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Cascaded chirped photon acceleration for efficient frequency conversion

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A cascaded sequence of photon acceleration stages using the instantaneous creation of a plasma density gradient by flash ionization allows the generation of coherent and chirped ultraviolet and x-ray pulses with independently tunable frequency and bandwidth. The efficiency of the cascaded process scales with $1/\omega$ in energy, and multiple stages produce significant frequency up-conversion with gas-density plasmas. Chirping permits subsequent pulse compression to few-cycle durations, and output frequencies are not limited to integer harmonics. *Published by AIP Publishing.*
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Lasers have historically developed most rapidly at the particular wavelengths for which gain media are readily available. As a result, the highest intensities, most complex waveforms, and largest pulse energies have been, with few exceptions, restricted to visible and near-infrared wavelengths. Plasma-based optical components, including amplifiers,[1–4] mirrors,[5] beam combiners,[6] and waveplates,[7,8] promise to overturn this paradigm by supporting the manipulation of high intensities with continuous scaling in wavelength. Frequency conversion is a particularly important optical technique to replicate in plasma, and has prompted a study of second harmonic generation from plasma mirrors,[9] third harmonic generation in underdense plasma,[10] plasma-based high-order harmonic generation,[11–14] and Doppler upshift from flying mirrors built with plasma surfaces.[15] We show here that flash-ionizing a gas density gradient during the traversal of a short light pulse to create a plasma gradient enables efficient frequency conversion with control over both central frequency and bandwidth. Practical limitations of frequency upshifting using a time-dependent index of refraction can be circumvented by cascading and chirping, permitting highly efficient frequency conversion from infrared to ultraviolet in gas-density media and the production of both broad and narrow bandwidth upshifted pulses.

The recent deployment of a technique for shaping focal spot dynamics with chirped pulses and chromatic focusing optics, i.e., the flying focus,[16–18] offers exquisite control over the evolution of intensity in space and time, allowing frequency manipulation based on controlled ionization. In particular, the flying focus permits the construction of a collinear flash-ionization scheme using a superluminal ionization front,[18] suggesting novel approaches for photon acceleration[19] or enhancing the performance of ionizing Raman amplifiers.[20,21] Previous study of plasma flash-ionization for photon acceleration used either high-voltage DC discharges[22] or ionizing microwave[23] or laser[24–29] pulses. High voltage discharges and ionizing pulses can rapidly produce a reasonably uniform plasma, reducing the index of refraction (n); if this change is homogeneous and fast, the photon wavenumber (k) remains constant and the frequency (ω) increases to maintain $\omega = ck/n$.[30–33] A relativistic ionization front propagating collinearly can further increase photon frequency using the double Doppler effect through temporal refraction or reflection[19,34–38] at the cost of a significant loss of energy. Although experimental upshifts have been demonstrated, substantial frequency conversion at optical wavelengths is limited by the available plasma densities.

In this paper, we propose a method of continuous electromagnetic wave frequency upconversion by cascading multiple flash-ionization stages. In each stage, a plasma density gradient is instantaneously created from a gas-density gradient. Ordinarily, a pulse exiting a plasma will stretch due to the lower group velocity inside a plasma; the use of a plasma density gradient creates a chirp which supports recompression of the upshifted pulse. In a cascaded process, short pulse durations can be maintained while the frequency is substantially upshifted in moderate-density plasmas.

