Sub femto-joule sensitive single-shot OPA-XFROG and its application in study of white-light supercontinuum generation

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Abstract: We report a new design of single-shot cross-correlation frequency-resolved optical gating (XFROG) with sub femto-joule sensitivity for complete field characterization of ultrashort optical pulse using a 400-nm-pumped type-I noncollinear optical parametric amplifier. Optical parametric gain as high as 10^8 with 0.8-0.9 femto-joules sensitivity had been demonstrated with an un-cooled CCD in this study. The experimental FROG traces have been successfully retrieved with an error no worse than 0.0014. The device had been to be useful for studying the generation mechanism of white-light supercontinuum (WLS).

OCIS codes: (320.0320) Ultrafast optics; (320.7100) Ultrafast measurements; (190.4410) Nonlinear optical parametric processes; (190.7110) Ultrafast nonlinear optics; (190.4970) Parametric oscillators and amplifiers; (300.6280) Spectroscopy, fluorescence and luminescence; (300.6420) Spectroscopy, nonlinear; (300.6530) Spectroscopy, ultrafast

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#3302 - $15.00 US Received 4 November 2003; revised 3 February 2004; accepted 7 February 2004

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1. Introduction

In the past decade, measuring the intensity and phase vs. time of femtosecond laser pulses has become a great issue due to its application in characterization of ultrafast signal. Basically, there are several families, including direct optical spectral phase measurement (DOSPM) [1], frequency-domain phase measurement (FDPM) [2], frequency-resolved optical gating (FROG) [3,4], and others [5], to characterize the amplitude and phase of electric field profile associated with the optical pulse. Techniques such as Frequency- Resolved Optical Gating (FROG) and Cross-correlation FROG (XFROG) [6,7] allow the measurement of a wide range of pulses. While these techniques have achieved fairly high sensitivity, they are not sensitive enough to measure extremely weak ultrashort light pulses [8]. Using spectral interferometry [9] trains of ~1-photon pulses could be measured was reported previously. Unfortunately, interferometric methods can be useful for aligning lasers and studying high-optical quality media, but they are of little value in more practical situations. So a non-interferometric technique lacking such restrictive coherence requirements and which achieves few-photon sensitivity is needed to help to elucidate fundamental weak-light emitting processes in many fields.

A new technique, which combines optical gating with high-gain optical parametric amplifier (OPA), had been recently demonstrated by one of us (Zhang) to be able to characterize weak spatially incoherent optical pulses with a multi-shot cross-correlated frequency-resolved optical gating (XFROG) [10]. The resulting OPA-XFROG technique had been proposed to measure ultrashort optical signal with extremely weak intensity. The detection sensitivity of OPA-XFROG is so high that the pulse energy of optical signal can be as low as a few atto-joules (<100 photons). The direct comparison of the optical field profiles deduced with OPA-XFROG and sum frequency generation (SFG) reveals that OPA-XFROG can be as accurate as SFG-XFROG.

Multi-shot OPA-XFROG measurement requires a scan of delay time between the pump and signal pulses. The acquisition of a typical OPA-XFROG pattern can take up to more than 20 minutes. This is particularly problematic when the signal pulses are either weak or irreproducible from shot to shot. Single-shot FROG was designed to conquer such difficulties [11,12,13]. A typical single-shot FROG setup maps delay time onto different spatial positions

#3302 - $15.00 US Received 4 November 2003; revised 3 February 2004; accepted 7 February 2004

by focusing two input beams with cylindrical lens. In order to minimize the group velocity dispersion (GVD) originating from dispersive optical components, the lenses had been replaced with two cylindrical mirrors in this study. The two beams cross each other in the vertical direction as they travel through a nonlinear optical medium. The line focusing geometry used in a single-shot FROG apparatus often leads to a detection sensitivity much lower than that of multi-shot FROG apparatus [11]. Therefore, single-shot FROG had only been employed to characterize laser pulses with sufficiently high intensity. In this article, we report a new design of single-shot cross-correlation frequency-resolved optical gating (XFROG) with sub femto-joule sensitivity for complete field characterization of ultrashort optical pulse using a 400-nm-pumped type-I noncollinear optical parametric amplifier. Optical parametric gain as high as 10^8 with 0.8-0.9 femto-joules sensitivity had been demonstrated with an un-cooled CCD in this study. The experimental FROG traces have been successfully retrieved with an error no worse than 0.0014. The device has been useful in the studying the generation mechanism of white-light supercontinuum (WLS).

