

# Shaping of wide bandwidth 20 femtosecond optical pulses

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We investigate the propagation of 20 fs pulses through a generalized pulse shaper/compressor and synthesize shaped waveforms with 20 fs features by linear spectral filtering using a generalized pulse shaper consisting of gratings and reflective optics. The use of reflective optics in the pulse shaper avoids cubic phase dispersion associated with lenses which significantly broaden short 20 fs pulses. As an example of our pulse-shaping capabilities, we generate pulse trains with repetition rates in excess of 12 THz using phase-only filtering.

In recent years, there has been considerable effort directed toward understanding the propagation of ultrashort pulses through generalized pulse compressors/shapers which employ spectrally dispersive elements (gratings or prisms) and temporally dispersive elements (lenses).<sup>1-5</sup> These studies have been motivated by a wide variety of applications, including the synthesis of arbitrarily shaped femtosecond pulses,<sup>1</sup> dispersion compensation in fiber-based high speed optical communications systems,<sup>2,3</sup> and stretching ultrashort pulses for high energy, chirped-pulse amplification (CPA).<sup>4</sup> By controlling the sign of group velocity dispersion in these systems, generalized compressor/shapers have been employed to stretch, compress, and perform linear filtering operations on pulses. However, little attention has been paid to higher order phase dispersion. In this letter, we investigate the propagation of 20 fs pulses through a generalized compressor/shaper in a regime where cubic phase dispersion (CPD) plays a dominant role. We show that by using a generalized pulse shaper consisting of all-reflective optics, severe CPD associated with the use of lenses can be avoided. This enables us to synthesize arbitrarily shaped optical waveforms with individual features as short as 20 fs, such as pulse trains with repetition rates exceeding 12 THz.

Our studies are also motivated by applications in molecular spectroscopy and control,<sup>6,7</sup> as well as in CPA. For example, theoretical simulations predict that designer fields suitable for controlling molecular motion will necessarily have features shorter than 10 fs. The ability to temporally tailor extremely short optical pulses therefore represents a significant advance toward achieving the more general goal of synthesizing optical fields suitable for molecular control. Furthermore, in high-power chirped pulse amplifiers, imperfect pulse recompression is often attributed to aberrations associated with refractive optics in pulse stretchers.<sup>5</sup> The use of reflective optics in generalized pulse stretchers may overcome this problem.

Our generalized pulse shaper, shown in Fig. 1, consists of a pair of 600-line/mm gratings placed at the focal planes of a unit-magnification confocal pair of concave 12.5 cm focal length spherical mirrors. Midway through the apparatus, the optical frequencies are spatially separated, with a spatial dispersion given by  $dx/d\lambda \approx f/(d \cos \theta_d) \approx 0.082$  mm/nm, where  $f$  is the focal length,  $d$  is the grating period,  $\theta_d = 24^\circ$  is the diffraction angle, and  $x$  is the position of each frequency component in the masking plane. Our

wide bandwidth shaper/compressor uses mirrors instead of achromatic lenses to avoid the deleterious effects of cubic phase dispersion which can distort short pulses and limit pulses shaping capability, discussed in more detail below. The angle between incident and reflected beams of each mirror is kept small ( $\sim 10^\circ$ ) to minimize astigmatism through the apparatus. For pulse shaping, a spatially patterned mask placed in the Fourier plane of the shaper modifies the amplitude and/or phase of spatially dispersed optical frequency components. The temporal profile of the output field is then given by the Fourier transform of the pattern transferred by the mask onto the pulse spectrum. A colliding-pulse mode-locked (CPM) ring dye laser<sup>8</sup> operating at 620 nm provides 80 fs pulses which are amplified to energies of 20  $\mu$ J in a two-stage copper vapor laser pumped amplifier.<sup>9</sup> Compression down to 20 fs is achieved using spatial soliton pulse compression,<sup>10</sup> a novel method which uses the stable, self-trapped propagation of bright, elliptically focused spatial solitons in a bulk nonlinear Kerr medium to achieve self-phase modulation ( $\Delta\lambda = 30$  nm), followed by single passing the pulses through a grating pair. Unlike fiber compression, in which pulse energies are limited to 10 nJ or less, we achieve compressed pulse energies up to 0.6  $\mu$ J. Pulse durations are measured in a conventional noncollinear autocorrelator before being sent to the shaper. A small portion of the compressed pulse is split off before entering the shaper and used as a reference for performing intensity cross correlation measurements of the shaped pulses.