Consider a laser pulse propagating through a transparent media (for time $t < 0$) instantaneously ionized at $t = 0$. The change in index of refraction from $n_0 \approx 1$ to $n_1 = \sqrt{1-(\omega_p/\omega_1)^2}$ shifts the pulse frequency; since the ionization is instantaneous, the wavevector may not change. This process can be understood as reflection and refraction due to a plasma-vacuum boundary in time. From the dispersion relation for electromagnetic waves in plasma: ($\omega^2 = c^2k^2 + \omega_p^2$) and $k_1 = k_0$, the upshifted frequency (ω_1) is related to the original frequency (ω_0) by

$$\tilde{\omega}_1 = \frac{\omega_1}{\omega_0} = \sqrt{1+N_{[0]}} = \frac{1}{\sqrt{1-N_{[1]}}}, \tag{1}$$

where $N_{[i]} = n_e/n_{\text{critical}}(\omega_i) = \omega_p^2/\omega_i^2$ and $\omega_p = \sqrt{4\pi n_e e^2/m_e}$ is the plasma frequency for electron density n_e. Note, $N_{[1]}/N_{[0]} = \omega_0^2/\omega_1^2$. In order to satisfy the conservation of energy and momentum, the energy in the original pulse is divided between two electromagnetic waves, one propagating

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in each direction, a static magnetic field and the kinetic energy of plasma electrons, as shown in Fig. 1(a). The relative energy fraction (W) in each mode depends on the magnitude of the upshift according to[39]

$$W^{\text{forw}}_{\text{back}} = \frac{1}{4}\left[1 \pm \frac{1}{\tilde{\omega}_1}\right]^2, \tag{2}$$

$$W_{\text{B}} = \frac{1}{2}\left[1 - \frac{1}{\tilde{\omega}_1^2}\right]^2, \tag{3}$$

$$W_{\text{kin}} = \frac{1}{2}\frac{1}{\tilde{\omega}_1}\left[1 - \frac{1}{\tilde{\omega}_1}\right]. \tag{4}$$

In the adiabatic limit of gradual ionization, the reflection becomes negligible and the energy of the forward mode decreases proportionally to $1/\tilde{\omega}_f$. It has previously been shown that the scale of energy density can be universally applied to all linear waves in a gradually ionizing plasma,[40,41] because the wave damping is caused by the ponderomotive potentials of the wave acting on the free plasma particles, which is irrelevant to the dispersion relations.

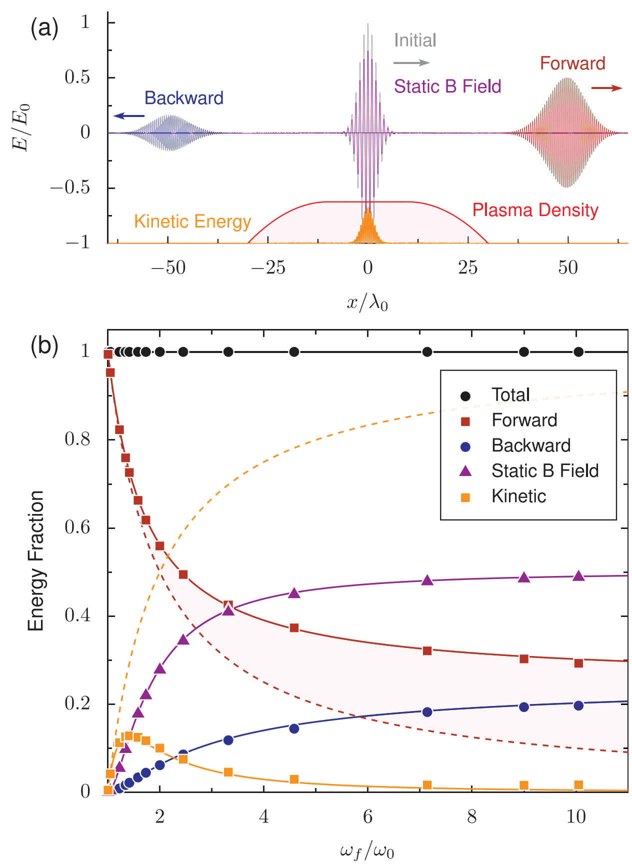

FIG. 1. (a) PIC simulation of frequency upshift for $\omega_1/\omega_0 = 2$, with initial field in gray, instantaneously created plasma density in red, and the forward (red) and backward (blue) propagating electromagnetic components, static magnetic field (purple) and electron kinetic energy distribution (orange) a short time interval after the ionization. (b) Energy fraction deposited in each mode as a function of frequency upshift magnitude, showing both the analytic predictions (lines) and PIC simulation results (points). The solid lines represent a single step upshift in a homogeneous plasma. The dashed lines show the limit of infinitely small sequential upshifts.

