Clean sub-8-fs pulses at 400 nm generated by a hollow fiber compressor for ultraviolet ultrafast pump-probe spectroscopy

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Abstract: Clean 7.5 fs pulses at 400 nm with less than 3% energy in tiny satellite pulses were obtained by spectral broadening in a hollow fiber and dispersive compensating using a prism pair together with a deformable mirror system. As an example, this stable and clean pulse was used to study the ultrafast pump-probe spectroscopy of photoactive yellow protein. Moreover, the self-diffraction signal shows a smoothed and broadened laser spectrum and is expected to have a further clean laser pulse, which makes it more useful in the ultrafast pump-probe spectroscopy in the future.

OCIS codes: (320.7150) ultrafast spectroscopy; (320.5520) pulse compression; (190, 7110) ultrafast nonlinear optics.

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1. Introduction

Laser spectroscopic techniques have been widely applied to all fields in science including chemistry, physics, and biology, providing microscopic insights into bulk materials, and molecules, and chemical and biochemical reaction processes [1–3]. The advances in ultrashort laser pulses technology make it possible to observe in real-time electronic, phonon, and vibrational and phonon dynamics in various molecular systems and bulk materials in femtosecond resolution [1–3]. To investigate the real-time dynamics of vibrational amplitude by pump-probe method, it is needed to use a pump pulse whose duration is shorter than the vibrational period of the mode of interest. The noncollinear optical parametric amplification (NOPA) made this real-time vibrational spectroscopy to be easily applied in visible spectral region in a sub-10-fs time resolution [2,3]. On the other hand, many basic molecules with conjugated π electron systems have absorption around the 400 nm wavelength. This makes it necessary to develop an ultrashort pulse in this spectral region. Sub-10-fs pulse around 400 nm has been developed by using a hollow-fiber compressor [4,5] or by broadband frequency doubling of a 15 fs pulse at 800 nm from Ti:sapphire laser [6]. A sub-15 fs pulse is not easy to be obtained directly from a Ti:sapphire amplifier and is usually obtained by another compressor [7]. The broadband frequency doubling method requires a configuration which inevitably induces angular dispersion of the output beam. The stability of spectral shape and intensity after the hollow fiber is poor in many cases because of the beam-pointing instability of the laser beam [8]. The compressed pulses usually have many parasitic pulses generated by the residual third-order dispersion from a prism pair and chirped mirrors [4,5]. In the pump-probe experiment, a clean-pulse without any satellite with the long-time stability of the intensity and spectral shape is of vital importance.

Recently, we obtained stable sub-10-fs pulses at 400 nm by spectral broadening in a hollow fiber and dispersion compensation with two pairs of chirped mirrors [5]. However, there remained still small satellites in the compressed output pulse and it is very inconvenient to tune the dispersion precisely to compensate the dispersion of different samples by using chirped mirror. In this paper, we replaced the chirped mirrors by a set of prism pair and a grating-deformable mirror dispersion compensation system. Through optimization using a deformable mirror, we obtained 7.5 fs clean pulse at 400-nm center wavelength with less than 3% energy in tiny satellite pulses. This stable and clean pulse was also used to preliminary study of the ultrafast spectroscopy of the photoactive yellow protein (PYP).

2. Experiment and results

The experimental setup was the same as in our previous work [5]. A beam-pointing stabilizer was used before a hollow-fiber compression system. About 900 μJ laser pulse after a Ti:sapphire laser system was frequency doubled in a 200-μm-thick beta barium borate (BBO, Type I, $\theta = 29.2°$) crystal. After some mirrors, about 90 μJ laser pulses at 400 nm were focused into a hollow fiber, which has a 140-μm inner diameter and a 60-cm length and was filled with 0.8-atm argon gas. The output pulse energy after hollow fiber was about 45 μJ with about 50% transmission efficiency. The output beam diameter was first reduced to about 2 mm by using two aluminum-coating concave mirrors. Then, the pulse was dispersion compensated by a pair of fused silica prisms with Brewster-angle cut and another dispersive compensating system composed of a grating (1200 groove/mm, blaze wavelength and angle: 400 nm, 13.88°), an aluminum-coating concave mirror (R = - 400 mm), and an aluminum-coating micromachined membrane deformable mirror (11 × 39 mm, OKO), as shown in Fig. 1(a). The distance between the prism pairs was about 16.8 cm. In this setup, the prism pair introduces negative second-order dispersion and negative third-order dispersion. The grating-deformable mirror compressor system will introduce negative second-order dispersion and positive third-order dispersion [8,9]. As a result, the combination of a prism pair and a

