High-Order Frequency Conversion in the Plasma Waveguide

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It is shown that a plasma fiber waveguide can provide phase matching over extended interaction lengths for the generation of high-order harmonics and difference frequencies through wave mixing or parametric amplification. An important consideration is that the plasma waveguide mode structure is independent of wavelength. Phase matching may be achieved through either frequency tuning or waveguide structure tuning.

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We have recently developed and characterized a method for optically guiding high-intensity laser pulses in a plasma fiber waveguide [1–3]. The extremely large product of interaction length and intensity made possible by this waveguide suggests its application to short wavelength generation by very high-order frequency conversion. An example of one of these processes under considerable recent study is high-harmonic generation by short intense laser pulses in gas jets of limited extent [4].

In this Letter several plasma waveguide-based schemes are proposed for the phase matching of high-order frequency conversion. Low-order wave mixing processes in plasma fibers are closely related to similar processes in conventional solid fibers [5], but in plasma fibers more degrees of freedom are available for phase matching the conversion process at all orders. Nonlinear optics in plasma fibers is distinguished by (1) the unique dispersion properties of the fiber, leading to among other things the wavelength independence of the transverse mode structure; (2) the confinement of extremely high propagating intensities, which gives rise to very high-order nonperturbative processes which can occur with efficiencies comparable to lower-order processes; and (3) the dynamic evolution of the plasma waveguide in time, so that the geometric contribution to its dispersion relation is tunable and offers a degree of freedom of phase matching not previously available.

In our guiding technique, an axicon lens [1] brings a laser pulse to a line focus in a gas. The shock expansion of the resulting spark forms a favorable refractive index profile into which a second laser pulse is injected after an adjustable delay. In experiments to date, pulses have been guided at distances of 2.2 cm (~70 Rayleigh lengths) at intensities greater than 10^{14} W/cm^2, sufficient to be in the regime of high-harmonic generation in atoms. In the plasma waveguide, the nonlinear polarization is induced by the input field(s) in ions and high ionization potential neutrals, which may be present. It has been shown [6] that the rate of harmonic emission is approximately proportional to the ionization rate; at sufficiently high intensity, harmonic emission is as efficiently generated from ions as from neutrals. Moreover, ions should produce a greater number of harmonics, since the maximum photon energy in the plateau is $E_{\max} \approx I_p + 3.2U_p$, where I_p is the ionization potential and U_p is the ponderomotive potential [5].

We consider the case in which the free electron contribution to the refractive index dominates the bound electron contribution (this is reasonable, provided all wave frequencies are low compared to the resonance frequencies of the background ions). The channel electron density profile is modeled to be axially invariant and monotonically increasing with radius $|\mathbf{r}_\perp|$ from the channel axis out to some boundary, outside of which it remains constant. Such a density profile, which we have called the "finite" profile [2], can support bound modes, and possesses a cutoff. In the real experimental profile, the electron density decreases beyond its peak at the position of the shock wave, leading to tunneling or leaking of field energy for waves which are near cutoff [2]. Here, however, we are concerned mainly with bound modes, so that the finite profile can be employed to good approximation. The transverse eigenmodes $u(\boldsymbol{r}_\perp)$ of the plasma waveguide are found from

$$[\nabla_\perp^2 - 4\pi r_e N_e(\boldsymbol{r}_\perp)]u = -\xi^2 u\,, \qquad (1)$$

