Frequency up-shifting of laser pulses by copropagating ionization fronts

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A method for up-shifting the frequency of a subpicosecond laser pulse that utilizes the interaction of the pulse with a co-propagating relativistic ionization front is examined. The induced frequency shift is found to initially scale linearly with the propagation time τ. Asymptotically, the frequency scales as $\tau^{1/2}$. Phase-slippage limitations may be overcome by appropriately increasing the plasma density as a function of τ, thus allowing for substantially higher-frequency shifts.

Recently, several methods for increasing the frequency of a laser pulse that utilize the interaction of the laser pulse with a plasma have been proposed [1–8]. For example, plasma simulation studies [2] indicate that frequency up-shifts may result when a laser pulse interacts with a plasma-wave wake field [9–11] (having a phase velocity near the speed of light) [5,6]. In this article, a method [4] for up-shifting the frequency of a laser pulse is proposed and analyzed, one which utilizes the interaction of a laser pulse with a relativistic ionization front. In such a scheme, an intense driving electron beam (or, alternatively, an ionizing laser beam) is used to generate an ionization front, propagating near the speed of light, with the plasma density rising from zero to some high value n_0 over a relatively short distance L. A "test" laser pulse of length $<L$, which is properly phased such that it copropagates with the ionization front in the region of the large negative local-density gradient, will be continuously up-shifted in frequency as it propagates. This mechanism may provide a practical and efficient method for tuning the frequency of a laser pulse in which the up-shift is adjusted by varying the ionization density and/or the interaction distance.

This mechanism also implies that a laser pulse producing ionization in a gas may be self-up-shifted as it propagates. Experimentally, this process has been observed as a blue frequency shift of the ionizing laser pulse [12]. This phenomenon was attributed to a plasma density increasing in time, and analytic estimates were obtained for the resulting linear frequency shift (valid for frequency shifts $\Delta\omega$ small compared to the initial frequency ω_0) [12]. In the following, a first-principles theory is derived that describes the evolution of the laser pulse and the resulting nonlinear frequency shift due to the interaction with the ionization front. This theory indicates that currently available ultrahigh-power, subpicosecond lasers [13] may be used to produce large frequency up-shifts ($\Delta\omega > \omega_0$) through the self-ionization process.

The physical mechanism by which an ionization front may be used to upshift the frequency of a laser pulse may be understood as follows. Consider a laser pulse of frequency ω and wave number k, copropagating in the region of the ionization front, with both the pulse and the front assumed to be moving at approximately the speed of light. Furthermore, assume that in the region of the laser pulse, the ionization front may be approximated by a linear density gradient of the form

$$n \simeq \begin{cases} \bar{n}, & \xi < -L \\ -\bar{n}\xi/L, & -L < \xi < 0 \\ 0, & \xi > 0 \end{cases} \tag{1}$$

where $\xi = z - ct$ and L is the length of the ionization front (see Fig. 1). For the present discussion, $\bar{n} = n_0$ and $L = L_0$ are assumed to be constant. Consider a laser pulse centered at $\xi_c = -L_0/2$, with a pulse length of l, where $L_0 > l \gg \lambda = 2\pi/k$, and with $\omega_0 \gg \omega_{p0} = (4\pi e^2 n_0/m_e)^{1/2}$. If the frequency shift remains small compared to ω_0, the local phase velocity of the pulse (via the dispersion relation $\omega^2 = \omega_p^2 + c^2k^2$, where $\omega_p^2 = 4\pi e^2 n/m_e$) is approximately $v_p/c \simeq 1 + \omega_{p0}^2 n/2\omega_0^2 n_0$. Local variations in the plasma density $n(\xi)$ lead to variations in the local phase velocity $v_p(\xi)$ of the laser pulse. For example, the local phase velocity near the front of the laser pulse $v_p(\xi_+)$ will be less than that near the back of the pulse $v_p(\xi_-)$ provided $n(\xi_+) < n(\xi_-)$. Hence, the individual phase peaks in the pulse $\sim\exp(ikz - i\omega t)$ may move relative to one another (i.e., closer together for the present example). The resulting change in the radiation wavelength may be estimated by $\lambda(\tau) \simeq \lambda_0 + \Delta v_p \tau$, where $\Delta v_p \simeq \lambda_0 dv_p/d\xi$ is the difference in phase velocity between adjacent phase peaks, $\lambda_0 \simeq 2\pi c/\omega_0$ is the initial wavelength, and $c\tau$ is the laser–ionization-front propagation distance. This gives $\lambda/\lambda_0 \simeq 1 - c\tau\omega_{p0}^2/2L_0\omega_0^2$, which corresponds to an increase in frequency of $\omega/\omega_0 \simeq 1 + c\tau\omega_{p0}^2/2L_0\omega_0^2$. By a similar mechanism, a plasma wave may be used to induce shifts in the frequency of a laser pulse [2,5,6].

