Demonstration of Single-Shot Measurements of $10^{13}$ Ultrahigh-Contrast Pulses by Manipulating Cross-Correlation

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In strong-field physics experiments with high-intensity lasers, single-shot characterization of the temporal contrast between the laser pulse peak and its temporal pedestal is important; this allows fast optimization of the pulse contrast and meaningful comparison with theory for each pulse shot. To date, high contrast ratios of $10^{10}$ have been demonstrated in single-shot measurements for petawatt (PW) lasers. However, ultrahigh contrast ratios of $\approx 10^{13}$, as required for the planned 200 PW lasers, pose challenges to high-intensity laser technologies and have thus far remained open for investigation. This article reports a pilot demonstration of ultrahigh-contrast measurements by adapting a single-shot cross-correlator (SSCC). An evaluation method for the SSCC detection limit is introduced. The strategy mimics the test beam with known spatial contrast, whose cross-correlation is equivalent to that of a test pulse with ultrahigh temporal contrast. The ultimate contrast measurement limit of $10^{13}$ is achieved, which corresponds to the highest pulse intensity by optical damage and the lowest temporal pedestal by single-photon detection. The photon noise of the detector is observed and becomes dominant as the temporal pedestal of the optical pulse decreases. The demonstrated detection ability is applied to a high-contrast laser system, suggesting the accessibility of ultrahigh-contrast measurements.

1. Introduction

Since the earliest demonstration of chirped-pulse amplification, femtosecond lasers are able to deliver ultrahigh peak powers beyond petawatt (PW) order, thereby paving the way for compact particle accelerators and X-ray sources.\[1\] One of the crucial parameters for strong-field physics experiments is the temporal intensity contrast between the laser pulse peak and its temporal pedestal, which should be sufficiently large to avoid early ionization of the plasma target before the arrival of the laser pulse.\[2\] Consequently, the present PW-class lasers with intensities of the order of $\approx 10^{21}$ \text{W cm}^{-2} are appropriate for high contrast ratios of $10^{10}$, whereas the contrast requirement may increase to $\approx 10^{12}$ for the planned 200 PW lasers (e.g., the Extreme Light Infrastructure) in the future, with extreme intensities of the order of $10^{24}$ \text{W cm}^{-2}.\[3\] Such a high contrast demand of $10^{13}$ is approximately five to six orders of magnitude greater than that of the femtosecond mode-locking lasers.\[4\]

Although femtosecond pulse measurements have developed well, ultrafast diagnosis of pulse contrast with an ultrahigh dynamic range imposes daunting technological challenges. The state-of-the-art delay-scanning cross-correlator (DSCC) provides a high dynamic range of $10^{12}$–$10^{13}$, but the scanning measurement is time-consuming and only suitable for high-repetition-rate lasers. For example, the front-end measurement of the Apollon 10 PW laser indicates a dynamic-range limited contrast as high as $10^{12}$;\[5\] however, the final output contrast of an ultrahigh-power laser is greatly inaccessible because of the low repetition rate. Furthermore, the DSCC measurements are the average results of repetitive detections, which may deviate from the real pulse contrast to some degree because the laser noise varies from one pulse shot to the next. A single-shot cross-correlator (SSCC) is therefore essential for diagnosing and optimizing the final contrast of a laser system.\[6\] Thus far, the SSCC has been successfully applied to several PW-class lasers with $\approx 10^{10}$ contrast as the result of a lengthy series of improvements over the past few decades.\[16\] It is highly desirable that SSCC measurements resolve ultrahigh contrasts of the order of $10^{13}$ for the expected 200 PW lasers in the future.

