Frequency-resolved optical gating and single-shot spectral measurements reveal fine structure in microstructure-fiber continuum

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Cross-correlation frequency-resolved optical gating with an angle-dithered nonlinear-optical crystal permits measurement of the intensity and the phase of the ultrabroadband (as much as 1200 nm wide) continuum generated from microstructure optical fiber. Retrieval revealed fine-scale structure in the continuum spectrum. Simulations and single-shot spectrum measurements confirmed that the fine structure does exist on a single-shot basis but washes out when many shots are averaged. © 2002 Optical Society of America OCIS codes: 320.0320, 320.7100, 190.5530, 190.5940, 190.7110.

It recently became possible to generate an ultrabroadband white-light continuum with a TEM_{00} spatial mode and a reportedly smooth stable spectrum extending throughout the entire visible and near-IR regions of the spectrum by propagation of subnanojoule 800-nm pulses through a microstructure fiber.¹

As numerous far-reaching applications for this light are envisaged, it is crucial to measure the continuum as well as possible, especially its intensity and phase versus time. Such a measurement involves many complications, however. First, a multishot measurement requires that all the continuum pulses in the train be identical. Second, the nonlinear-optical process used in making the measurement must have a massive phase-matching bandwidth that exceeds that of the continuum. Third, the continuum is the most complex ultrashort laser pulse ever generated.

In this Letter we report preliminary multishot cross-correlation frequency-resolved optical gating (XFROG) measurements² of the continuum made with a dithered-crystal technique. We find that the retrieved spectrum is not smooth or stable but instead contains fine structure on an ~1-nm scale, in disagreement with previous direct spectral measurements. In addition, we performed single-shot measurements of the continuum spectrum, which also revealed \sim 1-nm structure, in excellent qualitative agreement with the XFROG measurements. We also found that the continuum spectrum varies significantly from shot to shot, becoming smooth only in averages over several shots. Finally, it is interesting to speculate why our XFROG measurements-performed using more than 10^{11} shots—were able to see the spectral structure when simple spectral averages over as few as 100 pulses did not.

The big issue in our measurements is phase-matching bandwidth. A SHG-based measurement of a pulse with wavelengths from 400 to 1600 nm generates wavelengths from 200 to 800 nm. Because crystal dispersion increases drastically near 200 nm, phase matching these short wavelengths is especially problematic. This difficulty can be significantly reduced by performing XFROG, which uses sum-frequency generation (SFG) with a known narrower-band gate pulse (here an \sim 30-fs, 50-nm-bandwidth, 800-nm-wavelength pulse directly from a Ti:sapphire oscillator). SFG involving the continuum and the 800-nm gate pulse generates signal wavelengths from \sim 260 to \sim 550 nm, a considerably reduced signal-light bandwidth, which is, in fact, the more important quantity. Indeed, the required range of crystal phase-matching angles is much less (see Fig. 1).

The use of XFROG helps, but, by itself, is not sufficient for performing this measurement. Fortunately, in recent work we showed that, in a multishot measurement, it is in fact not necessary for crystal phase matching to achieve the full pulse bandwidth on every pulse; instead it is sufficient to do so over the measurement period—a much less strict condition. Because the crystal phase-matching angle varies with wavelength, angle dithering a relatively thick crystal will increase the bandwidth of an (autocorrelator or) FROG.

We combined the dithered-crystal technique with XFROG to measure the continuum generated by sending ~ 1 nJ of energy through 16 cm of microstructure fiber. We butt coupled the microstructure fiber to a 3.4- μ m mode-field-diameter standard step-index fiber, reducing the full divergence angle of the continuum from $\sim 60^{\circ}$ to $\sim 12^{\circ}$. Using a 40× reflective Cassegrain objective slightly off axis, we achieved good-quality collimation.

Our apparatus is shown in Fig. 2. We used a 1-mm-thick β -barium borate (BBO) crystal (whose group-velocity dispersion-induced chirp was small compared with that of the continuum) that was rapidly angle dithered by ~20°. The beam crossing angle was 2°, the delay increment was 3 fs, the



Fig. 1. (a) Normalized SHG phase-matching efficiency of $10-\mu$ m-thick BBO borate for angles ranging from 0° to 90°; (b) phase-matching angle tuning curve for SHG and SFG with an 800-nm gate pulse.



Fig. 2. Schematic diagram of the multishot XFROG measurement apparatus: BSs, beam splitters; μ -s, microstructure fiber; b-c, butt-coupling fiber.

wavelength increment was 1 nm, and the wavelength resolution was less than 1 nm. Our measured trace had dimensions 3000×1024 , which we interpolated and expanded to 8192×8192 for retrieval. We corrected traces for phase-matching efficiency versus wavelength, taking into account such effects as phase mismatch, nonlinearity dispersion, the ω factor in Maxwell's equations, our crystal-angle dithering, and efficiency of the grating-camera system.

