Characterization of Ultrashort Optical Pulses With Third-Harmonic-Generation Based Triple Autocorrelation

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***Abstract*—We present a method to obtain complete information of femtosecond pulses. By measuring triple-optical autocorrelation directly with third-harmonic generation, without spectral information, a temporal pulse shape can be obtained by analytical calculation without direction-of-time ambiguity. Combining the resulting optical pulse shape with its corresponding optical spectrum, the exact phase and color variations in time can all be recovered with a Gerchberg–Saxton algorithm through an iterative calculation with an $O(n)$ complexity.**

***Index Terms*—Pulse characterization, third-harmonic generation, triple correlation.**

I. Introduction

THE ULTRASHORT optical pulse is an increasingly important tool for many fields, from basic physics, chemistry, and biomedicine to applications including micro-machining and high-bandwidth optical communications. Its short optical duration allows us to study and utilize ultrafast time-domain events. Therefore, for most of these temporal applications, measuring the ultrashort optical pulse shape is a crucial diagnosis. Recent advances in ultrafast technology have pushed optical pulse duration below 5 fs, which is so fast that electronic devices (streak camera, etc.) are too slow to measure the evolution. Many techniques were developed to characterize the temporal pulse shape [1]–[5]. By far, the most common mean to evaluate femtosecond optical pulses is optical autocorrelation measurement based on second-harmonic generation (SHG) [1], [2]. An incident pulse is split into two replicas with relative temporal delay. These two pulses are then recombined in an SHG crystal. Thus, the generated SHG signal, with one photon contributed from each pulse, is usually detected with a photomultiplier tube and the variation of detected photocurrent with delay yields the pulse autocorrelation function

$$G^2(\tau) = \int I(t)I(t+\tau)\,dt. \tag{1}$$

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A proper measurement of autocorrelation can provide estimation on the pulse duration, and with the aid of high-order nonlinearity [3]–[5], some pulse asymmetry information can also be extracted without direction-of-time ambiguity. However, the complete temporal optical pulse shape cannot be determined directly from these previously proposed two-arm correlations [1]–[5] due to limited information in the measurements.

To obtain the exact optical pulse shape, an extra dimension of information is required for measurement. In the late 1960s, Blount *et al.* had proposed that by measuring the triple-correlation function, the temporal intensity of a pulse can be analytically calculated without direction-of-time ambiguity [6]. Moreover, computer simulation also showed that this suggested method is quite insensitive to noise [7]. This proposed triple-optical autocorrelation has been implemented previously with a cascade of SHG and sum frequency generation [8]. However, for pulse-formation dynamic study and other applications including optical coherent controls [9]–[11], we not only need the complete knowledge of pulse intensity variation in time, but also the color variation in time. As a Fourier correspondence in frequency-time transformation, determination of color variation in time means phase reconstruction of the measured optical pulses. Just like several other proposed methods (including frequency-resolved optical gating (FROG) [12], SPIDER [13], photodiode-based phase-retrieval ultrafast waveform measurements [14], [28] and phase and intensity from correlation and spectrum only (PICASO) [15], [16]), temporal data itself is not enough to provide color (or phase) variation information and extra spectral measurement is needed. In this paper, we add spectral information into triple-optical-autocorrelation measurements to make the triple-autocorrelation method capable of providing a complete knowledge of laser pulses. With the measured temporal intensity of an optical pulse and its corresponding spectral intensity obtained with a spectrometer, exact intensity and phase variations in time can all be recovered with the Gerchberg–Saxton (GS) algorithm through an iterative calculation with an $O(n)$ complexity. We have employed a single-staged third-harmonic-generation (THG) based triple-optical autocorrelation for direct pulse-shape measurement (TOAD), which is different from the two-staged implementation previously demonstrated [8], [17]. With these established procedures, optical pulse shapes can be directly calculated from TOAD measurement, and its phase variation can be exactly retrieved thereafter.

