Ultrafast spatiotemporal laser pulse engineering using chromatic dispersion

Yuelin Li\textsuperscript{1,3}, Sergey Chemerisov\textsuperscript{1} and Baifei Shen\textsuperscript{2}

\textsuperscript{1} Argonne National Laboratory, Argonne, IL 60439, USA
\textsuperscript{2} State Key Laboratory of High Field Laser Physics, Shanghai Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, PO Box 800-211, Shanghai 201800, China
E-mail: ylli@aps.anl.gov

\textit{New Journal of Physics 12} (2010) 123011 (17pp)
Received 21 July 2010
Published 8 December 2010
Online at \url{http://www.njp.org/}
doi:10.1088/1367-2630/12/12/123011

\textbf{Abstract.} We discuss a new method for manipulating the optical field distribution in time and space at the focal plane of a chromatic lens. Chromatic dispersion and aberration make it possible to control the photon distribution in both time and space by properly controlling the amplitude and phase of the incident laser pulse, including shortening a laser pulse or generating a specific time-dependent transverse distribution. As an example, we discuss the generation of a quasi-three-dimensional ellipsoidal photon distribution and a proof-of-principle experiment, where favourable agreement between theoretical calculations and the data is observed.
1. Introduction

Tailoring the geometry of a laser pulse is one of the important areas of laser research and applications. The most common form of a laser pulse is a Gaussian distribution in all dimensions. Transversely, this is the solution of the paraxial Helmholtz wave equation. A Gaussian distribution is invariant under Fourier transforms in both transverse and longitudinal dimensions.

Many applications only require shaping the transverse irradiance profiles. Those include material processing, medical procedures, lithography and optical data processing [1], mostly to generate a flat-top, homogeneous intensity distribution. A variety of aspheric optics designs exist to convert a Gaussian beam into a flat-top beam [1–3].

Precise, high-fidelity temporal shaping is demanded in applications such as coherence control in quantum systems, optical signal processing and laser–matter interaction for ultrafast electron and radiation sources and is made possible by the advent of ultrafast lasers [4]. Weiner [4] has given a comprehensive review of available techniques. The two main techniques are the spatial light modulator (SLM) [5] and the acousto-optic programmable dispersive filter (AOPDF) [6], which allows phase and amplitude tailoring in the time and frequency domains in ps or fs time.

Spatiotemporal control is intrinsically complex due to the difficulty in simultaneously controlling the spatial and temporal distributions. Examples of successful high-fidelity shaping are sparse. The techniques include the use of two-dimensional (2D) SLM to shape the waveform of the pulse at different spatial locations in a 2D manner [7], or the use of the spatial–temporal duality of light by transforming a 2D holographic image into a 2D spatial–temporal distribution [8]. 3D control has so far been achieved only by structured optics [9] or temporal multiplexing via volume holography [10].

One of the important applications of multiple dimension control of a laser pulse is for high-brightness electron beam generation in a photoinjector. This is critical for cost-effective x-ray free-electron lasers [11, 12] and other beam-based light sources [13], as well as ultrafast electron beam imaging and diffraction experiments [14]. To this end, viable solutions for a homogeneous cylindrical beam have been developed using a combination of transverse shaping and temporal phase control or with pulse stacking using multiple delay optics [15, 16] and birefringence crystals [17–20].
Figure 1. Schematic diagram of how phase tailoring can lead to control of the spatiotemporal distribution at the focal plane of a chromatic lens. Light rays of different colours are focused at different distances from the lens. In this case, the red rays are focused but the blue rays are not (left panel). Tailoring of the laser phase in time so that the blue is sandwiched between the red in time (middle panel), which results in the pulse going through a sequence of focusing–defocusing–focusing, thus a time-dependent transverse beam size (right panel).

For the more desirable homogeneous ellipsoidal beam shape due to totally linear space charge force [21], pulse stacking becomes very complicated, although theoretically still possible, for example, via temporal multiplexing via volume holography [10]. The drawback, besides the low efficiency, is the volatility of the hologram media, which degenerates during readout [22].

In this paper, we will discuss a new way of controlling the spatiotemporal distribution of photons via the well-known chromatic dispersion in an optical system [23–25]. Normally, such an effect causes adverse effects that distort both the temporal and the spatial fidelity of a laser pulse. In space, this gives rise to chromatic aberration, i.e. light of different wavelengths will focus at different distances from the lens. For ultrashort pulses, significant lengthening of the pulse has been predicted and observed [26–28].

However, due to the wavelength and phase dependence of the chromatic dispersion, pulse engineering is possible by coupling the space and time domain properly using available time and frequency domain phase- and amplitude-tailoring techniques.

We will start with a discussion of the chromatic aberration and its coupling to the time domain phase in a singlet lens, followed by Fourier optics of a laser pulse transmitted through a singlet lens with examples of temporal distortion due to chromatic dispersion for pulse with and without nonlinear phases. We will then give an example of using the aberration–phase coupling for an ellipsoidal pulse generation together with a proof-of-principle experiment. Future work will also be addressed.

2. Formulation of chromatic dispersion in an optical singlet lens

2.1. Chromatic aberration in Gaussian optics

Chromatic dispersion is the phenomenon in which the phase velocity and group velocity of a wave depends on its frequency, arising from the dependence of the refractive index upon the optical frequency [29].

