Phase matching for large-scale frequency upconversion in plasma

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We consider phase matching in the generation of very short-wavelength coherent radiation by nonlinear frequency upconversion in plasma and suggest some ways to improve phase matching in high-order harmonic generation. We obtain what are to our knowledge the first simple analytical expressions for a phase-matching factor in multiphoton mixing of an arbitrary order and demonstrate theoretically that high-order difference-frequency mixing in plasma could be, owing to its potential for phase-matching optimization, a more promising method of large-scale frequency upconversion than high-order harmonic generation.

Recently the generation of short-wavelength coherent radiation by large-scale nonlinear frequency upconversion has attracted much attention since very high-order odd harmonics have been observed, with output frequencies well inside the soft-x-ray domain.[1] The conversion efficiency, however, is disappointingly low. One of the main reasons for this is ionization of nonlinear media by intense radiation of a pumping laser: free-electron dispersion leads to positive mismatch in high harmonic generation (HHG) and therefore drastically lowers the conversion efficiency. Although HHG may improve to some extent in the strong-field regime,[2] plasma remains an inherently hostile medium for this process. On the other hand, recent results[3] suggest that ions have some advantages over neutrals for generating very short-wavelength coherent radiation.

In this Letter we consider dispersion properties of plasma as a nonlinear medium for large-scale nonlinear frequency upconversion. We demonstrate theoretically that, although there are some ways to improve phase matching further in HHG, high-order difference-frequency mixing (HDM) in plasma is a more promising method for generating very short-wavelength coherent radiation since it allows one to employ large free-electron dispersion for phase-matching optimization. We obtain what is to our knowledge the first simple analytical expressions for phase-matching factors and optimal phase-matching conditions in HDM. At this stage, we limit our calculations to a perturbation approximation; the nonperturbative regime will be addressed by us elsewhere.

For frequency transformations in ions, the wavelength of the incident radiation λ is usually much larger than the wavelength λ_0 of the transition from the ground level of the ion to its first excited level. At the same time, excited ionic levels are almost empty in plasma of moderate density. This means that bound-electron refraction is negligible for the fundamental harmonic. The same holds for higher harmonics, except for a sufficiently close resonance. In particular, for the qth-harmonic generation, if only one ionic transition from the ground level to a bound level is near resonant to the qth harmonic $\lambda_q \approx \lambda/q$ (but the detuning is still much larger than the Doppler width), the phase mismatch can be written as

$$\Delta k_{\mathrm{HHG}} \approx r_e q \lambda N_e \left[1 - 2^{-1} q^{-2} (N_i/N_e) \frac{(g_1 f_{12}/g_2)}{1 - \lambda_q/\lambda_0} \right]. \tag{1}$$

Here $r_e = e^2/m_0c^2$ is the classical electron radius, N_i (N_e) is the ion (electron) density, f_{12} is the oscillator strength of the resonant transition, and g_1 (g_2) is the degeneracy of the ground (excited) level. Relation (1) is derived from the conventional expressions for a free-electron refractive index[4] $n_e \approx 1 - (r_e/2\pi)N_e\lambda^2$ and for a resonant refractive index[5] simplified for detuning larger than Doppler width.

For the off-resonance case, the second term in relation (1) is negligibly small, and phase mismatch is positive. In a tight-focusing geometry, $b \ll L$, where $b = 4A/\lambda$ is the confocal parameter of a Gaussian beam with an effective area A and L is the length of a plasma cell, positive phase mismatch drastically reduces the conversion efficiency in both the weak- and strong-field regimes. If a beam is focused inside a medium, perturbation theory predicts that the output tends to zero for $\Delta k \geq 0$ as $b/L \to 0$. Looser focusing, $b > L$, improves phase matching but makes it necessary to have either a short medium or a large beam area (i.e., low intensity); both these conditions result again in low conversion efficiency.

