Generation of 8.5-fs pulses at 1.3 μm for ultrabroadband pump-probe spectroscopy

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Abstract: We report on a near-infrared non-collinear optical parametric amplifier (NOPA) based on periodically poled stoichiometric lithium tantalate. The NOPA generates μJ-energy pulses with spectrum spanning the 1-1.7 μm wavelength range, which are compressed to nearly transform-limited 8.5 fs duration by a deformable mirror. By synchronizing this source with a sub-10-fs visible NOPA, we demonstrate an unprecedented combination of temporal resolution and spectral coverage in two-colour pump-probe spectroscopy.

OCIS codes: (320.7110) Ultrafast nonlinear optics; (190.4970) Parametric oscillators and amplifiers.

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1. Introduction

Femtosecond pump-probe spectroscopy is a powerful technique for the investigation of electronic and vibrational dynamics in a variety of atomic, molecular and solid-state systems. The study of primary photophysical and photochemical relaxation processes often requires extreme temporal resolution, thus calling for ultrashort pump and probe pulses with durations down to few cycles of the carrier wave. In addition, the need to excite a system on resonance and probe optical transitions occurring at different photon energies requires independent frequency tunability of pump and probe pulses, calling for the generation of few-optical-cycle light pulses with broadly tunable carrier wavelength.

Optical parametric amplifiers (OPAs), combining nonlinear spectral broadening and broadband amplification, not only add frequency tunability to a fixed-wavelength pulse, but they also enable one to dramatically shorten its duration [1]. The phase matching bandwidth $\Delta\omega$ of an OPA depends on the group velocity mismatch (GVM) between signal and idler $\delta_{si} = 1/v_{gs} - 1/v_{gi}$, where $v_{gs}(v_{gi})$ is the group velocity of the signal(idler). To the first order one can write $\Delta\omega \propto 1/|\delta_{si}|$ [1], so that broadband gain is achieved when the group velocities of the signal and idler are matched (δ_{si}=0). Such a condition, for example, is satisfied in the case of type I phase matching at degeneracy ($\omega_s = \omega_i = \omega_p/2$) [2, 3]. If $v_{gi}>v_{gs}$ ($\delta_{si}>0$), however, group velocity matching can be achieved in a non-collinear interaction geometry, in which the angle Ω between signal and idler wave vectors is such that the signal group velocity is equal to the projection of the idler group velocity along the signal direction: $v_{gs} = v_{gi}\cos\Omega$. This concept, first recognized by Gale *et al.* [4], is exploited in the so-called non-collinear OPA (NOPA).

The well established visible NOPA is based on a β-barium borate (BBO) crystal pumped at 400 nm by the second harmonic (SH) of a Ti:sapphire laser and seeded by a white-light continuum (WLC) generated in a sapphire plate [5-7]. It allows the generation of visible pulses with duration down to 4 fs [8], energy up to 300 μJ [9] and repetition rate up to 1 MHz [10]. Until recently, however, the NOPA concept had not been applied to wavelengths longer than 1 μm [11]. The commonly used BBO crystal, in fact, displays in this range $v_{gi}<v_{gs}$ for both fundamental frequency (FF) and SH pumping. This is due to the fact that BBO has a zero dispersion wavelength, corresponding to a maximum of the group velocity, at 1.38 μm. Thus the signal wavelength lies in a region of high group velocity while the idler is slower. For crystals with higher refractive index the zero dispersion point shifts to longer wavelengths, allowing non-collinear group velocity matching. Examples of such crystals are periodically poled stoichiometric $LiTaO_3$ (PPSLT) and $KTiPO_4$ (KTP). In a previous work [12] we introduced a near-IR FF-pumped NOPA based on PPSLT and demonstrated compression of a portion of its spectrum to 16 fs by a simple prism pair. In this experiment, our compression capability was limited by uncompensated third order dispersion. Near-IR NOPAs exploiting

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the same concept were later reported for KTP [13-15]; in this case 25-fs pulsewidths could be achieved.

In this paper we harness the full potential of the near-IR NOPA based on PPSLT, by generating μJ-energy pulses with spectrum spanning the 1-1.7 μm wavelength range, which are compressed to nearly transform-limited (TL) 8.5 fs duration by an adaptive system based on a deformable mirror (DM). We also demonstrate two-colour pump-probe spectroscopy with an unprecedented combination of spectral coverage and temporal resolution, by synchronizing the near-IR NOPA with a sub-10-fs visible NOPA.

