Measurement of ultrashort optical pulses by third-harmonic generation

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Interferometric and intensity autocorrelations by third-harmonic generation for the measurement of ultrashort optical pulses are demonstrated. The third-harmonic signal is generated by tight focusing of the ultrashort pulses upon the surface of an ordinary glass slide, replacing the high-optical-quality nonlinear crystals used in conventional second-harmonic generation measurements. The method is shown to be applicable to real-time measurements. © 1997 Optical Society of America [S0740-3224(97)00608-5]

1. INTRODUCTION

Clever techniques have emerged recently for measuring the time-dependent amplitude and phase of ultrashort pulses,[1,2] yet second-harmonic generation (SHG) intensity and interferometric autocorrelation techniques first reported three decades ago[3] are still commonly employed. It is well known that SHG intensity autocorrelations cannot provide complete information about the pulse shape. SHG interferometric autocorrelations[4] are often used in the femtosecond-pulse regime to provide additional information about the pulse shape and phase, because chirped pulses are characterized by distinctive interferometric autocorrelation traces. Although they are not complete, SHG autocorrelation traces, together with spectral measurements, are used routinely for characterization of ultrashort optical signals for various purposes. These techniques are relatively simple and straightforward, and therefore it is expected that SHG interferometric and intensity autocorrelations will continue to be commonly employed for ultrashort-pulse characterization.

With the advent of ultrashort-pulse lasers, characterization of such pulses is needed. Because ultrashort pulses contain only a few optical cycles, care must be taken to ensure that the measurement process introduces no significant distortions. In particular, SHG autocorrelation measurements require careful phase matching, which is particularly problematic with ultrashort pulses in the 10-fs range. To obtain broadband SHG suitable for ultrashort-pulse measurements and to reduce material dispersion, very thin crystals, a few tens of micrometers in thickness, are usually used.[5]

We demonstrate measurements of ultrashort pulses by third-harmonic generation (THG). Our measurement scheme is similar to collinear SHG interferometric and intensity autocorrelations. However, in our setup the SHG nonlinear crystal is replaced by an ordinary microscope cover glass, and the THG signal of an ultrashort pulse, tightly focused upon the air–glass interface, is detected and analyzed. Our measurement scheme is significantly better than conventional SHG interferometric and intensity autocorrelations. Because the intensity of the third-harmonic signal is detected, the sixth power of the field is involved, and therefore the measurement process is significantly more sensitive to the particular pulse shape. THG interferometric autocorrelation analysis predicts a peak-to-background ratio of 32:1, compared with an 8:1 ratio in conventional SHG interferometric autocorrelations. Similarly, the predicted peak-to-background ratio for THG intensity autocorrelation is 10:1, compared with a 3:1 ratio in SHG intensity autocorrelation. THG, unlike SHG, does not require the use of noncentrosymmetric crystals because all materials have nonvanishing THG coefficients. Therefore the need for expensive high-optical-quality nonlinear SHG crystals, as thin as a few tens of micrometers, which are used for ultrashort-pulse measurements, is removed. Moreover, inasmuch as the effective interaction length in our THG measurement is just a few micrometers, phase matching is not a problem; thus this method can be used to characterize sub-10-fs pulses.

THG is a well understood and characterized nonlinear effect; however, it has found only limited applications because of the high light intensities that are typically involved. With the advances in ultrafast laser technology, THG as well as generation of higher harmonics can be realized with the now commonly available high-peak-power lasers. THG was used recently in a fringe-resolved optical gating measurement of short pulses[6] in which the advantages of surface THG were used. Fringe-resolved optical gating measurements are useful for complete temporal characterization of short pulses, but they involve a complex setup and postprocessing. In many experimental situations, complete characterization of phase and amplitude is not needed; interferometric or intensity autocorrelations are sufficient. Indeed, most experimentalists use SHG autocorrelations routinely to monitor their lasers. These real-time measurements can be simplified and improved with THG.