Chromatic dispersion, as a function of frequency, manifests itself into space as chromatic aberration in an optical lens, where light with different frequencies focuses at different locations.
For a thin, singlet spherical lens in air, the focal length $f$ is

$$\frac{1}{f} = [n(\omega) - 1] \left( \frac{1}{R_1} - \frac{1}{R_2} \right),$$

where $R_1$ and $R_2$ are the radii of curvature for the entrance and exit sides of the lens, respectively, and $n(\omega)$ is the frequency- or wavelength-dependent refractive index of the lens material. From equation (1), the change of the focal length due to a shift in frequency $\delta \omega$ is

$$\delta f = -\frac{f_0}{n_0 - 1} \beta \delta \omega,$$

where $f_0$ is the nominal focal length at $\omega_0$. We assume a constant $\beta = dn/d\omega$ for this analysis. At the nominal focal plane for $\omega_0$, this leads to a defocused beam or a larger focus size. In Gaussian optics, this new focus size can be easily calculated [29],

$$w \approx w_0 \left[ 1 + \left( \frac{\delta f}{z_R} \right)^2 \right]^{1/2} = w_0 \left[ 1 + \left( \frac{1}{z_R n_0 - 1} \beta \delta \omega \right)^2 \right]^{1/2}.$$ 

Here $w_0 = N\lambda_0/\pi$ is the beam waist at the nominal wavelength $\lambda_0$, with $N$ being the numerical aperture, and $z_R = \pi w_0^2/\lambda_0$ is the Rayleigh range. Note that both the beam waist and the Rayleigh length are a function of the wavelength. However, in our analysis, the effect is small and is henceforth ignored.

It is obvious from equations (2) and (3) that if one can manipulate $\delta \omega$ in time, a programmable time-dependent beam size can be achieved at a certain focal plane (see figure 1). At $\delta f \gg z_R$, one has

$$w(t) \approx w_0 \frac{1}{z_R n_0 - 1} \beta \delta \omega(t).$$

For a given $w(t)$, the needed phase of the chirped laser pulse can be obtained as

$$\phi(t) = \pm \int \delta \omega(t) \, dt = \pm \frac{n_0 - 1}{\beta} N \int w(t) \, dt.$$ 

For a desired time-dependent intensity $I(t)$, the amplitude of the laser should be

$$a(t) \propto I(t)^{1/2} w(t).$$

This is an example of how behaviour in time and space can be coupled, and it is only made possible through the interplay of chromatic dispersion and chromatic aberration. This forms the theoretical base for our spatiotemporal shaping method. It should be noted that the shaping is a localized time-dependent effect at a fixed plane in space.

As can be seen below, the final result is also significantly affected by the time domain effect and a certain diffraction due to the apodization of the beam.

### 2.2. Time domain effects using Fourier optics

The above discussion is based on geometry and Gaussian optics, where the effect due to dispersion is only considered in the space domain.

To treat the time domain effect, we base our discussion on the Fourier optics formula for a dispersive singlet lens elaborated by Kempe et al [28]:

$$U(r, \omega) = \int_0^\rho \rho \, d\rho \int_0^{2\pi} d\theta A(\rho, \omega) \Gamma_1(\rho, \omega) \exp \left( -jk_0 \sqrt{f_0^2 + \rho^2 + r^2 - 2\rho r \cos \theta} \right).$$

New Journal of Physics 12 (2010) 123011 (http://www.njp.org/)
\[ \Gamma_1(\rho, \omega) = \exp(jk_l d) \exp \left[ -j k_l k_a \frac{\rho^2}{2(n-1)f_0} \right]. \] (8)

Here \( U \) is the frequency domain representation of the field at the focal plane and \( f_0 \) is the nominal focal length of the lens. Here \( A(\rho, \omega) = F[a(\rho, t) \exp(-j\phi(t))] \) is the Fourier transform of the input pulse; and \( P, \rho \) and \( \theta \) are the lens radius or beam aperture, the ray entrance location from the axis and the azimuthal angle, respectively. \( \Gamma_1 \) is the lens transfer function, with \( k_l \) and \( k_a \) being the wave numbers in the lens and air, respectively. \( \Gamma_1 \) derives its \( \rho \) dependence from the thickness variation of the lens across the beam aperture. For a flat-top input beam, at the focal plane of the lens and on axis at \( r = 0 \), equation (7) can be rewritten as

\[ U(\omega) = 2\pi \int \rho \, d\rho A(\omega) \Gamma(\rho, \omega), \] (9)

\[ \Gamma(\rho, \omega) = \exp \left( jk_l d + \frac{k_a}{2f_0} \rho^2 \right) \exp \left[ -j (k_l - k_a) \frac{\rho^2}{2(n-1)f_0} \right]. \] (9a)

The time domain field can be obtained by an inverse Fourier transform of equation (9),

\[ u(t) = 2\pi \int \rho \, d\rho a^* \gamma, \] (10)

where the symbol \( * \) represents convolution, with \( \gamma = \gamma(\rho, t) = -F^{-1} \Gamma(\rho, \omega) \), respectively. Here \( F^{-1} \) denotes inverse Fourier transformation.