where $\underline{E}_\beta(\boldsymbol{r}_\perp, z, \omega) = u(\boldsymbol{r}_\perp)\exp[i\beta(\omega)z]$ is a pure propagating mode, $\nabla_\perp^2$ is the transverse Laplacian, $r_e = 2.82 \times 10^{-13}$ cm is the classical electron radius, $\xi^2 = k^2 - \beta^2$ is the eigenvalue, and k and β are the vacuum and waveguide wave numbers, respectively. Some general conclusions may be drawn without specifying the exact electron density profile $N_e(\boldsymbol{r}_\perp)$. The left side of Eq. (1) is independent of k, so that the eigenvalues ξ^2 and eigenmodes $u(\boldsymbol{r}_\perp)$ are *wavelength independent.* (The limits to this property are set by the nonpropagation of wavelengths longer than the plasma critical density cutoff [3] or the onset of relativistic electron dynamics through ultrahigh intensity $e|\mathbf{A}|/mc^2 \sim 1$ and/or short wavelength $2\pi/k = \lambda \approx \lambda_C$, where $\mathbf{A}$ is the vector potential and λ_C is the Compton wavelength.) Therefore, any light generated in the waveguide may be decomposed into the same set of modes as the driving wave(s). As an example, if only the lowest order transverse mode of the driving wave is bound by the finite guide,

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then only the lowest order mode of the generated wave is bound, and these transverse modes are spatially the same. High-order frequency conversion in a leaky guide [2] will be treated in future work.

Equation (1) may be rescaled to illustrate the dependence of the channel propagation wave number β on the channel structure. Consider a density profile $N_e(\boldsymbol{r}_\perp) = N_{e0} = \Delta N_e f(\boldsymbol{r}_\perp)$, where $f(\boldsymbol{r}_\perp) \to 0$ monotonically as $|\boldsymbol{r}_\perp| \to 0$. If we define the characteristic radius $w_{\rm ch}$ by the relation $\Delta N_e \equiv 1/r_e \pi w_{\rm ch}^2$, Eq. (1) becomes

$$\left[\tfrac{1}{4}\nabla_\perp^2 - f(\boldsymbol{r}_\perp)\right]u = -\eta u, \qquad (2)$$

where the transverse coordinate is now scaled to $w_{\rm ch}$, and we define a new eigenvalue $\eta = (\xi^2 - 4\pi r_e N_{e0})w_{\rm ch}^2/4$. Note that $\eta > 0$, since both $(\nabla_\perp^2 u)/u < 0$ and $f(\boldsymbol{r}_\perp) \to 0$ as $|\boldsymbol{r}_\perp| \to 0$; η is of order unity for low-order modes. For azimuthally symmetric profiles $f(\boldsymbol{r}_\perp) = f(r)$, the eigenvalue is $\eta = \eta_{pl}$, where p and l are radial and azimuthal mode indices. For the useful special case of an infinite parabolic profile $f(\boldsymbol{r}_\perp) = (r/w_{\rm ch})^2$ (with no limit in r), it can be shown [7] that $\eta_{pl} = 2p + l + 1$ and $u(r_\perp) = u_{pl}(r,\phi) = e^{il\phi}s^l L_p^l(2s^2)\exp(-s^2)$ (Laguerre-Gaussian functions), where $s = r/w_{\rm ch}$. Here, the lowest order mode u_{00} is a Gaussian with $1/e$ radius $r = w_{\rm ch}$, while $N_e(w_{\rm ch}) - N_{e0} = \Delta N_e$ $(\equiv 1/\pi r_e w_{\rm ch}^2)$, which is simply the density difference criterion for guiding. This criterion applies quite well to nonparabolic azimuthally symmetric profiles, for which it has been shown that the lowest order modes are nearly Gaussian with $1/e$ field radii given to good approximation by $w_{\rm ch}$ [2]. For general density profiles, the sets of u and η must be calculated numerically [3].

The waveguide propagation wave number is then given by $\beta^2 = k^2 - 4\pi r_e N_{e0} - 4\eta/w_{\rm ch}^2 = k^2[1 - N_{e0}/N_{\rm cr} - 4\eta/(kw_{\rm ch})^2]$, where $N_{\rm cr} = k^2/4\pi r_e$ is the critical density. The second and third quantities in parentheses are small [typical values of these terms for a guided optical driving wave are $N_{e0}/N_{\rm cr} \lesssim 10^{-3}$ and $(kw_{\rm ch})^{-2} \lesssim 10^{-3}$], so that

$$\beta \simeq k - r_e N_{e0}\lambda - \frac{\lambda}{\pi W_{\rm ch}^2}\,\eta\,. \qquad (3)$$

The propagation phase βz has two contributions: the plasma dispersion term $(k - r_e N_{e0}\lambda)z = k(1 - N_{e0}/2N_{\rm cr})z$, and the waveguide geometric contribution $\lambda\eta z/\pi w_{\rm ch}^2$. The latter replaces the free space focusing phase (which for Gaussian beams is $\tan^{-1}[\lambda(2p + l + 1)z/\pi w_0^2]$, where w_0 is the minimum $1/e$ beam radius).