One expects the above estimates to hold in the limit in which the frequency shifts remain small, $\Delta\omega = \omega - \omega_0 \ll \omega_0$. However, it is possible to analytically obtain expressions for large frequency shifts ($\Delta\omega \gtrsim \omega$) in the limit $\omega_p^2/\omega^2 \ll 1$ via the wave equation in one dimension (1D). The wave equation will be considered for two cases in which a test laser pulse interacts with (i) a wake field in a fully ionized plasma and (ii) an ionization front plasma. The transverse plasma response current, in the fluid approximation, is $J_\perp \simeq env_\perp$, where $v_\perp$ is the transverse electron fluid velocity. For a fully ionized plasma, conservation of canonical transverse momentum implies $v_\perp \simeq ca$, which is the quiver motion of the electrons in the laser field assuming $|a|^2 \ll 1$. Here, $a = eA_\perp / m_e c^2$ is the normalized vector potential of the laser. Using the variables $\xi = z - ct$ and $\tau = t$, the transverse wave equation for a fully ionized plasma is

$$\left[\frac{2}{c}\frac{\partial}{\partial \xi} - \frac{1}{c^2}\frac{\partial}{\partial \tau} \right] \frac{\partial}{\partial \tau} a = \frac{\omega_{p0}^2}{c^2} \frac{n(\xi,\tau)}{n_0} a(\xi,\tau) \ , \tag{2}$$

where $n(\xi,\tau)$ is the plasma electron density, which is assumed to be known and independent of the test laser pulse. Equation (2) has been used to study the frequency up-shifting of a laser pulse by a plasma-wave wake field [5].

For an ionization front plasma, the wave equation is modified[7,8,14–16] by the existence of the plasma

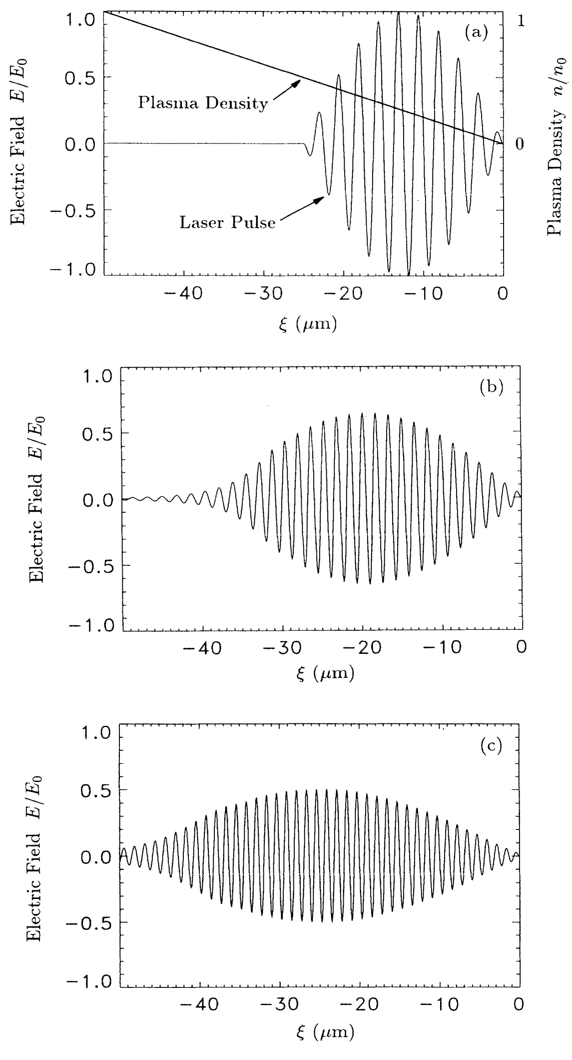

FIG. 1. The initial laser pulse and ionization density are shown in (a) as a function of $\xi = z - ct$. Numerical solutions of laser-pulse evolution after propagation 0.11 cm are shown in (b) for L and $\bar{n}$ constant, and in (c) for L constant and $\bar{n}$ an increasing function of τ.