The measured XFROG trace is shown in Fig. 3. The retrieved spectral phase was therefore mainly cubic, in agreement with the trace's parabolic shape. Although the retrieved spectrum had the same gross shape as the independently measured spectrum, we observed much fine-scale structure in the retrieved trace and spectrum that we did not see in the measured quantitites, a clear indication that something is amiss.

A possible explanation for these observations is that variations in the continuum pulse from shot to shot wash out the structure in directly measured spectra and in measured XFROG traces. That the information on the spectral structure remains in the multishot XFROG trace is possible because FROG traces contain much redundancy, and information washed out in one domain may remain in the other. Specifically, fine-scale frequency information is also present in the trace in the form of slow oscillations in delay, which are less likely to wash out.

We performed single-shot spectral measurements of the microstructure-fiber continuum. The output pulse from a Ti:sapphire oscillator was amplified by an adjustable-repetition-rate regenerative amplifier to ~100 μ J per pulse, which was then attenuated to ~1 nJ for input into 152 cm of 2- μ m-diameter microstructure fiber from OFS Fitel Laboratories. Using different amplifier repetition rates and camera exposure times, we could vary the number of shots in a measurement (see Fig. 4).

Figure 4 shows a 120-nm section of the continuum from 490 to 610 nm, with resolution of ~ 1 nm. Note that the fewer shots, the more spectral structure. Finally, the single-shot spectrum exhibits deep and fine oscillations, and each single-shot spectrum is different. If we manually take an average of just four successive single-shot spectra [Fig. 4(e)], the oscillation amplitude decreases greatly, and the spectrum approaches those averaged over many shots. Shot-to-shot energy jitter in our amplified pulses was $\sim 1\%$.

Theoretical simulations also confirmed our hypothesis (see Fig. 5). Using a cubic spectral phase and a smooth super-Gaussian spectrum, we generated a smooth parabolic XFROG trace [Figs. 5(a)-5(c)]. Imposing 100% multiplicative random structure on the spectrum, however, yielded a pulse with much structure in its trace [Figs. 5(d)-5(h)]. The XFROG algorithm retrieves both the smooth and the structured spectra from their respective traces. Averaging 100 such structured traces washed out the structure and generated an artificially smoothed trace, similar to what we measured in a multishot experiment [Figs. 5(i) and 5(l)]. Retrieval on the smoothed-out trace not only preseved the gross shape of the trace but also placed the fine structure back into the spectrum and the trace [Figs. 5(j), 5(k), and 5(m)]. Also interesting to note is that the spectral phase was well retrieved, whether we started from a smooth, a



Fig. 3. XFROG measurement of the microstructure-fiber continuum with an 800-nm 30-fs precharacterized reference pulse: (a) measured trace, (b) retrieved trace, (c) retrieved temporal intensity (solid curve) and phase (dashed curve), (d) retrieved spectral intensity (solid curve) and phase (dashed curve), (e) independently measured spectrum. The XFROG error was 0.012. The insets in (a) and (b) are higher-resolution sections in the traces. Both traces are 8192×8192 in dimension. SF, sum frequency.



Fig. 4. (a) Entire spectrum of the continuum, averaged over 10,000 pulses. Spectral section of the continuum exposed for (a) 10,000 shots, (b) 100 shots, and (d) a single shot. (e) Numerical average of single-shot measurements taken successively; dashed and dotted curves are two single-shot spectra taken seconds apart, and the solid curve is the average of four single-shot spectra. Our measured single-shot spectra contained $\sim 2 \times 10^5$ photons per pixel, so shot noise was negligible in these measurements. Averaged spectra were attenuated; single-shot spectra were not.

structured, or an artificially smoothed trace. This simulation imitates the real experiment and strongly supports our hypothesis.

Theoretical simulations of continuum generation have also predicted a deep and fine structure in the continuum spectrum.^{4,5} These simulations also predicte large variations in the spectral structure from small fractional input power changes, also in agreement with our measurements.

We conclude that, because any single continuum pulse spectrum is highly structured, any measured smooth multishot spectrum—from even the most stable unamplified oscillator—must have involved averaging over widely different highly structured individual spectra. However, the retrieved spectral phase appears to be relatively stable, consistent with the coherence measurements of Bellini and Hänsch related to bulk-media continuum generation.⁶ It will be interesting to see the implications of these variations for continuum applications.

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Fig. 5. Simulations: (a) theoretically generated and (b) retrieved XFROG traces. (c) Retrieved spectral intensity and phase of a pulse of a smooth super-Gaussian spectrum and cubic spectral phase; XFROG error, 0.00015. (d) Theoretically generated and (e) retrieved XFROG traces. (f) Retrieved spectral intensity and phase of a pulse with a structured XFROG error, 0.00031. (i) Average of 100 such structured traces. (j) Retrieved XFROG trace and (k) retrieved spectral intensity and phase from (i); XFROG error, 0.0027. (g), (h), (l), and (m) are higher-resolution sections in (d), (e), (i), and (j), respectively. All the traces are 1024×1024 in dimension. SF, sum frequency.

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