In Fig. 1(b), the energy deposited in each mode following the flash ionization process is shown as a function of the frequency upshift, based on both the analytic model (lines) and particle-in-cell (PIC) simulations (points). The dashed line indicates the energy deposited in the forward mode if the process is considered as an infinite number of infinitesimal steps, which gives $W_{\text{forw}} = 1/\tilde{\omega}_f$,[39] where ω_f is the final frequency. This decrease with increasing frequency is in contrast to the single-step case, where energy conversion to the forward-propagating electromagnetic field approaches 25% for large upshifts. The efficiency disadvantage is offset by the greater practicality and control over density available for the gas-density targets; a sequence of small upshifts may be more readily achieved and more controllable than a single large shift. For a finite number of steps, the energy conversion efficiency will lie in the shaded area between the two limits.

PIC simulations were carried out using the code EPOCH[42] in one dimension, with between 40 and 200 cells per initial wavelength and 10 to 50 particles per cell, depending on the degree of upshift. The simulations assumed infinitely heavy ions and took an electron temperature of 1 eV, although no significant differences were observed with slightly higher temperatures. The instantaneous flash-ionization process was approximated by initializing a simulation with the electric and magnetic fields of vacuum propagation overlapped with the finite plasma density associated with the completion of ionization. For the homogeneous plasma simulations shown in Fig. 1, the forward and backward propagating pulses passed out of the plasma through a quadratic density gradient to minimize the reflection at the plasma boundary. The energy and frequency of both forward and backward propagating pulses were then recorded in vacuum.

The use of a homogeneous plasma constrains the properties of the upshifted pulse. In particular, the lower group velocity in plasma $\left(v_g = c\sqrt{1 - N_{[1]}}\right)$ means that as the pulse is upshifted, it is also stretched in time, reducing its power by $1/\tilde{\omega}_f$. For the multi-step process, where forward-propagating energy scales as $1/\tilde{\omega}_f$, the pulse power then scales as $1/\tilde{\omega}_f^2$, which unfavorably compares with other possible mechanisms. The pulse stretching also limits the utility of flash ionization for large frequency upshifts; since even relatively short initial seeds will lengthen dramatically if upshifted to the x-ray regime; a 100 fs pulse with $\lambda = 1\,\mu$m will become a 100 ps pulse at $\lambda = 1$ nm.

In the frequency domain, the upshift process modifies not just the central frequency but also the pulse bandwidth ($\Delta\omega$). In the limit where the bandwidths are small compared to the central frequency ($\Delta\omega_i \ll \omega_i$), the upshifted bandwidth is related to the initial pulse relative bandwidth by[43]

$$\frac{\Delta\omega_1}{\omega_1} = \frac{\Delta\omega_0}{\omega_0}\frac{1}{1 + N_{[0]}} \tag{5}$$

or equivalently

$$\frac{\Delta\omega_1}{\Delta\omega_0} = \left[1 + N_{[0]}\right]^{-\frac{1}{2}} = \frac{\omega_0}{\omega_1} = \frac{1}{\tilde{\omega}_1}. \tag{6}$$

This decrease in absolute bandwidth exactly corresponds to the increase in pulse duration. Given a Fourier-transform-limited initial pulse, it is clear that upshift in a homogeneous plasma cannot support the bandwidth for shorter pulse durations.