source term S_0, where $\partial n / \partial t = S_0$. Assuming the source term to be unaffected by the presence of the low-amplitude test laser pulse, the linearized transverse momentum equation is given by $n \partial v_\perp / \partial t = -enE/m_e - v_\perp S_0$, where $E = -c^{-1} \partial A_\perp / \partial t$ is the transverse electric field of the laser pulse. Hence, $\partial J_\perp / \partial t = (e^2/m_e) nE$. Thus, when an ionization front is present, the transverse wave equation is given by

$$\left[\frac{2}{c}\frac{\partial}{\partial \xi} - \frac{1}{c^2}\frac{\partial}{\partial \tau} \right] \frac{\partial}{\partial \tau} E = \frac{\omega_{p0}^2}{c^2} \frac{n(\xi,\tau)}{n_0} E(\xi,\tau) \ . \tag{3}$$

Equation (3) describes the frequency up-shifting of a laser pulse by an ionization front. It is interesting to note that Eqs. (2) and (3) predict identical frequency up-shifts for given profiles of the laser pulse and the plasma density (in the limits $L > l \gg \lambda$ and $\omega_{p0}^2/\omega^2 \ll 1$), regardless of whether the plasma gradient arises from a plasma-wave wake field or an ionization front. However, Eq. (2) indicates that the laser pulse gains energy as it is up-shifted by a plasma-wave wake field [5], whereas Eq. (3) indicates that the laser pulse loses energy as it is up-shifted by an ionization front, as is discussed below.

Equation (3) may be solved analytically for the case of a square laser pulse of the form $E = \hat{E} \exp(i\omega\xi/c)$, for $-l < \xi < 0$, and $E = 0$, otherwise. Also, a linear ionization-density profile will be assumed, of the form given by Eq. (1). In general, the pulse amplitude $\hat{E}$, frequency ω, pulse length l, ionization density $\bar{n}$, and ionization-front length L are all functions of τ. In the limit $\omega_p^2/\omega^2 \ll 1$, the second-order τ derivative in Eq. (3) may be neglected, and one finds $\hat{E} = E_0 \omega_0 / \omega$ for $-l < \xi < 0$, where

$$\frac{\omega(\tau)}{\omega_0} = \left[1 + \frac{\omega_{p0}^2}{\omega_0^2} \int_0^\tau d\tau' \frac{c\bar{n}(\tau')}{n_0 L(\tau')} \right]^{1/2} , \tag{4}$$

with $n_0 = \bar{n}(\tau = 0)$, $E_0 = \hat{E}(\tau = 0)$ and $\omega_0 = \omega(\tau = 0)$. Notice that as the laser pulse evolves, $|\partial E / \partial \xi|^2$ is approximately constant in τ, i.e., frequency up-shifts correspond to decreases in the laser amplitude, $\hat{E}(\tau) \sim 1/\omega(\tau)$.

By integrating the wave equation in ξ over the longitudinal extent of the laser pulse, an equation for the quantity $U \equiv \int d\xi |\partial E / \partial \xi|^2$ may be derived. In the limit $\omega_p^2/\omega^2 \ll 1$, and for the linear density gradient given by Eq. (1), U evolves according to $\partial U / \partial \tau = \alpha \omega_{p0}^2 \bar{n} / 2 n_0 c L$, where $\alpha = \int d\xi |E|^2 > 0$. For the case of a square pulse of length l, one finds $l(\tau)/l_0 = \omega(\tau)/\omega_0$, where $l_0 = l(\tau = 0)$, which implies that the pulse length increases as the frequency is up-shifted. Hence the laser pulse energy decreases as it propagates, i.e., $lE^2 = l_0 E_0^2 \omega_0 / \omega$. The energy lost by the laser pulse appears as an effective transverse thermal energy of the plasma electrons behind the laser pulse.

For a fixed, externally generated ionization front (generated by an ionizing pump laser or electron beam) in which $\bar{n} = n_0$ and $L = L_0$ are independent of τ, Eq. (4) gives

