Phase-matching optimization of large-scale nonlinear frequency upconversion in neutral and ionized gases

P. L. Shkolnikov, A. E. Kaplan, and A. Lago*

Department of Electrical and Computer Engineering, The Johns Hopkins University, Baltimore, Maryland 21218

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We suggest and discuss two techniques that could radically improve the efficiency of large-scale nonlinear frequency upconversion: quasi-phase matching of high-order harmonic generation in density-modulated media and high-order difference-frequency mixing in plasma. We demonstrate theoretically that both techniques permit optimal phase matching with tight focusing of pump beams, which is detrimental to current approaches to frequency upconversion, with a potentially drastic increase in conversion efficiency. Optimal phase-matching conditions that we obtained in the perturbation limit are shown to be likely to hold for strong pumping fields as well. © 1996 Optical Society of America

1. INTRODUCTION

Bright, short-wavelength (shorter than 100 nm), coherent radiation would find numerous applications in areas as different as cell biology and material science. Two sources of such radiation were discovered almost simultaneously: x-ray lasing (see, e.g., the review in Ref. 1 and references therein) and high-order harmonic generation (HHG).[2,3] Unfortunately, remarkable progress in x-ray laser (XRL) research, which resulted in, e.g., near-diffraction-limited operation of a Ge XRL at $\approx$23 nm (Ref. 4) and ~30 MW output at $\approx$15.4 nm of an YXRL,[5] has yet to produce an XRL available outside national laser facilities. At the same time, HHG in gas jets[3] has been used successfully in generating harmonics of orders as high as 135[6] and at wavelengths as short as 7.6 nm.[7] The harmonic energy is, however, severely limited by a very low conversion efficiency (defined as the ratio of the harmonic power to the incident power); as a result, a noticeable output requires a very high pumping energy (~1 J in a subpicosecond pulse[8]). Such a low efficiency (below 10^{-7} for the highest applied power to date[8] is, to a large extent, due to poor phase matching. If HHG of widely available short-pulse high-intensity lasers were phase matched, it could become a convenient tabletop source of coherent, easily tunable short-wavelength radiation.

It follows from both perturbation theory and nonperturbative models that current experimental conditions of HHG are very far from optimal with respect to phase matching[9]; under similar conditions third-harmonic generation (THG), e.g., would be characterized as a non-phase-matched process. So far, to our knowledge, the only experimental way to improve HHG phase matching has been to focus the pump beam more weakly (see, e.g., Ref. 10), which carries the inherent flaw of lowering the incident intensity and thus decreasing overall conversion efficiency. Moreover, this technique can in principle have only a very limited success in approaching optimum phase matching. Indeed, the qth-harmonic generation,

$$\omega_q = q\omega , \tag{1}$$

is optimally phase matched if the harmonic wave (at the frequency ω_q), while propagating in a medium, remains, on average, in phase with nonlinear polarization induced by the fundamental frequency ω. The phase of this polarization experiences a large shift that is approximately equal, in the perturbation limit for a Gaussian fundamental beam focused in the middle of a medium of length L, to

$$\Delta\phi_{\text{geom}} \approx 2(q-1)\tan^{-1}(2L/b) , \tag{2}$$

where b is the beam confocal parameter. This so-called geometrical, or diffractive, phase shift should be approximately offset by the dispersion phase mismatch:

$$\Delta\phi_{\text{disp}} = L\Delta k, \qquad \Delta k = k_q - qk , \tag{3}$$

where k_q and k are the wave vectors of the harmonic and the fundamental beams, respectively (see, e.g., Refs. 11 and 12). For this compensation to be possible, $\Delta\phi_{\text{disp}}$ has to be large and negative. The actual Δk value in HHG experiments, however, is positive: a small positive value if the medium is a neutral gas, or, more likely, a large positive value if the medium becomes ionized. Indeed, the free-electron refractive index n_e of radiation with a wavelength λ is given by

$$n_e \approx 1 - (r_e/2\pi)N_e\lambda^2 , \tag{4}$$

where r_e is the classical electron radius and N_e is the free-electron density (see, e.g., Ref. 13). The resulting free-electron phase mismatch for the qth-harmonic generation,

$$\Delta k = q r_e N_e \lambda , \tag{5}$$

completely dominates the dispersive phase mismatch for even a small fraction of ionized atoms (see, e.g., Ref. 14). The disparity between $\Delta\phi_{\text{geom}}$ and $\Delta\phi_{\text{disp}}$ becomes more severe with tight focusing ($b \ll L$), the major reason for the use, in current experiments, of as weak a focusing as permitted by the intensity requirements for HHG (intensity $I > 10^{13}$ W/cm^2). Even with $b \gg L$, however, the best result that one can attain is to reduce the required Δk to almost zero, with the actual Δk still being a large positive value.

A similar problem has been successfully dealt with in THG.[12,15] Some approaches developed for THG were discussed recently in terms of their application to HHG. Although useful in principle, however, they either would be of no help for harmonics beyond the thirteenth to fifteenth orders (as with the use of resonant refraction; see, e.g., Refs. 16 and 17) or would yield phase-matching factors many orders of magnitude lower than optimal (as with the use of semi-infinite media; see, e.g., Ref. 12).

In the present paper we discuss two techniques that we believe can be used to solve the problem of phase matching a large-scale nonlinear frequency upconversion in a radical manner: quasi-phase matching (QPM) of HHG in media with modulated density[18] and high-order difference-frequency mixing (HDM) of two lasers.[17] The most important advantage of the proposed methods is that they make it possible to use tightly focused beams, with a strong potential for a much higher conversion efficiency. The feasibility of optimal phase matching in both techniques is demonstrated by us in the perturbation limit, for laser fields much weaker than the atomic (ionic) electric field. We show, however, that our conclusions are likely to remain valid for strong pumping fields as well.