However, if we consider the formation of a plasma density gradient (Fig. 2), either by modulating the intensity of the ionizing beam or by flash-ionizing a gas with a density gradient, the resultant pulse is chirped and may therefore be compressed by propagation through a dispersive medium or optical element, shortening its duration and increasing peak power to levels comparable to or greater than the original input pulse. Although there are many ways to compress a chirped pulse, here we use propagation through a preexisting plasma, which has the advantage of scaling well to short wavelengths. If the upshift occurs in a gradient which decreases in the direction of propagation, the pulse will be chirped from low to high frequency and the higher velocity of higher frequencies in an underdense plasma will allow compression.

The local frequency of the upshifted pulse will be a function of the local plasma density gradient. After the plasma is created with a density gradient profile $\omega_p(x)$, the instantaneous frequency of laser pulse at different positions is upconverted to $\omega_1(x) = \sqrt{\omega_0^2 + \omega_p^2(x)}$. Each frequency component propagates in the plasma at a different group velocity $v_g(\omega) = c\sqrt{1 - \omega_p^2(x)/\omega^2}$. Hence, the time for the frequency component $\omega(L) = \sqrt{\omega_0^2 + \omega_p^2(L)}$ generated at position L to exit the plasma is

$$t(L) = \int_L^{x_f} \frac{\mathrm{d}x}{c\sqrt{1 - \omega_p^2(x)/\omega^2(L)}}, \tag{7}$$

where x_f is the position of the plasma exit. For perfect pulse compression, all frequency components should reach distance x_f simultaneously, i.e., $t(L)$ is constant over a sufficient range of values for L. This approach can be used to estimate the final pulse duration for a given plasma shape and initial pulse duration. In principle, a single well-designed plasma shape can be used to provide the upshift as well as pulse compression, although here we will separate the upshift and compression into two separate steps for clarity in the analysis.

In Fig. 3(a), a PIC simulation of the upshift in a substantial plasma density gradient shows the resulting chirp in the output pulse. In (b), the chirped pulse then propagates through a homogeneous plasma, compressing until its duration decreases from 160 fs to 8 fs. The upshift by a factor of $\omega_1/\omega_0 \approx 1.4$ is associated with an immediate loss of about 25% of the pulse energy. However, as the pulse is compressed, its intensity substantially increases; the final pulse is 15 times more intense than the initial pulse. This method therefore allows the generation of a few cycle broad bandwidth pulse from a relatively long initial pulse duration, as demonstrated by the initial and final fields (c) and spectra (d).

Although single-step upshifts to extreme ultraviolet wavelengths are in principle possible using solid-density media, gas targets are substantially more practical, especially for producing density gradients. The lower electron density in gases reduces the frequency change which can be achieved in a single step. However, if chirped flash ionization and recompression are repeated in sequence, the resulting upshift cascade can produce relatively large changes in frequency using moderate plasma densities, while maintaining the power of the upshifted pulse. Figure 4 shows a PIC simulation of a pulse that is repeatedly upshifted in an exponential gradient and then recompressed in a homogeneous plasma (chirped points). The forward propagating field found in vacuum after each simulation was used as the initial field for the next flash-ionization step. In the chirped case, the initial pulse has a duration of 16 fs and propagates for $60\lambda_0$ in a homogeneous plasma with $N_{[0]} = 0.05$. In the unchirped case, the pulse is simply upshifted in a homogeneous plasma. For both the chirped and unchirped sets of simulations, the forward-propagating energy follows the expected $1/\tilde{\omega}_f$ scaling. In the unchirped case, the pulse intensity falls off with

