Diffraction of white-light supercontinuum by femtosecond laser-induced transient grating in carbon bisulfide*

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Experiments on fs laser-induced transient grating (LITG) in carbon bisulfide (CS_2) are carried out to explore the chirp characteristics of a white-light supercontinuum (SC) generated by a 800-nm, 160-fs laser pulse in a 4-mm thick Al_2O_3 crystal. Two orders of diffraction signals of SC by fs LITG in CS_2 are observed, demonstrating that both the third-order process and the fifth-order process are present simultaneously. The experimental results also imply that the formation of an fs transient refractive-index grating in CS_2 is mainly due to the electronic polarization process.

Keywords: laser-induced transient grating, white-light supercontinuum, chirp characteristic

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1. Introduction

When a powerful ultrafast pulse is focused into a transparent medium, it can undergo extreme spectral broadening and yield a white-light supercontinuum (SC). An SC, one of the most specular nonlinear-optical phenomena, was first observed from glass samples by Alfano and Shapiro in the 1960s.[1] Since then, enormous studies on generation and properties of SC in a wide variety of solids,[2] liquids,[3] and gases[4] have been carried out. As a source of tunable ultrafast pulses ranging from ultraviolet to infrared, white-light SC has been used widespreadly in fs time-resolved spectroscopy,[5] pulse compression of optical parametric amplifiers,[6] broadband spectrum lidar,[7] and so on.

The mechanism of SC generation remains far from well understood due to its inherent complexities, so generation and characterization of SC have been the subjects of many publications in recent years.[8−10] Chirp characteristic of SC has been investigated with frequency-resolved optical gating (FROG) technique and optical Kerr gate (OKG) technique.[11−15] FROG has evolved into a fairly general and powerful technique for measuring ultrafast laser pulses. But for measuring a white-light SC with FROG technique, which is based on second harmonic generation (SHG), there are two significant challenges: one is that the SC spectrum is too broad to phase match simultaneously; the other one is that the SC usually has no more than I nJ of energy, so the SC measurement technique must have high sensitivity. The OKG technique has many advantages, for example, high intensity of the probe pulse.[16] However, applications based on the OKG technique require optical materials to have large third-order nonlinearities, low optical loss, high transparency over large spectral ranges, high damage threshold to laser irradiation, good thermal stability and processability.[17] Such requirements are hard to meet in the fs optical OKG experiments. Thus it is important to develop effective and general techniques for white-light SC measurements in the fs region.

An ultrafast transient refractive grating, which process can phase-match a large bandwidth simultaneously, can be produced by interference of two synchronized pump pulses which are temporally and spatially overlapped in the materials. The probe SC pulse crosses the grating at an angle and the diffraction signal pulse emerges from the interaction region as the fourth beam in the rectangle formed by the three input pulses, so laser-induced transient grating (LITG) technique is background free and more sensitive. The time resolution of LITG technique is determined mainly by

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the width of laser pulses but not the optical materials used in the studies, and it has been regarded as a useful tool for studying molecular dynamics.[18−21]

In this paper, we report the experiments on the chirp characteristics of white-light SC with fs LITG in CS_2. The reason why we choose CS_2 here is not for its high nonlinearity, but for its high electronic delocalization and easy formation of transient grating in it. Two orders of diffraction signals of SC by fs LITG are observed and the chirp characteristics of fs SC are discussed. The experimental results are helpful for making the SC pulses more reproducible from pulse to pulse and more spatially uniform.

2. Experiments

Our fs LITG experimental arrangement is shown in Fig. 1. The fs laser pulse at 800 nm with a pulse duration of 160 fs was derived from a regenerative amplifier system working at 1 kHz (Spitfire, Spectra-Physics).

Two beam splitters (BS1 and BS2) split the output fs laser pulse into three beams whose wave vectors were denoted as $\boldsymbol{k}_1$, $\boldsymbol{k}_2$ and $\boldsymbol{k}_3$, respectively. The single pulse energy ranged from 1 μJ to 10 μJ, and the intensities of the three fs laser pulses are varied between 10^{10} W/cm^2 and 10^{11} W/cm^2 by using a set of matched neutral density filters. The two reflected beams of BS1 and BS2 ($\boldsymbol{k}_1$ and $\boldsymbol{k}_2$) were used as pump pulses to form an ultrafast transient grating in CS_2. The transmitted beam of BS2 focused into a 4-mm thick Al_2O_3 crystal to produce a white-light SC ($\boldsymbol{k}_3$). Then the SC was spatially crossed and overlapped in liquid CS_2 and diffracted by LITG to produce the signal pulses (see $\boldsymbol{k}_{s1}$ and $\boldsymbol{k}_{s2}$ in Fig. 3).