2. THEORY

Consider the case in which a laser beam, propagating along the z direction, is tightly focused within the interior of a nonlinear medium. In the theory of harmonic generation for focused Gaussian beams in the undepletedpump approximation, the third-harmonic power generated by the beam is given by[7]

$$P_{3\omega} = k_{3\omega} k_{\omega}^{3} \left(\frac{4\pi}{n_{3\omega} n_{\omega}^{2} c} \right)^{2} P_{\omega}^{3} |J|^{2}, \tag{2.1}$$

where P_{ω} is the fundamental beam power, k_{ω} and $k_{3\omega}$ are the fundamental and the third-harmonic wave numbers, respectively, and J is defined by

$$J = \int_{z_{L}'}^{z_{R}'} \frac{\chi(z')\exp(i\Delta k b z')}{(1 + 2iz')^{2}} \mathrm{d}z', \tag{2.2}$$

where χ is the third-order susceptibility; $\Delta k = k_{3\omega} - 3k_{\omega}$ is the phase mismatch; $b = k_{\omega} w_{0}^{2}$ is the confocal parameter of the fundamental beam with a waist radius of w_0; $z' = z/b$ is the normalized coordinate along the z axis, measured from the beam waist position; and z_{L}' and z_{R}' are the normalized input and output plane coordinates, respectively. In the case of an infinite uniform nonlinear medium this integral can be evaluated by contour integration. The result of this integration in this limit is that the efficiency of THG vanishes in the case of positive phase mismatch ($\Delta k > 0$), which is the case for normally dispersive materials, and even in the case of perfect phase matching ($\Delta k = 0$).[7]

When the nonlinear medium is not uniform, either in refractive index or in nonlinear susceptibility, the THG efficiency does not vanish, and significant THG output can be obtained. In the case of an air–glass interface, the third harmonic is generated efficiently when the beam waist is focused onto the interface, and it rapidly decreases as the waist–interface distance is increased to more than one confocal length. This phenomenon was studied recently in various solids,[8] a study that was motivated primarily by the quest for efficient THG sources, and was also used as the basis for a novel nonlinear microscope for observing transparent objects through variations in their third-order nonlinear properties.[9]

To analyze the THG interferometric and intensity autocorrelation signals, consider the interference of two identical, collinear pulses separated by a delay τ, tightly focused onto an air–glass interface. Because the linear index and the phase mismatch are discontinuous at the interface, the third harmonic is generated efficiently within one confocal parameter of the focused beam. The third-harmonic field is proportional to the third power of the fundamental field, and because in our measurement scheme the intensity of this signal is detected, the sixth power of the fundamental field is involved. Describing the time-dependent electrical field of each of the pulses by $E(t) = A(t)\cos(\omega t)$, where $A(t)$ is the amplitude and ω is the angular frequency, the THG interferometric autocorrelation intensity signal $I^{(3\omega)}$, as a function of the delay variable τ, is given up to a constant multiplicative factor by

$$I^{(3\omega)}(\tau) = \int |\{A(t)\cos(\omega t) + A(t-\tau)\cos[\omega(t-\tau)]\}^{3}|^{2}\mathrm{d}t, \tag{2.3}$$

where integration is carried over all times. It is straightforward to show, by using trigonometric identities, that Eq. (2.3) can be simplified to yield

$$\begin{aligned} I^{(3\omega)}(\tau) = {} & \frac{5}{8}\int A^{6}(t)\mathrm{d}t + \frac{45}{16}\int [A^{4}(t)A^{2}(t-\tau) \\ & + A^{2}(t)A^{4}(t-\tau)]\mathrm{d}t + \frac{15}{8}\cos(\tau\omega) \\ & \times \int [A^{5}(t)A(t-\tau) + A(t)A^{5}(t-\tau)]\mathrm{d}t \\ & + \frac{5}{8}[9\cos(\tau\omega) + \cos(3\tau\omega)] \\ & \times \int A^{3}(t)A^{3}(t-\tau)\mathrm{d}t + \frac{15}{8}\cos(2\tau\omega) \\ & \times \int [A^{4}(t)A^{2}(t-\tau) + A^{2}(t)A^{4}(t-\tau)]\mathrm{d}t, \end{aligned} \tag{2.4}$$

which involves calculations of overlap integrals of powers of the slowly varying amplitude of the pulse. For intensity autocorrelation, the last three terms in Eq. (2.4) are averaged out, and the THG intensity autocorrelation signal $\bar{I}^{(3\omega)}$, as a function of the delay variable τ, is given by

$$\bar{I}^{(3\omega)}(\tau) = \frac{5}{8}\int A^{6}(t)\mathrm{d}t \left\{ 1 + \frac{9/2}{\int A^{6}(t)\mathrm{d}t} \times \int [A^{4}(t)A^{2}(t-\tau) + A(t)^{2}A^{4}(t-\tau)^{4}]\mathrm{d}t \right\}. \tag{2.5}$$

As can be verified by inspection of Eqs. (2.4) and (2.5), the THG interferometric autocorrelation signal has a peak-to-background ratio of 32:1 and the THG intensity autocorrelation has a peak-to-background ratio of 10:1.