The combination of single-shot FROG with high-gain OPA could overcome the drawbacks encountered in both multi-shot FROG and single-shot FROG without gain. The weak optical pulse is amplified by optical parametric amplification process in a nonlinear crystal. The resulting line-shaped amplified signal is then imaged onto the entrance slit of a spectrometer and is detected with a 2D-CCD camera. The spectral content of the pulse is revealed along the horizontal axis of the OPA-XFROG pattern, and the vertical distribution presents the detailed information about the temporal profile. An entire FROG pattern can be acquired for each laser pulse. Note that single-shot scheme might also be valuable for probing few-cycles ultrashort pulses [14], since in this case pulse-to-pulse fluctuation becomes the major error source of pulse characterization.

There are two ways to implement single-shot OPA-FROG: First, when the duration of pump pulse is longer than that of signal pulse, one can use a collinearly phase-matched OPA with the weak signal as the seeding pulse. In this case, the signal beam is temporally and spatially overlapped with the pump in the nonlinear medium and no beam-crossing is needed. The amplified signal is then fed into a single-shot FROG to reveal the pulse characteristics of the signal [11,12,13]. The second approach can be employed when the duration of weak signal pulse is longer than that of the pump pulse. In this case two input beams shall cross each other as they travel through the nonlinear medium to have all temporal and spectral components of the signal pulse been amplified.

2. Experimental arrangement

The schematic of our single-shot beam-crossing OPA-FROG is presented in Fig. 1. A regenerative amplified Ti: sapphire laser provides output energy of more than 1 mJ/pulse at 810 nm with pulse duration of ~90 fs. The laser output is split into two parts: 90% of the energy is frequency-doubled to 405-nm with a 0.3-mm-thick type-I β-barium borate (BBO) second-harmonic generator (SHG). The SHG beam is then separated from the fundamental beam by dichroic mirrors and used to pump a type-I noncollinearly phase-matched BBO optical parametric amplifier (NOPA) with pump energy adjustable from 50 to 120 μJ. The rest 10% of the Ti:sapphire laser beam is used to generate white-light super continuum (WLS) with a 2-mm-thick CaF_2 plate [15]. The WLS generated is collimated and then focused by a cylindrical mirror onto the 2-mm long BBO NOPA crystal cut at $\theta=29^o$. The pump beam at 405-nm is horizontally focused with a cylindrical mirror to form a vertical line image. The pump and WLS are brought to the BBO crystal with a beam-crossing angle of about 5-6 degrees in the vertical direction.

The strong pump beam not only provides optical gain for the weak signal but also acts as an optical gating to resolve the signal pulse profile. During the amplification process, the signal pulse collides with the pump and is sampled along the vertical direction. The amplified signal was then imaged onto the entrance slit of a spectrometer with 0.15-m focal length. The spectral property of the amplified signal field is analyzed by the spectrometer in the horizontal

Received 4 November 2003; revised 3 February 2004; accepted 7 February 2004

direction while the vertical intensity distribution carries the temporal information. The FROG trace was acquired with a 2-D CCD camera without scanning any mechanical delay line.