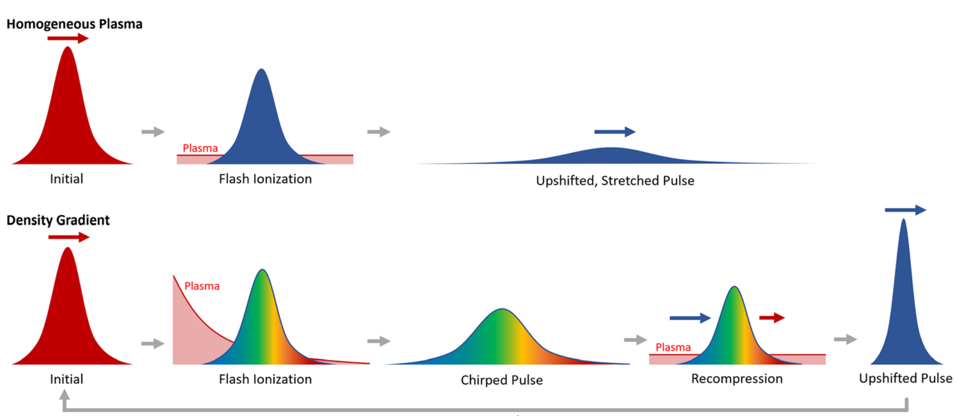

FIG. 2. Illustration of the frequency upshift process in (a) homogeneous and (b) inhomogeneous plasma. In the homogeneous case, ionization immediately upshifts the pulse frequency, and pulse stretches by a factor of ω_f/ω_0 as it leaves the plasma. In the inhomogeneous case, the plasma density gradient introduces a frequency chirp into the pulse. If the gradient is decreasing in the direction of the pulse propagation, the pulse stretch is slightly reduced due to the increased frequency and the higher group velocity of the trailing edge. Since the pulse is chirped, further compression may be achieved with traversal of a dispersive medium, compression gratings, or chirped mirrors. The final output pulse can be used as the seed for an additional step if further upshift is required.

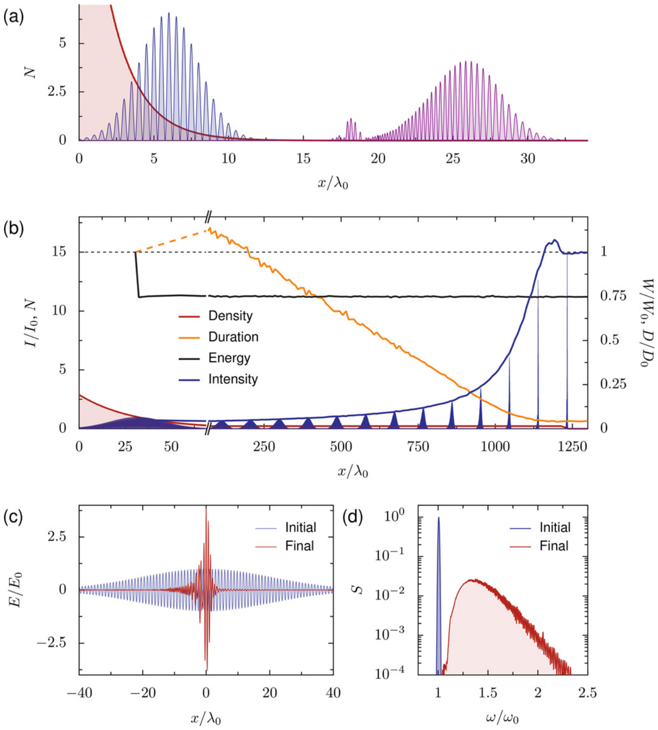

FIG. 3. (a) PIC simulation of flash ionization in an exponential density gradient, showing the chirp introduced into the pulse that propagates out of the plasma. (b) PIC simulation of flash ionization in an exponential density gradient [$N_{[0]}(x) \propto \exp(-x/L)$, where $L/\lambda = 30$] followed by recompression in a homogeneous plasma ($N_{[0]} = 0.2$). The initial pulse is shown at left, and the evolution of the pulse intensity, duration, and envelope are shown as it propagates through the plasma to the right. The final pulse has a duration of 8 fs, a peak frequency of $1.4\omega_0$, and 75% of the energy and 15 times the intensity of the original pulse. (c) The initial and final forward-propagating fields. (d) The initial and final spectra.

$1/\tilde{\omega}_f^2$, as expected from theory, but with chirping, the upshifted pulse almost maintains its initial intensity.