3. EXPERIMENTAL RESULTS

The basic elements of our THG interferometric autocorrelation setup are shown in Fig. 1. The laser source for our experiments was a Spectra-Physics synchronously pumped optical parametric oscillator (OPO) with an average power of 280 mW at a central wavelength of 1.5 μm, operating at a 80-MHz repetition rate. The input beam was split in the Michelson interferometer and recombined after passing through an adjustable relative delay by a computer-controlled stepper motor translator with 0.1-μm steps. The sum field was focused by a 60×, 0.85-N.A. microscope objective onto the front surface of an ordinary microscope cover glass, and the generated third-harmonic signal was collected by a 20×, 0.4-N.A. microscope objective. The fundamental signal was filtered out and the green third-harmonic intensity signal at 0.5 μm was measured with a photomultiplier and a lock-in amplifier.

A typical THG interferometric autocorrelation measurement of these pulses is shown in Fig. 2(a). The distinctive features of THG interferometric autocorrelation are clearly seen: peak-to-background ratio of 32:1, high

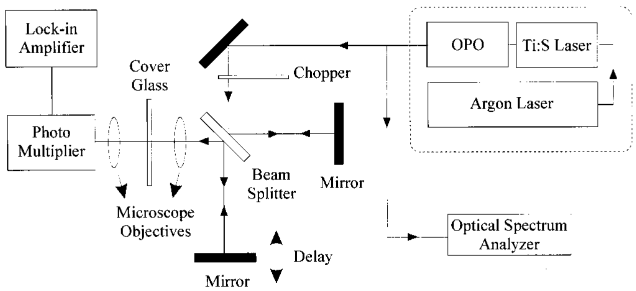

Fig. 1. Experimental arrangement for THG interferometric and intensity autocorrelation measurements. The relative delay between the two arms of the interferometer is set by a computer-controlled stepper motor with 0.1-μm steps. The combined field is focused by a 60×, 0.85-N.A. microscope objective onto the front surface of an ordinary microscope cover glass. The THG signal is collected by a 20×, 0.4-N.A. microscope objective.

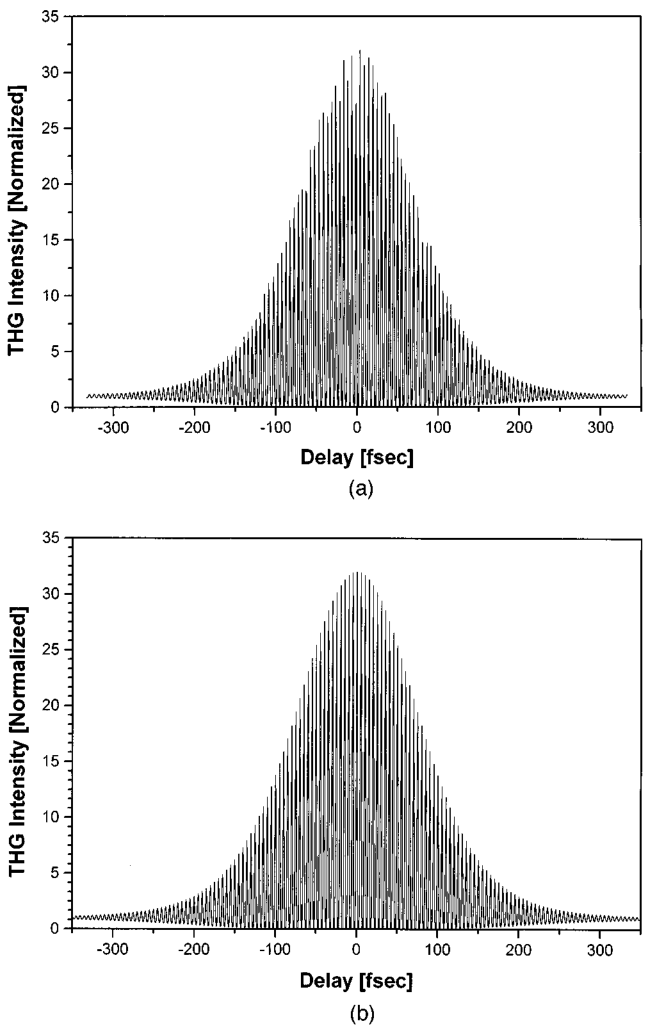

Fig. 2. (a) THG interferometric autocorrelation measurement. Note the 32:1 peak-to-background ratio and the high fringe contrast. (b) Theoretical calculations of THG interferometric autocorrelation of a Fourier-limited, 133-fs sech pulse centered at 1.5-μm wavelength.