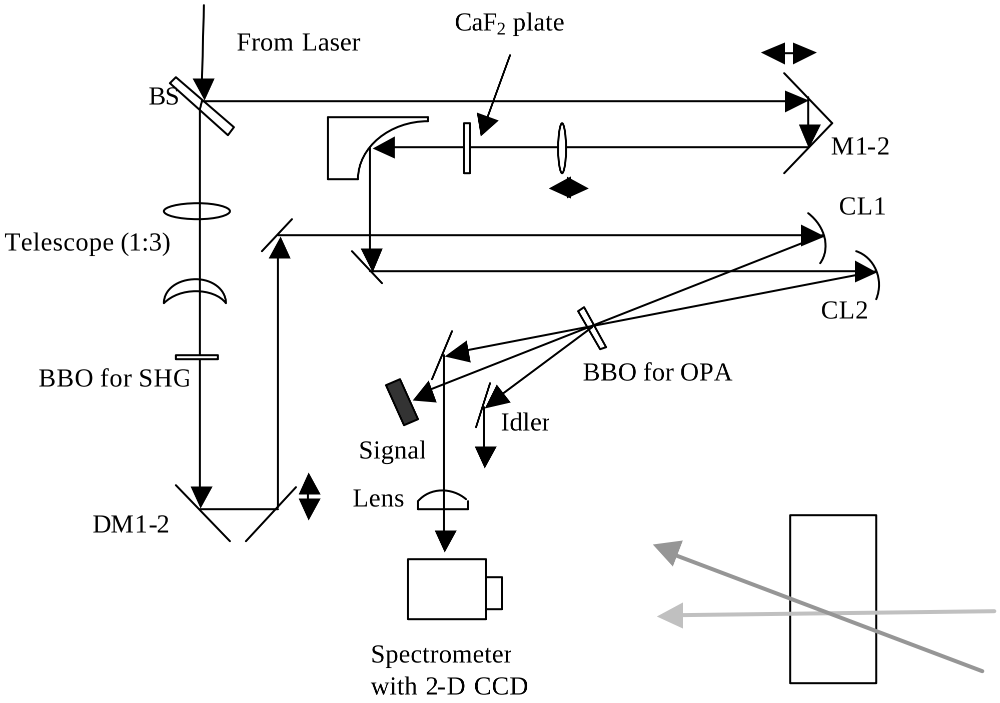

Fig. 1. Schematic setup of a single-shot XFROG apparatus for characterizing ultrashort optical field with extremely low intensity via high-gain optical parametric amplifier

OPA-XFROG apparatus can also be employed to characterize infrared pulses. For example, one can seed the OPA-FROG at the idler wave and then measure the corresponding signal-wave, which typically lies from the visible to the near IR spectrum region. Therefore the measurable wavelength range of OPA-FROG can cover the complete tuning range of the OPA used. For a typical 405-nm pumped type-I BBO NOPA, the measurable wavelength range can easily extend from 460 nm to 2.5 µm [16].

OPA-FROG involves a nonlinear-optical process with high optical gain. This feature allows us to increase the detection sensitivity by several orders of magnitude and to characterize extremely weak optical signals. With an uncooled CCD, we found the sensitivity of our single-shot OPA-XFROG to be 0.8-0.9 femto-joule. With a cooled CCD even higher sensitivity is expected. By modifying the conventional FROG algorithm we are able to retrieve the full field profile from a single-shot OPA-XFROG trace, the retrieved error is no more than 0.0014. The detailed theoretical analysis will be presented in a separated paper [17].

3. Results and discussions

The single-shot OPA-XFROG apparatus described above can be applied to characterize fairly complicated pulse profiles. To exhibit this ability, we present in Fig. 2 the OPA-FROG patterns with a seeding pulse from white light supercontinuum (WLS) and adjust the BBO crystal orientation to achieve the phase matching at 640-nm. The retrieved OPA-XFROG pattern, field profile, and temporal phase are shown in Figs. 2(b) and 2(c). As indicated by the retrieved temporal phase, at least 12π phase change is experienced within the pulse duration. In addition, the frequency is up chirping towards the positive delay time. This is reasonable in

Received 4 November 2003; revised 3 February 2004; accepted 7 February 2004

view that the frequency shifting from the pump wavelength (~800 nm) to 640-nm is quite large.

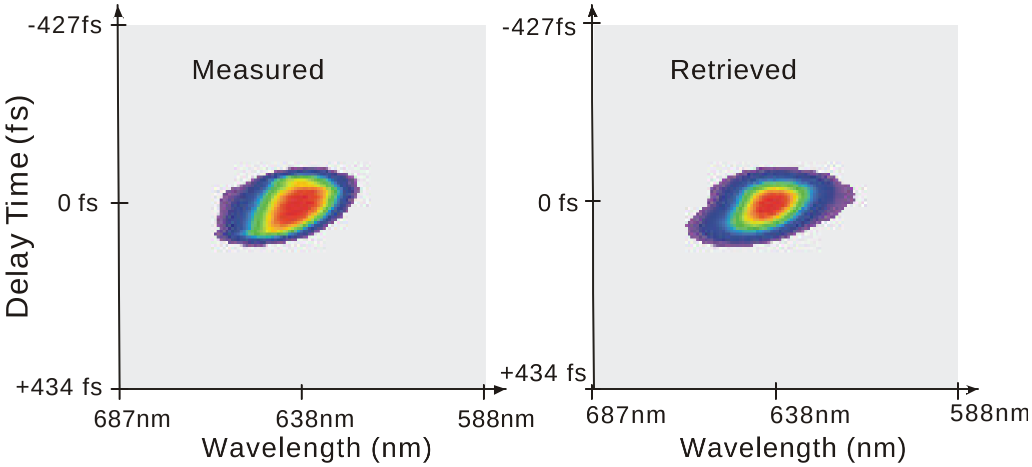

Fig. 2 (a) Measured and (b) retrieved OPA-FROG traces at a seeding wavelength of 640 nm. The retrieved error is about 0.0013