Coupling of the interacting and generated fields to the waveguide modes plays a crucial role. We can expand the bound portion of the nonlinearly generated field in waveguide eigenmodes according to $E(\boldsymbol{r},t) = \frac{1}{2}[\sum_j a_j(z,t)u_j(\boldsymbol{r}_\perp)\exp i(\beta_{j0}z - \omega_0 t) + \text{c.c}]$, where the mode amplitudes a_j are slowly varying in z and t, c.c. is the complex conjugate of the previous term, $\beta_{j0} = \beta_j(\omega = \omega_0)$, and ω_0 is the center frequency of the generated wave. We ignore the contributions of leaky and free waves, which will be considered in future work. Fourier transforming this expression and inserting the result into the Fourier-transformed wave propagation equation, projecting onto the jth bound channel mode u_j, neglecting group velocity dispersion (GVD), and returning to the time domain gives

$$\frac{\partial}{\partial z}a_j(z,\tau) = \frac{2\pi i\omega_0^2}{\beta_{j0}c^2}\,c^{-i\beta_{j0}z}\int d^2\boldsymbol{r}_\perp \underline{P}_{\rm NL}u_j^*(\boldsymbol{r}_\perp) \qquad (4)$$

for the growth of the amplitude a_j in the nonlinearly generated field of center frequency ω_0. Here $\tau = t - z/v_{g0}$ is a time coordinate local to the pulse ($\tau = 0$ corresponds, say, to the pulse peak), $v_{g0} = (\partial\omega/\partial\beta_j)_0$ is the group velocity of the jth mode at $\omega = \omega_0$, $\underline{P}_{\rm NL}$ is the nonlinear polarization (slowly varying in time, but not in space), and the integration is over the channel cross section. To evaluate the growth of the mode amplitude for a fixed position on the pulse (constant τ), Eq. (4) is integrated with respect to z.

If two interacting *modal* fields $\underline{E}_1$ and $\underline{E}_2$ are quasi-monochromatic with frequency ω_1 and ω_2, and have envelopes varying slowly in time compared to the medium response, the resulting nonlinear polarization at frequency ω can be written as

$$\underline{P}_{\rm NL} = P_{\rm NL}(E_1,E_2)e^{i\Psi(E_1,E_2,\varphi_1,\varphi_2)}, \qquad (5)$$

where $P_{\rm NL}(E_1,E_2)$ and Ψ are real; $P_{\rm NL}$, $E_1(\boldsymbol{r}_\perp,z,\tau_1,\omega_1)$, and $E_2(\boldsymbol{r}_\perp,z,\tau_2,\omega_2)$ are slowly varying amplitudes in time and space, $\tau_{1,2} = t - z/v_g(\omega_{1,2})$ are local time coordinates of the interacting fields [with group velocities $v_g(\omega_{1,2})$], and $\varphi_1(z)$ and $\varphi_2(z)$ are their modal propagation phases. If ω_1 and ω_2 are commensurate, then $P_{\rm NL}$ may also depend on φ_1 and φ_2, but we do not consider that case here. In the low intensity limit, where lowest-order perturbation theory applies, $\Psi(E_1,E_2,\varphi_1,\varphi_2) = \psi_0 + m\varphi_1 \pm n\varphi_2$ for sum or difference frequency generation $\omega = m\omega_1 \pm n\omega_2$ ($m + n$ odd, $\omega > 0$), and ψ_0 is constant. At higher intensities beyond the perturbation limit, it has been shown [8] that this can be generalized by writing $\Psi(E_1,E_2,\varphi_1,\varphi_2) = \psi(E_1,E_2) + m\varphi_1 \pm n\varphi_2$, where $\psi(E_1,E_2)$ allows for the possibility of an intensity-dependent phase. An intensity-dependent phase of the field-induced dipole moment has been predicted in calculations [9], with some supporting experimental observations [10]. Equation (4) then becomes

