Femtosecond pulses cleaning by transient-grating process in Kerr-optical media

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Received November 9, 2010; accepted January 3, 2011; posted online April 18, 2011

We use a transient-grating (TG) process in a Kerr bulk medium to clean a femtosecond laser pulse. Using the technique, the temporal contrast of the generated TG signal is improved by more than two orders of magnitude in comparison with the incident pulse in a 0.5-mm-thick fused silica plate. The laser spectrum is smoothed and broadened, and the pulse duration is shortened simultaneously. We expect to extend this technique to a clean pulse with broadband spectral bandwidth at a wide spectral range because it is a phase-matched process.

OCIS codes: 190.4380, 190.4410, 190.7110, 320.5520. doi: 10.3788/COL201109.051903.

In ultrafast spectroscopy and laser-matter interaction experiments, cleaning pulses with a smooth spectrum are considered to be vitally important in removing unwanted noise signals introduced by satellite pulses in an uncleaned pulse, which make the experiments complicated. In laser-matter interaction experiments in the relativistic dominated regime^[1,2] in particular, a pulse with high temporal contrast is necessary to prevent unwanted intense prepulses from generating preplasma before the main $pulse^{[3]}$. To improve the temporal contrast of this highly intense femtosecond pulse, several pulse-cleaning techniques have been developed. These methods include the use of saturable $absorbers^{[4]}$, a nonlinear Sagnac interferometer^[5], double chirped pulse amplification (CPA)^[6], plasma mirrors^[7], polarization rotation^[8], cross-polarized wave (XPW) generation^[9], and self-diffraction (SD) $process^{[10]}$. By using XPW in BaF_2 crystals, the temporal contrast was improved by about four orders of magnitude^[9]. However, the contrast enhancement was limited by the extinction ratio of the polarization discrimination device^[9]. Very recently, we demonstrated for the first time the application of a selfdiffraction process in a Kerr bulk medium to clean the pulse which does not need any polarizer. Through this method, it was proved that both prepulses and postpulses can be cleaned in one picosecond $region^{[10]}$.

As with the SD process, in which two beams are focused into a Kerr bulk medium with a small crossing angle to generate separated SD signals beside the incident beams, the generated signal by the transient-grating (TG) process is also spatially well separated from incident beams and there is no need for the polarization discrimination device. Both processes are third-order nonlinear optical processes. The TG process is a phase-matched process unlike the SD process; thus, the TG process can have a long nonlinear medium to enhance the signal and can be used in a wide spectral range from ultraviolet (UV) to mid-infrared (MIR) with a broadband bandwidth^[11]. Owing to its ability to operate with a long nonlinear medium, the TG process is more sensitive to the input pulse intensity and can be operated with nanojoule pulse energy input^[11]. Larger beam angles may also be used in the SD process, reducing the scattered-light background. All these advantages make the TG process another useful process for cleaning ultrashort laser pulses.

In this letter, we demonstrate experimentally the use of the TG process to clean a femtosecond laser pulse. Using this technique in a 0.5-mm-thick fused silica glass plate, the results indicate that the temporal contrast is improved by two orders of magnitude in comparison with the incident pulses. The laser spectrum is found to be smoothed and broadened simultaneously, and the pulse duration is shortened in this process.

The TG process is also a third-order nonlinear process and the intensity of the TG signal has a cubic dependence on the input intensities I_1 , I_2 , and I_3 . In the time domain, it can be expressed as

$$I_{\rm sd1} \propto I_1(t) I_2(t) I_3(t-\boldsymbol{\tau}), \tag{1}$$

where τ is a delay time between the grating formation pulse pair and the signal pulse to be diffracted. The SD process is a special case of τ being 0. This time-domain expression indicates that the pulse can be cleaned and that the pulse duration of the TG signal will be shortened in the TG process by controlling the delay time τ . The formation and disappearance of the grating in the TG process in non-resonant Kerr media is instantaneous^[12]. Therefore, even the satellite pulses in the picosecond range and the weak amplified spontaneous emission will separate the TG process from the main pulse in the time domain. In the frequency domain, the intensity of the generated TG signal can be described as^[11]

$$I_{\rm TG}(\omega_{\rm TG}) \propto \left| \iint d\omega_1 d\omega_2 \widetilde{E}_1^*(z,\omega_1) \widetilde{E}_2(z,\omega_2) \right|^2 \widetilde{E}_3(z,\omega_{\rm TG}-\omega_2+\omega_1) \exp[i(\omega_2-\omega_1)\tau] \right|^2, \quad (2)$$

where $\omega_1, \omega_2, \omega_3, \omega_{\text{TG}}$ are angular frequencies of three incident beams and the generated TG signal, respectively. The spectral intensity of the TG signal is given by an integral of the spectral intensity of three incident laser pulses. This means that the intensity of the TG signal at every wavelength is the average contribution of the entire spectral region of the incident pulses. As a result, the spectrum of the TG signal is smoothed automatically due to the third-order nonlinear process. Thus, the spectrum of the TG signal together with self-phase modulation and cross-phase modulation is broadened.

