Triple-optical autocorrelation for direct optical pulse-shape measurement

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Triple optical autocorrelation of femtosecond optical pulses was realized simply with third-harmonic-generation technique. This optical technique provides complete knowledge of transient pulse intensity variation directly in time domain. Only analytic calculation is needed to obtain the pulse-shape from data without direction-of-time ambiguity. Combined with a spectral measurement and the Gerchberg–Saxton algorithm, except for pulses with complete temporal and spectral symmetry that will cause a twofold ambiguity, exact phase variation in time can also be retrieved through an iterative calculation with an $O(n)$ complexity. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1501453]

In most time domain applications and studies, measuring the ultrashort optical pulse-shape is a crucial diagnosis. Because electronic devices are too slow to measure temporal evolution of femtosecond optical pulses, it is thus important to develop optical techniques based on slow electronic detectors in order to characterize these ultrashort pulses. In past decades there have been great advances in the techniques for measuring ultrashort optical pulses,[1–10] however, most of them cannot provide direct optical pulse shape measurement in time domain. Here we present a pure time-domain measurement technique, triple optical autocorrelation for direct pulse-shape measurement (TOAD), based on single-stage third-harmonic generation (THG). Only analytic calculation is needed to obtain the transient pulse-shape.

By far the most common mean to evaluate femtosecond optical pulses is optical autocorrelation measurement based on second harmonic generation (SHG).[1,2] A Michelson interferometer splits an incident pulse into two replicas with a relative temporal delay. These two pulses are then recombined in a 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 photocurrent with delay yields the autocorrelation trace. 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 complete temporal optical pulse shape cannot be determined directly from these previous proposed two-arm correlations[1–5] due to limited information in measurement. Some methods have been developed to obtain enough information on the measured pulse shape,[6–10] but they needed either iterative algorithm for multidimensional search or spectral information to finish this time-domain work. On the other hand, if another temporal dimension is added on to the two-arm correlation to form a triple correlation, thus obtained information will be not only sufficient to completely determine the pulse shape but also direct in time domain only. In previous studies, it had been shown that a triple correlation function is sufficient to determine the temporal intensity of a pulse with direct mathematical calculation.[11,12] Here we realize this technique for direct optical pulse-shape measurement based on simply one stage of a THG effect. According to this method, temporal intensity of optical pulses can be obtained directly without any spectral measurement or pulse-shape assumption. Moreover, computer simulation also showed that this suggested method is quite insensitive to noise.[13]

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

$$I(t)=\int \tilde{I}(\nu)\exp(-i2\pi\nu t)d\nu = \int |\tilde{I}(\nu)|\exp[i\alpha(\nu)-i2\pi\nu t]d\nu, \quad (1)$$

where $|\tilde{I}(\nu)|$ and $\alpha(\nu)$ are magnitude and phase of $I(t)$ in the frequency domain, respectively. After beam splitting, three replicas of laser pulses are focused on a THG crystal as shown in Fig. 1. If there are temporal overlaps among them, ten possible THG beams will be generated in different spatial directions due to different combinations of three individual photons, for example more than one photon can be contributed from one beam. The specific THG signal generated with only one photon from each replica is spatially selected with an iris according to momentum conservation law. By varying the time delay between pulses, τ_1 and τ_2, and recording the selected THG signals with a detector, the background-free triple correlation

$$G^3(\tau_1,\tau_2)=\int I(t)I(t+\tau_1)I(t+\tau_2)dt \quad (2)$$

can be directly obtained from the measurement. We can use it and its bispectrum $\tilde{G}^3(\nu_1,\nu_2)$, the Fourier transform of the triple correlation function, to calculate $|\tilde{I}(\nu)|$ and $\alpha(\nu)$[11] with

$$|\tilde{I}(\nu)|=\sqrt{\tilde{G}^3(\nu_1,0)/\tilde{I}(0)} \quad (3)$$

and

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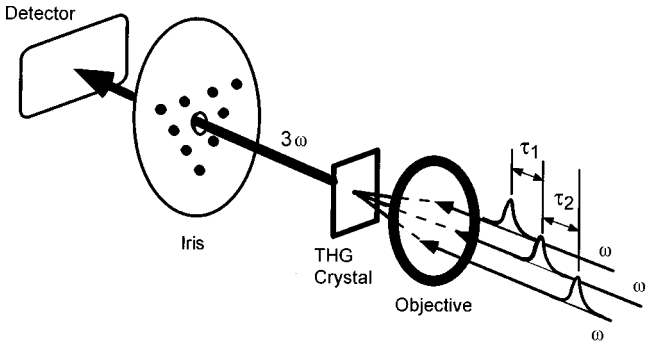

FIG. 1. Diagrammatic representation of THG based triple correlation.

$$\alpha(\nu)=\int_0^{\nu}\alpha'(\nu_1)d\nu_1$$
$$=-2\pi\int_0^{\nu}\Bigg[\int\tau_2\cos(2\pi\nu_1\tau_1)G^3(\tau_1,\tau_2)d\tau_1d\tau_2\Bigg/$$
$$\int\cos(2\pi\nu_1\tau_1)G^3(\tau_1,\tau_2)d\tau_1d\tau_2\Bigg]d\nu_1. \quad (4)$$

The temporal optical pulse shape $I(t)$ can then be fully determined by substituting them into Eq. (1).