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grating-deformable mirror compressor system help to reduce the third-order dispersion. The compressed pulse was guided into the pump-probe experiment setup which was used for pulse duration measurement by the self-diffraction frequency resolved optical gating (SD-FROG) method [9] in a 100-μm-thick CaF_2 crystal with a 1.2° crossing angle. In the experiment, to optimize the high-order spectral phase, the optimization of deformable mirror was act manually in two steps. At first, we manually set the deformable mirror based on legendre polynominals to optimize the SD signal roughly. Then, the deformable mirror was manually set pixel by pixel to further optimize the SD signal accurately. In this way, the high-order spectral phase was compensated.

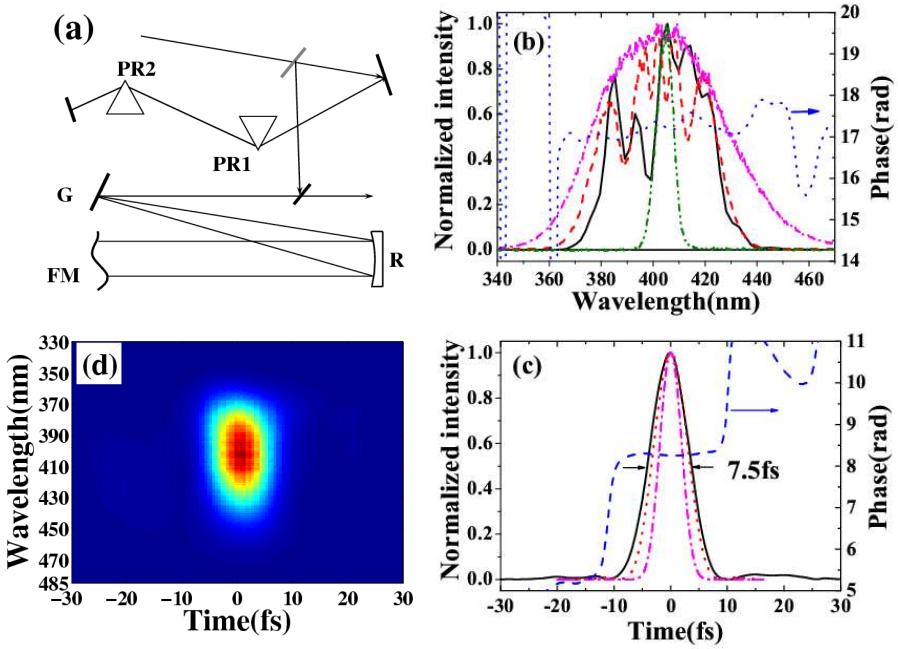

Fig. 1. (Color online) (a) Setup of the dispersive compensating system. PR1, PR2: Brewster-angle cut fused silica prisms; G: Aluminum-coating grating (1200 groove/mm, blaze wavelength and angle: 400 nm, 13.88°); R: Aluminum-coating concave mirror, R = - 400 mm; FM: deformable mirror. (b) Spectra of the incident laser pulse (olive dash-dot-dot line), the retrieved laser pulse (black-solid line), the compressed laser pulse (red-dash line), and the first-order SD signal (magenta-dash-dot line). The retrieved spectral phase is shown as blue-dot line. (c) The retrieved temporal intensity profile (black-solid line) and temporal phase (blue-dash line) of the compressed pulse. The red-dot line and magenta-dash-dot line show the transform-limited pulse of the compressed laser pulse and its first-order SD signal, respectively. (d) The measured two-dimensional SD-FROG trace.

Figure 1(b) shows the laser spectrum after the compressor system when the gas pressure of argon is 0.8-atm. The spectral width (FWHM, full width at half maximum) is broadened from about 7.5 nm to about 46 nm. The broadened spectrum can support 6.8 fs transform-limited pulse duration, as shown in Fig. 1(c). Figure 1(d) shows the measured SD-FROG trace with a 256 × 256 grid and a 0.5 fs delay time step. The laser spectrum, spectral phase, temporal intensity profile, and temporal phase are retrieved by using the commercial software (FROG 3.0, Femtosoft Technologies) with a 0.003 retrieval error. Figure 1(b) shows the retrieved spectrum and spectral phase with black-solid-line and blue-dot-line, respectively. The retrieved spectrum fits the laser spectrum very well. The temporal intensity profile and temporal phase are shown in Fig. 1(c) with black-solid-line and blue-dash-line, respectively. The pulse duration (FWHM) was 7.5 fs with less than 3% energy in tiny satellite pulses. The compressed pulse duration was close to the 6.8 fs transform-limited pulse duration. The

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spectral phase and temporal phase are flat owing to the optimization of the prism pair and the grating-deformable mirror dispersion compensation system [10].