Following the elaboration in Kempe et al [28] and ignoring the constant phase terms, we obtain from equation (9a) for \( \Delta \omega = \omega - \omega_0 \),

\[ \Gamma(\rho, \omega) = \exp \left[ jk(\beta T \Delta \omega + n_0 \chi T \Delta \omega^2) \right], \] (11)

with

\[ T(\rho) = d - \frac{\rho^2}{2(n_0-1)f_0}, \quad \beta = \frac{dn}{d\omega}, \quad \chi = \frac{\beta}{\omega} + \frac{1}{2} \frac{d^2n}{d\omega^2}. \]

Here \( d \) is the thickness of the lens at \( \rho = 0 \). Thus, we have

\[ \gamma(\rho, t) = F^{-1} \Gamma(\rho, \omega) = \frac{1}{2\sqrt{kn_0\chi T}} \exp \left[ \frac{j (t - k\beta T)^2}{4kn_0\chi T} \right]. \] (12)

For a Gaussian temporal profile,

\[ a(t) = a_0 \exp \left[ -2 \ln 2 \left( \frac{t}{\tau} \right)^2 \right], \] (13)

where \( \tau \) is the full-width at half-maximum (FWHM) of the pulse, we obtain from equations (10)–(12)

\[ u(t) = 2\pi a_0 \int \rho \, d\rho \frac{1}{(1 + \Lambda^2)^{1/4}} \exp \left[ -2 \ln 2 \frac{(t - k\beta T)^2}{\tau^2(1 + \Lambda^2)} (1 - j\Lambda) \right] \exp \left[ j \tan^{-1}(-\Lambda) \right], \] (14)

with

\[ \Lambda = \frac{4kn_0\chi T}{\tau^2}. \] (15)
In comparison with the input pulse, equation (14) shows two effects. The first is the group velocity delay (GVDE), which causes the rays at different radial locations to arrive at the focus at different times characterized by \( T(\rho) \) and has a quadratic dependence on the radius \( \rho \) of the entrance position. For short pulses, this leads to time broadening as first discussed by Bor \[26\].

The second effect is the common group velocity dispersion (GVDI), i.e. light waves with different frequencies propagate at different group velocities in a dispersive medium, which also leads to lengthening of the pulse characterized by \( \Lambda \) in (15).

3. Examples of pulse distortion and pulse shaping

3.1. Pulse shortening due to self-phase modulation

An interesting example is when the input pulse has a significant time/envelope-dependent nonlinear phase such as a third-order phase due to self-phase modulation (SPM) or B-integral. As we will see, this is the regime where our spatiotemporal shaping method resides, as described by equations (5) and (6), thus worth discussion in detail.

We assume that the pulse has a small enough bandwidth; hence, the second-order terms in equation (7) can be neglected; therefore

\[ \gamma(\rho, t) = \delta(t - k\beta T), \] (16)

and the nonlinear Schrodinger equation (NLSE) \[30\] governing the propagation of a laser pulse in a nonlinear medium can be analytically solved to give the pulse right after the lens as

\[ a(\rho, t) = a_0 \exp\left(-2 \ln 2 \frac{t^2}{\tau^2}\right) \exp\left[j \mu \exp\left(-4 \ln 2 \frac{t^2}{\tau^2}\right) \zeta\right], \] (17)

\[ \mu = k n_2 a_0^2 d. \]

Here \( \zeta \) is a phase modulation parameter, \( \zeta = T/d \) if the SPM is generated in the lens and \( \zeta = L/d \) if the self-modulated phase is accumulated before arriving at the lens through an effective medium path of \( L \). The pulse at the focus is then

\[ u(t) = a_0 \int \rho \, d\rho \exp\left(-2 \ln 2 \frac{(t - k\beta T)^2}{\tau^2}\right) \exp\left[j \mu \exp\left(-4 \ln 2 \frac{(t - k\beta T)^2}{\tau^2}\right) \zeta\right]. \] (18)

We can see that similar to equation (14), the integrand in equation (18) shows that the field at the focus is the superposition of pulse slices with shifted arriving time. However, the temporal dependence of the phase now causes the superposition to be either constructive or destructive, and in general with small enough \( \mu \), shortens the pulse. This is depicted in figures 2(a) and (b), showing the real part of the integrand in equation (18) as a function of \( t \) and \( \rho \). The drifting of the field pattern as a function of \( \rho \) is clear, resulting in shortened pulses \( u(t) \), as shown in figures 2(c) and (d). The pulses are shortened to 0.21 and 0.2 ps from that of the input pulse of 1 ps for \( \zeta = T/d \) and \( \zeta = L/d = 1 \), respectively, a factor of 5 in reduction. In the calculation, we use a fused silica lens of \( f_0 = 150 \) mm, \( d = 5 \) mm, and an aperture of \( P = 12 \) mm in radius. The laser wavelength is 249 nm. An intensity of \( 5 \times 10^{11} \) W cm\(^{-2}\) is used resulting in a \( \mu = 15 \) rad. We use an \( n_2 = 2.38 \times 10^{-16} \) W cm\(^{-2}\) \[31\]. This pulse shortening is in contrast to the lengthening effect discussed by previous authors \[26–28\]. Under the same condition, a 50 fs pulse would be stretched to 0.5 ps without the SPM. Because of the similarity, we limit our discussion to cases with \( \zeta = 1 \) in the following.
Figure 2. The real part of the complex field at the focus as a function of time and radius for $\zeta = T/d$ (a) and $\zeta = 1$ (b), and the corresponding intensity of the integrated field (solid) and the input pulse (dashed) as a function of time for the same cases ((c) and (d)). The input pulse is shortened from 1 ps to 0.21 ps (c) and 0.20 ps (FWHM) (d) at the focus, a reduction by a factor of 5. The calculation assumes an $f = 150$ mm lens with $P = 12$ mm and $d = 5$ mm. The laser wavelength is 249 nm with $\mu = 15$ at a laser intensity of $5 \times 10^{11}$ W cm$^{-2}$.