2. Two-cycle pulse generation at 1.3 μm

Figure 1 shows a scheme of the near-IR NOPA and the adaptive compressor. The system is pumped by 80-μJ, 150-fs pulses at 785 nm and 1 kHz from a regeneratively amplified Ti:sapphire laser. A 2-μJ fraction of the pump is focused in a 2-mm-thick sapphire plate to generate a single-filament WLC, used as a seed. The PPSLT crystal (HC Photonics), doped with 1% MgO to prevent photorefractive damage, is 1.2-mm long and 1-mm thick and has a fan-out design, with poling period Λ varying from 20.5 to 23.7 μm over the 10-mm crystal width. This allows fine-tuning of the quasi-phase-matching condition to maximize the gain bandwidth. The crystal length is chosen to be equal to the pulse splitting length between pump and signal/idler. Pump and signal have the same extraordinary polarization, as required for a quasi phase-matched interaction in this crystal. The external pump-signal angle is set to $\alpha \approx 4.1°$, corresponding to an internal value ≈1.9°. The single-stage NOPA produces pulse energies in excess of 2 μJ, which are sufficient for applications to time-resolved spectroscopy. The rms pulse-to-pulse fluctuations were lower than 1% and no optical or photorefractive damage were observed during long-term operation.

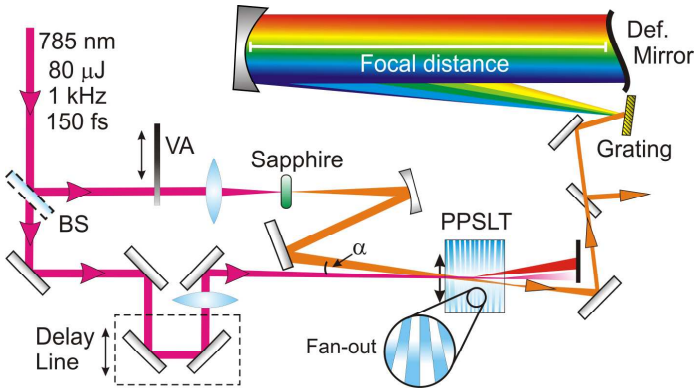

Fig. 1. Experimental setup of the near-IR NOPA based on PPSLT. BS: beam splitter; VA: variable attenuator.

Figure 2 shows, as a dash-dotted line, the frequency-dependent parametric gain of PPSLT, calculated in the plane wave approximation with monochromatic and undepleted pump, for poling period $\Lambda \cong 20.9$ μm and internal pump-signal angle 1.9°. The same figure shows as a solid line the typical spectrum of the pulses generated by the NOPA. It extends from 1 to 1.7 μm and corresponds to a 8.2 fs TL duration (less than two cycles of the 1.3 μm carrier wavelength). Its coverage corresponds quite well to the calculated gain spectrum; the reduced intensity at longer wavelengths is due to the lower energy density of the WLC. The spectrum shows a reproducible dip around 1.16 μm, which is due to a parasitic sum-frequency generation process with the 800-nm pump, phase-matched to the second order in the PPSLT crystal. The near-IR NOPA has a bandwidth comparable to that of the degenerate collinear OPA based on BBO, that we recently reported [3], but its spectrum is blue-shifted by ≈200 nm. In order to achieve dispersion control over the ultrabroad NOPA bandwidth, we implemented an adaptive compressor (see Fig. 1), in which a DM is placed in the Fourier

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plane of a 4*f* zero-dispersion pulse shaper [16, 17] consisting of a near-IR diffraction grating (Newport, 150 lines/mm, gold coated) and a spherical gold mirror (R=500 mm). The overall throughput of the compressor is 65% and the transmitted spectrum (dashed line in Fig. 2) loses some intensity in its red wing due to the wavelength dependent diffraction efficiency of the grating.

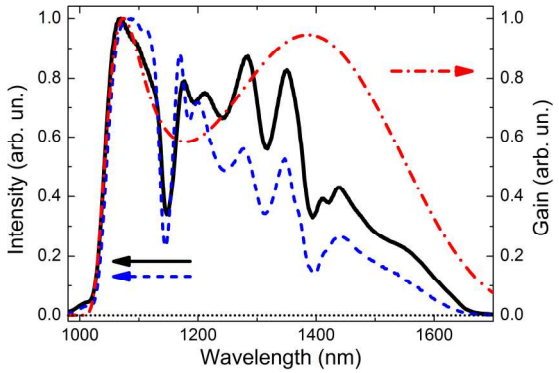

Fig. 2. Dash-dotted line: calculated gain bandwidth of the near-infrared NOPA; solid line: output spectrum of the NOPA; dashed line: spectrum after the compressor.