$$\omega(\tau)=\omega_0(1+c\tau\omega_{p0}^2/L_0\omega_0^2)^{1/2} . \quad (5)$$

Equation (5) indicates that asymptotically, for $c\tau \gg L_0\omega_0^2/\omega_{p0}^2$, the frequency scales as $\omega\simeq\omega_{p0}(c\tau/L_0)^{1/2}$, where $c\tau$ represents the distance the pulse has propagated through the plasma. Notice that frequency shifts $\Delta\omega\sim\omega_0$ require $c\tau\simeq L_0\omega_0^2/\omega_{p0}^2$. In principle Eq. (5) indicates that there is no upper limit to how far the frequency may be upshifted (assuming that the ionization front and laser pulse may be sustained over a sufficiently large distance within the plasma); however, this is a result of assuming $v_0\simeq v_g\simeq c$, where v_0 is the velocity of the ionization front and v_g is the group velocity of the laser pulse. In practice, $v_0\neq v_g$, which implies that the laser pulse will "phase slip" in ξ out of the region of the ionization front $(-L_0<\xi<0)$ and, thus, will no longer be frequency upshifted. Consider a laser pulse with a pulse center ξ_c initially located at $\xi_{c0}=\xi_c(\tau=0)$, where $-L_0<\xi_{c0}<0$. Phase slippage will cause ξ_c to evolve in τ according to $\partial\xi_c/\partial\tau=v_g-v_0$. Assuming $v_0\simeq c$, $v_g/c\simeq 1-\omega_{p0}^2 n(\xi_c)/2\omega^2 n_0$, and $n(\xi_c)=-n_0\xi_c/L_0$, where $\omega=\omega(\tau)$ is given by Eq. (5), give $\xi_c(\tau)/\xi_{c0}=\omega(\tau)/\omega_0$. The detuning distance $c\tau_d$, defined to be the propagation distance required for the laser pulse to phase slip out of the ionization front, $\xi_c(\tau_d)=-L_0$, is given by $c\tau_d=L_0(\epsilon^{-2}-1)\omega_0^2/\omega_{p0}^2$, where $\epsilon\equiv|\xi_{c0}/L_0|$. Inserting this into Eq. (5) gives a maximum frequency up-shift of $\omega(\tau_d)=\omega_0/\epsilon$. For example, a pulse initially centered at $\xi_{c0}=-L_0/4$ $(\epsilon=\frac{1}{4})$ gives a final frequency of $\omega(\tau_d)=4\omega_0$ after a detuning distance of $c\tau_d=15L_0\omega_0^2/\omega_{p0}^2$.

The above analysis has assumed that (i) the velocity of the ionization front is constant, $v_0\simeq c$, as may be the case for an ionization front produced by a relativistic electron beam, and (ii) the density profile of the ionization front $n(\xi)$ was assumed independent of the propagation time τ. The limit placed on the frequency shift as a result of phase slippage may be circumvented by relaxing either of these two conditions.

If the ionization front is directly produced by the laser pulse itself, then the condition $v_0(\tau)=v_g(\tau)$ may automatically be satisfied. When $v_0=v_g$, there is no slippage and, hence, the frequency of an intense laser pulse may be "self-upshifted" as a result of the interaction of the pulse with its self-generated ionization front. For a square pulse that generates an ionization front with a uniform gradient $dn/d\xi=-n_0/L_0$, the frequency evolves according to Eq. (5). Since $v_0=v_g$, the absence of slippage implies that the up-shifting process will be limited by some other mechanism, such as laser-pulse diffraction. As an example, consider a 100-fs ($l_0\simeq 30$ μm), KrF laser pulse ($\lambda_0=2\pi c/\omega_0\simeq 0.25$ μm) ionizing hydrogen gas at 1 atm ($\lambda_{p0}=2\pi c/\omega_{p0}\simeq 4.5$ μm). In order to double the laser frequency, an interaction distance of $c\tau\simeq 3.0$ cm is required. Assuming that the laser-pulse propagation distance is limited by diffraction implies $c\tau\simeq 2Z_R$, where $Z_R=\pi r_s^2/\lambda_0$ is the vacuum Rayleigh length and r_s is the laser spot size. Setting $2Z_R\simeq 3.0$ cm gives $r_s\simeq 34$ μm. The energy required to ionize this volume of hydrogen at 1 atm is about 11 mJ. Hence, a laser-pulse energy on the order of 100 mJ should be sufficent [13]. This process of frequency self-up-shifting may be of practical significance; however, a detailed analysis of this process requires that the dynamics of laser-induced ionization be included self-consistently. (Such a formalism has been attempted in Ref. [7]).