As the pulse shortening is due to localized destructive superposition of the field, firstly, the phase slip between the pulse slices should be large enough so that destructive superposition dominates and, secondly, the phase slippage should be small enough so that the phase span is limited and the final pulse will not become the sum of a set of quasi-random phases, which will result in a thermal light [32]. Let the group delay between the centre and edge be $\Delta t$, the above statement can be expressed as $\Delta t \mu / \tau \approx \text{constant}$ or

$$\eta \mu \approx \text{constant}, \quad (19)$$

with

$$\eta = \frac{\beta R^2}{2(n_0 - 1) f_0 \tau} = \frac{1}{N} \frac{\beta P}{\tau}.$$  \quad (20)

The scaling in equation (20) is qualitatively demonstrated in figure 3, where the ratio of the FWHM at the focus to that of the input pulse is plotted as a function of $\eta$ and $\mu$, where the maximum shortening centres around $\eta \mu = 2\pi - 3\pi$. Although the plot in figure 3 is for an input pulse duration of 1 ps, it is verified that it is universal and covers a large range of input pulse duration from a few fs to several ps if the GVDI effect is ignored.

To gain more insight, a set of numerical integrations for equation (7) is carried out with SPM effect included in the input pulse and with GVDI in the lens considered. Figure 4(a) shows the on-axis pulse envelope as a function of the distance from the focus. In figures 4(b) and (c), the intensity distribution as a function of radius and time of the 1 ps pulse at the focus is compared with and without SPM. Clearly, the pulse is significantly shortened and maintains its short duration in a large space range, and its spatial fidelity is well maintained. The rest of
Figure 3. Output/input pulse duration ratio as a function of the time shift parameter $\eta$ and the phase shift parameter $\mu$ for a $\tau = 1$ ps Gaussian pulse for $\zeta = 1$. The two dotted lines are $\eta\mu = 2\pi$ and $3\pi$. Calculation for pulse duration ranging up to a few nanoseconds gives identical distributions.

Figure 4. (a) On-axis laser pulse envelope as a function of the distance from the focus and temporal and spatial distributions of the focus for SPM shortened pulse (b) and a pulse without SPM effect ($n_2 = 0$) (c). The calculation assumes an $f_0 = 150$ mm lens with $P = 12$ mm and $d = 5$ mm, $\mu = 15$ at a laser intensity of $5 \times 10^{11}$ W cm$^{-2}$. The pulse is shortened to about 20% the input pulse duration and retains about 20% of the total pulse energy.

the pulse is ‘scattered’ away from the focal region due to the wavefront distortion. In the case shown in figure 4, the shortened pulse retains about 22% of the total pulse energy, proportional to its pulse duration.

It should be mentioned that the modulated phase can also be acquired through cross-phase modulation (XPM) when multiple laser beams overlap in time and space in the same medium, and can be more severe than SPM.

Note that equation (18) is valid only when $\Lambda^2 \ll 1$ for both the input pulse and the pulse with the modulated phase, this eventually limits the initial pulse duration, the medium length and the laser pulse intensity. The laser intensity is in addition limited by the damage threshold of fused silica, which is a few times of $10^{12}$ W cm$^{-2}$ [33, 34]. For the examples in this paper, the validity is checked by solving the NLSE [30] numerically.
This pulse shortening due to SPM is only one of the examples of how dispersion can affect the performance of an optical lens and needs to be carefully examined for many applications involving manipulating intense UV beams, such as focusing the multi-kilojoule UV laser beam into a holrum in inertial confinement fusion experiments and in shaping and delivering a high-quality UV pulse for a modern photoinjector, which we will discuss below. In these applications, the modulated phase may accumulate during the laser transport and frequency conversion. The remedy is to use achromatic optics when possible, which has been shown to be effective in mitigating the pulse lengthening effect. On the positive side, in laser plasma experiments, it is possible that the phase slip between the pulse slices is large enough so that the final pulse becomes the sum of a set of quasi-random phases, which can be useful in suppressing certain plasma instabilities.

3.2. Pulse engineering: a quasi-light ellipsoid

3.2.1. Numerical results. A practical example of the spatiotemporal shape is the uniform ellipsoidal (UE) pulse desired for modern photoinjectors [21, 23, 35] to generate high-brightness electron beams. Such a beam profile provides the possibility of maximizing the beam brightness with high efficiency suitable for more cost-effective x-ray free-electron lasers and other electron beam-based light sources [11–13]. The envelope of the electron beam is defined by an ellipsoid surface.

\[ R(t) = R_0 \sqrt{1 - \left(\frac{t}{\Theta}\right)^2}. \]  

(21)

Here 2\(\Theta\) is the full temporal width and \(R_0\) the maximum radius and \(t \leq \Theta\). Within the envelope, the electron distribution should be homogeneous, and outside the envelope, there should be no electrons at all. The scheme for generating such an electron distribution via dynamic self-evolution [36, 37] is limited to low charge cases [25]; thus to generate an ellipsoidal laser beam remains the ultimate solution and challenge.