The use of a prism [3] instead of a grating as dispersive element in the pulse shaper would have the advantage of an higher throughput; however, because of its lower angular dispersion, a prism would require a spherical mirror with much longer radius (R=2000 mm) to completely fill the DM. For our near-IR NOPA, compensation of the large second order dispersion introduced by the sapphire plate and the PPSLT crystal (≈160 fs^2 at 1.3 μm), requires a strong parabolic curvature of the DM surface. In the prism-based shaper this curvature is sufficient to break the symmetry of the 4*f* scheme, thus distorting the pulse wavefront, preventing perfect recombination of the frequency components of the ultra-broadband pulse and causing a significant spatial chirp of the compressed pulses. This effect is stronger, for a given deformation, for longer focal length *f*. With the grating this problem is negligible, because of the much shorter focal length of the mirror compared with the DM deformation and because the additional dispersion introduced by the prism material is removed, with a further reduction of the needed DM curvature.

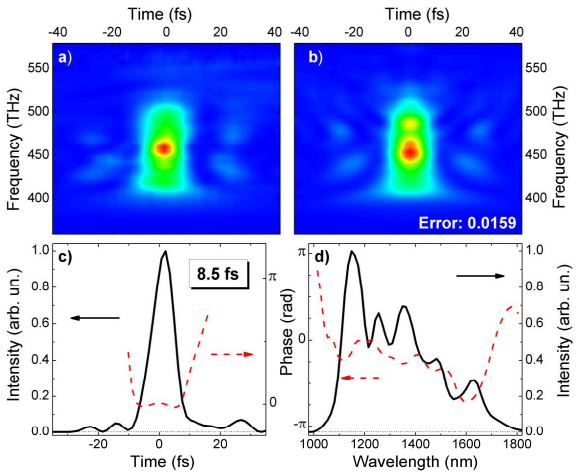

Fig. 3. FROG measurement of the compressed NOPA pulses. (a) intensity of the measured FROG trace (128×128 pixels); (b) retrieved FROG trace in intensity with 0.0159 reconstruction error; (c) intensity and phase profile as a function of time; (d) spectral intensity and phase.

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Our home-made DM is a 45×15 mm silver coated rectangular nitrocellulose membrane, 5-μm thick. The mirror is deformed by the electrostatic pressure between the membrane and a series of 30 electrodes (each 1.4 mm wide, 15 mm long and 0.3 mm spaced) independently polarized from 0 to 295 V. In order to prevent spatial beam quality degradation by the transverse curvature of the mirror surface, the electrodes have a rectangular hole in the center of 2×1.2 mm. This electrode design, in comparison with filled one, allows the reduction of the transverse curvature by nearly two orders of magnitude. The DM response resulting from each actuator was numerically modeled and experimentally calibrated by means of a Twyman-Green interferometer [18] allowing the definition of the influence functions matrix [19]. The mirror shape for optimum pulse compression was obtained by measuring the pulse spectral phase with Second Harmonic Generation Frequency Resolved Optical Gating (SHG-FROG) and introducing the additional phase required to correct it. The set of mirror actuator voltages corresponding to the targeted deformation was computed by performing a pseudo inversion of the influence function matrix [19]. Such process was iterated, usually two or three times, in order to obtain a finer phase correction.

Figures 3(a) and 3(b) show the experimental and retrieved SHG-FROG traces for the compressed NOPA pulse, measured with a 20-μm-thick BBO crystal, while Figs. 3(c) and 3(d) report the retrieved temporal and spectral intensity and phase profiles. The measured 8.5 fs pulsewidth is very close to the TL duration and has very low side lobes, demonstrating that the DM based compressor allows dispersion compensation to all orders. This pulse duration corresponds to only two cycles of the 1.3 μm carrier wavelength (4.3 fs period).

3. High time resolution pump-probe spectroscopy

The ultra-broadband near-IR NOPA has been absolutely synchronized with a 6 fs visible NOPA [20], driven by the same amplified Ti:sapphire laser. The unique combination of high temporal resolution and broad tunability provided by this system enables studying a variety of primary relaxation events of photoexcited molecules, such as internal conversions through conical intersections, charge photogeneration and coherent vibrational motion in the excited state.

