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# Experimental study on the chirped structure of the white-light continuum generation by femtosecond laser spectroscopy

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The chirped structure of the white-light continuum generation (WLCG) pulse produced by focusing 800nm laser pulse with a pulse duration of 150fs (FWHM: full-width-at-half-maximum) onto a 2.4 mm thick sapphire plate was investigated by the optical Kerr gate technique with normal hexane as the optical Kerr gate medium. The observed WLCG was positively chirped, the measured anti-Stokes spectrum of WLCG ranges from 449 to 580nm with a temporal span of 2.56ps. When using metal reflecting mirrors to eliminate the group velocity dispersion (GVD) effect, we found that a span of 1.3ps still remained, indicating that the chirped pulse cannot be accounted for simply by GVD of the pulse propagation in the dispersive media. Our results suggest that the light-induced refractive index change due to the third-order nonlinear optical effect leads to an additional positive group velocity dispersion, which contributes to an important portion of the observed temporal broadening of the chirped WLCG. In addition to using reflective optical elements instead of dispersive optical elements, an effective way of reducing the chirp is to minimize the optical path length of the WLCG medium.

**Keywords:** white-light continuum generation, chirped structure
**PACC:** 4265, 7847

## 1. Introduction

When focusing ultrafast laser pulses of high power density onto nonlinear optics media, by adjusting laser power and the focusing point, the input laser pulse will have a remarkable broadening in the spectral region associated with a chirped structure. This phenomenon is known as the white-light continuum generation (WLCG). With the rapid advance in femtosecond laser in the recent years, WLCG has been used in many fields,[1] especially acting as the probe pulse of the ultrafast time-resolved difference absorbance spectrum[2] or the seeding laser pulse in an optical parametric amplifier (OPA).[3] WLCG was first observed in crystals and glass by Alfano *et al* in 1970,[4] and later it was observed in the liquid media in 1977[5] and in the gas phase in 1986.[6] After the experimental observation, there have been several different mechanisms proposed to explain the WLCG phenomenon, including stimulated Raman scattering by Smith *et al*,[5,10] self-phase modulation by Yang *et al*[4,7–9] and four-wave mixing mechanism by Penzkofer *et al.*[10–12] However, in real cases all the above mechanisms can be involved simultaneously in WLCG, depending on the media as well as the power and the pulse width of the incident laser. In each case, one of the mechanisms would play the primary role.[13]

In the study of the ultrafast time-resolved difference absorbance spectroscopy, WLCG is generally employed as the probing beam for the photo-induced transient absorbance change because of its wide spectral range extending from a wavelength shorter than 400nm to that longer than 1000nm. However, owing to its chirped structure, i.e. within the temporal envelope of WLCG, light of different wavelengths arriving at different time, the different absorbance spectra cannot be acquired simultaneously, thus additional data processing such as chirp correction is necessary to re-

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construct the early time spectra.[14,15] The prevailing account of the chirped structure of WLCG is based on the group velocity dispersion theory,[20] which states that when a light pulse transmits through dispersive optical elements or media, a group velocity dispersion (GVD) effect is expected. Because of the positive GVD, low-frequency components within the WLCG pulse precede temporally over the high-frequency components, which leads to a broadening of the WLCG pulse. In this way the frequency distribution within the WLCG pulse follows a time sequence that forms a chirped structure.[16,20]

In 1995, Yamaguchi *et al*[16] measured the chirped structure of WLCG generated in water by use of Kerr gate technique.[17] In this paper, we measure the chirped structure of WLCG generated in a sapphire plate by the same method. We further compare the chirped structures of WLCG measured under two different conditions, i.e. by use of non-reflective optical elements and reflective optical elements respectively. In the latter case, the GVD effect during the light propagation can be eliminated. The experimental results free of GVD effect are compared with those with GVD correction in theory. We find that the temporal broadening of WLCG cannot be accounted for simply by GVD of the pulse propagation in the dispersive media. Our results suggest that the light-induced refractive index change due to the third-order nonlinear optical effect leads to an additional positive group velocity dispersion, which contributes to an important portion of the observed temporal broadening of the chirped WLCG pulse.

