Increased-bandwidth in ultrashort-pulse measurement using an angle-dithered nonlinear-optical crystal

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Abstract: We show that the usual phase-matching-bandwidth constraint in ultrashort-laser-pulse measurement techniques is overly restrictive. Specifically, the phase-matching bandwidth need not exceed the pulse bandwidth on every pulse. Instead, only the phase-matching bandwidth *integrated over the measurement period* need exceed the pulse bandwidth. We show that angle-dithering a second-harmonic-generation crystal that is otherwise too narrowband (that is, too thick) can yield sufficient phase-matching bandwidth and an accurate pulse measurement. We apply this technique to frequency-resolved optical gating (FROG) and show that accurate pulse measurements can be made using a comparatively very thick and hence narrowband crystal. An additional advantage of using a thick crystal is increased signal strength.

OCIS codes: (320.7100) Ultrafast measurements

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1. Introduction

As ultrashort light pulses shrink in length and grow in utility, the ability to measure them increases in importance. Fortunately, remarkable progress has recently occurred in the development of such techniques, and it is now routine to completely measure the time dependence of these pulses.[1-5] With the most commonly used new method, frequency-resolved optical gating, (FROG),[2,5-12] it is now possible to measure pulses on a single shot; over a wide range of wavelengths, pulse lengths, and complexities; and to do so in a manner that is general, robust, accurate, and rigorous. Furthermore, FROG measurements are accurate in the presence of both random[13] and nonrandom[12] error.

Almost all such techniques, from autocorrelation to FROG, require the use of a nonlinear-optical process, which has a finite phase-matching bandwidth, potentially limiting the accuracy of the measurement. If only a portion of the pulse spectral content achieves phase-matching, then an inaccurate pulse measurement will result.[14] This problem is especially severe for extremely short pulses, whose spectrum can extend over many nanometers. In general, the nonlinear-optical phase-matching bandwidth is inversely proportional to the crystal thickness, so very thin crystals are required. For example, the measurement of sub-10-fs pulses typically requires a ~10-μm-thick second-harmonic-generation (SHG) crystal. As the SHG efficiency scales as the square of the crystal thickness, use of such an extremely thin crystal yields drastically reduces the sensitivity of any technique.

It has long been assumed that the phase-matching bandwidth must exceed the pulse bandwidth for every pulse in a multishot pulse measurement. We point out here that this assumption is overly restrictive in multishot pulse measurements. In fact, only the phase-matching bandwidth *integrated over the measurement period* need exceed the pulse bandwidth. As a result, only a fraction of the pulse spectrum need be phase-matched on one shot as long as the remaining portion of the pulse spectrum achieves phase-matching on other shots during the measurement. Because the range of wavelengths that achieve phase-matching depends sensitively on angle, we can *angle-dither* a SHG crystal that is otherwise too narrowband (that is, too thick) to yield a significantly increased effective phase-matching bandwidth for a given crystal thickness. Because the SHG efficiency scales as the square of the crystal thickness, angle-dithering also yields significantly greater signal strength.

In this letter, we demonstrate the use of angle-dithering in SHG FROG measurements of a 70-fs, 50-nm bandwidth pulse. We use a 1-mm-thick BBO crystal, which, when stationary (in the usual configuration), yields insufficient phase-matching bandwidth to measure such a pulse. However, when we angle-dither this crystal, the time-integrated phase-matching bandwidth increases significantly, and we achieve a near-perfect measurement of the pulse, in excellent agreement with a reference measurement made using a much thinner, stationary 100-μm-thick KDP crystal with sufficient bandwidth.

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We should mention that angle-dithering a crystal in this manner to increase phase-matching bandwidth in SHG FROG measurements was suggested earlier, by Nacké,[15] et al., although this idea does not appear to have been demonstrated or analyzed.

2. Theoretical Considerations

In ordinary FROG measurements, the experimental trace is given by:

$$I_{FROG}(\omega,\tau) = \left|\tilde{E}_{sig}(\omega,\tau)\right|^2 \tag{1}$$

where:

$$\tilde{E}_{sig}(\omega,\tau) = \mathrm{F}\left\{E_{sig}(t,\tau)\right\} \tag{2}$$

and F{ } represents the one-dimensional Fourier Transform from the time to the frequency domain. In SHG FROG measurements,[2,3,16] the signal field is given by:

$$E_{sig}(t,\tau) = E(t)\,E(t-\tau) \tag{3}$$

where *E(t)* is the pulse electric field.