The optical pulses used to demonstrate this concept came from a mode-locked Cr:forsterite laser operating with a center wavelength of 1230 nm and a bandwidth of 18 nm. The average output power was 280 mW with 110 MHz repetition rate. We then misaligned the laser cavity in order to distort the output pulse-shape for TOAD measurement. We split 180 mW of the laser output beam into three mutual parallel and equal distance replicas with beam splitters and used the computer-controlled translation stages to introduce delays among them. After focusing into a 0.45-μm-thick *p*-type gallium nitride, which served as the THG crystal,[14] spatially selected THG signals can be easily detected by a CCD based spectrometer around 410 nm. Usually more than 6000 counts can be detected with an integration time of 0.01 s at 0 time delays. Our previous experiments using the same Cr:forsterite laser demonstrated enhanced THG in GaN thin film due to close resonance of defect/doping levels and band-tail states.[14] With a sample thickness less than the THG coherent length, which is estimated to be around 1 μm for GaN with our specific experimental wavelengths,[15] we can neglect the problems of phase matching angle and phase matching bandwidth. The use of a spectrometer helped us to distinguish the THG signals from other multiphoton excited photoluminescence in GaN.[15]

Figure 2 shows two examples of TOAD traces. The TOAD trace in Fig. 2(a) corresponds to normal laser operation without cavity misalignment, where a single-peak pulse can be observed. The TOAD trace in Fig. 2(b) corresponds to the cavity-misaligned operation, where multiple-peaked structure for the laser pulse can be directly found in the trace. It is important to notice the diagonal symmetry in the measured traces, which is the characteristic of triple correlation. Following Eqs. (1)–(4) without iterative calculations, their corresponding temporal pulse-shapes can then be directly obtained. Figures 3(a) and 3(b) show the recovered temporal intensity profiles (solid curve), which correspond to the

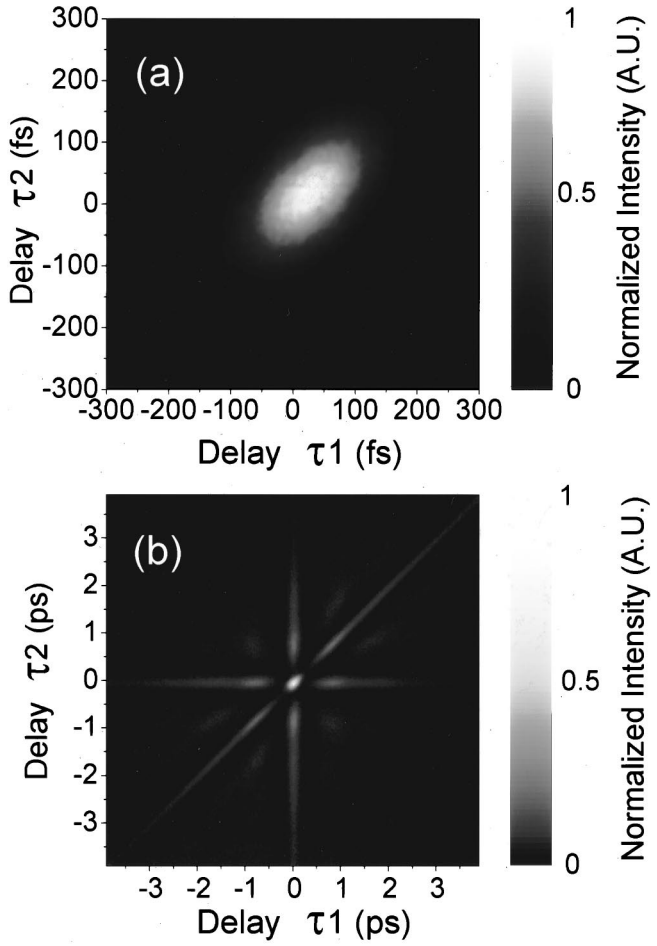

FIG. 2. Two THG-based triple correlation traces for pulses from a mode-locked Cr:forsterite laser.

TOAD traces in Figs. 2(a) and 2(b), respectively. Please notice that, besides noise induced measurement errors, Fig. 3 presents the exact pulse-shape (intensity, not field) without direction-of-time ambiguity. Corresponding magnitude distribution of the electric field can be obtained from Fig. 3 by taking a square root.

