Generation of ultra-intense single-cycle laser pulses by using photon deceleration

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A scheme to generate single-cycle laser pulses is presented based on photon deceleration in underdense plasmas. This robust and tunable process is ideally suited for lasers above critical power because it takes advantage of the relativistic self-focusing of these lasers and the nonlinear features of the plasma wake. The mechanism is demonstrated by particle-in-cell simulations in three and 2½ dimensions, resulting in pulse shortening up to a factor of 4, thus making it feasible to generate few-femtosecond single-cycle pulses in the optical to IR domain with intensities $I > 10^{20}$ W/cm² by using present-day laser technology.

The quest for attosecond laser pulses is at the forefront of research in laser physics (1–3). Pulses in the attosecond range may give rise to the development of attoelectronics, making it possible to study the dynamics and to control electronic processes in biology, chemistry, and solid-state physics, in the same way femtosecond laser technology led to femtochemistry (1). On the other hand, state-of-the-art ultra-intense lasers can deliver up to 1 PW, with pulse durations from 500 fs down to 18 fs, at 800 nm to 1 μ m (4). Two paths toward attosecond pulses can be identified; the first one, associated with solid-state laser oscillator technology (5), has pushed the limit of the shortest laser pulse down to 4.5 fs in the near-IR to visible domain. At these wavelengths, breaking the attosecond threshold implies the generation of subcycle pulses (6, 7). The other path is based on the careful combination of some of the short wavelength harmonics generated in the ionization of a rare gas by intense femtosecond laser pulses (8), leading to 100-as extreme UV pulses (3). The possibility of producing even shorter single-cycle, ultra-intense pulses opens the way to new unexplored physics and the possibility of generating ultra-intense attosecond pulses (3).

Current methods for ultra-short pulse generation and compression already push the limits of the linear and nonlinear optics of conventional materials (5). Further developments on ultra-intense lasers must then be based on the nonlinear optics of plasmas (the medium capable of handling high-power densities and heat loads) at relativistic intensities (9). An example is, for instance, the plasma equivalent of the optical parametric amplifier (10), recently introduced by Shvets *et al.* (11).

In this article, we propose a method to further shorten the existing shortest pulses to ultra-intense single-cycle pulses. This method is based on the frequency downshift (or photon deceleration) experienced by a laser pulse in a plasma because of the combined self-interaction with the relativistic mass nonlinearity and the laser wake field (12). The photon frequency downshift is accompanied by the conservation of the total wave action, leading to a strong enhancement of the laser field vector potential (13). Relativistic self-focusing also provides an additional amplification of the peak laser field. Using three-dimensional (3D) and two-dimensional (2D) particle-in-cell (PIC) simulations we find that this method works for a wide range of parameters for pulse widths, laser frequencies, laser intensities, and plasma densities. The method is general and robust because the plasma density can be tuned to generate pulses at a wide range of frequencies and pulse durations. Although previous works (6, 7) on the generation of single-cycle

radiation have relied on one-dimensional (1D) theory and numerical solutions of the Maxwell-Bloch equations, we use full-scale 3D PIC simulations to demonstrate our method.

A plasma can be used in several ways to compress a pulse. To illustrate these possible methods, we start from the 1D quasi-static equations (14) for a laser and the wake it creates

$$-2\partial_{\xi}\partial_{\tau}a = \frac{a}{\chi}, \quad \partial_{\xi}^2\chi + \frac{1}{2}\left[1 - \frac{1+a^2}{\chi^2}\right] = 0, \quad [1]$$