To generate an ellipsoidal envelope described by equation (21), substitute \(w(t)\) with \(R(t)\) in equation (5); then the desired phase becomes

\[ \phi(t) = -\omega_0 t \pm \frac{\Delta \omega}{2} \left[ t \left( 1 - \left( \frac{t}{\Theta} \right)^2 \right)^\alpha + \Theta \sin^{-1} \left( \frac{t}{\Theta} \right) \right], \]  

(22)

where \(\alpha = 1/2\), and \(\Delta \omega = (n_0 - 1)N R_0/\beta f_0\) is the maximum frequency shift. To keep the laser flux \(|a(t)|^2 / R(t)^2\) constant over time, we have

\[ a(t) = a_0 \left[ 1 - \left( \frac{t}{\Theta} \right)^2 \right]^{\xi}, \]  

(23)

with \(\xi = 1/2\). Equations (22) and (23) describe a pulse that can form a spatiotemporal ellipsoid at the focus of a singlet lens.

As mentioned earlier, the Gaussian optics method is used to obtain equations (5) and (6); thus equations (22) and (23) do not treat the effects of diffraction due to beam apodization and the time domain effect of dispersion described in section 3.1. These effects are numerically evaluated using a Fourier optics model described by equations (7) and (8).

In fact, the GVDE and diffraction effects prevent us from generating a perfect UE pulse, thus \(\alpha\) and \(\xi\) in equations (22) and (23) are adjusted for better emittance in accordance with the particle simulation [25]. The time and frequency domain representations of a pulse with
Figure 5. (a) Time and (b) frequency domain representation (solid line: intensity; dashed line: phase), and (c) the spatiotemporal intensity distribution of a laser pulse that gives an excellent emittance performance in beam simulation ($\alpha = 1/2$ at $t < 0$, $\alpha = 1$ at $t \geq 0$, and $\xi = 1/2$ in equations (22) and (23)). The pulse has a 5% full bandwidth at 249 nm (about 1% FWHM). A $P = 25 \text{ mm}$ and $f_0 = 150 \text{ mm}$ fused silica lens is used. The spatiotemporal distribution in (c) represents the laser pulse to be applied to the cathode.

Figure 6. Numerically calculated spatiotemporal laser profiles with different input bandwidths with $P = 25 \text{ mm}$ input beam (flat top) using the same $\alpha$ and $\xi$ as in figure 5(c). From left to right: $\Delta \omega/\omega = 8$, 4, 2, 1 and 0.5%. Note the different scales in $r$ as noted in each panel.

Excellent emittance performance are shown in figures 5(a) and (b), with $\Theta = 6 \text{ ps}$, $\lambda_0 = 0.25 \mu \text{m}$ and $\Delta \omega/\omega = 8\%$. The spatiotemporal flux at the focal plane of an $f_0 = 150 \text{ mm}$ fused silica lens is given in figure 5(c). In particle tracking simulation, the emittance performance of the beam was found to closely mimic that of an ideal ellipsoidal beam [25, 38].

3.2.2. Physics limitations. The intensity in figure 5(c) displays the basic features of a UE pulse but with noticeable distortions, which also highlights the limitation of how the scheme can be applied. The distortion has three components representing three different physics limitations, which can be more clearly observed in the dependence of the spatiotemporal profile on the laser and lens parameters in figures 6 and 7. Figure 6 shows simulated spatiotemporal profiles of the laser pulse as a function of the laser bandwidth $\Delta \omega/\omega = 8$, 4, 2, 1 and 0.5% with a lens radius of $P = 25 \text{ mm}$. Figure 7 shows profiles as a function of the lens radius $P$ at a fixed bandwidth $\Delta \omega/\omega = 8\%$.

The first limitation is the GVDE shown in equation (14), which generates the prominent recess in the leading edge and the protrusion at the trailing edge. This is due to the group delay between rays traversing the lens at different radii, with the maximum delay determined by the
Figure 7. Numerically calculated spatiotemporal laser profiles with different input beam sizes (flat top) using the same $\alpha$ and $\xi$ as in figure 2(c). From left to right: $P = 25, 12, 6, 4$ and 2 mm. Note the different scales in $r$ as noted in each panel. $\Delta \omega/\omega = 8\%$.

The second limitation is the fringes caused by diffraction, mostly clear at smaller beam apertures. The detail of the diffraction structure depends on the laser bandwidths and other factors (figures 6 and 7). Even in this application the effect is detrimental and needs further analysis; this complicated diffraction pattern can provide another dimension for spatiotemporal control of the pulse. Using apodization to manipulate the depth of focus was explored decades ago [39] and the effect of apodization of an ultrafast laser pulse remains an interesting research topic [40].

The third limitation is due to the fact that the phase in equation (22), although apparently complex, is dominated by the common time-dependent third-order phase. This is discussed in section 3.1, where temporal modulation due to superposition of the field will occur, thus limiting the homogeneity of the intensity distribution.

As expected, as the bandwidth decreases, the maximum radius of the beam decreases proportionally, and the diffraction structure becomes more dominant. Smaller beam sizes also make diffraction more dominant. Although the effect on the beam needs to be evaluated further, it is clear that larger beam size and larger bandwidth are preferred. Various bandwidth broadening methods can be utilized for generating laser pulses with large bandwidth and one of them can be the recently proposed flying plasma mirror compression scheme [41].