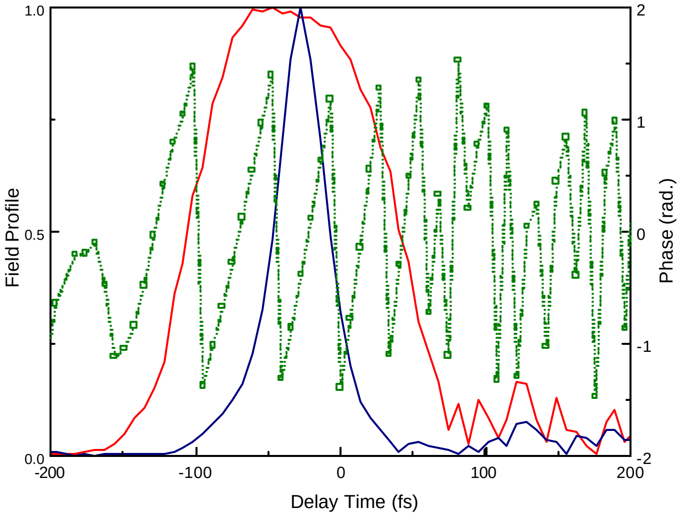

Fig. 2. (c) Field profile of the seeding pulse at 640 nm (solid curve with red color) and the corresponding temporal phase (open symbols) are retrieved from the OPA-XFROG trace shown in Fig. 2(a). The pump field profile (solid curve with blue color) at 400 nm with 52-fs FWHM pulse duration is included for comparison

The temporal phase distortion in WLS shall become smaller at a spectral position near to the pump wavelength. To confirm this, we further characterize WLS pulse by adjusting BBO crystal orientation to achieve the phase matching at 840-nm. The results are presented in Fig. 3. The temporal phase shown in Fig. 3(c) is indeed close to the case with a near-constant phase within the pulse duration.

Received 4 November 2003; revised 3 February 2004; accepted 7 February 2004

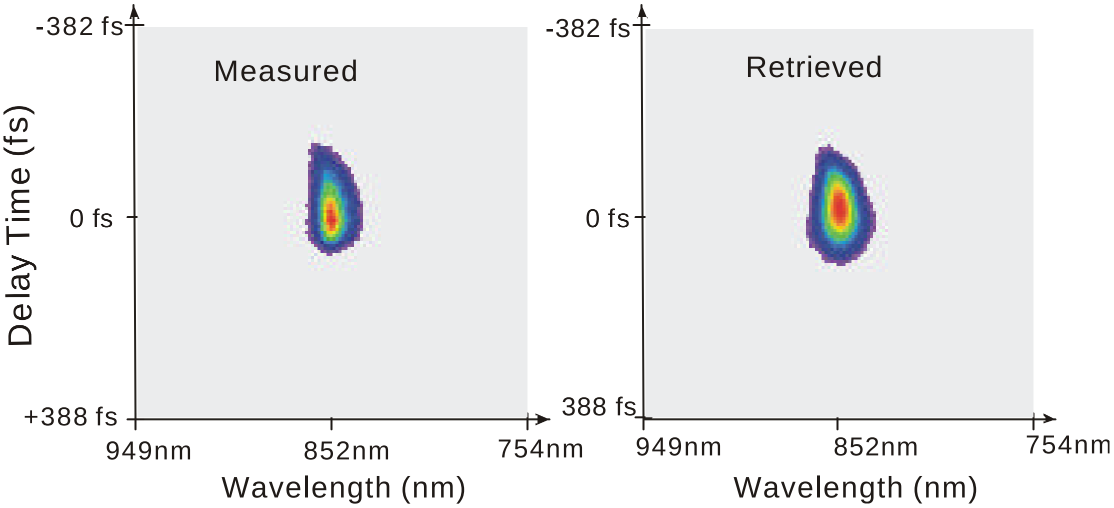

Fig. 3. (a) Measured and (b) retrieved OPA-FROG traces at an idler wavelength near 840 nm. The retrieved error is about 0.0014.