where $a = eA/mc^2$ is the normalized vector potential, $\xi = \omega_p(t - x/c)$, $\tau = \omega_p x/c$, are the normalized speed of light frame variables, and $1/\chi$ is the plasma susceptibility. For long pulses, the susceptibility can be approximated as $1/\chi = (1 + |a|^2/2)^{-1/2}$. In this limit, the pulse undergoes relativistic self-phase modulation, causing a local change in the laser frequency of the form, $\partial_{\tau}\hat{\omega} = -1/(2\hat{\omega})\partial_{\xi}1/\chi$ (12, 15, 16), where $\hat{\omega} \equiv \omega/\omega_p$ is the laser frequency normalized to the plasma frequency, $\omega_p = (4\pi e^2 n_{e0}/m_e)^{1/2}$. This results in a positive frequency chirp (the front is downshifted while the back is up-shifted) and the negative group velocity dispersion (17) ($dv_g/d\lambda < 0$) of the plasma leads to pulse compression. This mechanism would be the plasma equivalent of pulse compression in optical fibers by the soliton effect (18–20). However, in reality a long pulse still creates a plasma wave wake that modulates the susceptibility, and hence the index of refraction, $\eta = [1 - (\hat{\omega}^2\chi)^{-1}]^{1/2}$, at the plasma frequency. This leads to the pulse breaking up into beamlets spaced at the plasma wavelength caused by Raman forward scattering type instabilities (9). To avoid this, another possibility would be to start with a short pulse, $\sim \lambda_p$ long, and try to compress it. This is illustrated in Fig. 1*a* where numerical solutions to Eq. 1 are plotted for a pulse length $L_0 = \lambda_p$ (we define L_0 as the full width at half-maximum of a) and a peak value of $a \equiv a_0 = 0.5$ (we use linearly polarized light). As shown, the pulse completely sits inside the first oscillation of the wake. As before, the front of the pulse will be downshifted while the back of the pulse will be upshifted. Eventually, the pulse will compress, leading to the case in Fig. 1*b* where the numerical solutions to Eq. 1 are shown for a pulse for which $L_0 = \lambda_p/2$. In this case the pulse completely resides in a negative index of refraction gradient (Fig. 1*b*), and the whole pulse is downshifted. It should be noted that in any scheme that relies on the wake to compress a pulse, the pulse is gradually losing energy as well. As a result, although pulse compression occurs, the overall energy in the pulse decreases as well. We have found that by simply using a linear wake pulse compression over Rayleigh length distances cannot be achieved nor can it lead to single-cycle pulses.

However, if the pulse intensity is increased the index of refraction gradient increases dramatically. This is illustrated in Fig. 1*c* and *d* where a_0 is increased from 0.5 to 1.4 and to 4. This

Abbreviations: 3D, three-dimensional; 2D, two-dimensional; 1D, one-dimensional; PIC, particle-in-cell.

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Fig. 1. Normalized absolute vector potential of laser pulse ($\omega_0 = 5$) and the plasma susceptibility χ^{-1} as determined from the quasi-static equation for χ : (a) $L_0 = \lambda_p$, $a_0 = 0.5$, while $L_0 = \lambda_p/2$ for (b) $a_0 = 0.5$, (c) $a_0 = 1.4$, and (d) $a_0 = 4.0$.

leads to a strong frequency downshift of the pulse front on a time scale t_{down} much faster than the compression time, t_{comp} , of the pulse because of the strong group velocity variation along the pulse: the front is strongly downshifted to almost zero wave-number, and it quickly falls behind the rest of the laser pulse. In this manner, the front of the pulse is etched away by the strong frequency downshift, whereas the main part of the pulse is continuously downshifted, or photon decelerated, at a lower rate.

The laser pulse's energy decreases as the wake is generated but the total number of photons $N\gamma$, or equivalently, the classical action of the electromagnetic wave, is conserved. Photon deceleration will continue until the laser central frequency ω approaches a few ω_p , at which time only a single cycle of radiation is left. $N\gamma$ can be written as $N\gamma \propto a^2 \omega L_0 W^2$, where L_0 is the pulse duration, and W is the laser spot size. Because the number of photons $N\gamma$ is conserved and ω is decreasing then a increases: this mechanism acts as an amplifier for the vector potential. These qualitative arguments were first given in ref. 13.

These qualitative arguments can be supplemented by semi-quantitative estimates for important time scales based on the quasi-static equations for the laser and the plasma wake (14). We first assume that the local frequency along the pulse can be determined by the linear dispersion relation $\hat{\omega}(\xi, \tau) = \sqrt{\hat{k}^2 + 1/\chi}$ where $\hat{\omega} = \omega/\omega_{p0}$, and $\hat{k} = kc/\omega_{p0}$. The maximum frequency rate of change can then be calculated from the condition $\partial_{\xi}^2 [1/\chi(\xi = \xi_M)] = 0$ along with the equation for χ (Eq. 1). Exact analytical results can be obtained for a square-shaped pulse, for which the maximum gradient is $\Theta(a_0) = (\partial_{\xi} 1/\chi)_{\text{max}} = (1/\chi_M^2)(a_0^2 + 2 - \chi_M - (1 + a_0^2/\chi_M)^{1/2})$, where $\chi_M = \chi(\xi_M) = 2/3(a_0^2 + 1) - 1/3(4a_0^4 + a_0^2 + 1)^{1/2}$ is evaluated at the position of maximum gradient $\xi = \xi_M$. The local frequency at $\xi = \xi_M$ evolves as $\hat{\omega}^2 \approx \hat{\omega}_0^2 - t\Theta$. The photon deceleration time t_{down} is then defined as the time necessary for the frequency at the front of the pulse to decrease to $\omega/\omega_{p0} \leq 2$: $t_{\text{down}} = (\hat{\omega}_0^2 - 2)/\Theta(a_0)$, which scales as a_0^{-1} for $a_0 \gg 1$. Examining Fig. 1 c and d, it is clear that the very front part of the pulse is propagating at the linear group velocity in the plasma (because $1/\chi \approx 1$), while the back of the pulse is propagating in an almost vacuum-like region where $1/\chi \ll 1$. The compression time is the time necessary for the back of the pulse to reach the front of the pulse $t_{\text{comp}} \approx \hat{\omega}_0^2 \bar{L} \sqrt{a_0^2 + 1/\sqrt{a_0^2 + 1} - 1}$, where the velocity of the back of the pulse was assumed to move at the nonlinear group velocity (21), and $\bar{L} = L_0 \omega_p/c$. We will refer back to these expressions shortly. We also note that the typical time for linear dispersion of the pulse scales as $t_{\text{disp}} \approx 1/2\hat{\omega}_0^3 \bar{L}^2$ (22, 23), much longer than t_{comp} for typical parameters of ultra-intense $a_0 > 1$, short pulses $\bar{L} \approx \pi$.