Figure 1(b) also shows the spectrum of the SD signal at zero delay time with magenta-dash-dot line. It can be obviously found that the spectrum of the SD signal was broadened and smoothed in comparison with the laser spectrum after the gas-filled hollow fiber that strong modulated by the self-phase modulation effect. The smoothed spectrum of the SD signal can support about 4.5 fs transform-limited pulse duration, which is shorter than that of the compress pulse 6.8 fs, as shown in Fig. 1(c). When both the incident pulse energies for SD signals generation were 200 nJ and the beam diameters on the 100-μm-thick CaF_2 crystal were 90 μm, there are four SD signals on both sides at zero delay time. The pulse energy of the first-order SD signal on every side was 14 nJ. The energy transfer efficiency from incident laser pulses to the first-order SD signals was about 7%. It is also expected that the SD signal has a shorter and cleaner pulse [11]. The same as another third-order nonlinear process, cross-polarized wave (XPW) generation [12], the pulse shortening and cleaning can be easily understood by the expression in the time domain: $I_{SD} \propto I^2(t)I(t-\tau)$. The SD signal with smooth spectrum and clean pulse would be more useful in the ultrafast pump-probe spectroscopy research in the future. Here, the further study of the SD signal is limited by the pulse energy in the experiment.

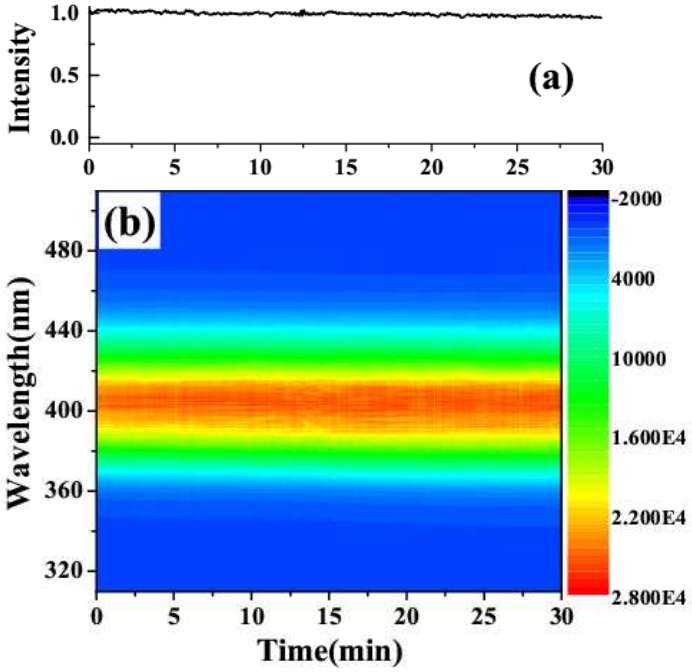

Fig. 2. (Color online) The stability of the spectral intensity (a) and the spectral profile (b) of the first-order SD signal in half an hour.

The spectral intensity stability and the spectral profile of the first-order SD signal was monitored with a spectrometer (USB4000, Ocean Optics) for half an hour, as shown in Fig. 2. It shows that even the SD signal keeps the spectral profile and has power stability of less than 1.5%RMS in half an hour. The excellent stability owing to the pointing stabilizer system we used in the hollow fiber system [5]. The clean-pulse with long-time stability of the intensity and spectral shape provides reliable data in the pump-probe experiment.