A few ways to circumvent the problem of HHG phase matching in plasma can be derived from the approaches used in nonlinear optics of gases.[6] One is to use ions with a transition from their ground level to an excited level being near resonant to the qth harmonics, so that $\lambda_q \lesssim \lambda_0$. Then, if

$$q^{-2}(N_i/N_e)(g_1 f_{12}/g_2)(1 - \lambda_q/\lambda_0) > 2, \tag{2}$$

plasma mismatch would be negative and, therefore, more favorable for HHG. Optimal phase mismatch $\Delta k_{\mathrm{opt}} = -2(q-2)/b$ (Ref. 6) (perturbation-theory re-

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sults assumed) would then take place if

$$2^{-1}q^{-2}(N_i/N_e)\frac{(g_1f_{12}/g_2)}{1-\lambda_q/\lambda_0} = 1 + \frac{2(q-2)}{br_eq\lambda N_e}. \quad (3)$$

Phase matching may also be attained by adding some buffer ions with a density N_{buf} determined by Eq. (3), where N_{buf} is used instead of N_i (for $q = 3$, see Ref. 7).

While resonant refraction is fairly efficient for not very large q ($q < 15$), its advantage diminishes for higher harmonics since optimal negative phase mismatch grows in modulus with q, thus making the requirements [relations (2) and (3)] more stringent. Moreover, using this approach, one may attain a favorable phase matching for only one harmonic at a time. To improve phase matching (to some degree) for a number of harmonics, one has to use semi-infinite media,[6] that is, to focus a driving laser at the edge of a sufficiently long medium. In this case, substantial (although much lower than optimal) output is feasible in the tight-focusing geometry even for positive mismatch.

So far, we have considered HHG. There are, however, nonlinear processes for which plasma dispersion can be favorable, in contrast to HHG, for which it is detrimental. In particular, from the point of view of phase matching, HDM in plasma for biharmonic pumping with two frequencies ω_1 and ω_2,

$$\omega = m\omega_1 - l\omega_2, \quad (4)$$

where m and l are integers, $m >> 1$, presents much better potentials for large-scale nonlinear upconversion of the frequency ω_1 than does HHG. Indeed, in the absence of resonances close to both incident and generated radiation, Δk for HDM is determined by free-electron dispersion only and can be written for collinear beams as

$$\Delta k \approx r_eN_em[\lambda_1 - (l/m)\lambda_2 - \lambda/m],$$
$$\lambda = \lambda_1\lambda_2/(m\lambda_2 - l\lambda_1). \quad (5)$$

By choosing (or tuning) the second laser as well as adjusting plasma density and the confocal parameters, one can in principle optimize phase matching [relation (5)] for both positive and negative optimal Δk. Moreover, since in most of the cases [see relations (13) and (14) below] the optimal $|\Delta k|$ is large for high-order mixing of focused beams, it may be necessary to have large dispersion to optimize phase matching. From now on we assume that $\omega_2 << \omega_1$ and $m > l$, so that shorter wavelengths could be attained by HDM of a given order $(m + l)$. (The same choice of frequencies is necessary if one wants to obtain short-wavelength output by mixing loosely focused beams, since the optimal Δk is close to zero for such a process.)

To our knowledge, no closed analytical expressions for HDM output have ever been published (Ref. 6 contains only some general suggestions regarding location of phase-matching optima, whereas the extensive general analysis in Ref. 8 yields complicated integrals in the complex plane). Following the procedure developed in Ref. 9, we have derived analytical expressions for the phase-matching factor determining the HDM output for tightly focused collinear beams in homogeneous media. These expressions are simple enough to allow for analytical approximations of optimal phase matching. We consider general multiphoton mixing,

$$\omega = \sum_{j=1}^{m}\omega_j - \sum_{j=1}^{l}\tilde{\omega}_j,$$

with all the beams being the first-order cylindrically symmetrical Gaussian beams propagating along the z axis with coinciding waist locations, so that their electric-field amplitudes can be written as[9]

$$E_j(x,y,z) = E_{j0}\exp(izk_j)(1 + i\epsilon_j)^{-1} \times \exp[-k_jR^2/b_j(1 + i\epsilon_j)]. \quad (6)$$