Emphasizing the potential frequency-filtering effect of the crystal, we can write the frequency-domain electric field, $\tilde{E}_{sig}(\omega,\tau,\theta)$, produced by a SHG crystal in a FROG device as:[14]

$$\tilde{E}_{sig}(\omega,\tau,\theta) \propto \mathrm{F}\left\{E(t)\,E(t-\tau)\right\} F(\omega,\theta) \tag{4}$$

where $F(\omega,\theta)$ represents the frequency filter that results from phase-matching, and which is a function of crystal angle, θ. We have emphasized the potential dependence of the signal field on crystal angle. Also, this expression holds only when the group-velocity dispersion can be neglected. Typically, this filtering takes the form:

$$F(\omega,\theta) \propto \mathrm{sinc}\left\{\frac{[n_i(2\omega,\theta)-n_j(\omega,\theta)]\,\omega L}{2c}\right\} = \mathrm{sinc}\left\{\beta L[\omega-\omega_0(\theta)]\right\} \tag{5}$$

where $n_i(2\omega,\theta)$ and $n_j(\omega,\theta)$ are the refractive indices as a function of frequency and potentially crystal angle for the two beams, c is the speed of light, L is the crystal thickness, β is nearly a constant, and $\omega_0(\theta)$ is the phase-matching frequency for the crystal angle, θ. For very broadband pulse measurement problems, additional effects,[17] such as the dependence of β on frequency, frequency-dependent mode size, the frequency-dependent strength of the nonlinear-optical susceptibility, and the factor of ω that appears in the slowly varying envelope equation also appear in $F(\omega,\theta)$, but the above expression suffices for our purposes at the moment. We will, however, consider these factors later.

Usually, one requires that $F(\omega,\theta)$ be essentially constant over the spectral range of the signal pulse (i.e., that of the pulse to be measured). In this case, the measured signal field will be precisely the usual measured field that is assumed in SHG FROG measurements:

$$\tilde{E}_{sig}(\omega,\tau,\theta) \propto \mathrm{F}\left\{E(t)\,E(t-\tau)\right\} \tag{6}$$

Of course, this is not always possible—or even desirable. If, however, we vary the crystal angle during the measurement in such a manner that it includes at some point every frequency of the pulse to be measured, then an accurate measurement of the pulse can be obtained. Figure 1 is a simple animation picturing the essential idea of phase matching part of the pulse at a given angle.

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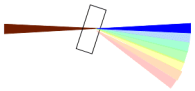

Fig. 1. (116K) Animation of angle dithering to increase the effective bandwidth. While a "thick" SHG crystal may not phasematch the entire pulse at once, dithering the crystal can cover the full bandwidth of the pulse. Dithering much more quickly than the integration time of the measurement can increase the effective overall bandwidth of otherwise too-thick crystals.

Mathematically, suppose that we vary the crystal angle so that it has an angular distribution over the measurement period of, say, $g(\theta)$. The signal field will then be the weighted average of the mag-squared signal field over the various crystal angles seen by the device:

$$I_{FROG}^{dither}(\omega,\tau) = \int_{-\pi}^{\pi} \left|\tilde{E}_{sig}(\omega,\tau,\theta)\right|^2 g(\theta)\, d\theta \tag{7}$$

$$= \int_{-\pi}^{\pi} \left|\mathrm{F}\{E(t)\,E(t-\tau)\}\,F(\omega,\theta)\right|^2 g(\theta)\, d\theta \tag{8}$$

Since only the frequency filter, $F(\omega,\theta)$, and angular distribution, $g(\theta)$, depend on the crystal angle, only they remain inside the integral:

$$I_{FROG}(\omega,\tau) = \left|\mathrm{F}\{E(t)\,E(t-\tau)\}\right|^2 \Phi(\omega) \tag{9}$$

where:

$$\Phi(\omega) \equiv \int_{-\pi}^{\pi} \left|F(\omega,\theta)\right|^2 g(\theta)\, d\theta \tag{10}$$

Thus, the integral, $\Phi(\omega)$, yields a new, considerably broader frequency-dependent filter, which can be made fairly constant over the range of frequencies of the pulse. Indeed, if the above sinc dependence for $F(\omega,\theta)$ is accurate (and β is in fact a constant), then, as long as the crystal angle-dither range is large, $\Phi(\omega)$ is a constant over the entire spectrum of the pulse and so it can be neglected. A more rigorous examination of the phase-matching efficiency vs. wavelength, accounting for variation in height and width of the efficiency curves as a function of angle, ω-dependent spot size, the frequency-dependent strength of the nonlinear-optical susceptibility, and the usual factor of ω that results from the slowly varying envelope approximation, still yields only a small variation. For example, for a fundamental wavelength range 750 nm to 850 nm in BBO, the variation in efficiency is linear and small, 0.0018/nm, and accounting for this factor resulted in no perceptible variation in the retrieved answer for our ~40 nm (FWHM) pulses. For measuring very broadband pulses, $\Phi(\omega)$ will likely have larger variations that must be taken into account. This is not difficult, however, as the FROG trace need simply be divided by $\Phi(\omega)$ before utilizing the pulse-retrieval algorithm. In other words, this easily implemented correction occurs outside the pulse-retrieval algorithm.