Once the temporal pulse-shape is obtained, according to Gerchberg–Saxton (GS) algorithm,[16,17] phase variation in time can be determined with the help of the corresponding spectrum. Although there are ambiguities in the GS algorithm,[18] for optically realizable functions, they rise only for pulses with complete temporal and spectral symmetry that will cause a twofold ambiguity.[17] Therefore the temporal phase variation can be uniquely determined except for symmetric pulses with an $O(n)$ complexity in contrast to other SHG-based techniques.[6] If the twofold ambiguities occur due to a completely symmetric pulse shape or spectral shape, they can be distinguished with an auxiliary measurement. Based on the GS algorithm, we developed a phase retrieval program for phase distribution determination.[19] With the TOAD recovered intensity and a measured spectrum, the retrieved phases are shown in Fig. 3 (dotted curve). These results indicated that the method of TOAD can obtain complete information of the laser pulses. To investigate this capability of TOAD, we performed our TOAD measurements on the output pulses of a mode-locked Ti:sapphire laser and introduced dispersion by placing a 2-cm-thick BK7 window

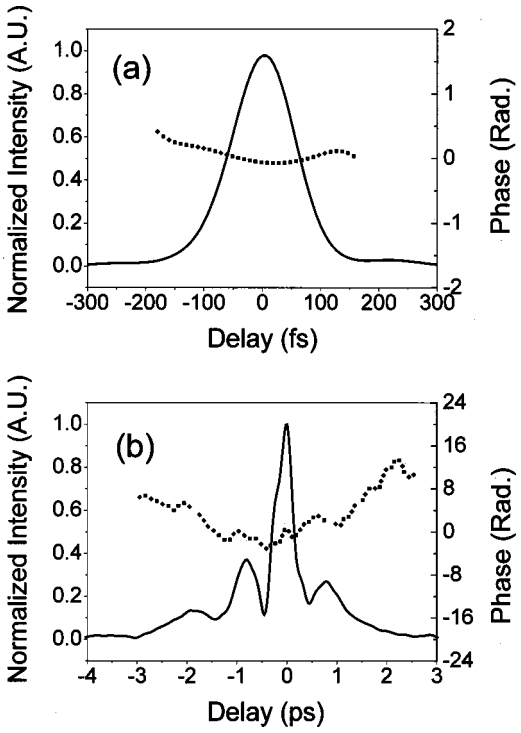

FIG. 3. Recovered temporal intensity (solid curve) and phase (dotted curve), corresponding to the THG triple correlation traces in Fig. 2.

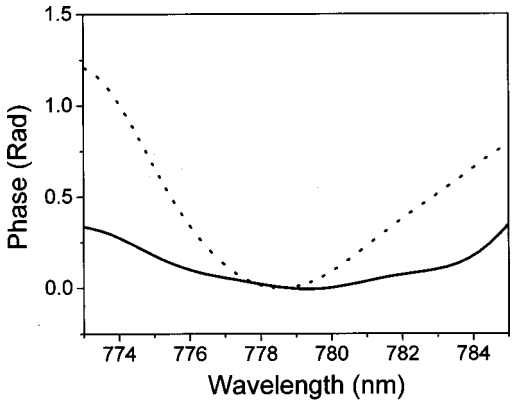

FIG. 4. TOAD retrieved phase spectra of the output pulses from a mode-locked Ti:sapphire laser, centered at 780 nm, with (dotted curve) and without (solid curve) a BK7 window.

before the TOAD measurement system. According to the dispersion relation given by Schott Glass,[19] the corresponding second-order dispersion evaluated at a wavelength of 780 nm is 930 fs^2. The retrieved spectral phase (Fig. 4) shows that the second-order dispersion was increased from 870 fs^2 (solid curve) to 1800 fs^2 (dotted curve) after introducing the dispersive window, with a positive difference agreed well with the expected value.

Compared to SHG, THG can be generated in any material, taking advantage of the interface inhomogeneity. Although the THG efficiency is $<10^{-6}$ in our case with a GaN thin film, the signal can already be easily detected by an unamplified Si photodetector. Moreover, some efficient THG crystals have been reported recently.[20,21] These progresses in THG crystals can improve the THG efficiency in a TOAD measurement, therefore the signal-to-noise ratio or the cost of TOAD.

In conclusion, we demonstrated a triple optical autocorrelation technique for direct optical pulse-shape measurement based on third-harmonic generation. 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 band gap materials. This time-domain technique thus allows quick and easy determination of the measured optical pulse shape without iterative algorithm. Combined with a spectral measurement and Gerchberg–Saxton algorithm, except for pulses with complete temporal and spectral symmetry that will cause a twofold ambiguity, exact phase in time can also be retrieved, through an iterative calculation with an $O(n)$ complexity.

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