Here $R^2 \equiv x^2 + y^2$, $\epsilon_j = 2(z - f)/b_j$, and f is the z coordinate of the focus ($z = 0$ at the input window). For the sake of simplicity, let us consider first (similarly to Ref. 9) equal confocal parameters for all the beams; a more general expression will follow. According to the perturbation theory, the envelope of the driving polarization $P(x,y,z)$ at the output frequency ω is proportional to the product of driving electric-field amplitudes:

$$P(x,y,z) = S\chi N_i\prod_{j=1}^{m}E_j\prod_{j=1}^{l}\tilde{E}_j{}^*, \quad (7)$$

where the $\tilde{E}$'s correspond to frequencies $\tilde{\omega}$. The numerical factor S is determined by m, l and depends on the way the susceptibility χ is defined. Following the procedure,[9] one arrives at the general expression for the electric-field amplitude at ω:

$$E(x,y,z) = S\chi N_i\prod_{j=1}^{m}E_{j0}\prod_{j=1}^{m}\tilde{E}_{j0}{}^*(ik_0{}^2k^{-1}b)\exp(izk') \times \int_{-\zeta}^{\epsilon} d\epsilon' \frac{\exp[-(b\Delta k/2)(\epsilon - \epsilon')]}{(1 + i\epsilon')^{m-1}(1 - i\epsilon')^{l-1}(k'' - i\epsilon'k')H} \times \exp\left(\frac{-R^2}{bH}\right). \quad (8)$$

Here k_0 and k are the wave vectors (of the generated radiation) in vacuum and in a nonlinear medium, respectively, $k' = \sum_{j=1}^{m}k_j - \sum_{j=1}^{l}\tilde{k}_j$, $k'' = \sum_{j=1}^{m}k_j + \sum_{j=1}^{l}\tilde{k}_j$, $\Delta k = k - k'$, and $H = (1 + \epsilon'^2)(k'' - i\epsilon')^{-1} - i(\epsilon' - \epsilon)k'^{-1}$. We assume that plasma is contained in a cell of length L, so that $z = L$ and $\epsilon = 2(L - f)/b$ in Eq. (8). For the processes of interest for large-scale frequency transformations, $\sum_{j=1}^{m}k_j >> \sum_{j=1}^{l}\tilde{k}_j$, so that

$$k' \approx k''. \quad (9)$$

As a result, $H \approx (1 + i\epsilon)k'^{-1}$, and $E(x,y,z)$ corresponds, consistently with Ref. 9, to a lowest-order Gaussian beam. The intensity of this beam is obtained by evaluation of an integral

$\int_0^\infty 2\pi R|E(R,z)|^2 \, dR$ and is proportional to the phase-matching factor F,

$$F = \left| \int_{-\zeta}^{\epsilon} d\epsilon' \frac{\exp(-b\Delta k\epsilon'/2)}{(1 + i\epsilon')^{m-1}(1 - i\epsilon')^l} \right|^2 . \qquad (10)$$

F defined by Eq. (10) reduces to the respective phase-matching factors in Ref. 9 if $m + l = 3$.