The chirped process is stable for incomplete compression, as evidenced by the oscillations in peak power. If the pulse is under-compressed, in the next stage, it is spread over a larger region of the gradient when upshifted, receives a larger bandwidth, and as a result is proportionally more compressed on the following step. The cascaded upshift is therefore tolerant to small deviations in plasma properties.

The general problem of determining the ideal $N(x, t)$ for a sequence of upshifts to produce a particular final pulse from a given initial pulse is complex, with potentially many viable solutions for particular desired results. As an example, we consider the problem of upshifting a many-cycle Gaussian pulse with initial duration (FWHM) τ ($\tau\omega_0 \gg 1$) from ω_0 to $\omega_f = 4\omega_0$, while maintaining the pulse duration ($\tau_f = \tau_0$). We constrain the problem to a 10-step cascaded process and use pure flash ionization, i.e., in each step ionized and H is the Heaviside step function.

$$N(x, t) = N(x)H(t - t_0), \tag{8}$$

where t_0 is the time at which the medium is fully ionized. In the homogeneous upshift case, independent of the number of steps, the bandwidth would decrease by a factor of $\omega_f/\omega_0 = 4$ [Eq. (6)] and the pulse duration would increase by the same

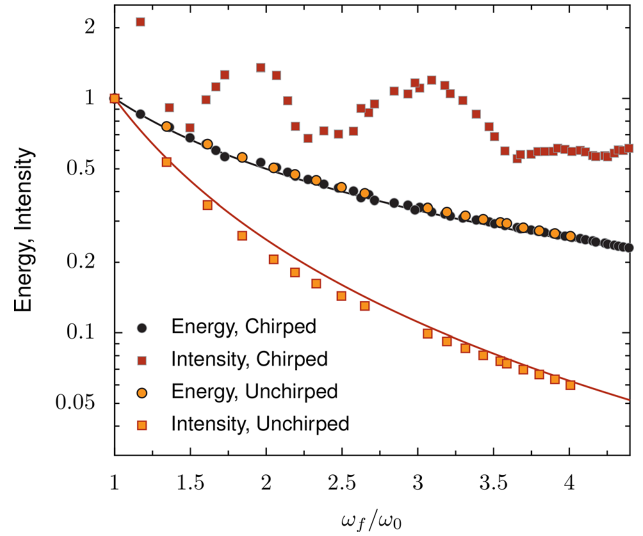

FIG. 4. Energy and intensity of pulses at each step in an upshift-compression scheme from PIC simulations. The solid lines represent the energy and intensity scaling for infinitely small steps in a homogeneous plasma. The points are taken from PIC simulations of a cascade chirped scheme, where the pulse is upshifted in an exponential gradient and recompressed in a homogeneous plasma. In the inhomogeneous simulations, the pulse energy follows the same line as for a homogeneous plasma, but the chirping and recompression keep the peak pulse power substantially above the homogeneous case.

factor. To maintain $\tau_f = \tau$, the ionization process must itself introduce a bandwidth $\Delta\omega_f = \Delta\omega_0$.

There are three obvious approaches for dividing the total upshift process into 10 steps: the density gradient may be the same in each step $n_e(x)_{[i]} = n_e(x)_{[0]}$, which is experimentally simplest, the frequency upshift may be proportionally constant ($\omega_{n+1}/\omega_n = \text{constant}$), which is theoretically simplest but requires an increasing real density, or each stage may be independently optimized to find the highest-efficiency path through the conversion process. For simplicity, here we will limit ourselves to the second approach, which allows us to simply treat the cascaded upshift as ten identical steps with $\omega_{n+1}/\omega_n = 4^{1/10} \approx 1.15$.