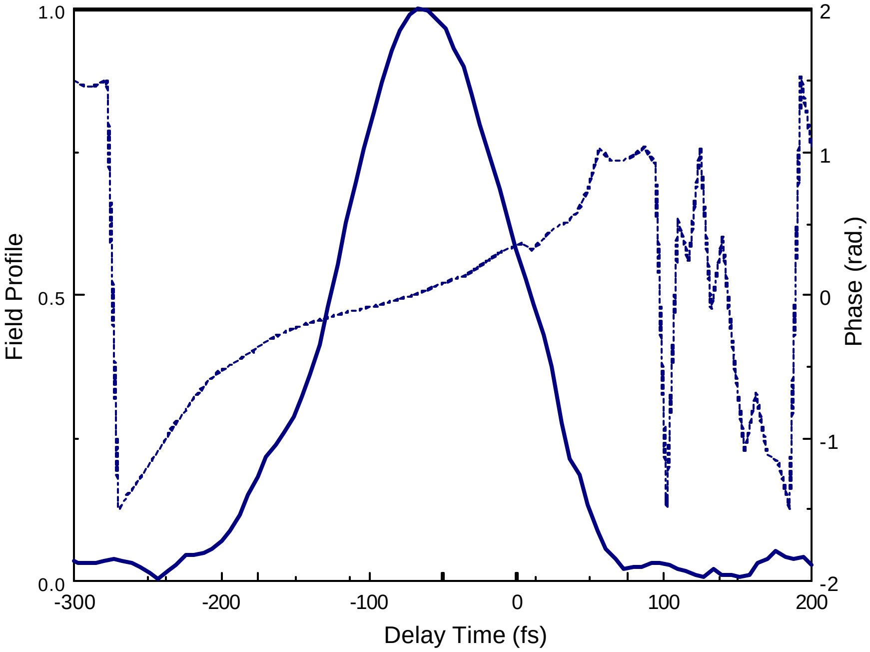

Fig. 3. (c) Field profile of the seeding pulse at 840 nm (solid curve) and the corresponding temporal phase (dashed curve) are retrieved from the OPA-XFROG trace shown in Fig. 3(a)

When the pump intensity of WLS is increased, the spectrum of the idler pulse of OPA at 840 nm further extends at the short-wavelength side. The slight tilting in the OPA-XFROG trace shown in Fig. 4 indicates the pulse experiences a non negligible quadratic phase distortion.

#3302 - $15.00 US
Received 4 November 2003; revised 3 February 2004; accepted 7 February 2004

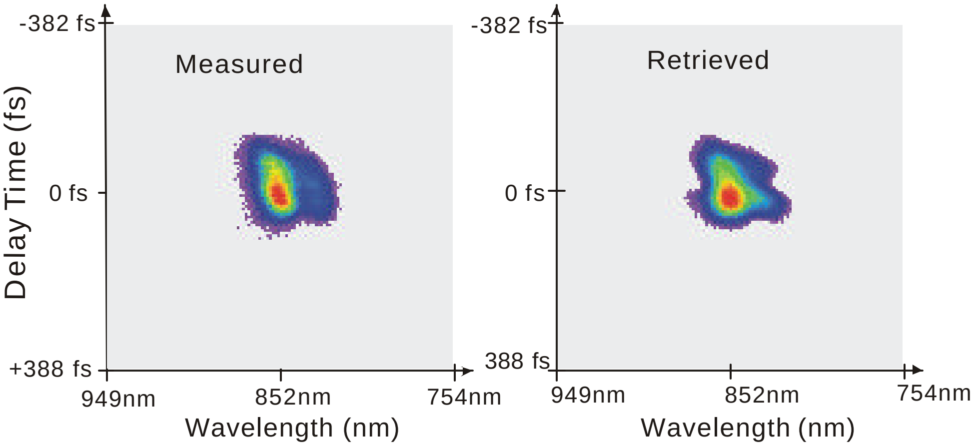

Fig. 4. Measured and retrieved OPA-FROG traces at an idler wavelength of 840 nm from a supercontinuum white light generator pumped by 1-kHz 52-fs pulses @ 800-nm with an averaged power of 1.5 mW.

By bringing the pump intensity of WLS above the threshold of multifilament formation (~9×10^{11} W/cm^2) [18], temporal pulse breakup is expected to appear [19, 20]. In Fig. 5, we present a single-shot OPA-FROG trace of the WLS generator pumped at a fairly high intensity of I_p=2×10^{13} W/cm^2.