II. TOAD: Principles

For an optical pulse, time-dependent intensity $I(t)$ can be expressed in the frequency domain ν as

$$I(t) = \int \tilde{I}(\nu)\exp(-i2\pi\nu t)\,d\nu = \int \left|\tilde{I}(\nu)\right|\exp(i\alpha(\nu) - i2\pi\nu t)\,d\nu \tag{2}$$

where $|\tilde{I}(\nu)|$ and $\alpha(\nu)$ are magnitude and phase of $I(t)$ in the frequency domain. After beam splitting, three replicas of the laser pulse could be focused on a THG crystal. By varying the time delays τ_1 and τ_2 between three replica pulses and recording the THG signal generated with one photon from each replica, the background-free triple-optical autocorrelation

$$G^3(\tau_1, \tau_2) = \int I(t)I(t+\tau_1)I(t+\tau_2)\,dt \tag{3}$$

can be directly obtained. Then we can use it and its bispectrum $\tilde{G}^3(\nu_1, \nu_2)$, the Fourier transform of the triple-optical-autocorrelation function, to calculate $|\tilde{I}(\nu)|$ and $\alpha(\nu)$ [6] with

$$\left|\tilde{I}(\nu)\right| = \left[\tilde{G}^3(\nu_1, 0)\Big/\tilde{I}(0)\right]^{1/2} \tag{4}$$

and

$$\alpha(\nu) = \int_0^\nu \alpha'(\nu_1)\,d\nu_1 = -2\pi\int_0^\nu \left(\int \tau_2\cos(2\pi\nu_1\tau_1)G^3(\tau_1, \tau_2)\,d\tau_1\,d\tau_2 \Big/ \int \cos(2\pi\nu_1\tau_1)G^3(\tau_1, \tau_2)\,d\tau_1\,d\tau_2\right) d\nu_1. \tag{5}$$

The temporal optical pulse shape $I(t)$ can then be fully determined by substituting them into (2).

III. Obtaining Pulse Shape With THG-Based Triple Autocorrelation

The optical pulses used to demonstrate this concept came from a mode-locked Cr : forsterite laser operating around 1230 nm with a typical bandwidth of 12 nm [Fig. 2(a)]. The average output power was 280 mW with a 110-MHz repetition rate. By using beam splitters, the laser output beam was split into three mutual parallel and equal-distance replicas. Computer-controlled translation stages were used to introduce delays among them. Through an objective (54-18-15, Special Optics), each 60-mW beam was subsequently focused into the THG crystal with a 5-μm spot size. The THG crystal we employed was a 0.45-μm-thick p-type gallium nitride, which was grown on double-side polished sapphire and buffered with 1.45-μm-thick AlGaN. Our previous experiments had employed the same Cr : forsterite laser to demonstrate enhanced THG in the GaN thin film due to close resonance of defect/doping levels and bandtail states [18]. With the help of interface and defect resonant-enhanced THG virtual transitions [18] in the p-doped GaN thin film, 2 nW of THG power can be generated with 180 mW of incident power. If there are temporal overlaps among each pulse, ten possible THG beams will be generated in different spatial directions due to different combinations of three individual photons. The middle THG beam will be the signal generated with one photon from each pulse replica according to momentum conservation law. As shown in Fig. 1, the THG beam that contained triple-optical-autocorrelation information was spatially selected with an iris. The generated THG signals were so strong that they can be directly observed with naked eyes and can be directly detected with a Si-based photodetector [18]. Finally, we used an optical filter to block any possible fundamental and SHG [18] lights and the generated THG signal was detected with a CCD-based spectrometer. The use of a spectrometer helped us to distinguish the THG signals from other multiphoton photoluminescence [18]–[20]. Fig. 2(b) is the measured THG spectrum, which centered at 410 nm with 4-nm full-width at half-maximum (FWHM). We integrated the spectral energy of THG signal as the intensity data point of TOAD. An example of the measured TOAD trace is shown in Fig. 3(a), which is a single-peaked pulse under typical laser

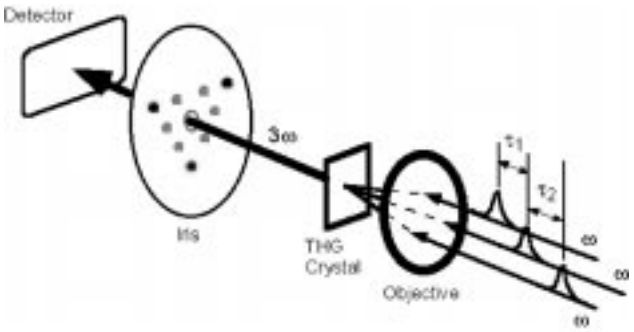

Fig. 1. Diagrammatic representation of THG-based triple-optical autocorrelation.