$$\frac{\partial}{\partial z}a_j(z,\tau) = \frac{2\pi i\omega_0^2}{\beta_{j0}c^2}\,e^{i\Delta kz} \times \int d^2\boldsymbol{r}_\perp\, P_{\rm NL}(E_1,E_2)u_j^*(\boldsymbol{r}_\perp)e^{i\psi(E_1,E_2)}, \qquad (6)$$

where $\Delta k = m\beta_1(\omega_1) \pm n\beta_2(\omega_2) - \beta_j(\omega_0)$ for the case where the interacting modal fields are channel eigenmodes η_1 and η_2 with propagation phases $\varphi_1 = \beta_1(\omega_1)z$ and $\varphi_2 = \beta_2(\omega_2)z$.

The intensity-dependent phase $\psi(E_1, E_2)$ contributes to the modal overlap of the nonlinear polarization through the transverse integration in Eq. (6). It may also contribute to the phase mismatch if there is a significant difference in the group velocities among the generated and driving waves. For sufficiently small GVD however, $\tau \approx \tau_1 \approx \tau_2$, and $\psi(E_1, E_2)$ does not contribute to the z integration of Eq. (6), and that case is assumed here. We therefore identify ΔkL as the phase mismatch in a plasma waveguide of length L, with optimum phase matching for $|\Delta kL| \ll 2\pi$. Light is generated in any channel mode u_j to which $P_{\rm NL}$ couples; the efficiency of generation depends on Δk and on the overlap of $P_{\rm NL}$ with u_j. Regarding GVD, the maximum envelope slip for similar modes η over a channel of length L is $\Delta s \sim (N_{R2}\eta/2\pi)\lambda_2$, where $N_{R2} = L/(\pi w_{\rm ch}^2/\lambda_2)$ is the number of Rayleigh lengths of channeled propagation of the lowest frequency driving wave. If $N_{R2} \sim 25$ and $\eta \sim 1$, then $\Delta s \sim 5\lambda_2$, which is small compared to envelope widths $\gtrsim 100$ fs, although the numerical degree to which envelope slip affects phase matching requires knowledge of the detailed functional dependence of $\psi(E_1, E_2)$ [9].

We first consider the process of lowest-order nth harmonic generation, $\omega = n\omega_1$, for which $\Delta k = n\beta_1(\omega_1) - \beta_j(\omega)$, or, using Eq. (3),

$$\Delta k = -n\lambda_1\left[r_e N_{e0}\left(1 - \frac{1}{n^2}\right) + \frac{1}{\pi W_{\rm ch}^2}\left(\eta_1 - \frac{\eta}{n^2}\right)\right], \tag{7}$$

where η corresponds to mode u_j. Note that if the driving and generated waves are in the same channel mode ($\eta_1 = \eta$), $\Delta k < 0$ for $n > 1$ and phase matching is impossible. For $\eta_1 \neq \eta$, $\Delta k = 0$ gives the *modal* phase matching condition,