The actual scenario is highly nonlinear and multidimensional. A comprehensive theory is therefore difficult to obtain so we rely on full-scale, fully explicit PIC simulations. We used OSIRIS[§],

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Fig. 2. Time evolution of the normalized laser electric field, $eE/(mc\omega_p)$ for two laser amplitudes (a–c) and (d–f).

which is an object-oriented, parallel, electromagnetic PIC code. The simulations are performed in both 2+1/2D (two dimensions in real space and three dimensions in momentum space) and 3D (three dimensions in both real and momentum space). This code uses a moving window that moves at the speed of light, so that in all figures the laser is nearly stationary.

We begin by self-consistently illustrating the generation of single pulses with 1D simulations. In these 1D simulations, the frequency ratio is 5, the laser pulse length is $L_0 = \lambda_p/2$, and the peak amplitude is either 1.4 or 4.0. We find from these 1D simulations that $a_0 \gg 1$ for this scheme to work. This is shown in Fig. 2, where a sequence of the laser electric field is presented: for $a_0 = 4$ (Upper), the front of the pulse is strongly downshifted, and the resulting very long wavelength quickly falls behind the main part of the pulse (Fig. 2c) while the rear of the pulse is compressed. For lower intensities (Fig. 2 Lower), the downshift of the front of the pulse is not strong enough, and the downshifted part of the pulse does not slip back fast enough, thus resulting in a stretched pulse with a negative chirp (Fig. 2f). These simulations therefore indicate that t_{down} needs to be $< t_{\text{comp}}$ for pulse compression and single-cycle generation. Using the expressions for a square pulse and assuming $\bar{L} = L_0 \omega_p/c = \pi$, and $\hat{\omega}_0 = 5$, implies that $a_0 > 3$ so that $t_{\text{down}} < t_{\text{comp}}$. We

Fig. 3. Snapshots of the laser's electric field for the short-pulse case. The isosurfaces are taken at $\pm 0.1 a_0(\omega_0/c)$.

Fig. 4. Spectrum of the laser electric fields for a short-pulse 2+1/2D run: (a) for $t = 0$, and (b) for $t = 70.0/\omega_p$ (x is the propagation direction).

therefore, use $a_0 = 4$ for most of the 2+1/2D and 3D simulations to be discussed next.

Multidimensional simulations are important because one might expect that each transverse slice of the pulse might evolve similarly to a 1D simulation with the corresponding value of $a_0(x_\perp)$. If this were the case the pulse would have large phase variations across the wave front. Furthermore, new effects occur in 2D and 3D such as self-focusing and ponderomotive blow-out. In a typical 3D run, the system size is $480 \times 160 \times 160$ cells (the physical size of the 3D simulation is $30c/\omega_p \times 40c/\omega_p \times 40c/\omega_p$), with eight particles per cell; while in a typical 2D simulation the system size is 750×160 cells. The physical size of the 2D simulations is typically $45c/\omega_p \times 80c/\omega_p$. Although the particles are only loaded in the region $5c/\omega_p < (y,z) < 35c/\omega_p$, and the 3D system still has more than 55 million particles. The cell size was always $\leq 0.3c/\omega_0$ in the propagation direction.

Fig. 5. Time evolution of the laser fields in the longer-pulse case. (a–c) The laser electric field at $t = 0\omega_p^{-1}$, $t = 70\omega_p^{-1}$, and $t = 140\omega_p^{-1}$. (e and f) The vector potential at the same times as b and c, respectively. (d) The compressed vector potential (at $t = 70\omega_p^{-1}$) in the short-pulse case. (g–i) The center lineouts of d–f, respectively.

Fig. 6. Snapshots of the laser's vector potential for the longer-pulse case.