A commercial Ti:sapphire CPA laser system (Legend-USP, Coherent) producing 35-fs, 2.5-mJ maximal pulse energy at 800-nm center wavelength and running at 1-kHz repetition rate was used in the experiment (Fig. 1). Four 1-mm-thick fused silica beamsplitters (BSs) with 80/20, 80/20, 80/20, and 50/50 transmission/reflection ratios were used to reduce the pulse energy to about 100 μ J and to introduce prepulses and postpulses through the back-and-front reflections of the BSs. After a variable neutral-density (VND) filter, the laser pulse was split into three beams by a 67/33 and a 50/50 BS. One of the laser beams can tune the time delay through a motor-controlled stage with a 10 nm/step. The three beams were focused with a BOXCARS arrangement^[13] using a



Fig. 1. Schematic of the experimental setup. BS1–BS7 are seven 1-mm-thick fused silica beamsplitters with 80/20, 80/20, 80/20, 50/50, 67/33, and 50/50 reflection/transmission ratios, respectively. SM1: spherical mirror, R=-600 mm. SM2: spherical mirror, R=-500 mm. L: focal lens with 50-mm focal length.

concave mirror with a 300-mm focal length into a 0.5mm-thick fused silica (FS) glass plate located after the focal point. The beam diameters on the glass plate were about 400 μ m. Transmission pulse energies of beam1, beam2, and beam3 were 32, 37, and 20 μ J, respectively. The pulse energy of the generated TG signal was about 1.2 μ J.

We performed a second-order autocorrelation to measure the temporal contrast which needed much lower incident pulse energy^[14]. The measurement procedure was almost the same as our previous work^[10]. In the secondorder autocorrelation measurement, a 1-mm-thick 50/50 beamsplitter was used to split the laser beam. A 170- μ mthick beta barium borate (BBO, Type I, θ =29.2°) crystal was used to generate a sum-frequency (SF) signal. After an aperture, a low-wavelength-pass filter (LPF) cutting at 440 nm, and a 1000-times-decreased natural-density (ND) filter, the SF signal was focused into a fiber and detected using a multi-channel spectrometer (USB4000, Ocean Optics) with 200 ms integration time.

The intensity of the SF signal was obtained by integrating the spectral intensity over the spectral range from 370 to 430 nm to reduce other noises. In the delay time range when the SF signal was very strong to saturate the spectrometer, a 1000-times ND-filter was added (for TG signal from -100 to 100 fs, for incident pulse from -1000 to 1000 fs). In this case, the delay time dependent SF intensities of incident laser pulses and the TG signal were obtained (Fig. 2). In the figure, autocorrelation data of incident laser pulse and TG signal in the delay time show a range of -6 to 6 ps. The data were obtained with a 10-fs delay time step. The temporal contrast of the TG signal was clearly improved by about two orders of magnitude compared with incident pulses. The two peaks in the TG signal autocorrelation at ± 1.8 ps were due to the back-and-front reflections of the $170-\mu$ m-thick BBO crystal used in the autocorrelator.

The pulse durations of the input pulse and TG signal were measured using second harmonic generationfrequency-resolved optical grating SHG-FROG with the same setup as autocorrelation measurement and replacing the beamsplitter by a half-reflective mirror. The spectral and temporal profiles, as well as the phase were retrieved using a commercial software (FROG 3.0,



Fig. 2. Normalized SAC intensities of the incident pulse (upper curve) and that of the TG signal (lower curve) in the delay time from -6 to 6 ps with a 10-fs/step resolution.



Fig. 3. (a) and (b) are the measured spectrum, the retrieved spectrum, and the retrieved spectral phase of the incident pulse and the TG signal, respectively; (c) and (d) are the retrieved temporal profile and the phase of the incident pulse and the TG signal, respectively. The inset patterns in (c) and (d) are the measured 2D SHG-FROG traces of the incident pulse and the TG signal, respectively.

Femtosoft Technologies) with a 256×256 grid. The retrieval errors were smaller than 0.005. Figure 3(a) shows the retrieved laser spectrum, retrieved spectral phase. and measured spectrum. The measured spectral intensity of the TG signal, retrieved spectral intensity profile, and retrieved phase are shown in Fig. 3(b). The TG signal spectrum was obviously smoothed and broadened from the input laser spectrum. The center wavelength of the TG signal was red-shifted for several nanometers, which could be due to the crossing phase modulation (XPM)^[15]. Even though the input laser spectral phase showed a positive chirp, the obtained TG signal showed a small negative chirp. This is likely due to the XPM process and suitable delay among the incident $pulses^{[10]}$. The retrieved temporal profile and temporal phase of the input laser pulse and the TG signal are shown in Figs. 3(c) and (d), respectively. Pulse duration of the TG signal was shortened from the input 74 fs to a compressed 51 fs. Retrieved pulse of the TG signal also showed that the satellite pulses were removed. The two inset patterns shown in Figs. 3(c) and (d) are the measured twodimensional (2D) SHG-FROG traces of incident pulse and TG signal, respectively.

In conclusion, we demonstrate femtosecond pulse cleaning using the TG process in Kerr bulk media. Similar to the SD process, there is no need for a polarizer device in this method because the generated TG signal is well separated from the incident laser beams. Simultaneously, the laser spectrum is smoothed and broadened, and the pulse duration is shortened for the generated TG signal in comparison with the incident laser pulse. Although its improvement and efficiency are currently low compared with those of the SD process, the TG process is a phase-matched process, which makes it useful for the optimization of temporal and spectral characteristics by cleaning ultrashort laser pulses in a wide spectral range from UV to MIR with a broadband bandwidth.

This work was partially supported by the 21st Century COE program on "Coherent Optical Science" and supported by the grant from the Ministry of Education (MOE) under the ATU Program at National Chiao Tung University. A part of this work was performed under the joint research project of the Laser Engineering, Osaka University, under contract subject B1-27. Dr. Jun Liu also thanks the support of the Opening Funds of the State Key Laboratory of High Field Laser Physics in China.

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