Herein, we demonstrate an ultrafast method for evaluating the contrast measurement ability of an SSCC and highlight the...
ultimate measurement limit set by the photon noise of the detector. While the dynamic range of the DSCC measurements can easily be tested by adjusting the optical attenuation during the corresponding long scanning times, it is assumed that the dynamic-range test of the SSCC measurements must rely on ultrashort pulses with known contrasts better than $10^{13}$. In addition, the test pulse must have a sufficiently high power, e.g., $\approx 100$ GW, to achieve high intensities within a correlation area of $\approx 10 \times 10 \text{mm}^2$. Such a high-power ultrahigh-contrast pulse has not yet been available; therefore, all SSCC designs have not been tested in practice. In the current work, we mimicked a test pulse in the spatial domain based on time-space mapping and evaluated the efficacy of the SSCC device to measure contrast of the order of $10^{13}$.

2. SSCC Test Strategy

The principle of SSCC measurements can be explained simply as follows: a femtosecond test pulse with a picosecond noise pedestal is recorded using an oscilloscope with an adapter. Here, the adapter plays a key role in temporal magnification and converts the test pulse into a series of nanosecond-spaced temporal slices. As shown in Figure 1a, the adapter relies on a correlation unit of noncollinear third-harmonic generation (THG) and a detection unit of a fiber-array-mediated photomultiplier tube (PMT) (see details in Section 3.2). If the test pulse has a uniform beam profile in the noncollinear plane ($x \approx 2$), then the noncollinear THG allows time-space mapping to enable a test pulse $I(t)$ represented by the correlation function $A^{(2)}(x) = A^{(0)}(x) = I(t) \otimes P(t - \tau)$, where the delay $\tau$ has a linear dependence on the transverse variable $x$ as $\tau = \gamma x$ and the coefficient $\gamma$ of the time-space mapping can be determined by the noncollinear angle $\alpha$.[17,19] With the parallel detection of a 100-pixel fiber array along $x$, the transversely distributed $A(x)$ is further mapped to a series of temporal slices spanning $500 \text{ ns}$.[18,19] Such a temporally magnified signal series can be resolved using the PMT and oscilloscope: this is the typical procedure for single-shot contrast measurement.

If the test laser beam $I(\xi)$ is spatially nonuniform and has an ultrahigh spatial contrast between the beam center and surrounding areas, the THG correlation function becomes $A^{(0)}(x) = I(\xi) \otimes \delta(\xi - x).$[17,20] Therefore, a spatially shaped beam $I(\xi)$ can also produce the same correlation function as the test pulse $I(t)$ does (Figure 1b). This suggests an alternative scheme to test the device dynamic range. As shown in Figure 1c, a test beam $I(\xi)$ is synthesized by spatially packaging a high-intensity narrow beam at the center of a continuous-wave (CW) wide beam. Given the measurement frame of time-space mapping, the high-intensity femtosecond beamlet mimics a peak pulse under test, and the weak CW beamlet mimics a temporal noise pedestal. In other words, the synthesized beam acts as a test pulse with a known contrast set by the intensity ratio of the two beamlets.

In the temporal cross-correlation (Figure 1a), the nonlinear angle $\alpha$ determines the coefficient $\gamma$ of time-space mapping, so an increase in $\alpha$ can enlarge the temporal window of the single-shot measurements.[17,19] In the spatial cross-correlation (Figure 1b), a nonlinear angle $\alpha$ must be chosen for group velocity matching between the interacting waves, as typically performed in a noncollinear optical parametric amplifier (OPA).[21] In the case of unmatched group velocities, the femtosecond pulse peak and the CW pedestal in the synthesized beam will have different temporal overlaps with the sampling pulses in the correlation crystal, which degrades the THG correlation and renders the contrast measurements unfaithful. For convenience, the temporal cross-correlation in Figure 1a also adopts the noncollinear angle $\alpha$ for group velocity matching.

![Figure 1](https://example.com/figure1.png)

**Figure 1.** Temporal magnification and SSCC test strategy. a) The test pulse $I(t)$ is represented by the THG correlation function $A^{(2)}(x)$ via time-space mapping; the temporal magnification $M$, based on two mappings between $t$ and $x$, produces a test pulse spanning $M \Delta t$ and allows accommodation with the PMT and oscilloscope. b) A test beam $I(\xi)$ is represented by $A^{(2)}(x)$ without time-space mapping. c) Mimicking the high-contrast pulse $I(t)$ with $A^{(2)}(x)$ from a delicately synthesized beam $I(\xi)$. 