A detailed analysis of the HDM phase matching factor (including its generalization to inhomogeneous media) is not given here. Here we consider only beams tightly focused ($b << L$) inside the cell. Then $\epsilon, \zeta \to \infty$, and the integral [Eq. (10)] can be solved in closed form by use of Ref. 10:

$$\begin{aligned} F_{r,s} &= u_{r,s}{}^2(b\Delta k), \quad \text{if } \Delta k > 0 , \\ &= u_{s,r}{}^2(b|\Delta k|), \quad \text{if } \Delta k < 0 , \end{aligned} \qquad (11a)$$

where

$$u_{r,s}(x) \equiv \frac{\exp(-x/2)\pi}{r!\,2^{r+s}} \sum_{k=1}^{s} \frac{(r+s-k)!}{(s-k)!\,k!} x^k ,$$
$$r \equiv m - 2, \quad s \equiv l - 1 . \qquad (11b)$$

In particular, if $l = 1$ [HDM, Eq. (4), with $l = 1$ could be called near-mth-harmonic generation, m even],

$$\begin{aligned} F_{r,0} &= \pi^2 2^{-2r} \exp(-b\Delta k), \quad \Delta k > 0 , \\ &= \pi^2 2^{-2r} \exp(-b|\Delta k|) \left[\sum_{k=0}^{r} (b|\Delta k|)^k / k! \right]^2 , \\ &\qquad \Delta k < 0 . \end{aligned} \qquad (12)$$

The maximal phase-matching factor $F_{r,0}$ corresponds to

$$(b\Delta k)_{\text{opt}} \approx -2r^2/(r + 1), \qquad r = m - 2 . \qquad (13)$$

For $r >> 1$, relation (13) is almost an equality and could be further simplified to $(b\Delta k)_{\text{opt}} \approx -2r$. If $l > 1$, the sign of Δk_{opt} depends on whether $r > s$ or $r < s$. For instance, if $r > s$, $\Delta k_{\text{opt}} < 0$, and for $r, s >> 1$,

$$(b\Delta k)_{\text{opt}} \approx -2(r - s) . \qquad (14)$$

The next, more accurate approximation is $(b\Delta k)_{\text{opt}} \approx -2(r - s)[1 - 1/4(s + 1)]$.

The phase-matching factor for HDM of beams with two different confocal parameters (b_1 and b_2 for ω_j and $\tilde{\omega}_j$, respectively) can now be obtained from Eqs. (11) by (i) adding the factor $(b_2/b_1)\phi^{r+s+1}$ to Eq. (11b), where $\phi \equiv (b_1 + b_2)/2b_1$; (ii) replacing $b\Delta k$ with $\phi b_1 \Delta k$ in the polynomials; and (iii) replacing b with b_1 in the exponent.

For an illustration, let us consider HDM of KrF 248.4-nm (ω_1) and Nd 1.064-μm (ω_2) laser beams with equal confocal parameters in plasma. For near-fourth-harmonic generation of KrF 248.4 nm (λ = 66 nm), relation (5) yields $\Delta k \approx -3.8 \times 10^{-18} N_e$ cm^{-1} and, therefore, close to the optimal product $b\Delta k \approx -3$ for $b = 1$ cm and $N_e \approx 1.2 \times 10^{19}$ cm^{-3}. The maximum of the phase-matching factor for HDM with $m = 66$, $l = 17$ ($\approx$4-nm output) corresponds to $b\Delta k \approx -95$, which leads through relation (5) to the optimal plasma density $N_e \approx 2 \times 10^{18}$ cm^{-3} for $b = 1$ cm.

In the loose-focusing limit, $b >> L$, where optimal Δk is equal to zero for any mixing, a CO_2 laser, instead of a Nd laser, can be used to optimize phase matching in KrF near-40th; near-42nd; and near-44th-harmonic generation in plasma, with the output wavelength as short as 5.6 nm.

It is worth noting that noncollinear phase matching is also theoretically possible for HDM in both tight- and loose-focusing geometries.

In conclusion, we derived phase-matching conditions for generation of very short-wavelength coherent radiation by means of large-scale nonlinear frequency upconversion in plasma. We suggested ways to improve phase matching in upconversion experiments already under consideration (high-order odd harmonic generation) and demonstrated that high-order difference-frequency mixing in plasma is a more promising method of large-scale frequency upconversion owing to its potential for phase-matching optimization.

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