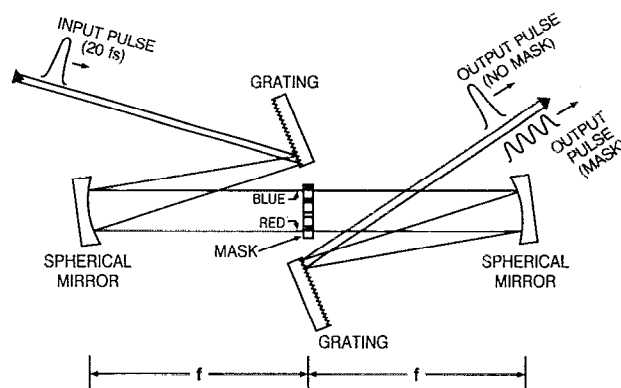


FIG. 1. Dispersion-free, wide-bandwidth femtosecond pulse shaping apparatus.

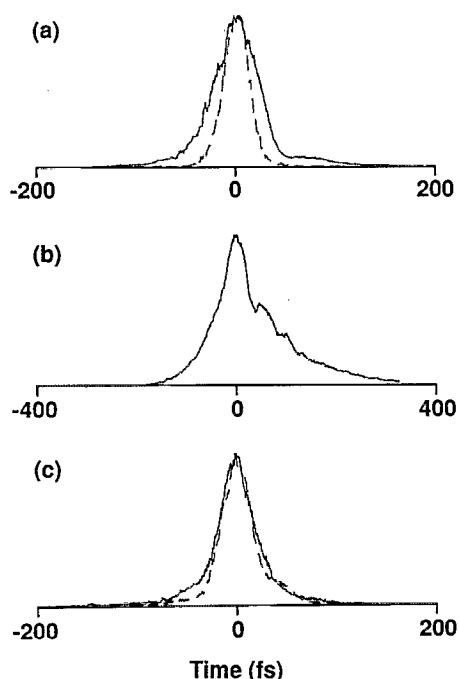


FIG. 2. (a) Cross correlation (solid line, FWHM=51 fs) of unshaped output pulses from a pulse shaper containing achromatic lenses with 18 fs  $\text{sech}^2$  reference pulses (autocorrelation shown as dashed line, FWHM=28 fs) with grating separation set for minimum cross correlation width; (b) cross correlation of unshaped output pulses from a pulse shaper containing achromatic lenses with 18 fs  $\text{sech}^2$  reference pulses with grating moved 3 mm off minimum cross correlation width setting; (c) cross correlation (solid line, FWHM=37 fs) of unshaped output pulses from the pulse shaper shown in Fig. 1 with 21.5 fs  $\text{sech}^2$  reference pulses (autocorrelation shown as dashed line, FWHM=33 fs).

We first investigate the effects of higher order phase dispersion on pulses propagating through a generalized compressor/shaper consisting of gratings and 50 mm focal length achromatic lenses (such as that described in Refs. 1 and 2). In Fig. 2(a), we display the cross correlation of the output of the shaper (solid line) with an 18 fs input pulse [autocorrelation full width at half maximum (FWHM) of 28 fs, shown as dashed line]. The cross correlation FWHM has nearly doubled to 51 fs, corresponding to a deconvolved pulsewidth of 48 fs. Furthermore, the cross correlation is noticeably asymmetric, with a slight tail appearing after the main peak. We believe that uncompensated cubic phase dispersion (CPD) is responsible for these effects. Normally, the grating separation in a grating/lens shaper can be set such that a pulse suffers no dispersion at any order,<sup>2</sup> assuming thin lenses are free of aberrations. This, however, neglects any contribution of material dispersion introduced by the achromats, which can be significant for pulses much shorter than 100 fs. The separation of the gratings can be adjusted to provide negative group velocity dispersion to compensate for any linear chirp (positive quadratic phase dispersion) introduced by lens material; however, this introduces phase dispersion at *all* orders (in particular, positive CPD) due to the gratings in the shaper.<sup>3</sup> Additionally, any positive CPD from the lenses themselves will add to the total CPD. We have estimated the broadening from each source using formulas in Ref. 3