It should also be mentioned that the 3D distribution is not a pulse that can propagate in free space, but a spatiotemporal distribution at a certain plane that mimics such a pulse. The 3D distribution can in general be image-relayed using achromatic optics to maintain the temporal–spatial fidelity, and the associated dispersion can be pre-compensated, as can be seen in the proof-of-principle experiment in the next section.

3.2.3. Experiment and results. The phase in equation (22) is dominated by the common third-order phase that can be generated via SPM/XPM and is exploited in various laser applications, in particular in a few cycle pulse generation for the large bandwidth it generates during propagation. For a precise control, one of the practical solutions is the AOPDF [6]. AOPDF uses the transient Bragg effect in a crystal induced by an acoustic wave to manipulate the phase and amplitude of a laser pulse.

A proof-of-principle experiment to demonstrate the dual time–space control is carried out. A schematic diagram of the experiment is shown in figure 8. A pair of Pockels cells is used...
Figure 8. Schematic diagram of the experiment. Key: PP: pulse picker; D: DAZZLER; SF: achromatic spatial filter; ZSL: ZnSe lens; AL: achromatic image relay lens; ODL: optical delay line; C: camera; I: iris.

to reduce the repetition rate of a Ti:sapphire oscillator from 90 MHz to 1 kHz. The 40 nm bandwidth pulse is stretched to 135 fs after the Pockels cells. It is split into two arms. One traverses a delay line to serve as a probe beam. The other, denoted as the main beam, is sent through an AOPDF and is modulated in phase and amplitude. It is then spatially filtered to generate a Gaussian beam using a pair of achromatic lenses and a pinhole. A plano-spherical ZnSe lens (25 mm diameter, 88.9 mm radius of curvature and 2.9 mm centre thickness, Janos Technology, A1204-105) is used for its high dispersion (250 fs$^2$ mm$^{-1}$ at 800 nm) to form the desired spatiotemporal distribution at its focal plane. The focal plane is image relayed by an achromatic lens onto a CCD camera to interfere with the probe beam. The interference fringes as a function of delay between the two beams are recorded on a 12 bit camera and are used to extract the spatiotemporal intensity distribution of the main beam. The imaging system is aligned to focus at 845 nm, accomplished by generating an 845 nm beam via the AOPDF.

For the delay scan, the AOPDF is set up with $\Theta = 1$ ps and a full bandwidth from 845 to 790 nm. The spectrum modulation function is calculated using the native spectrum of the laser to generate those specified by equations (22) and (23). At the focus of the ZnSe lens, this pulse is expected to generate a tightly focused spot at the beginning and end of the pulse, but be defocused between the ends. Unless otherwise specified, all the second-order dispersion in the optics, including the third- and fourth-order dispersion in the AOPDF crystal, is cancelled by properly setting the AOPDF. The calculated amplitude and phase in the time and frequency domains are given in figures 9(a) and (b), together with a spectrum measured in the experiment. Although the measured spectrum closely matched the theoretical one, some deviation is evident and is expected due to the limited crystal length and slightly nonlinear response across the spectrum of the AOPDF. The transverse beam profile is given in figure 9(c) with a 1/e$^2$ radius of 6 mm. To avoid potential saturation effect of the AOPDF, the power level is set at 20%.

Figure 10 shows the spectrum at several power settings of the AOPDF where the variation is clearly visible.

To extract the spatiotemporal intensity of the main beam, we start with the signal recorded on the camera:

$$I(r) = I_m(r) + I_p(r) + 2 \cos(\omega[\tau + \delta(r)])$$

$$\times \int a_m(t, r) a_p(t - \delta(r) - \tau, r) \cos[\psi_m(t) - \phi_p(t - \delta\psi(r) - \tau)] \, dt,$$

(24)
Figure 9. Laser pulse amplitude $a$ (bold solid lines) and phase $\phi$ (dashed lines) calculated from equations (5) and (6) for $\alpha = \xi = 1/2$ in the time (a) and frequency (b) domains, and the measured spectrum amplitude (thin solid line in (b)). The transverse profile of the laser pulse after the spatial filter and before the ZnSe lens is shown in (c) with a slight asymmetry.

Figure 10. (a) Variation of measured spectra as functions of AOPDF settings and (b) the detail of (a) from 790 to 810 nm. The target spectrum is the same as in figure 9(b), with power level of AOPDF adjusted as noted in the figure. The dark green curve is the target spectrum.

where $a(t, r), \psi(r)$ and $I$ are the amplitude, phase and integrated intensity of the laser beams; the subscripts m and p denote the main and probe beam, respectively; $\tau$ is the delay; and $\delta \psi(r)$ is the phase variation due to the angle between the two laser beams. The phase term in the integral, although impossible to evaluate for each location, only causes the interference fringes at the detector to shift. Therefore, if the probe pulse is much shorter than the main pulse, equation (24) can be reduced to

$$I(r) \approx I_m(r) + I_p(r) + 2 \cos (\omega[\tau + \delta(r)]) \sqrt{\Delta t_p i_m(\tau, r)} \sqrt{I_p(r)}.$$

Here $\Delta t_p$ is the duration of the probe pulse, and $i_m$ is the time-dependent intensity distribution. The second term describes the fringes as functions of delay and location, from which one can extract the contrast ratio $C(\tau, r)$,

$$C(\tau, r) = \frac{4 \sqrt{\Delta t_p i_m(\tau, r)} \sqrt{I_p(r)}}{I_m(r) + I_p(r)}.$$
Figure 11. Measured (top row) and simulated (middle row) spatiotemporal distributions with different linear chirp in the main beam, and the intensity as a function of time at \( r = 0 \) (bottom row; measured: bold lines; simulated: thin lines). Striations in the experiment data are due to the fluctuation of the laser pointing.

which can also be measured in the experiment, and in turn gives

\[
i_m(\tau, r) \propto C^2(\tau, r) \frac{[I_m(r) + I_p(r)]^2}{I_p(r)},
\]

(26)

where \( I_p(r), I_m(r) \) can also be measured independently in the experiment.