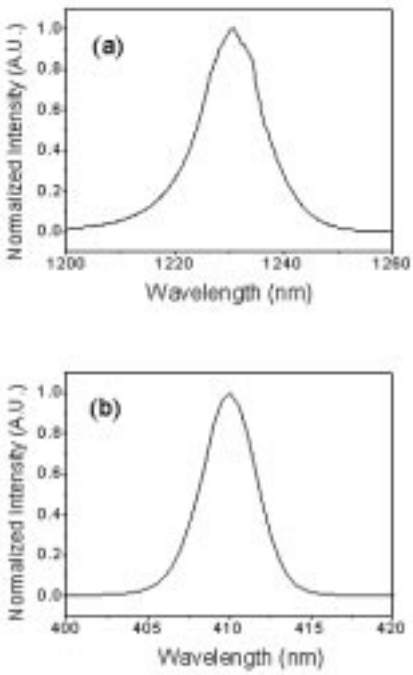

Fig. 2. (a) Fundamental spectrum of the mode-locked Cr : forsterite laser pulses. (b) The corresponding THG spectrum.

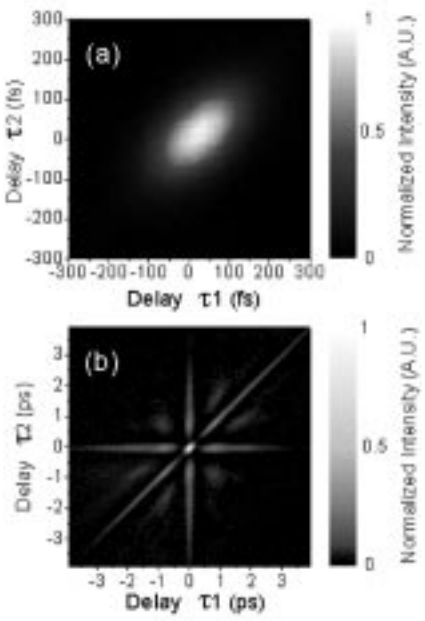

Fig. 3. Two THG-based TOAD traces for pulses from a modelocked Cr : forsterite laser. (a) Single-peaked pulse under typical laser operation. (b) Misaligned laser cavity—distorts the output pulse shape into a multiple-peaked structure.

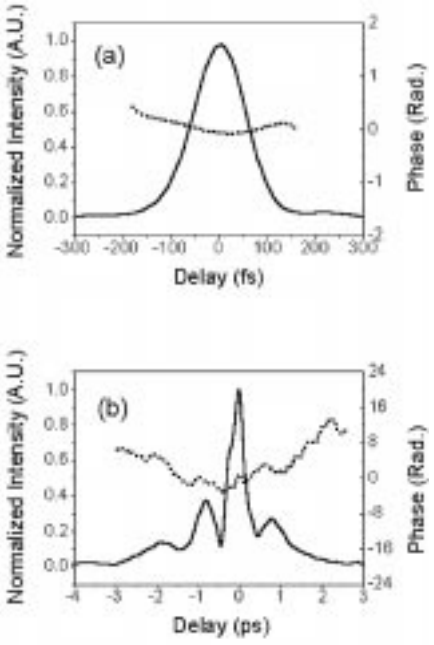

Fig. 4. Recovered temporal intensities (solid curves) and phases (dotted curves) corresponding to the THG-based TOAD traces in Fig. 3.

operation. As a comparison, we misaligned the laser cavity in order to distort the output pulse shape into a multiple-peaked structure. Its TOAD trace is shown in Fig. 3(b), which extends to several picoseconds and shows more features than the previous case. Both of the results have a diagonal symmetry, which is the characteristic of the TOAD-autocorrelation function. Following (2)–(5), their corresponding temporal pulse shapes can then be directly obtained and are displayed in Fig. 4 (solid curve). We have also measured the background-free SHG autocorrelation for the typical pulse by using the same GaN sample as the SHG crystal. The measured autocorrelation trace (solid curve in Fig. 5) agrees well with the reconstructed autocorrelation trace made from the TOAD obtained $I(t)$ (dotted curve in Fig. 5).

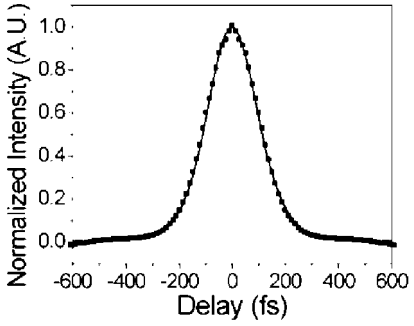

Fig. 5. Measured SHG autocorrelation trace (solid curve) and the reconstructed autocorrelation trace (dotted curve) made from TOAD-obtained $I(t)$ in Fig. 4(a).