## 2. Experimental arrangement

The experimental set-up is shown in Fig.1. The 800nm fundamental frequency was delivered from a regenerative Ti: sapphire femtosecond laser (Hurricane, Spectra Physics Inc.) with a repetition rate of 1kHz and a pulse duration of 150fs (FWHM). The output laser beam was split into two. One beam after passing through an optical delay translation stage was focused onto a second harmonic generation (SHG) BBO crystal of 1 mm thick to generate 400nm laser. The other with a power of 6$\mu$J per pulse was focused onto a sapphire plate of 2.4mm thick to generate WLCG. Polarization of WLCG was determined to be the same as that of 800nm laser. The beam of WLCG passed through a quartz polarizer and an analyser, in between them a glass cuvette with a path length of 2 mm filled with normal hexane was placed as the medium for the optical Kerr gate. The 400nm laser beam was employed as the pump beam for the optical Kerr effect. Its polarization was set to 45° with respect to the WLCG by a 1/2$\lambda$ wave plate. Both the pump and the WLCG beams were focused individually into the normal hexane, with their focal points overlapped inside the medium. The time delay between the pump beam and the WLCG was realized by a computer controlled translation stage. Since normal hexane has a quick second-order nonlinear optical response,[16] the Kerr effect acts as a time gate with a gate width comparable to that of the laser pulse. The Kerr effect induced transmission of WLCG was detected by a CCD assisted spectrometer interfaced to a computer for data handling. The data were collected and analysed by a computer.

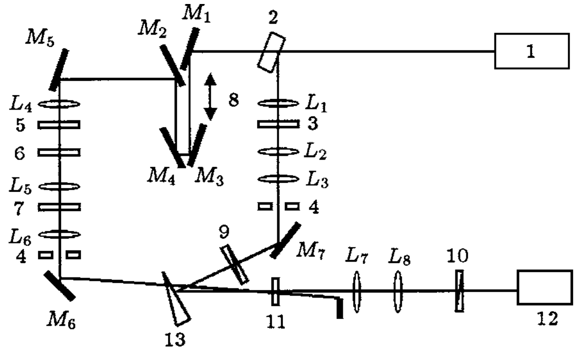

**Fig.1.** Experimental set-up for the measurement of chirped WLCG using optical Kerr gate with non-reflective optical elements: 1. Ti:sapphire laser; 2. beamsplitter; 3. sapphire plate; 4. electronic shutter; 5. BBO crystal; 6. 800nm filter; 7. 1/2$\lambda$ wave plate; 8. translation stage; 9. polarizer; 10. analyser; 11. cuvette filled with normal hexane; 12. CCD equipped spectrometer; 13. optical wedge. $L$: lenses; $M$: reflecting mirrors.

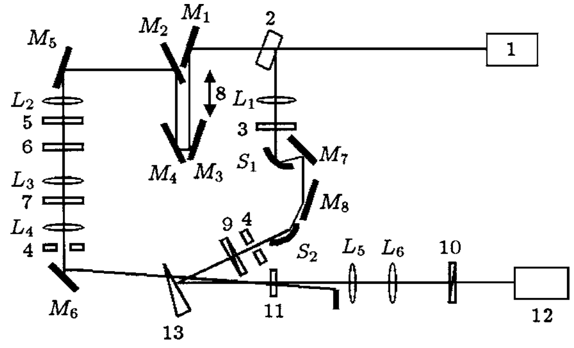

**Fig.2.** Experimental set-up for the measurement of chirped WLCG using optical Kerr gate with reflective optical elements for elimination of GVD effect: $S$: spherical metal reflecting mirrors. All the other symbols are same as in Fig.1.

To minimize the temporal broadening caused by GVD, metal concave reflecting mirrors were employed instead of lenses, and the chirped structure of WLCG were recorded accordingly. The experimental arrangements are shown in Fig.2.

## 3. Results

Figure 3 shows a series of gated spectra of WLCG detected at different time delay of the pump laser with the lenses as the focusing elements. Obviously at the leading edge of the WLCG pulse the lower-frequency spectral components are dominant, while the trailing edge consists of the high-frequency spectral components, i.e. the WLCG pulse is positively chirped.