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[17,19] Source: [Advanced Photonics](https://www.adpr-journal.com)
3. Experimental Section

3.1. Light Sources

As shown in Figure 2, two types of 1054 nm test lasers obtained by second-harmonic generation (SHG) of 2108 nm lasers were alternately applied as inputs to the SSCC in the experiments. One was a high-contrast spatial test beam comprising a high-intensity femtosecond narrow beam and low-intensity CW wide beam, and the other comprised a high-contrast temporal test pulse from the SHG of a single-shot optical parametric chirped-pulse amplification (OPCPA) laser. Each test laser is independently coupled to the SSCC using the translation stage TS-1. A 1 kHz femtosecond OPA (OPera SOLO, Coherent) pumped by a Ti:sapphire regenerative amplifier (Astrella, Coherent) delivered 330 μJ, 50 fs pulses at 2108 nm that were further converted to 120 μJ femtosecond pulses at 1054 nm by SHG with a 1 mm-thick LiNbO3 crystal. This 1 kHz, 1054 nm femtosecond laser and a single-frequency CW fiber laser (Rock single-frequency laser, NP Photonics) were synthesized into a high-contrast spatial test beam. The second test laser source relies on the SHG of a 2108 nm OPCPA system. The OPCPA pump laser comprises the aforementioned CW fiber laser with a 2 ns waveguide modulator, a 10 Hz Nd:YLF regenerative amplifier, and a single-shot three-stage Nd:glass amplification system that delivers a 1 J, 2 ns pulse at 1054 nm. The 2108 nm femtosecond OPCPA pulse was stretched to 1.5 ns using a pulse stretcher, then amplified to 80 mJ by an OPCPA with a 22 mm-thick LiNbO3 crystal and 1 CW cm⁻² pump intensity, and finally compressed into a 30 mJ, 300 fs pulse using a pulse compressor. This 2108 nm femtosecond OPCPA beam was further converted to a 10 mJ femtosecond pulse at 1054 nm by SHG with a 4 mm-thick β-BBO crystal, which served as the high-contrast temporal test pulse.

3.2. SSCC Device

One of the key components of the SSCC is an adaptor that temporally magnifies the test pulses and allows accommodation with the PMT and oscilloscope. Such an adaptor consists of a correlation unit of noncollinear THG, a detection unit of fiber array, and a PMT. In the THG correlation unit using in this work (Figure 2), the 1054 nm test pulse was doubled in frequency using a 4 mm-thick β-BBO crystal (θ = 22.8°) to produce a 527 nm sampling pulse. Owing to the nonlinear process of SHG, this sampling pulse will be cleaner than a test pulse. The remaining test pulse at 1054 nm, after passing through a half-wave plate, serves as another incident pulse to the THG correlation. The angle of intersection between the test and sampling beams was 33.6°, and the THG correlation adopted a type-I β-BBO crystal (θ = 61°) of size of 38 (x) x 10 (y) x 2 (thickness) mm³. The laser beams of both the test and sampling pulses were nearly uniform in the x domain and covered the entire crystal. The noncollinear THG performed time-to-space mapping and induced a temporal window of 120 ps. The generated 351 nm correlation signal A(3)(x) with a beam width of W = 38 mm was imaged onto a 38.5 mm-wide fiber array by a cylindrical lens (f = 100 mm). In the SSCC detection unit, the fiber array consisted of 100 UV fibers (OPTRAN UVNS, CeramOptec) with incremental lengths of 1 m, in which the 1st and the 100th fibers were 2.6 and 101.6 m long, respectively. The UV fiber with a core diameter of 105 μm had a high transmission (≈10 dB km⁻¹) at 351 nm. The correlation signal A(3)(x) was coupled to the fiber array by a cylindrical lens with f = 30 mm, and collected by a fiber bundle, which outputs a series of 5 ns-delayed temporal slices spanning 500 ns. Consequently, serial detection can be used instead of parallel detection. In this study, a PMT (H10721-113, Hamamatsu) was used as the detector. To accommodate a high dynamic range and ensure a linear response of the PMT, a series of variable fiber