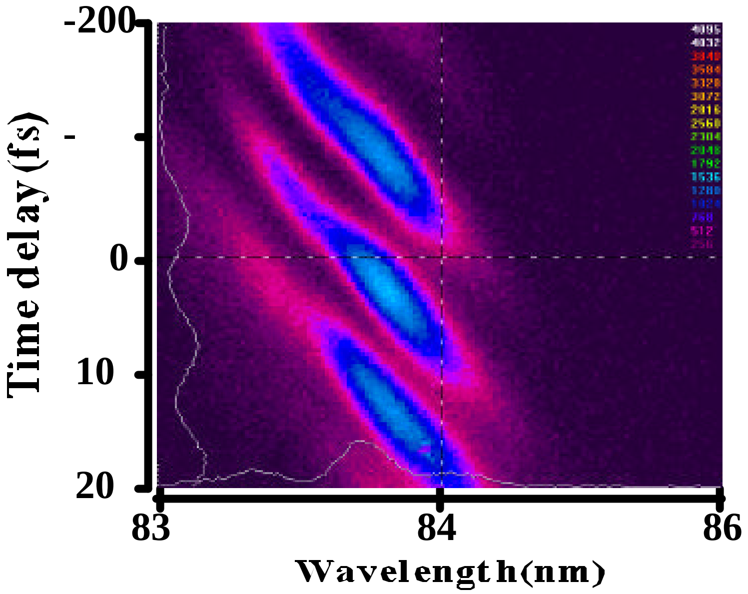

Fig. 5. Measured OPA-FROG trace at an idler wavelength of 840 nm from a highly excited super continuum white light generator pumped by 1-kHz 52-fs pulses @ 800-nm with an averaged power of 4 mW

Three tilting stripes can be observed. Each stripe in Fig. 5 is found to be separated along the time-delay axis by 100 fs. Assuming the multiple stripes appearing in Fig. 5 to be originated from that different index of refraction have encountered at different position on the pump beam cross section. Therefore the temporal separation of the breakup pulses can be expressed as $\Delta t = n_2 I_p / L$. Note that to generate WLS we use a 2-mm thick CaF_2 plate.

#3302 - $15.00 US Received 4 November 2003; revised 3 February 2004; accepted 7 February 2004

With L=2 mm, I=2×10^{13} W/cm^2, and $\Delta\tau$=100 fs, we calculate n_2 of CaF_2 to be 7.5×10^{-16} cm^2/W. This value is about two times larger than sapphire [21] and remains a reasonable estimate in view of the high uncertainty of pump intensity in WLS medium. It has been known that multifilament formation in WLS is extremely unstable and unrepeatable. In fact, this has been the major cause for our poor knowledge on the generation mechanism of WLS. Single-shot OPA-FROG design reported in this Letter can be a useful apparatus for probing such weak optical pulse generated from ultrafast and unrepeatable process.

There is a detection limit on the pulse duration for the single-shot XFROG measurement. The maximum pulse width that the device can handle depends on the beam-crossing angle α between the pump and the seeding pulses, the thickness *d* and the index of refraction of the nonlinear crystal. With *v* denoting the speed of light in the medium, the maximum measurable signal pulse width can be estimated to be: [11]

$$\Delta t = d(1-\cos a)/v \ .$$

With a 2-mm thick BBO crystal and the visible signal wavelength, the value of $\Delta\tau$ is found to be a few hundreds femtoseconds. For short signal pulses, thinner BBO with a thickness less than 100 μm shall be used to reduce GVD of the crystal. Since no dispersive components are involved in our design, we believe our setup could be useful in measuring optical pulses down to a few optical cycles using an improved design. Our recent study on white-light OPA showed that an extremely broad bandwidth from the visible to the near-IR can be achieved. In addition, special mode-matching scheme, spatial coherence, and highly stable absolute phase of the optical pulse are not needed in our single-shot OPA-XFROG technique.

4. Summary

In summary, we have demonstrated, for the first time, a new single-shot XFROG technique for characterizing ultrashort optical pulses with extremely weak intensity. By using beam-crossing OPA-FROG with all reflective optics, our design can be used to measure optical pulse with pulse energy less than femto-joule and pulse width as short as a few femtoseconds. The measurable wavelength range of the light pulses can cover from the visible to the IR, depending on the phase-matching of nonlinear crystal used, the pump wavelength, and the detailed configuration of OPA. The device has been used in study of mechanism of white-light supercontinuum at high pumping intensity above the threshold of multifilament formation where the signal is irreproducible fromshot to shot.

Acknowledgments

The authors acknowledge the sponsorship of National Science Council under various grants and the Pursuit of Academic Excellence Program of the Ministry of Education, Republic of China. Dr. J. -Y. Zhang thanks the support of Georgia Southern University during his sabbatical leave.

#3302 - $15.00 US Received 4 November 2003; revised 3 February 2004; accepted 7 February 2004