IV. Phase Retrieval

As was theoretically proven by Huiser, Drenth, and Ferwerda in 1976, with exact knowledge of the temporal intensity of an optical pulse (that can be obtained by TOAD) and its corresponding spectral intensity (that can be easily obtained with a spectrometer), its phase and color variations in time can be determined with a GS algorithm [21], [22]. Although there are ambiguities in the GS algorithm [23], for optically realizable functions, only pulses with complete temporal and spectral symmetry will cause a twofold ambiguity [22]. Excluding this ambiguity, the temporal phase variation can be uniquely determined with an $O(n)$ complexity in contrast to other SHG-based techniques [12]. If the twofold ambiguity occurs due to a completely symmetric pulse shape or spectral shape, they can be distinguished with an auxiliary measurement such as insertion of an optical window with known dispersion. In our developed GS algorithm, the TOAD-recovered temporal intensity $I_{\mathrm{TOAD}}(t)$ and an initial guessed temporal phase form the initial condition of the iteration loop. For each iteration, the temporal electric-field signal (including magnitude and phase) is first Fourier transformed to frequency domain with its spectral magnitude replaced by tje square root of the measured optical spectrum, then it is inverse transformed to time domain with its temporal magnitude replaced by square root of $I_{\mathrm{TOAD}}(t)$. At the end of the loop, the program evaluated the cost of the data $\varepsilon_{\mathrm{GS}}^{(k)}$ by

$$\varepsilon_{\mathrm{GS}}^{(k)} \equiv \left\{ 1/N \sum_{i=1}^{i=N} \left[I_{\mathrm{GS}}^{(k)}(\tau_i) - I_{\mathrm{TOAD}}(\tau_i) \right]^2 \right\}^{1/2} \tag{6}$$

which is the root-mean-square between $I_{\mathrm{GS}}^{(k)}$ (estimated intensity of the present iteration) and I_{TOAD}. As iteration loops go on, if the cost stays at a steady value, the present estimated temporal data will be registered and the phase will be rolled away from the original value by several times the phase difference between the present and the previous ones. This phase-rolling procedure acts as mutation in Genetic algorithm and prevents guessed data

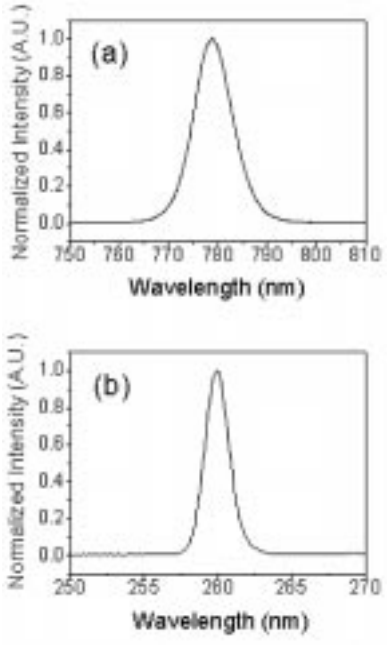

Fig. 6. (a) Fundamental spectrum of the mode-locked Ti : sapphire laser pulses and (b) the corresponding THG spectrum.

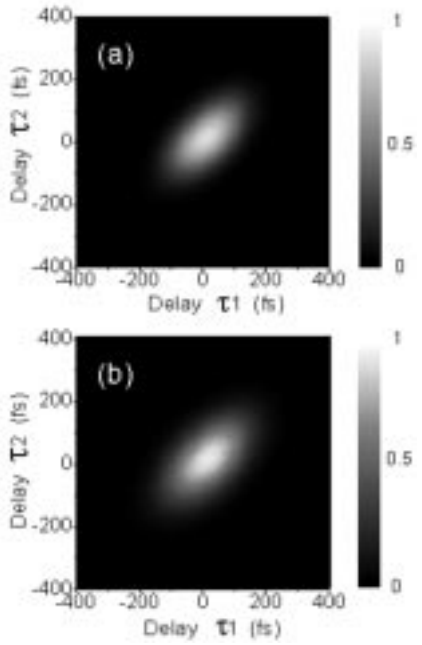

Fig. 7. THG-based TOAD traces for output pulses from a mode-locked Ti : sapphire laser (a) with and (b) without BK7 window.

from being trapped in local minima [24]. This process goes on after several mutations until the phase is recovered with an acceptable cost.