Two sets of experiment data were presented. In the first set, while maintaining the spectrum, we control the linear chirp of the main pulse using the AOPDF. Due to the specific phase of the pulse, this change will shift the ‘waist’ (the fattest part of the spatiotemporal distribution of the beam) in time. A comparison is given in figure 11 between the experiment measurement and simulation with linear chirp set at different values from the fully compensated case. Other than the striations due to shot-to-shot laser fluctuation, the agreement is excellent. The input beam is a Gaussian beam with a 1/e² width of 3.9 mm. No aperture is used in this part of the experiment.

The Fourier model also predicts that the fine structure of the beam is highly sensitive to the beam apodization as shown in figures 6 and 7. This is measured using a beam with 1/e² width of 6 mm. In the measurement an iris located directly in front of the ZnSe lens is adjusted to different sizes. The measured spatiotemporal intensity distributions are given in the top row of figure 12. The corresponding distributions from the Fourier model are given in the middle row of figure 12. The second-order dispersion is set at zero. An iso-intensity surface plot comparison is given in figure 13 for the iris radius \( P = 3 \) mm case.

As predicted by the Gaussian beam optics, the pulse shows generally an ellipsoidal envelope, but with dramatic variation in the internal structure due to diffraction at the iris. The diffraction pattern changes as a function of time due to both the changing wavelength and the changing focusing condition. With larger aperture size, the internal structure acquires higher and higher spatial frequency and eventually flattens out, as shown in figures 5–7.
Figure 12. Measured (top row) and simulated (middle row) spatiotemporal intensity distributions with different iris radii $P$ using the experiment condition. The bottom row shows a comparison of the intensity at $r = 0$ extracted from the top and middle rows (measured: bold lines; simulated: thin lines).

Figure 13. Cut-away view along the $t$–$r$-plane of the measured (right) and calculated (left) spatiotemporal iso-intensity surface plot of the $P = 3$ mm case in figure 12.

Although the agreement between the simulation and experiment is generally good, several discrepancies can be noticed. The first is that better agreement between experiment and calculation is achieved at small aperture sizes. This can be partially attributed to the limited dynamic range of the probing system, which makes the extraction of signals difficult at low-intensity wings of the distribution. In addition, the measurement suffers from the pointing stability of the laser, which causes shot-to-shot fluctuation of both beams and thus fluctuation of the measured intensity.

The temporal resolution of the measurement is limited by the probe pulse duration at about 130 fs; a shorter probe pulse would demand a higher dynamic range for data recording.
4. Conclusion

We discussed the well-known effect of chromatic dispersion of a singlet lens on the spatial–temporal behaviour of an ultrafast laser pulse. A pulse engineering scheme exploiting these chromatic aberrations is analysed with an example of generating quasi-ellipsoidal pulses. A proof-of-principle experiment was carried out, with results confirming the theoretical and numerical models. Further investigation is planned to establish the adaptive control as well as preservation of the phase in frequency conversion. We also discussed the limitation of the shaping scheme as presented by the inherent physics that enables this shaping method, i.e. the chromatic dispersion.

Acknowledgments

We thank K-J Kim and K Harkay for support. This work was supported by the US Department of Energy, Office of Science, Office of Basic Energy Sciences, under contract no. DE-AC02-06CH11357. BS is also supported by the 973 Program (project no. 2006CB806000), the National Natural Science Foundation of China (project nos 10834008 and 60921004), the Shanghai Natural Science Foundation (10ZR1433800) and the Program of Shanghai Subject Chief Scientist (09XD1404300).

© US Government.

