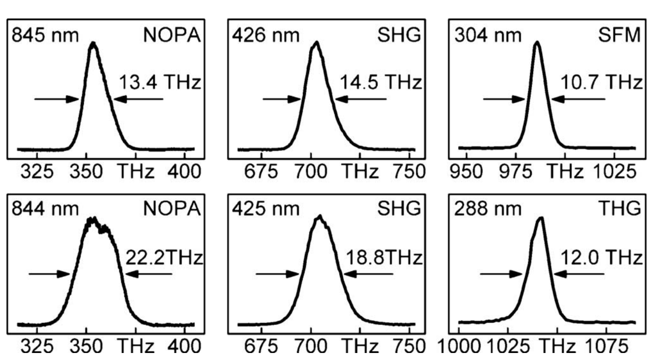

Fig. 4. Spectra of the NOPA (first column), SHG (second column), SFM (top right graph), and THG (bottom right graph) pulses.

NIR should be ideal for nonlinear imaging techniques. Compared with a MHz oscillator pump system [5], the setup is simplified, the output energy is increased by more than an order of magnitude, and the tuning is largely improved. The obtained pulse duration is similar to recent optical parametric chirped pulse amplifiers (OPCPA) in the TW regime [9,10] relying on the same amplifier concept. However, continuum seeding allows extended tunability compared with seeding with a large-bandwidth Ti:sapphire oscillator. The presented nonlinear conversion experiments demonstrate that the whole range down to below 300 nm is accessible and the system should therefore become a most valuable alternative for spectroscopic applications in femto-science. We expect to further increase the bandwidth of the pulses by reducing the chirp in the seed light (e.g., by chirped mirrors) and thus shorten their temporal width. By pumping with the tripled fiber-laser output, the full spectral range of the seed continuum will become available for amplification. In this way the primary NOPA tuning range would be widened to well below 500 nm, and UV pulses could then be generated directly by simple frequency doubling.

Note added in proof: Very recently a NOPA pumped by a 2 MHz Yb-doped fiber amplifier was reported. It delivers sub-20 fs pulses at a fixed wavelength of 800 nm [11].

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