3. Experiment and Analysis

In order to demonstrate this technique, we modified an existing calibrated single-shot FROG by simply replacing the thin 100-μm KDP crystal, (which had sufficient phase-matching bandwidth) with a 1-mm BBO crystal (which, undithered, had insufficient phase matching

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bandwidth). The thick BBO crystal was mounted on an Intelite CLS-200 galvanometer, hooked to allow us to dither the crystal. The input signal to the galvanometer was a Stanford Research Systems DS345 function generator, which allowed us to modulate the amplitude and frequency of the dithering. The input pulses were generated by a K-M Labs Ti:Sapphire oscillator, with a central wavelength of 800 nm and approximately 40 nm of bandwidth. The pulses were split and recombined in the SHG crystal with a tunable delay as a pair of crossed-line foci using a cylindrical lens (f = 20 cm, input spot size ~7 mm, crossing angle ~6º), as per the usual single-shot FROG geometry.[5] Here, "single-shot" refers to the mapping of delay onto position by crossing at a line focus, but the measurement is still "multishot" in the sense that the measurement is not of a single pulse, but of a number of identical pulses. The resulting autocorrelation signal was imaged onto the slit of a Acton SpectraPro150 spectrometer. The spectrometer output was sent to a Cohu 2122 CCD camera and Spiricon LBA-PC video capture card, and the intensity and phase retrieved from the resulting FROG traces using Femtosoft's commercially available FROG code. The only difference between the thin KDP experiments and the thick BBO experiments was the addition of more neutral density filters in front of the spectrometer to keep from saturating the camera, since the BBO produced about 100 times more SHG signal.

We first measured our oscillator pulses using the above standard single-shot FROG setup, using the 100-μm KDP crystal, undithered. We then replaced the KDP crystal with our 1-mm BBO crystal and made a second measurement. As expected, the BBO crystal had insufficient bandwidth to phase-match the entire pulse, resulting in a FROG trace that was "clipped" in frequency. We then dithered the BBO crystal at 62 Hz, and observed the CCD camera output. As we increased the amplitude of the dithering via the output voltage function generator (using the built in ramp function), the trace expanded along the frequency axis, until a "threshold voltage," after which further increases in voltage only decreased the overall amplitude of the FROG trace. To insure that the phase matching was even across all wavelengths, we took our measurement at approximately twice the threshold voltage, corresponding to a dither angle range of ~11˚(190 mrad). To just barely dither enough to phase-match all the wavelengths is unadvisable, as it would be analogous to using a crystal just *barely* thin enough, causing an artificial weighting function favoring central wavelengths and suppressing the highest and lowest wavelengths. One could imagine, however, cleverly dithering the crystal in a nonlinear manner, emphasizing wavelengths in the wings of the pulse spectrum (and correcting for this bias later) in order to more accurately measure the pulse in these spectral regions. We have chosen not to do this for this experiment, as FROG does very well at measuring the phase even when the intensity is low.