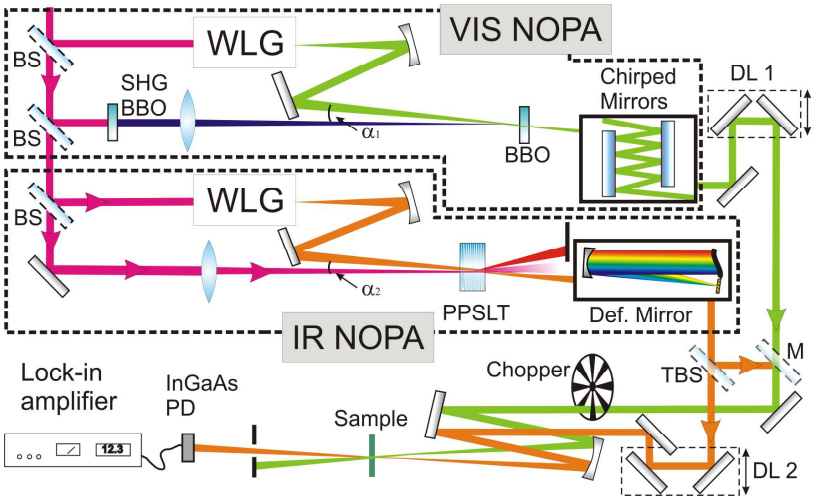

Fig. 4. setup for the generation of two-colour sub-10-fs pulses. BS, beam splitters; WLG, white-light generation; DL1, DL2, delay lines. A thin beam splitter (TBS) and a mirror (M) can be added to change the system to a degenerate setup.

The experimental pump-probe setup is shown in Fig. 4. The visible NOPA is pumped by 20 to 30 μJ of the SH of Ti:sapphire and amplifies a WLC seed produced in a 1-mm-thick sapphire plate in a single pass of a 1-mm BBO crystal (type I phase matching, $\theta = 32°$). Pump and WLC seed form an angle $\alpha_1 = 6°$, which corresponds to 3.7° inside the BBO crystal,

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giving broadband phase matching in the visible (500-700 nm). The 1 to 2 μJ pulses are compressed to nearly TL 6 fs duration by 10 bounces on custom designed chirped mirrors, which provide highly controlled negative group delay over bandwidths approaching 200 THz [21]. The outputs of the two NOPAs are synchronized by delay line DL1 and focused on the sample in a non-collinear geometry; after the sample the near-IR probe beam, spatially filtered by an iris, is focused on a monochromator and an InGaAs detector. The system can be easily made degenerate, for pump-probe experiments in the near-IR, by inserting a thin beam splitter and a mirror (transparent in Fig. 4) realizing a Michelson interferometer.

To assess the temporal resolution of the two-colour pump-probe configuration, we performed studies on the excited state dynamics of a film of poly-phenylene vinylene (PPV), solution cast onto a thin glass slide. In such blends, the visible pump pulse excites the first singlet transition ($S_0 \rightarrow S_1$). Figure 5 shows a $\Delta T/T$ dynamics pumping with a sub-10-fs visible pulse and probing with a sub-10-fs near-IR pulse. At 1.21 μm probe wavelength, we observe a photoinduced absorption (PA) with pulsewidth-limited risetime (10 to 90% in 14.5 fs). The PA is assigned to a transition from the photoexcited S_1 state to an higher-lying S_n state [22]; the partial decay with 70-fs time constant is assigned to intramolecular vibrational relaxation within S_1.

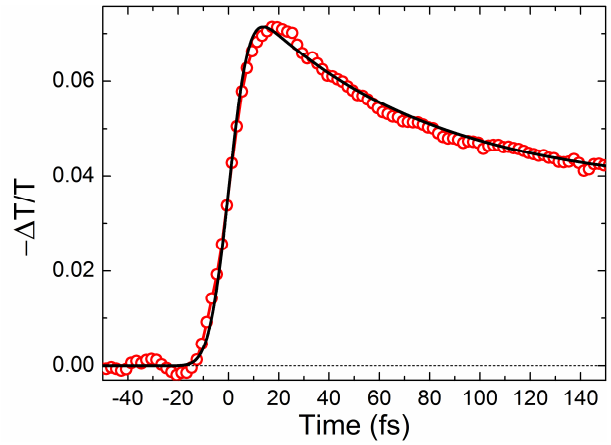

Fig. 5. Red dots: $\Delta T/T$ dynamics for a PPV film excited by the 6-fs visible NOPA and probed at 1.21 μm wavelength. Black solid line: fit using pump-probe cross-correlation with 14.2 fs FWHM.

4. Conclusions

In this paper we have demonstrated the generation of 8.5 fs pulses at 1.3 μm from a near-IR NOPA based on PPSLT, using a DM based adaptive compressor. We achieve a shortening by nearly a factor of 20 of the 150-fs pump pulses. This result is a further proof of the power of OPAs for the generation of few-optical-cycle pulses tunable over a broad frequency interval, provided that the proper broadband phase matching conditions are identified. Due to its modest pump energy requirements, the near-IR NOPA can be easily synchronized to other broadband OPAs, allowing the study of previously unexplored primary photoinduced dynamical processes over a very broad spectral range.

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