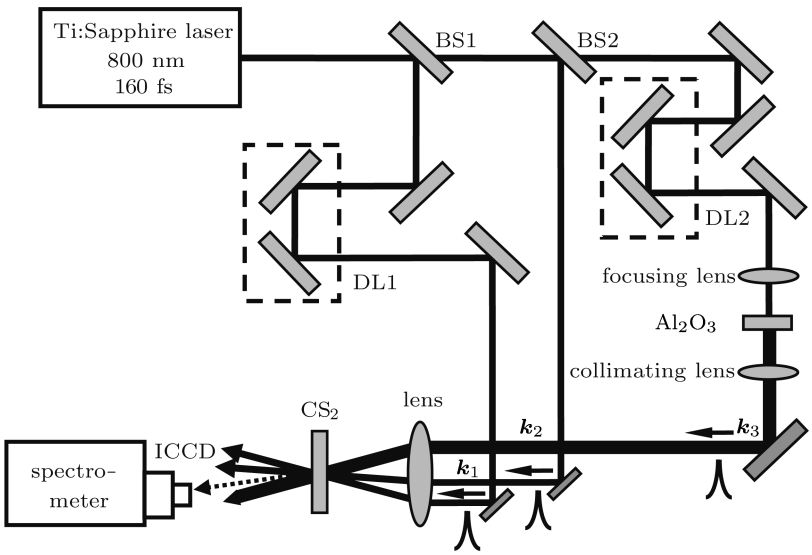


Fig. 1. Schematic diagram of the experimental setup. BS: beam splitter; DL: delay line. In our experiments, all beams were horizontally polarized.

A delay line (DL1) in Fig. 1 was used to adjust the delay time between two pump pulses $\boldsymbol{k}_1$ and $\boldsymbol{k}_2$. The DL1 was fixed when two pump pulses ($\boldsymbol{k}_1$ and $\boldsymbol{k}_2$) were overlapped in time and focused in space to form a refractive-index LITG in CS_2. The other delay line (DL2) was used to adjust the delay time between two pump pulses ($\boldsymbol{k}_1$ and $\boldsymbol{k}_2$) and the SC ($\boldsymbol{k}_3$) to observe the diffraction signals of SC at different delay times. Figure 2 illustrates the timing of two pump pulses ($\boldsymbol{k}_1$ and $\boldsymbol{k}_2$) and SC pulse ($\boldsymbol{k}_3$) to measure the chirp structure of SC with femtosecond LITG technique. The time "zero" was defined as the moment when two pump pulses ($\boldsymbol{k}_1$ and $\boldsymbol{k}_2$) are incident into the CS_2.

The transparent liquid CS_2 used in the experiment had a purity of 99.9% and was contained in a 1-mm thick quartz cell; the freshness of CS_2 must be ensured during the experiment. The detection setup composed of a spectrometer (Optics 250IS/SM, Bruker) and an intensified charge coupled device

(ICCD) camera (IStar 740, Andor).

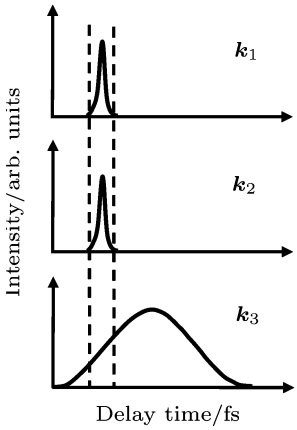


Fig. 2. Schematic of two pump pulses ($\boldsymbol{k}_1$ and $\boldsymbol{k}_2$) and SC pulse ($\boldsymbol{k}_3$) sequence in fs LITG experiments.

3. Results and discussion

As shown in Fig. 3(a), the three fs laser pulses $\boldsymbol{k}_1$, $\boldsymbol{k}_2$ and $\boldsymbol{k}_3$ were arranged in folded BOXCARS configuration.[22] The beam-crossing angle between adjacent beams was about 3°. According to the phase-matching, this configuration can be used to achieve background-free detection of diffraction signals at the inferior right corner of the folded box. Two orders of diffraction signals ($\boldsymbol{k}_{s1}$ and $\boldsymbol{k}_{s2}$) of SC pulse ($\boldsymbol{k}_3$) were defined by the wave vector $\boldsymbol{k}_{s1} = \boldsymbol{k}_1 - \boldsymbol{k}_2 + \boldsymbol{k}_3$ (first order diffraction) and $\boldsymbol{k}_{s2} = 2(\boldsymbol{k}_1 - \boldsymbol{k}_2) + \boldsymbol{k}_3$ (second order diffraction). Figure 3(b) shows the images of beam spots of two pump pulses ($\boldsymbol{k}_1$ and $\boldsymbol{k}_2$), SC pulse ($\boldsymbol{k}_3$) and the diffraction signals of two orders ($\boldsymbol{k}_{s1}$ and $\boldsymbol{k}_{s2}$). Practically, the spatial distribution of fs laser pulse induced white-light SC had a radial structure with a number of coloured rings around a common white centre.