Figure 2. Schematic of the experimental setup. In brief, the setup consists of two laser sources and the SSCC. The roles of the four translation stages are as follows: TS-1 and TS-4, selecting and adjusting the test laser; TS-2, adjusting the delay between test and sampling pulses; TS-3, spatially scanning the sampling laser.
Attenuators were applied to reduce the signal intensities in the different fiber channels. In addition, a bandpass filter with high transmission for a 351 nm correlation signal was added between the fiber bundle and PMT to suppress the noise interference from the scattering of both the 1054 nm test and 527 nm sampling lasers. These two types of laser pulse peak scattering typically have intensities of approximate $10^{-6}$ relative to the 1054 and 527 nm lasers,[23] which are larger than the 351 nm correlation signal for the temporal pedestal of the test pulse and will therefore severely degrade the pulse contrast measurements.

The SSCC device was slightly modified to evaluate the contrast measurement limit (Figure 3). In the first modification, the spatially uniform test beam was replaced with a synthesized high-contrast beam. A 1054 nm mirror with 40% reflectivity and 60% transmission was used to combine the femtosecond narrow beam and CW wide beam. The transmitted femtosecond narrow beam had 25 μJ pulse energy, 150 fs duration, and 0.38 mm (x) × 1.8 mm (y) beam spot, providing an intensity of 30 GW cm⁻². The reflected wide CW beam had 30 mW full power and 38 mm (x) × 2.5 mm (y) beam spot, rendering a full intensity of 40 mW cm⁻². In the second modification, the 50 μJ femtosecond sampling laser at 527 nm was focused onto the correlation crystal to produce a beam spot similar to that of the femtosecond narrow beam at 1054 nm and a high intensity of 60 GW cm⁻². To achieve the THG correlation function $A^{(3)}(x)$, the sampling laser was scanned in the x direction using the translation stages TS-2 and TS-3. The other parameters of the synthesized test beam and sampling beam are as previously noted. The third modification reduced the noncollinear THG angle from 33.6° to 13.8° and the β-BBO crystal orientation (θ) from 61° to 36.5° to match the velocities of the two incident waves. By scanning the sampling laser beam along the x direction, we obtained a maximum THG signal peak of ≈9 μJ. This position, corresponding to the THG signal peak, is defined as $x = 0$. The THG efficiency relative to the narrow-beam femtosecond pulse at 1054 nm was 36% for the pulse energy and 12% for the photon number. The THG signal peak produced was imaged into the 50th channel of the fiber array. The total transmission from the correlation crystal to the PMT was measured to be 25%.

4. Results

4.1. Evaluation of Dynamic Range with a Spatially High-Contrast Test Beam

In the test experiment for the 1054 nm SSCC, the correlation unit adopted a β-BBO crystal of size of 38 (x) × 10 (y) × 2 (thickness) mm³ and noncollinear angle $\alpha = 13.8^\circ$. A temporal window of ≈50 ps, the maximum attainable delay between the test and sampling pulses deduced from the noncollinear angle of 13.8° and clear aperture of 3.75 cm in the x direction, was obtained.[19] Consequently, the delay per unit crystal size is 13.3 ps cm⁻¹, which defines the coefficient of time-to-space mapping. In addition, the fiber-pixel time was 500 fs because the entire fiber array with 100 pixels is matched to the temporal window of 50 ps. A 1 kHz repetition rate of the femtosecond beamlet with a width of $W_x = 0.38$ mm was obtained by SHG of the 2108 nm OPA system and was maintained at a fixed intensity of 30 GW cm⁻² at the crystal surface, while the CW beamlet with a width of $W_x = 38$ mm was varied in intensity. Because the high-intensity (≈60 GW cm⁻²) sampling beam at 527 nm in the SSCC also had a narrow width of $W_x = 0.38$ mm, the test experiments were performed by beam scanning along the x direction. As shown in Figure 3a, the contrast measured by our device agrees well with the intensity ratio of the two beamlets. The detectable CW beamlet intensity can be as low as 3.5 mW cm⁻², indicating an ultra-high dynamic range of ≈10³.