With this developed phase-retrieval method and a spectral measurement, we obtained the temporal phases of laser pulses in previous cases (see dotted curves in Fig. 4). The misaligned case [Fig. 4(b)] shows stronger second-order dispersion and higher order dispersion than typical case [Fig. 4(a)] in time domain.

V. Characterizing the Dispersion of Pulses

Utilizing the developed GS algorithm, we performed a dispersion measurement with laser pulses passing through a dispersive window. The study was performed on a mode-locked Ti : sapphire laser (Tsunami, Spectra-Physics). The wavelength of laser output pulses was centered at 780 nm with a typical bandwidth of 9 nm [Fig. 6(a)]. The average output power was 650 mW with a 82-MHz repetition rate. For the Ti : sapphire output pulse-shape measurement, an objective (54-17-30, Special Optics) focused each laser beam (70 mW) to 6-μm spot sizes and an interference filter, centered at 260 nm, was used to block fundamental, SHG, and multiphoton-induced photoluminescence. Other setups of TOAD measurements were the same as those in the Cr : forsterite output pulse-shape measurement. The THG signal was generated with the same p-doped GaN thin film described above for Cr : forsterite laser characterization utilizing surface THG effects in AlGaN/sapphire interface [25]. Finally, the collected THG spectrum showed a center wavelength of 260-nm, one third of its fundamental wavelength, with 2-nm FWHM [Fig. 6(a)]. The measured THG-based TOAD trace for typical pulses from a mode-locked Ti : sapphire laser is shown in Fig. 7(a). In order to introduce the optical dispersion for investigation, we placed a 2-cm-thick BK7 window before the TOAD measurement system. According to the dispersion relation given by Schott Glass, the corresponding second-order dispersion

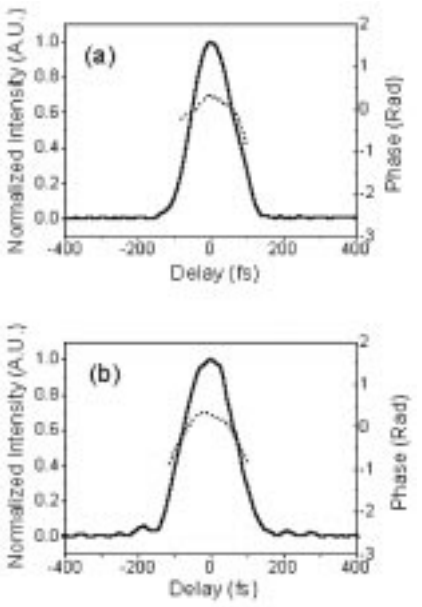

Fig. 8. Recovered temporal intensities (solid curves) and phase profiles (dotted curves), corresponding to the TOAD traces in Fig. 7.

evaluated at a wavelength of 780 nm is 930 fs^2. Fig. 7(b) shows the measured TOAD trace after introducing the dispersion window with a shape similar to but larger than Fig. 7(a), which implies pulsewidth broadening due to introduced positive optical dispersion. Fig. 8(a) and (b) shows their corresponding optical pulse shapes directly obtained from the measured TOAD traces in Fig. 7(a) and (b). Pulsewidth broadening from 130 fs [solid curve in Fig. 8(a)] to 162 fs [solid curve in Fig. 8(b)] can be found. After applying the phase retrieval program with the measured optical spectrum [Fig. 6(a)] to both cases, their corresponding temporal phases can also be determined (dotted curves in Fig. 8).

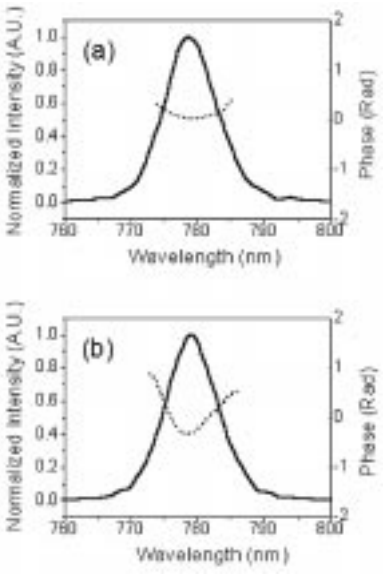

Fig. 9. Fourier-transformed optical intensity (solid curves) and phase (dotted curves) spectra, corresponding to the temporal traces in Fig. 8.