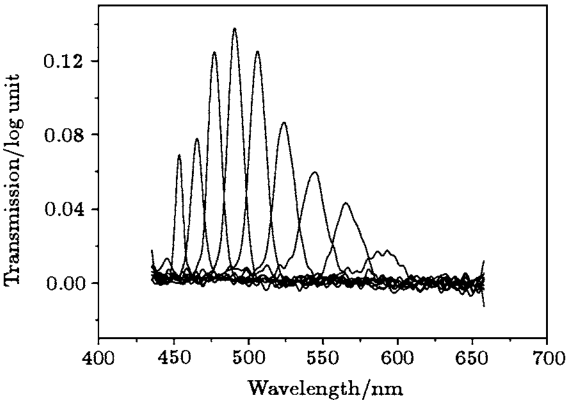

**Fig.3.** A series of the gated spectra of WLCG probed at different time delay of Kerr gate using lenses. From left to right, the relative time delay is from −20.3 to −17.6ps with an interval of 0.3ps.

Figure 4 presents the relative delay time versus the various peak wavelength of the gated spectra.

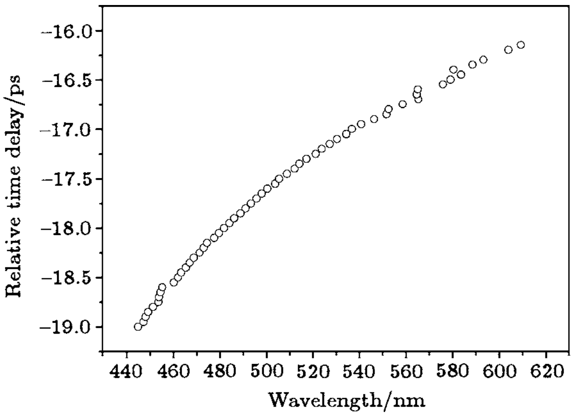

**Fig.4.** Plot of relative time delay against the peak wavelength of the gated spectra of chirped WLCG for non-reflective optical elements.

Figure 5 shows some selected gated spectra at four different wavelengths. It clearly demonstrates that the WLCG is positively chirped, since the low-frequency components of the pulse precede ahead of the high-frequency components. Figure 6 is the auto-correlation curve of the 800nm laser acquired by second harmonic generation in a BBO crystal of 1mm thick. Comparing Figs 5 and 6, it is concluded that the pulse shape of the individual spectral component is similar to that of the 800nm laser pulse, and the width of the Kerr gate is comparable to that of the laser pulse.

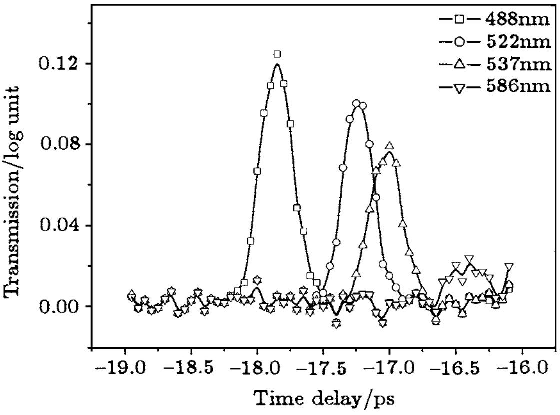

**Fig.5.** Line shapes of a single wavelength pulse within the WLCG at four selected wavelengths for non-reflective optical elements.

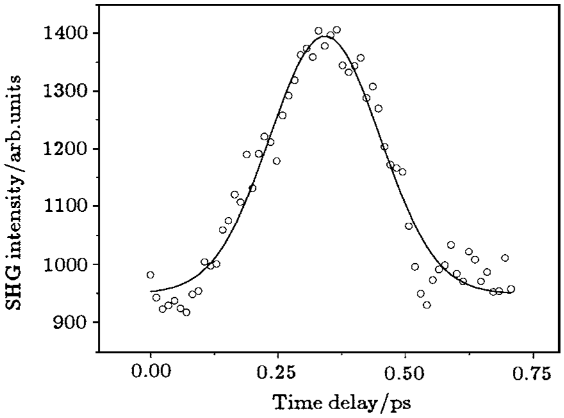

**Fig.6.** Auto-correlation curve of the fundamental 800nm laser pulse.

Figure 7 gives an envelope of the WLCG pulse constructed by plotting the integrated intensity at different time delay against the delay time. The lineshape can be fitted by a Gaussian type of curve with a FWHM of 1.2ps.