The input/output relationship of the PMT is briefly introduced herein. The PMT (H10721-113, Hamamatsu) used in the SSCC has an anode radiant sensitivity of $2.2 \times 10^5$ A W⁻¹. A photon at

![Figure 3. Dynamic-range tests with synthesized high-contrast beams. a) Measured THG correlation function $A^{(3)}(x)$ at two CW intensities. Each dataset was averaged over 16 shots with the oscilloscope. b,c) Recorded 5000-shot PMT photovoltages at the 51th fiber channel (corresponding to the THG signal at $x = 0.38$ mm) and two CW intensities of 3.5 mW cm⁻² and 40 mW cm⁻². d,e) Histograms of the photon numbers detected per shot corresponding to (b) and (c), respectively. In (d), the first bin has a large number of shots (3878), which has been purposely divided by 20 to obtain a clear plot.](image-url)
351 nm has an energy of $5.66 \times 10^{-19}$ J, corresponding to $2.8 \times 10^{-10}$ W in the PMT characteristic time of 2 ns. According to these specifications, a single photon of the THG correlation signal triggers an anode current of $6.16 \times 10^{-3}$ A, and is further converted to a voltage of 3 mV with a coupling resistance of 50 Ω. Therefore, the recorded average photocurrent of 3.1 mV at the CW intensity of 3.5 mW cm$^{-2}$ corresponds to only one THG photon from a fiber pixel (Figure 3b,d). In this situation, the shot-to-shot fluctuations are of the order of 25%, which is typical for single-photon detection with the characteristics of white noise.[24] At higher THG photon numbers (>10), the fluctuations are greatly reduced (Figure 3c), and the distribution of the detected photon numbers differs slightly from that of a Poisson distribution having the same average flux (Figure 3e). Consequently, the observed severe fluctuations due to photon noise eventually limit the SSCC measurement for ultrahigh-contrast pulses. At this point, it is also clear that the DSCC measurements may not be reliable owing to the fluctuations of photon and laser noises. The ultimate measurement limit of $10^{13}$ contrast achieved here nearly corresponds to the temporal pedestal from the intense pulse peak as well as consider two fluctuations, a single photon of the THG correlation signal triggers an anode current of 6.16 A, limited SSCC measurements for an ultrahigh contrast of $10^{12}$.[22] However, the full-window (120 ps) single-shot measurement by the SSCC showed a ratio of only $3 \times 10^{10}$ around −105 ps (black curve in Figure 4). This measurement-limited intensity ratio of $3 \times 10^{10}$ can be attributed to the low intensity of the test pulse. The intensity of the total test pulse was only $10^2$ W cm$^{-2}$ because the test beam with a limited energy of $\approx 10$ mJ and limited power of $\approx 40$ W was transformed to a large beam size that spanned the entire correlation crystal in the x direction in the full-window measurement. To enhance the dynamic range of the single-shot full-window measurement to up to $10^{13}$, a 1054 nm test pulse with a high power of $200$ GW was necessary but unavailable in our experiments. Therefore, we reduced the beam size in the x direction by a factor of five to obtain a high intensity of $50$ GW cm$^{-2}$. However, this compromised approach reduces the single-shot temporal window to only 24 ps, which is linearly proportional to the beam width in the x direction.[19] In this case, six-shot measurements with a small window (24 ps) were applied to form a full-window (120 ps) plot. Each pair of successive measurements was equally delayed by 20 ps by adjusting the translation stage TS-2, which resulted in a 4 ps temporal overlap between two successive measurements for data normalization. A real contrast as high as $0.5 \times 10^{13}$ was achieved by the six equally delayed single-shot measurements (red curve in Figure 4). The improved contrast measurements over two orders of magnitude are consistent with the theoretical expectation that the THG correlation signal has a cubic dependence on the total intensity of the test pulse.