With a complete knowledge of the pulses, we can then apply the Fourier transformation on these temporal optical signals to obtain their corresponding spectral distribution information (as shown in Fig. 9), including magnitude and phase information, and retrieve their dispersions from the phase-determined spectra (dotted curve in Fig. 9). We found that the second-order dispersion was increased from 870 to 1800-fs^2, with a positive difference which agreed well with the expected value.

Compared to SHG, THG can be generated in any material, taking advantage of the interface inhomogeneity. Here we picked a GaN thin film grown on top of a sapphire substrate as THG crystal due to its wide bandgap and its availability in our laboratory just to demonstrate the feasibility of complete pulse shape and phase determination of femtosecond optical pulses with the THG-based TOAD. Although the THG efficiency is $<10^{-6}$ in our case with the GaN thin film, the signal can already be easily detected by a Si-based photodetector without amplifiers. Moreover, some efficient THG crystals have been reported recently. Yelin *et al.* focused 130-fs pulses at a wavelength of 1.5 μm on nematic liquid crystal and achieved 10^{-5} THG efficiency in a volume as small as 5 μm^3 [26]. Zhang *et al.* applied the technique of quasiperiodic optical superlattice (QPOS) on $LiTaO_3$ and got 27% THG at a wavelength of 0.48 μm [27]. These progresses in THG crystal can improve the THG efficiency, and therefore the signal-to-noise ratio of TOAD.

VI. Phase Verification With FROG

To verify the above demonstrated phase-retrieval method with an independent technique, we characterize the pulses shown in Fig. 9(a) right after the TOAD measurements by using a SHG FROG. The SHG crystal we used in the FROG system was a 0.3-mm-thick BBO (CASIX), which is cut at a 29.2° phase-matching angle for a wavelength of 800 nm. Its phase-matching bandwidth (16 nm) at 780 nm is much larger than the bandwidth (9 nm) of our mode-locked Ti : sapphire

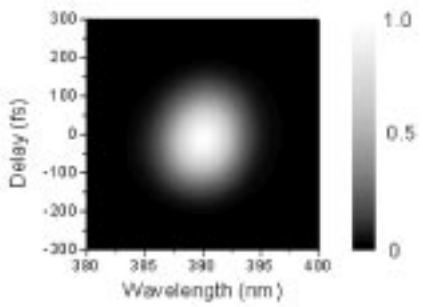

Fig. 10. FROG trace of laser pulses corresponding to Fig. 7(a).

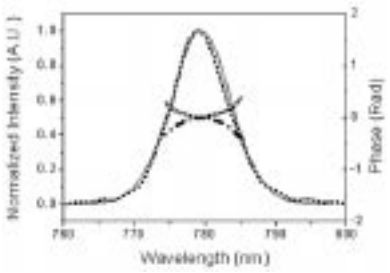

Fig. 11. FROG-recovered spectral intensity (Gaussian-like solid curve) and its corresponding twofold degenerate phases (solid curve and dash-dotted curve). These results are compared with data in Fig. 9(a) (dotted curves).

laser. With this BBO, we measured the SHG-spectra of background-free autocorrelations [(1)] at different delay τ to form a FROG trace (Fig. 10). Thus, retrieved optical intensity and phase spectra are shown in Fig. 11 as solid curves. Because the autocorrelation function has direction-of-time ambiguity, there are twofold degenerate phase solutions with the method of SHG-FROG. The intensity and the phase with positive dispersion agree well with the recovered results by using THG-based triple autocorrelation (shown as dotted curves in Fig. 11). This result also supports the capability of accurate phase retrieval by the developed GS algorithm.

VII. Summary

In conclusion, we demonstrated a method to obtain complete information of femtosecond pulses. First, we realized THG-based triple-optical autocorrelation for direct optical pulse-shape measurement. Since THG is allowed for any materials, our developed technique can be easily realized with little modification on the common autocorrelation setup with a thin film of wide-bandgap materials. This time-domain technique allows quick and easy determination of the measured optical pulse shape without iterative algorithms. For further information on the color or phase variation in time, with the help of a measured optical spectrum, a simple GS algorithm (with $O(n)$ complexity) can uniquely determine it except for pulses with complete temporal and spectral symmetry that will cause a twofold ambiguity.

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