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3. Example of application

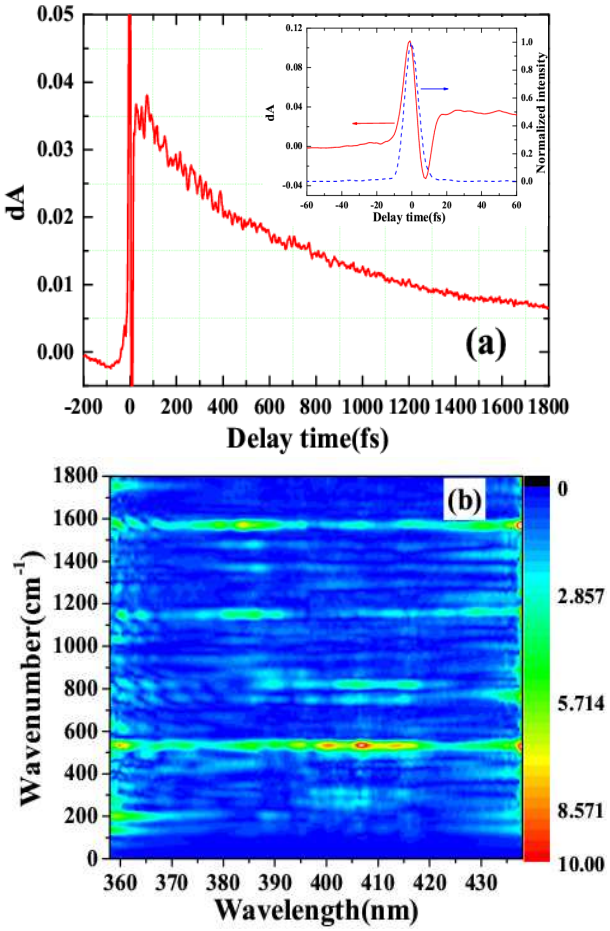

Fig. 3. (Color online) (a) The total absorbance dA *vs.* the pump-probe delay time in the spectral range from 374 nm to 399 nm. The inset is the enlarged curves of dA between –20 and 20 fs (red-solid curve) and the SD autocorrelation curve (blue-dashed curve); (b)The two-dimensional FT amplitude spectra of the normalized difference transmittance spectra$\Delta T/T$ at the probe spectral range from 358 to 438 nm.

As in our previous visible-pump/visible-probe experiments [2], the ultrafast pump-probe experiment equipment is a combined system of a polychromator and a multichannel lock-in amplifier (MLA). The reference and probe pulses were angular dispersed by the polychromator (600 grooves/mm, 300 nm blazed) and the light at different wavelength was guided by a 128-channel bundle fiber to the 128 photo-detectors before the MLA. The spectral resolution of the system was about 0.8 nm. In the experiment, the beam diameters (at $1/e^2$) and pulse energies of the pump and probe beam were 90 μm and 37 nJ and 85 μm and 3.7 nJ, respectively. The sample is PYP dissolved in water with some buffer composed of Tris-HCl at PH 7.3, which is circulating in a flow cell (46mL/min) with 0.5-mm light path length. The absorbance at 446 nm was ~0.98 at 0.5-mm light path length. The experiment was performed at room temperature (295 ± 1K).

Time trace of the total absorbance dA (dA = -log (1 + $\Delta T/T$)) was obtained as a function of the pump-probe delay time from –200 to 1800 fs with 1-fs step in the spectral range from 374 to 399 nm, as shown in Fig. 3(a). The positive value of dA in the spectral region indicated a photoinduce absorption process. It also clearly shows the decay process and the vibrations

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from the curve. The inset in Fig. 3(a) shows the enlarged curves of dA between −20 and 20 fs and the SD autocorrelation curve, which shows that the decay even begins at about 15 fs. Figure 3(b) shows the two-dimensional Fourier transform (FT) amplitude spectra of the normalized difference transmittance spectra*ΔT/T* at the probe spectral range from 358 to 438 nm. It shows clearly several vibration modes of the PYP around 1570, 1150, 820, 750, and 535 cm^{-1} at different probe wavelengths [13]. A detail analysis of this pump-probe experiment will be described elsewhere. This experiment proved that the obtained pulse is useful for the UV ultrafast pump-probe spectroscopy.

4. Conclusion

In summary, we obtained stable and clean 7.5 fs laser pulses with less than 3% energy in satellite pulses at 400 nm center wavelength by spectral broadening in a hollow fiber and dispersive compensating using a prism pair together with a deformable mirror system. PYP was used as an example to demonstrate the application of this stable and clean pulse in the ultrafast pump-probe spectroscopy. It is also expected that the SD signal would be more useful in the ultrafast spectroscopy research owing to it has a smoother spectrum and is expected to has a cleaner pulse in compare with the compressed pulse after hollow fiber.

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