References

[1] Dickey F M and Holswade S C 2000 Laser Beam Shaping: Theory and Techniques (New York: Marcel Dekker)
[2] Hoffnagle J A and Johnson C M 2000 Design and performance of a refractive optical system that converts a Gaussian to a flattop beam Appl. Opt. 39 5488–99
[3] Zhang S, Neil G and Shinn M 2003 Single-element laser beam shaper for uniform flat-top profiles Opt. Express 11 1942–8
[4] Weiner A M 1995 Femtosecond optical pulse shaping and processing Prog. Quantum Electron. 19 161–237
[5] Weiner A M 2000 Femtosecond pulse shaping using spatial light modulators Rev. Sci. Instrum. 71 1929
[6] Verluise F, Laude V, Cheng Z, Spielmann Ch and Tournois P 2000 Amplitude and phase control of ultrashort pulses by use of an acousto-optic programmable dispersive filter: pulse compression and shaping Opt. Lett. 25 575–7
[7] Vaughan J C, Feurer T and Nelson K A 2002 Automated two-dimensional femtosecond pulse shaping JOSA B 19 2489–95
[8] Nuss M C and Morrison R L 1995 Time-domain images Opt. Lett. 20 740–2
[9] Piestun R and Miller D A B 2001 Spatiotemporal control of ultrashort optical pulses by refractive–diffractive–dispersive structured optical elements Opt. Lett. 26 1373–5
[10] Hill K B, Purchase K G and Brady D J 1995 Pulsed-image generation and detection Opt. Lett. 20 1201–3
[11] Brinkmann R, Materlik G, Rossbach J and Wagner A 1997 Conceptual design of a 500 GeV e+e linear collider with integrated x-ray laser facility DESY Report No. DESY97-048 (Hamburg: Deutsches Elektronen-Synchrotron)
[12] Cornacchia M et al 1998 Linac coherent light source (LCLS) design study report SLAC Report No. SLAC-R-521 (Stanford, CA: Stanford Linear Accelerator Center)
[13] Gruner S M, Bilderback D, Bazarov I, Finkelstein K, Krafft G, Merminga L, Padamsee H, Shen Q, Sinclair C and Tigner M 2002 Energy recovery linacs as synchrotron radiation sources Rev. Sci. Instrum. 73 1402
[14] Zewail A H 2010 Four-dimensional electron microscopy Science 328 187
[15] Sider C 1998 Pulse-train generation using a 2n-pulse Michelson interferometer Appl. Opt. 37 5302
[16] Tomizawa H, Dewa H, Taniuchi T, Mizuno A, Asaka T, Yanagida K, Suzuki S, Kobayashi T, Hanaki H and Matsui F 2006 Adaptive shaping system for both spatial and temporal profiles of a highly stabilized UV laser light source for a photocathode RF gun Nucl. Instrum. Methods Phys. Res. A 557 117–23
[17] Dromey B, Zepf M, Landreman M, O’Keeffe K, Robinson T and Hooker S M 2007 Generation of a train of ultrashort pulses from a compact birefringent crystal array Appl. Opt. 46 5142–6
[18] Will B I and Klemz G 2008 Generation of flat-top picosecond pulses by coherent pulse stacking in a multicrystal birefringent filter Opt. Express 16 14922–37
[19] Bazarov I V, Ouzounov D G and Dunham B M 2008 Efficient temporal shaping of electron distributions for high-brightness photoemission electron guns Phys. Rev. ST Accel. Beams B 11 040702
[20] Sharma A K, Tsang T and Rao T 2009 Theoretical and experimental study of passive spatiotemporal shaping of picosecond laser pulse Phys. Rev. ST B 12 033501
[21] Brady D and Psaltis D 1992 Control of volume holograms J. Opt. Soc. Am. A 9 1167
[22] Li Y and Crowell R 2007 Shortening of laser pulse with self modulated phase at the focus of a lens Opt. Lett. 32 93–5
[23] Li Y and Chemerisov S 2008 Manipulation of spatiotemporal photon distribution via chromatic aberration Opt. Lett. 33 1996–8
[24] Bor Z 1989 Distortion of femtosecond laser pulses in lenses Opt. Lett. 14 119–21
[25] Kompe M, Stamm U, Wilhelmi B and Rudolph W 1992 Spatial and temporal transformation of femtosecond laser pulses by lenses and lens systems J. Opt. Soc. Am. B 9 1158–65
[26] Born M and Wolf E 2003 Principles of Optics (Cambridge: Cambridge University Press)
[27] Agrawal G P 1995 Nonlinear Fiber Optics (New York: Academic)
[28] Taylor A J, Rodriguez G and Clement T S 1996 Determination of n2 by direct measurement of the optical phase Opt. Lett. 21 1812–4
[29] Goodman J W 1985 Statistical Optics (New York: John Wiley)
[30] Tien A, Backus S, Kapteyn H, Murnane M and Mourou G 1999 Short-pulse laser damage in transparent materials as a function of pulse duration Phys. Rev. Lett. 82 3883–6
[31] Limborg-Deprey C and Bolton P 2006 Optimum electron distributions for space charge dominated beams in photoinjectors Nucl. Instrum. Methods Phys. Res. A 557 106–16
[32] Luiten O J, van der Geer S B, de Loos M J, Kiewiet F B and van der Wiel M J 2004 How to realize uniform three-dimensional ellipsoidal electron bunches Phys. Rev. Lett. 93 094802
[33] Veeveti S P, Vijayan C D, Sharma K, Schimmel H and Wyrowski F 2006 Diffraction induced space-time splitting effects in ultra-short pulse propagation J. Mod. Opt. 53 1819–28
[34] Li Y, Chemerisov S and Lewellen J W 2009 Laser pulse shaping for generating uniform three-dimensional ellipsoidal electron beams Phys. Rev. ST Accel. Beams B 12 020702
[35] Welford W T 1960 Use of annular apertures to increase focal depth J. Opt. Soc. Am. 50 749–52
[36] Ji L L, Shen B F, Li D X, Wang D, Leng Y X, Zhang X M, Wen M, Wang W P, Xu J C and Yu Y H 2010 Relativistic single-cycled short-wavelength laser pulse compressed from a chirped pulse induced by laser-foil interaction Phys. Rev. Lett. 105 025001

New Journal of Physics 12 (2010) 123011 (http://www.njp.org/)