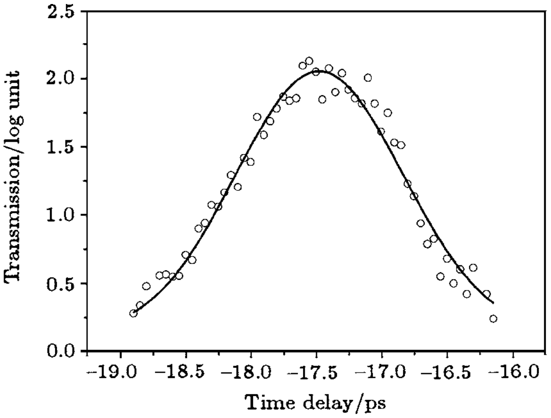

**Fig.7.** Line shape of the WLCG wavepacket constructed from the integral intensity of the gated spectra at different time delay for the non-reflective optical elements.

When replacing lenses with metal concave reflecting mirrors, the corresponding relative delay time versus the various peak wavelength of the gated spectra is shown in Fig.8.

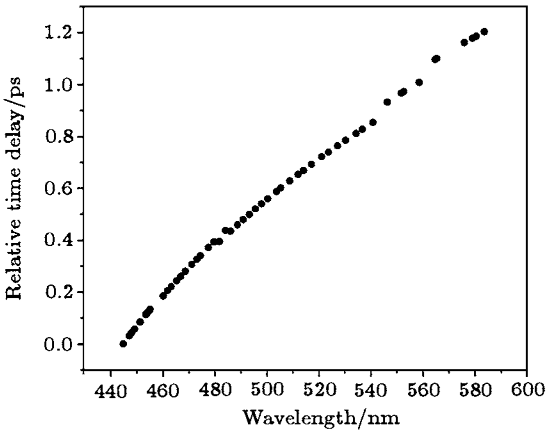

**Fig.8.** Plot of relative time delay against the peak wavelength of the gated spectra of the chirped WLCG for reflective optical elements.

## 4. Discussion

Under our experimental condition, the WLCG is generated mainly by the self-phase modulation mechanism, while the spectrum investigated lies in anti-Stokes area.[7] When focusing ultrafast laser pulses of high power density into the media, the third-order nonlinear optical effect will induce a transient refractive index change in the media, giving rise to a transient change in the phase of the laser pulse propagating in the media in response. Such an effect is called the self-phase modulation. It is generally believed that the WLCG produced by the self-modulation effect would cause a chirped structure of the propagating wavepacket, i.e. the leading edge of the WLCG pulse comprises the red spectral component while the trailing edge comprises the blue component, and the self-modulation effect may not necessarily lead to the broadening of the WLCG pulse. The temporal broadening of the WLCG pulse is caused by GVD effect when it transmits through the media, air and non-reflective optical elements.[16] Thus, the broadening of WLCG caused by GVD effect can be calculated in principle.

Considering the case when non-reflective optical elements are used. In such a case, the GVD effect arises mainly from the light propagation in the air, lenses, sapphire plate that generates WLCG and normal hexane used as Kerr effect media, among which the GVD effects in air and normal hexane are small and can be neglected in the calculation. In the light path for WLCG shown in Fig.1, there are two quartz lenses, one quartz polarizer and analyser mounted to a total thickness of 7.5mm between the sapphire plate and the Kerr gate medium.

The dispersion formula for quartz is as follows[18]

$$\begin{aligned} n^2-1=&\frac{0.6961663\times\lambda^2}{\lambda^2-0.0684043^2}\\ &+\frac{0.4079426\times\lambda^2}{\lambda^2-0.1162414^2}\\ &+\frac{0.8974794\times\lambda^2}{\lambda^2-9.896161^2}; \end{aligned}$$

and that for sapphire reads[18]

$$\begin{aligned} n^2-1=&\frac{1.023798\times\lambda^2}{\lambda^2-0.00377588}\\ &+\frac{1.058264\times\lambda^2}{\lambda^2-0.0122544}\\ &+\frac{5.280792\times\lambda^2}{\lambda^2-321.3616}. \end{aligned}$$