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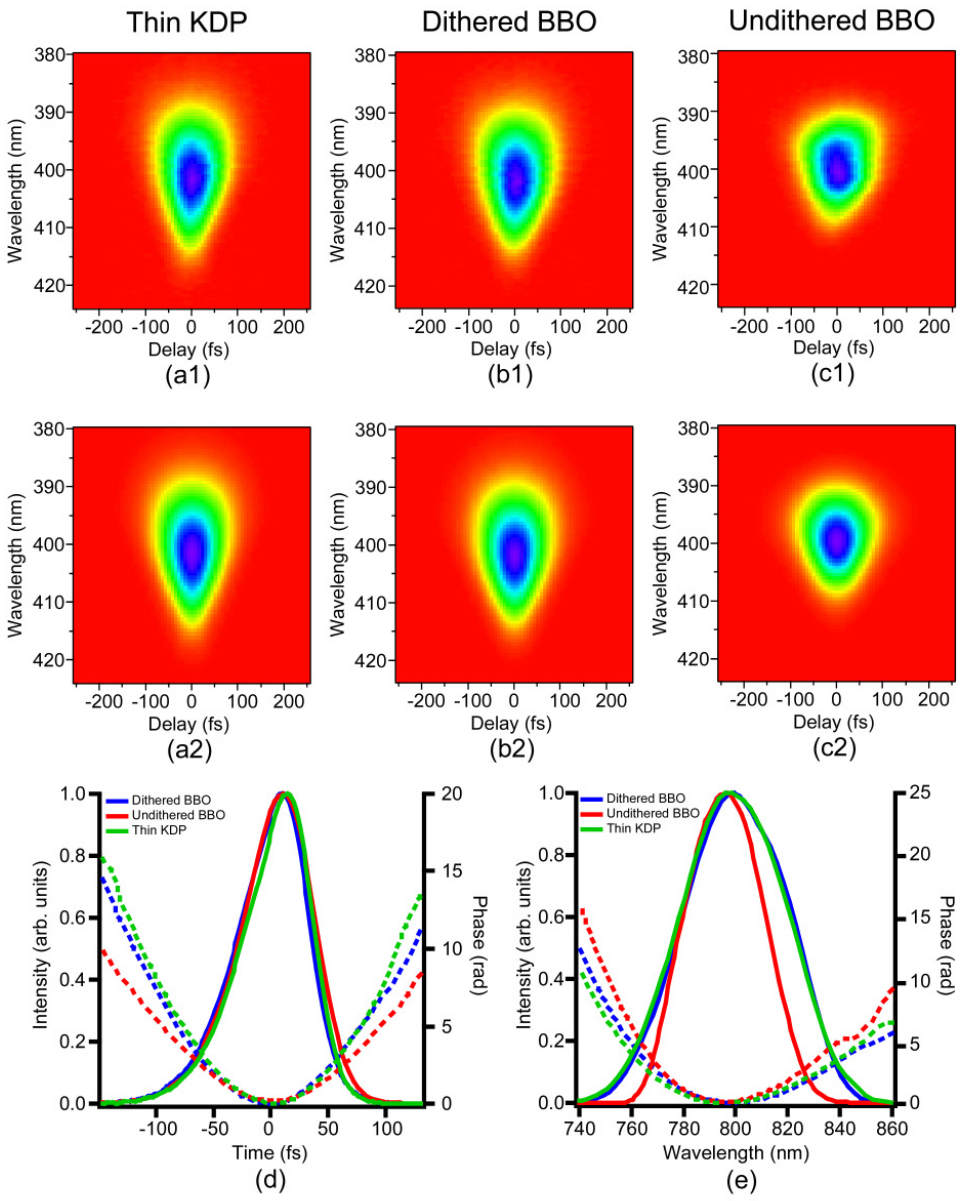

Fig. 2. The measured and retrieved FROG traces and retrieved intensities and phases for 3 different setups. (a1), (b1), and (c1) are the measured FROG traces for the 100 μm KDP crystal, the dithered 1 mm BBO crystal, and the undithered BBO crystal, respectively. (a2), (b2), and (c2) are the corresponding retrieved traces. Note the clipping of frequencies in the undithered BBO crystal. Shown in (d) and (e) are the temporal and spectral retrieved intensities and phases of all 3 cases (solid lines are intensities, dashed lines are phases). Note the undithered BBO retrieval obtains a clipped spectrum and an incorrect temporal and spectral phase, despite the appearance of the temporal intensity being correct.

Figure 2 shows the measured FROG traces and the retrieved intensities and phases vs. time and frequency, where the pulse time-domain electric field is defined as:

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$$E(t) = \mathrm{Re}\left\{\sqrt{I(t)}\,\exp\left(\mathrm{i}\omega_0 t - \mathrm{i}\varphi(t)\right)\right\} \tag{11}$$

where $I(t)$ and $\varphi(t)$ are the time-dependent intensity and phase of the pulse, and ω_0 is a carrier frequency. The frequency-domain quantities are defined similarly. The agreement between the pulse measurements made using the thin KDP crystal and the angle-dithered thick BBO crystal is excellent. For comparison, the FROG code was run to "retrieve" the intensity and phase of the erroneous undithered BBO trace. The FROG errors, pulse widths, spectral widths, and time-bandwidth products (TBP) are shown in Table 1.