for CPD contributions from the grating separation and the standard Sellmeier equations for material CPD contributions from the lenses. We find that the total broadening  $\Delta\tau_{\text{CPD}}$  is  $\sim 43$  fs for pulses with 30 nm bandwidths, in reasonable agreement with the observed broadening of 30 fs. When the grating separation is adjusted slightly in order to obtain the sharpest central peak, the cross correlation [Fig. 2(b)] becomes markedly asymmetric with a long oscillatory tail, further evidence for the presence of CPD in grating/lens shapers. Replacing the achromats with single element lenses results in similar amounts of temporal broadening, now due to chromatic aberration. Due to the variation in focal lengths of different frequency components of the pulse, each frequency sees a different grating separation and therefore different amounts of phase delay. Note that grating/lens pulse shapers work well with 75 fs input pulses. The CPD observed for 20 fs pulses is a direct consequence of the increased bandwidth associated with shorter pulses.

In contrast, a short pulse which propagates through our grating/mirror shaper suffers negligible broadening. Figure 2(c) shows the cross correlation of the output of the shaper (solid line) with a 21.5 fs reference pulse (autocorrelation FWHM of 33.3 fs, shown as the dashed line). Dispersive effects due to the mask substrate, ordinarily present in shaping experiments, were included by placing an unpatterned 3 mm thick glass blank in the masking plane. The cross correlation is slightly broadened from 33 to 37 fs, demonstrating the essentially dispersion-free nature of the shaper. The slight temporal broadening is possibly due to a slight cubic phase dispersion introduced by the substrate, although 4 fs is within our experimental measurement error, due to amplitude fluctuations of the amplified pulses. We believe that this result bears directly upon a problem that has recently plagued workers in the ultrafast high-energy amplifier community; namely, that of stretching and recompression of femtosecond pulses for chirped-pulse amplification. Many groups<sup>4</sup> report on the lack of complete recompressibility after stretching 60–100 fs pulses in multipass pulse stretchers which use gratings and achromats in a dispersive configuration. Lens aberrations have been suggested as the cause of this distortion.<sup>5</sup> The use of mirrors in pulse stretchers may circumvent this problem.

As a demonstration of our pulse shaping capability, we have synthesized variable, high repetition rate pulse trains using phase-only masks<sup>11</sup> based on so-called maximal length sequences ( $M$  sequences).<sup>12</sup> These masks are pseudorandom binary phase masks whose phase response is periodic with a period corresponding to a frequency  $\delta F$ . Each period is divided into  $P$  pixels, with the phase of each pixel given by either zero or  $\Delta\phi = 2\pi d(n-1)/\lambda$ , as determined by the  $M$  sequence, where  $d$  is the etch depth of the pixel and  $n$  is the index of the substrate. In this case, we chose a depth corresponding to a phase shift of  $0.84\pi$  for  $\lambda = 620$  nm. The output pulse train then consists of a series of  $\sim P$  individual pulses with repetition rate  $\delta F$ . Because we use phase-only filtering, all of the energy of the incident pulse is used to form the pulse train, resulting in intensities