The group velocity of a wavepacket centring at different wavelengths is

$$v_{\rm g}(\lambda)=\frac{c}{n(\lambda)-\lambda\dfrac{{\rm d}n(\lambda)}{{\rm d}\lambda}}.$$

Temporal broadening of the WLCG pulses caused by a positive GVD is defined as follows

$$\Delta t=\frac{L}{v_{\rm g}(\lambda_i)}-\frac{L}{v_{\rm g}(\lambda_0)},$$

where $L$ is the path length in the media, subscript 0 refers to the starting reference wavelength and subscript $i$ indicates the wavelength discussed. The calculated relative delay time at the corresponding wavelength caused by the GVD effect is plotted in Fig.9.

By subtracting the calculated time delay from the observed one, the contribution from the high-order nonlinear optical effect, i.e. the GVD corrected delay time can be obtained, as shown in Fig.9.

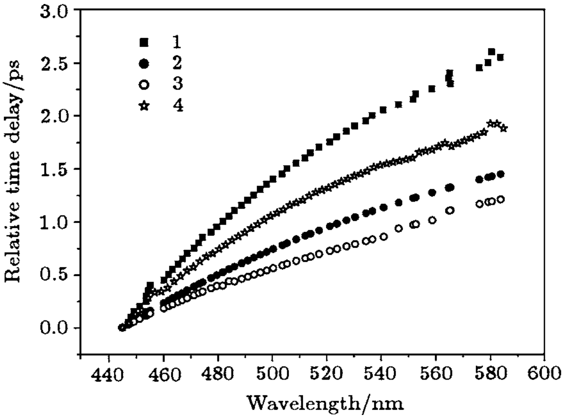

**Fig.9.** Comparison of the chirping effect of WLCG with and without the GVD effect: 1. With GVD for non-reflective optical elements; 2. our result of elimination of GVD with reflective optical elements; 3. the result of elimination of GVD with reflective optical elements in Koyama's laboratory; 4. result for non-reflective optical elements corrected for the GVD effect by calculation.

When the lenses are replaced by metal concave reflective mirrors, the GVD effect can be partially eliminated. The experimentally observed temporal broadening under such a condition is thus supposed to be caused only by the nonlinear optical effect inside the sapphire plate. Our experimental result and that of Koyama's laboratory in Kwansei Gakuin University, Japan, by use of metal reflective mirrors are also given in Fig.9 for comparison.

As shown in Fig.9, the measured anti-Stokes spectrum of WLCG ranges from 449 to 580nm with a temporal span of 2.56ps for non-reflective optical elements. When using metal reflecting mirrors to eliminate the GVD effect, we found that a span of 1.3ps still remains, which is comparable to the GVD corrected value. When using metal reflecting mirrors, because the optical path departed from the optical axis and WLCG has a chromatic aberration, which accounts for the discrepancy between our result and that of Koyama's group. Our result unambiguously suggests that a substantial amount of chirped pulse broadening is caused by high-order nonlinear optical effect in the WLCG-generating medium which cannot be removed by use of reflective optical elements. We believe that a thinner sapphire plate would be helpful in further reduction of the temporal broadening of the WLCG pulse. In fact, we have tried to use a 500$\mu$m thick sapphire plate to generate WLCG, however, the WLCG produced in this way was quite unstable. Attempt to characterize the chirped structure of WLCG in a 500$\mu$m thick sapphire plate is failed.

## 5. Conclusion

The temporal broadening of the chirped WLCG pulse is proved to be associated with the generation of WLCG by self-phase modulation. The third-order nonlinear optical effect changes the transient refractive index of the media causes self-phase modulation effect which results in WLCG. The change of the transient refractive index of the media also leads to the temporal broadening of the chirped WLCG pulse and induces the temporal broadening of WLCG pulses. Such a temporal broadening of the WLCG pulse cannot be eliminated by using metal reflecting elements. However, it might be compensated by other means. Two methods may be used to minimize the temporal broadening of the WLCG pulse. One is to reduce the thickness of the WLCG media, and the other is to utilize optical elements such as chirp mirrors or a pair of prisms to compensate the chirped structure.

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