According to the group velocity dispersion theory, the light beams of different wavelengths in an SC pulse arrives at CS_2 at different times.[23] By varying the delay time of SC ($\boldsymbol{k}_3$) while maintaining the two pump pulses fixed, different wavelengths in the SC were diffracted by the LITG induced by two pump pulses at different delay times. Thus the LITG acted as a "gate" to measure the chirp characteristics of the SC pulse. The time duration of LITG in CS_2, which was close to the duration of the two pump pulses, determined the time resolution of the fs LITG technique.[24] In contrast, the OKG signal of CS_2 was generally as long as several hundreds of fs, which was based on the birefringent effect due to the reorientation of CS_2.[25,26]

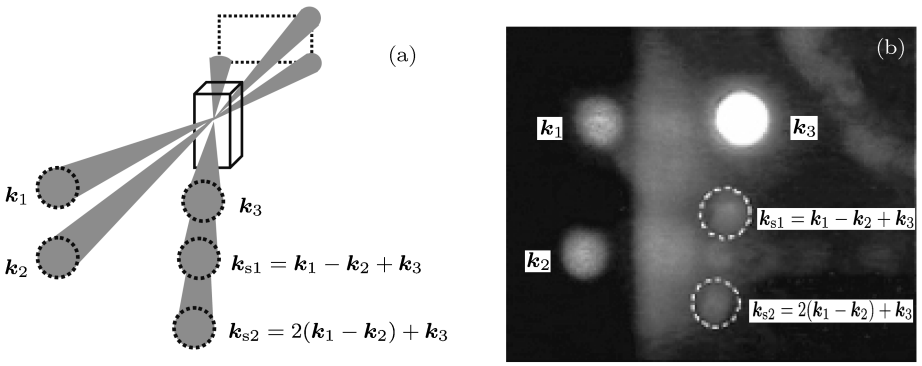


Fig. 3. (a) Beam arrangement in BOXCARS configuration in experiment, in which two pump pulses ($\boldsymbol{k}_1$ and $\boldsymbol{k}_2$) were overlapped and fixed in time and the time delay between two pump pulses ($\boldsymbol{k}_1$ and $\boldsymbol{k}_2$) and the SC ($\boldsymbol{k}_3$) was varied to obtain the time-dependent chirp structure of a white-light SC; (b) spatial profiles of two pump pulses, SC pulse and diffraction signals of two orders.

It is worth while mentioning that two fs pump pulses which coincided in both time and space and formed the fs LITG, provided a strong off-resonance ultrafast laser field on CS_2 molecules. The mass of an electron was so small that the response time of electronic phase varied with the fs pulse. Previous studies have proved that the dephasing time of an electron is on the order of several fs at room temperature.[27] Therefore the formation of fs LITG in CS_2 was mainly attributed to the polarization of electrons but not to the reorientation of molecules.[28]

Figure 4 shows the SC spectrum generated in Al_2O_3 crystal by an fs laser pulse. Its wavelengths ranged from 450 nm to 780 nm and the intense 800-nm

portion of the spectrum was filtered out by a low-pass filter. As shown in the inset of Fig. 4, the full width at half maximum (FWHM) of fs laser pulse measured with Positive Light Single Shot Autocorrelator (Positive Light, Inc.) in the experiment was 160 fs, and it was estimated to be the time duration of fs LITG in liquid CS_2.

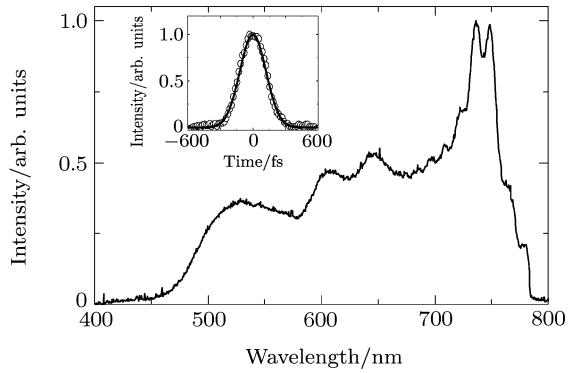


Fig. 4. Spectrum of SC generated with 800 nm, 160-fs laser pulse in 4-mm thick Al_2O_3 crystal. The inset shows the autocorrelation temporal trace of a pump laser pulse (represented by circles) and Guassian fits to the data (solid line).