Since both the initial pulse shape and the final desired spectrum are Gaussian, it is natural to use a linear function form $\omega_1(x) - \omega_1 = a(x - x_0)$ to imprint the desired chirp and then match the initial spatial extent ($\Delta x = \tau c$) to the desired bandwidth ($\Delta\omega_1 = \text{TBP}/2\pi\tau$), where TBP is the time bandwidth product (0.44 for Gaussian pulses), giving the desired frequency distribution as

$$\omega_1(x) - \omega_1 = \frac{0.44}{2\pi c\tau^2}(x - x_0). \tag{9}$$

Using Eq. (5), the density gradient will therefore have the form

$$N(x) = \left[\frac{\omega_1}{\omega_0} + \left(\frac{0.44}{2\pi\omega_0\tau}\right)\frac{x - x_0}{c\tau}\right]^2 - 1. \tag{10}$$

Note that $2\pi\omega_0\tau$ is the FWHM number of cycles in the initial pulse. Neglecting the compression that occurs as the pulse traverses the flash-ionized gradient, this produces a linearly chirped pulse with a Gaussian spectrum centered at ω_1. For a single step, with $\omega_1/\omega_0 = 1.15$ and $2\pi\omega_0\tau = 100$, the density gradient required to produce a pulse-duration-maintaining bandwidth will almost linearly span $N = 0.3$ to $N = 0.35$ over $4\tau c$.

Provided that $\Delta\omega_1/\omega_1 \ll 1$, recompression of a linearly chirped pulse can be achieved in an underdense plasma; the specific parameters necessary can be found by expanding the group velocity around the central frequency to first order and solving for the constant plasma density (N^c) and length (x_L) that cause all frequency components to exit the compression stage at the same time. The result of this single upshift and compression is a pulse with $\omega_1/\omega_0 = 1.15$, flat phase, and the same duration as the original pulse. Nine further steps with appropriately shifted densities will produce a final pulse with $\omega_f = 4\omega_0$ and $\tau_f = \tau$. The form of the full plasma distribution is therefore

$$N(x,t) = \sum_{i=1}^{10}\left[N_i^u(x - x_i^u) + N_i^c(x - x_i^c)\right]H(t - t_{0,i}), \quad (11)$$

where

$$N_i^u(x) = \left(\left[\left(\frac{\omega_1}{\omega_0}\right)^i + \left(\frac{0.44}{2\pi\omega_0\tau}\right)\frac{x}{c\tau}\right]^2 - 1\right)H(2c\tau - |x|) \quad (12)$$

and

$$N_i^c(x) = N_i^c H(0.5x_L - |x|). \quad (13)$$

The first term (N_i^u) is for the upshift and the second (N_i^c) is for compression, where x_i^c and x_i^u must be large enough to prevent the overlap between subsequent plasma sections and, $t_i \approx x_i/c$. Increasing the bandwidth of the upshift by increasing the gradient will allow more robust recovery of the original pulse duration, since retrieving the original pulse duration will not require perfect compression.

The proposed process has several limitations which must be considered. The initial pulse intensity must remain below the ionization threshold of the material, so this mechanism may not be suitable for extremely high peak power sources. Resonances in the initial gas may prevent certain wavelength bands from being used and a large number of sequenced steps may prove experimentally difficult to implement. However, for moderate energy pulses, this mechanism offers a highly controllable method for precise manipulation of short-wavelength light.

In conclusion, we have suggested that a cascaded scheme of chirped photon acceleration stages allows efficient upshifting of moderate energy pulses, with the potential to produce few-cycle EUV and x-ray pulses and a high degree of control over bandwidth and frequency independently. Cascading enables the process to be conducted in gas-density targets, and, with appropriate chosen gradients and recompression, the chirped upshift process is stable, converging to an average value even if over or under-compressed in a particular stage.

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[43]This relation may be derived from the plasma dispersion relation by taking $(\omega_1 + \Delta\omega_1)^2 = (\omega_0 + \Delta\omega_0)^2 + \omega_p^2$, assuming $\Delta\omega_i/\omega_i \ll 1$, and simplifying to get $\omega_1\Delta\omega_1 = \omega_0\Delta\omega_0$. Using Eq. (1), this can be rewritten in either the form of Eq. (5) or (6).