Crystal	Temporal Width (fs)	Spectral Width (nm)	FROG error	TBP
100 μm KDP	67.3	49.1	0.00478	1.56
1 mm BBO (dithered)	68.8	48.7	0.00434	1.57
1 mm BBO (undithered)	72.1	37.0	0.00697	1.26

Table 1. The measured temporal and spectral widths, FROG errors, and time-bandwidth products (TBP) for FROG measurements made using three different nonlinear-optical crystals. Note the excellent agreement between measurements made using the thin KDP crystal and the angle-dithered thick BBO crystal. Note also the significantly reduced spectrum and TBP obtained using the undithered thick BBO crystal.

As expected, the spectral width of the pulse measured using the undithered BBO is considerably less, and the FROG error is higher, as the FROG trace does not correspond to a real pulse. However, the error is only slightly higher here, and still less than 1%. This could fool an experimentalist into thinking his or her FROG was correctly set up, and the crystal sufficiently thin. This problem is easily discovered, however, by manually rotating the crystal about its phase-matching axis; if new portions of the trace are "introduced" (or enhanced) as the crystal is rotated, the crystal is clipping off (or unevenly weighting) frequencies, and is too thick for undithered usage.

Another consideration is that, when any beam is brought to a focus into a SHG crystal, the beam is actually entering the crystal at a finite range of angles, which increases the phase-matching bandwidth of the process. This is analogous to the dithering, except that all the wavelengths are phase-matched at once. This effect almost certainly plays a positive role in most pulse measurements, whether FROG or autocorrelation or another technique. And, indeed, it is playing a role in our experiments. From the Sellmeier equations, one would expect 1-mm BBO to have ~2.5 nm of phase-matching bandwidth (FWHM), but we instead have almost 40 nm. But focusing easily explains this effect; to phase-match 800±19nm requires only a 1.4º (24 mrad) range of angles, which, with our 200-mm focal length lens and accounting for refraction at the crystal face corresponds to our input spot size of about 7 mm. However, focusing alone should *not* be used to increase the phase-matching bandwidth. When the line focus is imaged onto the spectrometer slit, SHG will enter the spectrometer at a range of angles, which will alter the direction of diffraction of the gratings and result in errors. The range of angles must be kept small, or systematic error will be introduced.

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Also, the variations of the crystal length seen by the beams as the crystal angle varies have a very small effect. Our dithering of 11º (190 mrad) introduces in BBO at most a factor of 1/cos(.057)=1.0016 of extra crystal length, which is negligible.

In addition, we should point out that dithering the crystal angle walks the signal beam in space. In FROG measurements, where the signal beam must then be spectrally resolved, it is crucial that the signal beam does not walk off the spectrometer slit. Thus, it may be helpful to dither the crystal angle so that the beam walks in the direction *along* the spectrometer slit, and not perpendicular to it, although this was not essential in our experiment, as the beam walk was fairly small at the near-normal-incidence angles used in our experiment. The walkoff introduced in dithering 1 mm of BBO, even dithered as much as ± 90 mrad, is less that 40 μm, and we are deliberately over-dithering to keep the phase-matching relatively flat, so the walkoff of actual signal is closer to less than 20 μm.

Angle-dithering the crystal angle allows the use of a significantly thicker crystal for FROG measurements. In effect, angle-dithering removes phase-matching-bandwidth effects from pulse-measurement devices. Stated equivalently, it removes group-velocity-mismatch (GVM) effects from consideration. However, this now allows group-velocity *dispersion* (GVD) to be the next strongest distorting effect. GVD is almost always much smaller than GVM, however, and in previous ultrashort-pulse measurements, it could always be neglected. Taking advantage of the possibility of using a dithered thick crystal in the manner described herein, GVD must now be considered. Fortunately, it yields very little distortion in our measurements, (our retrieved pulse using the dithered BBO is only 1.5 fs longer than the retrieved pulse from the stationary 100 μm KDP crystal) but it must be considered in measurements of extremely short pulses when taking advantage of angle-dithering of a relatively thick crystal.

4. Conclusions

We have shown experimental evidence that a SHG crystal that is otherwise too “thick” for sufficient phase-matching bandwidth can be angle-dithered to obtain the bandwidth necessary for multishot FROG measurements. Use of such a thick crystal gives the advantage of increased signal strength (over two orders of magnitude in our experiment.) These thick crystals are as easily obtained as standard crystals, easier to handle, and are easily implemented in existing experimental setups. This technique overcomes the limitation of group-velocity-mismatch (GVM), and introduces considerations of group-velocity-dispersion (GVD) , but proper crystal selection keeps pulse spreading small or negligible.

Finally, we point out that angle-dithering the SHG crystal for increased bandwidth can be used in other pulse-measurement techniques, such as autocorrelation, collinear techniques,[18] and frequency-domain phase measurement.[1]

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