The nonlinear diffraction signals of different wavelengths of SC by fs LITG in CS_2 were measured by changing the delay time between the SC pulse ($\boldsymbol{k}_3$) and the two pump pulses ($\boldsymbol{k}_1$ and $\boldsymbol{k}_2$), and the chirp characteristics of SC were obtained.

Figure 5 shows the contour maps of the first order and the second order diffraction signals of the white-light SC, showing the most of the wavelength components of SC in the time domain. The correlation between wavelength and delay time in SC can be clearly observed in the contour maps of two orders of diffraction signals, and there is an obvious difference between the first and the second order diffraction signals in the detected spectral contents. Since the two pump femtosecond laser pulses ($\boldsymbol{k}_1$ and $\boldsymbol{k}_2$) and the probe SC pulse ($\boldsymbol{k}_3$) were arranged in folded BOXCARS configuration, only when the probe wavelength phase matched, can it be diffracted by LITG. The first and the second order signal phases matched at different probe wavelengths respectively, so the first order diffraction signal of SC ranged from 450 nm to 600 nm (see Fig. 5(a)) and the second order diffraction signal of SC ranged from 480 nm to 680 nm (see Fig. 5(b)). Furthermore, at the wavelength ranging from 450 nm to 560 nm, the difference in refractive index between different corresponding wavelengths is small, so the difference in velocity between different corresponding wavelengths is also small. Wavelengths in the above region are much closer to each other than the wavelengths in a range from 560 nm to 680 nm, so the SC has lower chirp in the wavelength range from 450 nm to 560 nm as shown in Fig. 5. The measured chirp characteristics accord well with previous experimental results detected by OKG technique.[15,29]

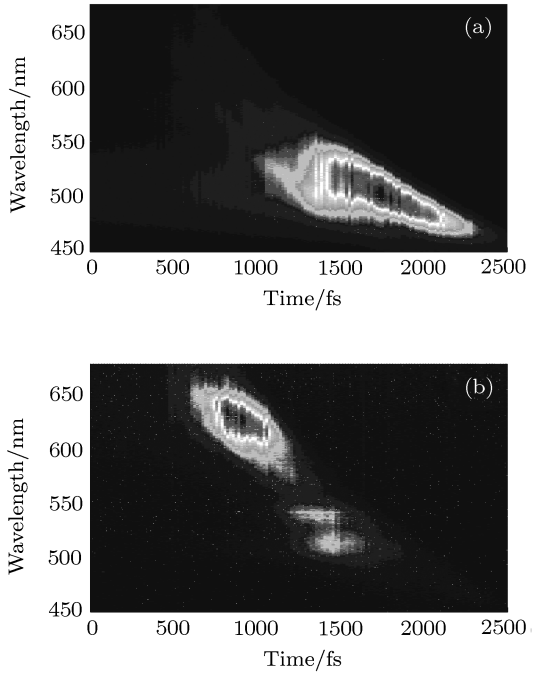


Fig. 5. Contour maps of the first (a) and second (b) order diffraction signals of the white-light SC, which was generated by a 800-nm, 160-fs laser pulse in 4-mm thick Al_2O_3 crystal.

The long-wave part ranging from 680 nm to 780 nm in fs SC pulse was not found in either of the two orders of diffraction signals, and it can be inferred that the long-wave part in fs SC was diffracted by fs LITG in CS_2 with lower diffraction efficiency, compared with the short-wave part ranging from 450 nm to 680 nm in fs SC. It has been demonstrated in experiment and theory that the diffraction efficiency decreases as the wavelength of probe pulse increases.[30]

In addition, the wavelength dependence of the fs LITG diffraction efficiency is also attributed to the nonlinear optical properties of liquid CS_2. The first order diffraction signal corresponds to the third order nonlinear optical process, whereas the second order diffraction signal corresponds to the fifth order optical process. It has been demonstrated experimentally that when the fs laser pulse intensity is in a range of $7.5\times10^{10}\sim2.2\times10^{11}$ W/cm^2, the third-order and the fifth-order process coexist in CS_2 due to the three-photon absorption.[31]

4. Conclusion

The fs LITG technique is used to achieve background-free detection of two orders of diffraction signals of SC generated by a 800-nm, 160-fs laser pulse in 4-mm thick Al_2O_3 crystal. The time duration of fs transient refractive-index grating in CS_2, which is formed due to the electronic polarization process, is close to the FWHM of fs pump pulses. It acts as a "gate" to measure the chirp structure of SC pulse. The first order and the second order diffraction signals of SC by fs LITG reveal the correlation between wavelength and delay time in a white-light SC, and it is demonstrated that the third-order process and the fifth-order process coexist in CS_2 at high fs laser intensity.

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