$$n^2 = \frac{\Gamma + \eta}{\Gamma + \eta_1}, \tag{8}$$

where $\Gamma = r_e N_{e0}\pi w_{\rm ch}^2 = N_{e0}/\Delta N_e$, and Γ^{-1} is a measure of the channel depth. Therefore, phase matching for lowest-order harmonic generation can occur only through coupling of the harmonic light to channel modes higher than that of the driving wave. For N_{e0} in the range $10^{17}-10^{19}$ cm^{-3} and $w_{\rm ch}$ in the range 10–50 μm, Γ is in the range 0.1–100. Equation (8) suggests that if the driving wave is coupled to a low-order channel mode ($\eta_1 \sim 1$) and the harmonic to a high-order mode ($\eta \gg \eta_1$), and Γ is of order η_1, then a harmonic of order $n \sim \eta^{1/2}$ could be phase matched. Since the mode η must be bound, this requires, for a symmetric channel, an approximate density difference $N_e(r_m) - N_{e0} = \Delta N_e(r_m) > \eta^2/\pi r_e r_m^2 = n^4/\pi r_e r_m^2$, where $r = r_m$ is the radius of peak electron density [2]. For large n, a very deep channel is required. Such a channel, which may be difficult to produce in any case, will support many modes such that the transverse spatial overlap of the nonlinear polarization $P_{\rm NL}$ with mode η will likely be small.

A scheme ensuring better overlap is difference frequency generation. This has been suggested as a means to compensate the phase shift for tightly focused beams in plasmas [11]. In the plasma waveguide, $\Delta k = m\beta_1(\omega_1) - n\beta_2(\omega_2) - \beta_j(\omega)$, or

$$\Delta k = [-m\lambda_1(\Gamma + \eta_1) + n\lambda_2(\Gamma + \eta_2) + \lambda(\Gamma + \eta)]\frac{1}{\pi W_{\rm ch}^2}, \tag{9}$$

using Eq. (3). If we assume $m, n \gg 1$ and $\omega_1 \geq \omega_2$, most of the phase matching occurs through the first two terms in this equation, since the third term is down by $\sim 1/m^2$ from the first two and may be ignored if the residual phase mismatch it produces is small. This short wavelength limit (SWL) requires $|\Delta kL| = (\Gamma + \eta)N_R \ll 2\pi$, where $N_R = L/(\pi w_{\rm ch}^2/\lambda)$ is the number of Rayleigh lengths of propagation of the *generated* light. For $\Gamma \approx \eta \approx 1$, $L = 1$ cm, and $w_{\rm ch} = 10$ μm, the SWL occurs for $\lambda < 100$ nm. In the SWL, phase matching can occur over a *range* of channel modes η of low order, effectively increasing the coupling of $P_{\rm NL}$.

First, consider the case where m, n, and Γ are given, and either ω_1 or ω_2 is tuned to achieve phase matching. This analysis also applies to high-order parametric amplification, with ω as the signal and either ω_1 (ω_2) or ω_2 (ω_1) contributing to idler (pump) waves, and with the signal and/or idler amplified from noise such as plasma radiation. In the SWL, $\Delta k = 0$ yields $\omega_1/\omega_2 \approx m(\Gamma + \eta_1)/n(\Gamma + \eta_2)$, giving phase matched output at frequency $\omega \approx m\omega_1[1 - (n/m)^2(\Gamma + \eta_2)/(\Gamma + \eta_1)]$ for the case where ω_1 is fixed and ω_2 is tuned. For the case where all fields are in the same mode $\eta_1 = \eta_2 = \eta$, phase matching is *independent of waveguide parameters* (same-mode phase matching), and is therefore insensitive to variations in Γ and $w_{\rm ch}$ along the channel as long as the fields stay in comparable modes. Owing to the wavelength independent mode structure of the plasma waveguide, this is likely. Equation (9) then gives $q - \delta + 1/q = 0$, where $q = \omega_1/\omega_2 > 1$ and $\delta = (m^2 + n^2 - 1)/mn$, and either ω_1 or ω_2 is tuned to achieve phase matching. For $m \gg 1$ and $n \gg 1$, $\delta \approx m/n + n/m$ gives $q_{\rm opt} \approx m/n$ and phase matched output at $\omega \approx m\omega_1[1 - (n/m)^2]$. Given the requirement $|\Delta kL| < \pi$, phase matched difference frequency generation occurs for both different-mode and same-mode cases over a bandwidth $\Delta\omega_2/\omega_2 \approx 2\pi^2/nN_{R2}(\Gamma + \eta_2)$ if ω_1 is fixed and ω_2 is tuned [or $\Delta\omega_1/\omega_1 \approx 2\pi^2/mN_{R1}(\Gamma + \eta_1)$ for the case of ω_2 fixed and ω_1 tuned]. This is rather substantial: For example, for $N_{R2} \sim 25$ and $\Gamma \sim \eta_2 \sim 1$, $\Delta\omega_2/\omega_2 \approx 0.4/n$, so that low orders of subtracted photons ensure phase matching over an extremely wide tuning range. Since $\Delta\omega/\omega \sim 0.01$ for a 100 fs optical pulse, it is also seen that ultrashort pulses can be phase matched to high order over their full bandwidths.