The multidimensional simulations confirm the qualitative behavior observed in the 1D simulations: for $a_0 > 3$, and $\hat{\omega}_0$ between 5 and 7, the photon deceleration mechanism still compresses the pulse to a single cycle. This is illustrated in the 3D results of Fig. 3 ($a_0 = 4$, $\hat{\omega}_0 = 5$, $\hat{L} = \pi$, $W_0 = 7.6c/\omega_p$), where a pulse initially with five cycles is compressed to a single cycle after propagating ≈ 0.3 Rayleigh lengths. An important reason that the scheme still works in multidimensions is that at high intensities and for the chosen spot sizes, the transverse ponderomotive force expels the electrons, creating a well-defined cavity in which the trailing part of the pulse resides.

It is also of interest to examine the evolution and the formation of a single-cycle pulse in k -space because it provides signatures for the fundamental multidimensional features involved in this scheme, i.e., photon deceleration and relativistic self-focusing. To illustrate this, a time sequence of the spectrum of the laser electric field is plotted in Fig. 4, from a 2D simulation with otherwise identical parameters as that in Fig. 3. The main part of the laser spectrum is strongly downshifted from $k_x \approx 5$ to $k_x \approx 3.5$. This downshift is the signature for photon deceleration.

Furthermore, the spectrum of the pulse is broadened in the transverse direction that corresponds to the relativistic self-focusing of the laser ($\Delta k_y \approx \max |k_y|$ goes up). For the parameters in the simulations, the laser power is well above the relativistic self-focusing threshold for almost the entire pulse.

It is instructive to consider the actual physical conditions that meet the normalized parameters used in our simulations; with $a_0 = 4$ and $\hat{\omega}_0 = 5$, for 800-nm pulses, the peak intensity is $\approx 3 \times 10^{19} \text{ W/cm}^2$, and the background plasma electron density is $n_e = 7 \times 10^{19} \text{ cm}^{-3}$, while the plasma period is $\tau_p \approx 13 \text{ fs}$. The transverse width of the laser is $W_0 = 7.6c/\omega_p$, which corresponds to $W_0 \approx 4.8 \text{ }\mu\text{m}$, and the pulse duration is $\tilde{L} = \pi$, or 6.5 fs. Even though such laser parameters might be generated in the near future, state-of-the-art ultra-intense laser pulse durations are in the 15-fs range, corresponding to the normalized pulse duration $\tilde{L} = 7.5$.

Next, we present simulations for $\tilde{L} = 7.5$ to show that photon deceleration can still compress existing pulses. We first stress that for such intensities ($a_0 \gg 1$) the nonlinear plasma wavelength is significantly increased (as shown in Fig. 1 c and d), and a longer ultra-intense pulse with $\tilde{L} \leq (\text{nonlinear plasma wavelength})/2 = \pi a_0/\sqrt{2}$ can still completely reside in a region of negative index of refraction gradient. We begin by showing 2D simulation results for these longer pulses in Fig. 5 a–c). We present the time evolution of the laser electric field from a simulation with the same parameters as in Fig. 3, except that now $\tilde{L} = 7.5$. This sequence clearly illustrates that both self-focusing and pulse compression/shortening occur within relatively short distances, i.e., ≈ 1 Rayleigh length. A compression/shortening factor of 4 is observed.

Furthermore, these 2D simulations clearly show the predicted amplification of the vector potential. In Fig. 5d, we present the

vector potential for the shorter pulse case $\tilde{L} = \pi$, corresponding to the electric field in Fig. 4b, while in Fig. 5 e and f the vector potential for the longer pulse is shown at the same time steps as in Fig. 5 b and c). Both the shorter-pulse scenario and the longer-pulse case lead to a_0 amplification, and to the formation of relatively clean single-cycle pulses, as demonstrated by the lineouts on axis of the vector potential in Fig. 5 g–i). The long wavelength structure observed behind the single-cycle is removed once the pulse is taken out of the plasma because of the very small group velocity associated with their wavelengths.

We conclude by presenting results from a full-scale 3D simulation for a technologically feasible 15-fs laser pulse. The simulation parameters were again $\tilde{L} = 7.5$, $a_0 = 4$, $W_0 = 7.6c/\omega_p$, and $\hat{\omega}_0 = 5$. In Fig. 6, a time sequence of the isosurface contours, $a_0 = \pm 0.1$, and lineouts on axis of the vector potential are plotted. This figure clearly shows that within a propagation distance $\omega_p x/c = 174 = 1.2$ Rayleigh lengths, the pulse is shortened by a factor of ≈ 4 and the peak vector potential increased by a factor ≈ 3 (see Movie 1, which is available as supporting information on the PNAS web site, www.pnas.org). By the end of the simulation the pulse has evolved to nearly a single cycle. These results appear very promising and we hope to further optimize the pulse shortening scheme in future work.

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