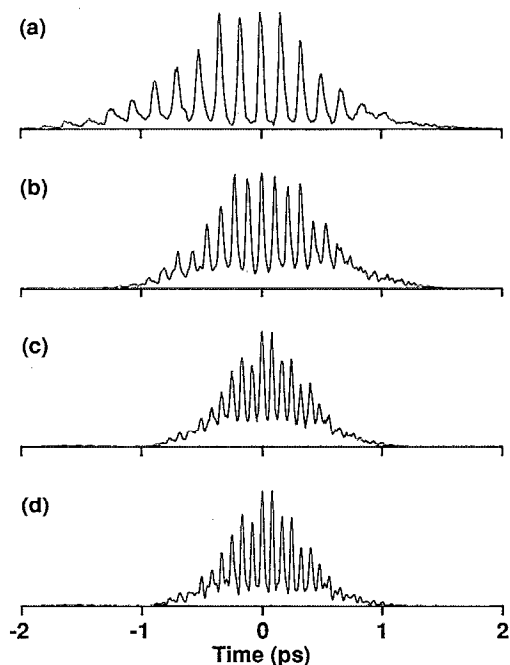


FIG. 3. Cross correlations of (a) 6.0 THz, (b) 9.3 THz, and (c) 12.5 THz pulse trains generated using phase-only filtering; (d) deconvolution of the measured cross correlation shown in (c) with a 22 fs reference pulse.

greater than those obtained using amplitude filtering. Figure 3 shows cross-correlation measurements of pulse trains generated for  $\delta F = 6.0$  THz [Fig. 3(a)], 9.3 THz [Fig. 3(b)], and 12.5 THz [Fig. 3(c)] with  $P = 15$ . In each case, we observe a series of well-defined pulses under a fairly smooth envelope. For the 12.5 THz train, peak-to-valley ratios are in the 70% range, with a modest degradation of the baseline. By comparison, as observed in previous experiments, 5.85 THz pulse trains produced using phase-only filtering of 75 fs pulses experienced severe degradation due to overlap and interference of adjacent pulses in the train.<sup>1</sup> In Fig. 3(d), we display the actual intensity profile of the 12.5 THz train, obtained by deconvolving a 22 fs sech<sup>2</sup> reference pulse from the experimental cross correlation trace shown in Fig. 3(c). The peak-to-valley ratio now approaches 85%. Without the dispersion-free performance of the reflective optics pulse shaper, adjacent pulses would overlap, and well-defined pulse trains at these repetition rates would not be possible. High repetition rate trains such as these may prove useful for selective excitation of large amplitude phonon modes in III-V and II-VI semiconductors via multipulse impulsive stimulated Raman scattering.<sup>6</sup>

Other challenges faced when shaping ultrashort pulses are the variation of phase and spatial dispersion ( $dx/d\lambda$ ) over the pulse spectrum. For example, the linear spatial dispersion  $dx/d\lambda$  and phase difference  $\Delta\phi$  will vary by 0.7% and 4.6%, respectively, for a pulse having a 30 nm bandwidth (similar to that used in our experiments), due to higher order spatial dispersion from the gratings ( $dx^2/d^2\lambda$ ) and phase variation caused by fixed etch depths (since  $\phi \sim 1/\lambda$ ) in phase masks. We have numerically ex-

amined how these variations affect the fidelity of shaped pulses in our current experiments by computing theoretical intensity profiles of high repetition rate pulse trains for bandwidths up to  $\Delta\nu = 50$  THz ( $\Delta\lambda = 65$  nm). We assume pulse trains are generated using phase masks patterned according to periodic repetitions of  $M$ -sequences as in Ref. 11. For these calculations, both the variation of linear spatial dispersion and the exact phase difference  $\Delta\phi(\lambda)$  in the masking plane were computed exactly by determining  $x(\lambda)$  (directly from the grating equation) and phase at the masking plane. Our results indicate a remarkable insensitivity to these variations and show that, even with pulses as short as 9 fs, well-defined pulse trains with repetition rates up to 20 THz are possible, with peak-to-valley contrast ratios approaching 100% and no temporal distortion of the individual features. These results indicate that the temporal resolution achievable in these experiments is determined by the duration and spectral bandwidth of the input pulses rather than variations of phases and linear spectral dispersion.

In summary, we have demonstrated the shaping of pulses with features as short as 20 fs using linear spectral filtering in a modified, reflective optics pulse shaper which minimizes the effects of cubic phase dispersion. To show our capability, we have generated high energy bursts of 20 fs pulses with repetition rates up to 12.5 THz using phase-only filtering. Other arbitrarily shaped pulses, such as square pulses, are also possible. Generalized pulse compressors/shapers such as these may also play an important role in stretching pulses for high energy, chirped pulse amplification.

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