If ω_1 and ω_2 are given, the values m, n, η_1, η_2, and η may be chosen to minimize Δk, but since these may be changed only incrementally, the channel inverse depth Γ must be tuned to achieve phase matching. This is accomplished by varying the gas density and delay between channel formation and injection. We have already demonstrated the ability to continuously tune Γ and control the mode structure [2]. In the SWL, the optimum inverse depth yielding $\Delta k = 0$ is $\Gamma_{\text{opt}} \approx (r\eta_1 - q\eta_2)/(q - r)$, where $r = m/n$ and $q = \omega_1/\omega_2 \gtrsim 1$. It is notable that for the case of $q = 1$, difference wave mixing can take place with *one color* if $\eta_1 \neq \eta_2$, or coupling occurs to more than one mode, where the subtracted photons are provided by the higher-order mode η_2 ($\omega = m\omega_1 - n\omega_1$ may be viewed as a higher-order process of harmonic generation than the lowest-order process $\omega = n\omega_1$). In this case, once the phase mismatch has been minimized via the tuning of Γ, ω_1 may be changed without affecting Δk, allowing for a tunable harmonic source.

Note that for phase matched difference wave mixing (either with tuned $\omega_{1,2}$ or tuned Γ), it is not necessary to have a deep channel (small Γ) or a high mode number η_2. Therefore, the transverse spatial overlap of P_{NL} with phase matched modes is better than that for the case of lowest-order harmonic generation.

For phase matched difference frequency generation over long channels (except for the same-mode case), the channel inverse depth Γ must not vary appreciably. In the SWL, the phase mismatch resulting from an average deviation $\Delta\Gamma$ along a channel of length L is $\Delta kL \approx -\Delta\Gamma L(m\lambda_1 - n\lambda_2)/\pi w_{\text{ch}}^2$. This gives approximately

$$\left|\frac{\Delta\Gamma}{\Gamma}\right| < \frac{2\pi}{|\eta_1 N_{R1} - \eta_2 N_{R2}|} \tag{10}$$

for the tolerable level of inverse depth variation. For example, if $\omega_1/\omega_2 \sim 1.8$, $N_{R1} \sim 25$ and $N_{R2} \sim 45$, with $\eta_1 \sim 1$ and $\eta_2 \sim 3$, it is required that $|\Delta\Gamma/\Gamma| < 0.05$. This sensitivity indicates the importance of producing a uniform plasma waveguide. Waveguide creation by ionizing a gas with a moderately short, intense laser pulse is well suited for this [3].

We have shown that the plasma waveguide can be used to achieve phase matching in both harmonic generation and difference wave generation (or parametric amplification). Lowest-order harmonic generation ($\omega = n\omega_1$) is phase matched by generating the harmonic in a higher waveguide mode than the driving wave. In difference wave generation ($\omega = m\omega_1 - n\omega_2$, where $m > n$ and $\omega_1 \gtrsim \omega_2$), phase matching is achieved by dividing the driving wave(s) over more than one guide mode or maintaining a common mode for all waves. This offers the possibility of significantly better spatial overlap of the driving polarization with the channel modes than in lowest-order harmonic generation. Because high-order processes at high intensity can occur with efficiency comparable to lower-order processes, difference wave generation may be the more efficient route to high-order frequency conversion.

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