Alternatively, the limitations resulting from phase slippage, which occur for the case of an externally generated ionization front, may be eliminated by gradually increasing the plasma density of the ionization front as a function of the propagation time τ. By correctly tailoring the ionization density as a function of τ, it may be possible to control the laser group velocity such that $v_0=v_g$, thus eliminating phase slippage. The requirement that $v_g=v_0$ is constant independent of τ implies that density must be tailored such that $n(\tau)\sim\omega^2(\tau)$, i.e., $L_0\bar{n}(\tau)/L(\tau)n_0=\omega^2/\omega_0^2$. Solving for the ratio $R=\bar{n}(\tau)/L(\tau)$ gives $R/R_0=\exp(c\tau\omega_{p0}^2/L_0\omega_0^2)$, where $R_0=R(\tau=0)$. Hence, the ionization profile must be tailored such that $\bar{n}/L$ is an exponentially increasing function of τ, which may be achieved by using additional lasers to control the ionization rate. For such a density profile, Eq. (4) indicates that the frequency is exponentially up-shifted:

$$\omega(\tau)=\omega_0\exp(c\tau\omega_{p0}^2/2L_0\omega_0^2) . \quad (6)$$

Zero slippage further requires that the ionization-front velocity be held constant at $v_0/c=1-|\xi_{c0}|\omega_{p0}^2/2L_0\omega_0^2$, which may be satisfied by adjusting the energy of the driving electron beam.

To validate the analytical results, the full wave equation, Eq. (3), is solved numerically for a laser pulse having a half-sine initial profile $|E|=|\sin(\pi\xi/l_0)|$ for $-l_0<\xi<0$, as shown in Fig. 1(a), for the parameters $\lambda_0=2.5$ μm, $\lambda_{p0}=10$ μm, $l_0=25$ μm, $L_0=50$ μm, and $v_0=c$. Equation (3) is solved for two cases: (i) $\bar{n}=n_0$ and $L=L_0$ are constant, which is assumed in deriving Eq. (5), and (ii) $L=L_0$ is constant and $\bar{n}=n_0\exp(c\tau\omega_{p0}^2/2L_0\omega_0^2)$, which is assumed in deriving Eq. (6). The results, after propagating a distance $c\tau=(2\ln 2)L_0\lambda_{p0}^2/\lambda_0^2\simeq 0.11$ cm, are shown in Fig. 1(b) for case (i) and in Fig. 1(c) for case (ii). The results show excellent agreement with the analytic predictions (which assumed a square pulse) to within 5%. Specifically, Fig. 1(b) indicates that $\omega\simeq 1.5\omega_0$ $(\lambda\simeq 0.65\lambda_0)$, $|E|\simeq 0.65|E_0|$, and $l\simeq 1.5l_0$, in agreement with the analytic theory, $E\sim 1/\omega$ and $l\sim\omega$, where ω is given by Eq. (5). Likewise, Fig. 1(c) indicates that $\omega\simeq 2\omega_0$ $(\lambda\simeq 0.5\lambda_0)$, $|E|\simeq 0.5|E_0|$, and $l\simeq 2l_0$, in agreement with the analytic theory where ω is given by Eq. (6). The profile distortion and the "tail" observed behind the pulse in Figs. 1(b) and 1(c) may be attributed to longitudinal dispersion arising from the initial half-sine profile.

The above analysis indicates that the use of a relativistic ionization front to up-shift the frequency of a laser pulse may be of practical significance. Physically, frequency up-shifts arise from the pulse propagating in the region of the ionization front in which the local-density gradient is negative. This produces a negative gradient in the local phase velocity within the pulse, which allows the laser wavelength to decrease and the frequency to increase as the pulse propagates. Interaction with an ionization front having a uniform gradient,

$dn/d\xi = -n_0/L_0$, would give the frequency described by Eq. (5), $\omega \sim \tau^{1/2}$. The laser pulse evolves such that the amplitude decreases, $|E| \sim 1/\omega$, the pulse energy decreases, $lE^2 \sim 1/\omega$ and the pulse length increases, $l \sim \omega$. Furthermore, phase-slippage limitations may be overcome by appropriately increasing the plasma density as a function of propagation time τ, which may lead to substantially higher frequencies, i.e., as described by Eq. (6). Hence, the use of ionization fronts, together with varying the ionization density and/or the interaction length, may provide a convenient method for up-shifting and tuning laser-pulse frequencies. In addition to this mechanism, an ionization front may also be used as a relativistic plasma mirror which may reflect an oppositely directed laser pulse [4,8,14]. The reflected radiation will be up-shifted by the relativistic Doppler factor, $\omega_s \simeq 4\gamma_0^2\omega_i$, where ω_i is the frequency of the incident radiation, ω_s is the frequency of the scattered (reflected) radiation, and $\gamma_0^2 = 1/(1-v_0^2/c^2)$ is the relativistic factor associated with the ionization front. This process is currently being investigated and will be the subject of future publication.

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