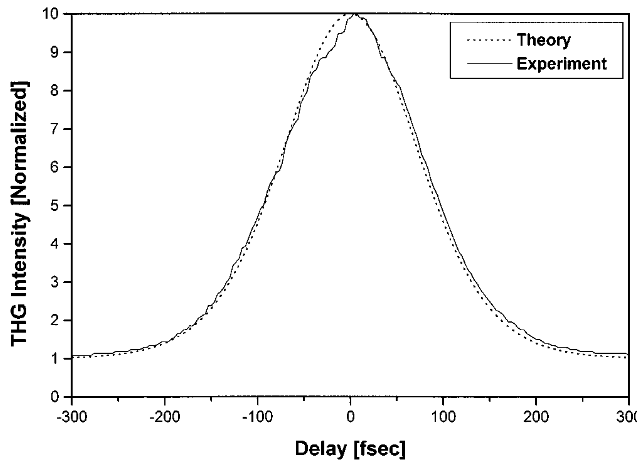

Fig. 3. THG intensity autocorrelation corresponding to a 133-fs sech pulse.

fringe contrast ratio, and smooth variation of the bottom envelope. For comparison, a theoretical curve of THG interferometric autocorrelation of a Fourier-limited, 133-fs sech pulse at 1.5 μm is shown in Fig. 2(b).

To measure the THG intensity autocorrelation we averaged out the cosine terms in Eq. (2.4) by setting a long integration time constant of the lock-in amplifier and increasing the scan rate of the stepper motor. The results are shown in Fig. 3 and compared with a theoretical calculation of THG intensity autocorrelation of a 133-fs sech pulse. Note that for this measurement the peak-to-background ratio is 10:1, as expected.

Finally, to complete the measured data set, we measured the pulse power spectrum, using an optical spectrum analyzer. The FWHM of the power spectrum, centered at 1.5 μm, was 19.5 nm, from which we conclude that the pulses were ~10% wider than transform-limited sech pulses. This chirp is not obvious in Fig. 2(a), but it is well known that small chirps cannot easily be detected in SHG interferometric autocorrelation, and THG measurements show similar behavior.

We have also verified the applicability of our measurement scheme for fast-scan, real-time interferometric and intensity autocorrelation measurements. For this purpose the chopper and the lock-in amplifier were removed from the setup, and the output signal of the photomultiplier was measured directly by an oscilloscope. The measured THG signal levels were sufficient to yield autocorrelation traces with no further amplification.

4. CONCLUSIONS

The information about the pulses measured with interferometric and intensity THG autocorrelations is essentially similar to that from SHG measurements; however, THG measurements have several advantages. THG, unlike SHG, does not require the use of noncentrosymmetric crystals, because all materials have nonvanishing THG coefficients. Therefore the need for the expensive high-optical-quality nonlinear SHG crystals, as thin as few tens of micrometers, that are used for ultrashort-pulse measurements is removed. In our setup the THG source was an ordinary glass slide, which replaced the SHG crystals. SHG autocorrelation measurements require careful phase matching, which is particularly problematic with ultrashort pulses near 10-fs.[10] When THG is used, the effective interaction length is approximately one confocal parameter of the beam, which can be of the order of the optical wavelength. Therefore phase matching is not a problem, and our method is inherently wideband and can readily be used to characterize ultrashort pulses.

In conclusion, we have demonstrated that one can use THG in glass to obtain interferometric and intensity autocorrelations of ultrashort pulses. The method combines the simplicity of the SHG interferometric and intensity autocorrelation techniques with the properties of third-order correlations, using common glass as the nonlinear medium. Because the sixth power of the field is involved, the measurement process is significantly more sensitive than SHG to the particular pulse shape.

With a slight modification, our method could also be applied for THG background-free autocorrelations and cross correlations. It is expected that this technique will prove useful for ultrashort-pulse characterization, especially for pulses in the 1–1.5-μm wavelength range, where the THG signal can easily be detected with conventional, inexpensive optical components.

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