4.2. SSCC Measurements for a Temporally High-Contrast Test Pulse

Figure 4 shows two SSCC measurements for a real 1054 nm ultrashort pulse with $\approx 40$ GW peak power. The temporal window of the SSCC was increased to 120 ps using a larger noncollinear angle of $\alpha = 33.6^\circ$. As noted in Section 3.1, the 1054 nm temporally high-contrast test pulse was generated through SHG of the 2108 nm OPCPA output, which is expected to have an ultrahigh contrast beyond $10^{12}$.[22] However, the full-window (120 ps) single-shot measurement by the SSCC showed a ratio of only $\approx 3 \times 10^{10}$ around −105 ps (black curve in Figure 4). This measurement-limited intensity ratio of $\approx 3 \times 10^{10}$ can be attributed to the low intensity of the test pulse. The intensity of the total test pulse was only $10^2$ W cm$^{-2}$ because the test beam with a limited energy of $\approx 10$ mJ and limited power of $\approx 40$ W was transformed to a large beam size that spanned the entire correlation crystal in the x direction in the full-window measurement. To enhance the dynamic range of the single-shot full-window measurement to up to $10^{13}$, a 1054 nm test pulse with a high power of $200$ GW was necessary but unavailable in our experiments. Therefore, we reduced the beam size in the x direction by a factor of five to obtain a high intensity of $50$ GW cm$^{-2}$. However, this compromised approach reduces the single-shot temporal window to only 24 ps, which is linearly proportional to the beam width in the x direction.[19] In this case, six-shot measurements with a small window (24 ps) were applied to form a full-window (120 ps) plot. Each pair of successive measurements was equally delayed by 20 ps by adjusting the translation stage TS-2, which resulted in a 4 ps temporal overlap between two successive measurements for data normalization. A real contrast as high as $0.5 \times 10^{13}$ was achieved by the six equally delayed single-shot measurements (red curve in Figure 4). The improved contrast measurements over two orders of magnitude are consistent with the theoretical expectation that the THG correlation signal has a cubic dependence on the total intensity of the test pulse.

5. Conclusion

We demonstrated the ability of a single-shot resolution of $10^{13}$ ultrahigh-contrast pulses by manipulating the cross-correlation. Based on the equivalence of the cross-correlation function, the detection limit of the SSCC was studied using a spatially high-contrast test beam synthesized by spatially packaging a high-intensity narrow beam at the center of a CW beam. By setting the high intensity of the narrow beam close to the SSCC damage threshold and the low intensity of the CW beam down to the order of a few photons, we observed detection-induced photon noise and demonstrated the photon-noise-limited SSCC measurements for an ultrahigh contrast of $10^{13}$. Although the temporally high-contrast test pulse was limited in power, multishot measurements of such real test pulses also verified the ability of the SSCC. A true single-shot measurement for an ultrahigh contrast of $10^{13}$ can thus be anticipated with a test pulse having a high power of $\approx 200$ GW, which should be feasible with SHG from a terawatt-class laser. The demonstrated ability is ready for a broad diagnosis from current PW lasers to the expected 200 PW lasers in the future and beyond. To further enhance the measurement ability of the SSCC beyond $10^{13}$, one may apply the plasma-mirror technique to separate the weak temporal pedestal from the intense pulse peak as well as consider two SSCC devices to separately measure the temporal pedestal and peak pulse.[25] This route is promising for relaxing the limitation of the SSCC damage threshold because the intense pulse peak

![Figure 4. Two SSCC measurements for a real ultrashort pulses. The black curve represents a full-window (120 ps) single-shot measurement at a low intensity (10 GW cm$^{-2}$), whereas the red curve consists of six equally delayed small-window (24 ps) measurements at a high intensity (50 GW cm$^{-2}$).](image-url)
can be attenuated drastically without impeding the temporal pedestal.

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Conflict of Interest
The authors declare no conflict of interest.

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Research data are not shared.

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