In the issues under consideration here we use several simplifying assumptions. First, throughout the paper, we assume that the laser beams whose frequency transformations are considered do not alter the electron density, the degree of ionization, or the ionization stage of the nonlinear medium. The plasma, therefore, should be prepared before the pump beams arrive, and the ionization potential of the plasma ions should be high enough to withstand the strong field of those beams. Such a medium could be, e.g., a plasma of He-like Li ions (ionization potential, ~75 eV), such as that used by a RIKEN group in their HHG research[19] and created by a long pulse from an auxiliary laser. In accordance with the HHG experimental conditions, we also assume that harmonics are generated by bound electrons; we therefore neglect harmonic generation by relativistic electrons,[20] which is, in any case, much weaker. Absorption at both the fundamental and the harmonic frequencies is neglected, as is the depletion of the pump beams.

This paper is structured as follows. Section 2 is devoted to the QPM optimization of HHG in neutral and ionized gases with a modulated density. In Section 3 we discuss HDM as a potentially better alternative to HHG, with respect to phase matching, for large-scale nonlinear frequency upconversion. Finally, Section 4 addresses the applicability of our results to strong pumping fields.

2. OPTIMAL QUASI-PHASE MATCHING FOR HIGH-ORDER HARMONIC GENERATION IN GASES AND PLASMA

Recently Rax and Fisch[21] suggested plasma density modulation as a method for optimizing phase matching for THG by relativistic plasma electrons. Their idea is essentially a ramification of the well-known nonlinear optics method of QPM first proposed in 1962 (Ref. 22) and extensively studied in the ensuing years—but almost exclusively for second-harmonic generation in solids (see, e.g., Ref. 23 and references therein). This lack of interest in QPM in higher-order (until recently, largely third-order) harmonic generation is most likely due to the existence of potentially much better and less cumbersome methods to optimize phase matching (see, e.g., Refs. 12 and 24). As we have already pointed out in Section 1, however, many phase-matching techniques successful in THG are of much less use for HHG, which makes QPM more attractive. At the same time the discussion in Ref. 21 is limited to the relativistic THG in plasma in the plane-wave approximation.

In the present paper, we consider QPM of HHG with a focused beam in a medium of plasma or a gas whose nonlinear susceptibility and refractive index are spatially modulated, particularly through medium density modulation, and demonstrate that QPM optimization is feasible with currently available laser and plasma technologies.[18] To obtain analytic results, we rely on perturbation-theory expressions for the phase-matching factor. Our results remain valid beyond the perturbation limits as well, if some general assumptions hold regarding the nonlinear polarization induced by a strong laser field (see Section 4). Accurate quantitative estimates of the improvement in HHG that is due to QPM require nonperturbative calculations of HHG in homogeneous media with a large negative $b\Delta k$ value; to our knowledge, no such calculations have yet been published. On the basis of a nonperturbative model described in Ref. 9, however, we might expect the QPM-optimized harmonic intensity to be as much as 2 orders of magnitude larger, with the available depth of plasma density modulation, than the output intensity attainable under currently employed techniques involving loose focusing and poor phase matching. An even more important advantage of QPM optimization is the opportunity to use the tight focusing deemed deleterious for HHG thus far. As a result, the intensities high enough for a strong single-atom response would be provided at a much lower incident power as compared with the loose-focusing regime and would increase the power conversion efficiency by many orders of magnitude.

In the perturbation limit the power of the qth harmonic generated by the lowest-order Gaussian fundamental beam, which propagates along the z axes in a homogeneous medium and is tightly focused ($b \ll L$) in the middle of the medium, is proportional to the phase-matching factor $|F_0|^2$ (see, e.g., Refs. 11 and 17):

$$F_0 = I(p) = \int_{-\infty}^{\infty} \mathrm{d}u \, \exp(iup) f_q(u), \quad f_q(u) = (1 + iu)^{1-q},$$
$$u = 2z/b, \quad p = b\Delta k/2. \tag{6}$$

Inasmuch as $I(p \geq 0) = 0$ and $|I(p)|^2$ is maximal at $p = -(q-2)$ (see, e.g., Ref. 12), harmonic generation is strongly inhibited in positively dispersive media ($\Delta k \geq 0$) such as rare gases and plasma. One should bear in mind though that Eq. (6) is a simplification; in reality, infinite limits should be replaced by finite numbers. As a result, perturbation theory permits some harmonic output even in positively dispersive media, although this output is low and drastically decreases with increasing Δk.

Equation (6), which is the basis of our QPM model, corresponds to the direct generation of the qth harmonic by the fundamental beam in the process $\omega_q = q\omega$. In perturbation theory the same frequency can also be generated by cascade processes, such as $\omega_{q-2} = (q - 2)\omega$, and then $\omega_q = \omega_{q-2} + \omega + \omega$. Beyond the perturbation limits, however, the distinction between direct and cascade processes becomes meaningless. At the same time, if some general assumptions hold, the phase-matching factor would still look similar to that in Eq. (6), so the optimal values of the parameter p would be approximately the same as for perturbation HHG (see Ref. 25 and Section 4 below). This is consistent with the experimental fact that both HHG by strong fields and THG by weak fields are inhibited in positively dispersive media in essentially the same way. In particular, HHG output drastically drops with tight focusing and with the onset of ionization, just as it would happen in THG.

A. Modulation of Nonlinear Susceptibility

In the derivation of Eq. (6), both the nonlinear susceptibility responsible for the qth-harmonic generation and the linear susceptibility that determines the refractive index are assumed to be independent of z. We begin our departure from Eq. (6) by modulating only the nonlinear susceptibility, with the refractive index remaining constant. One way to achieve this experimentally would be to shift the resonant level if the HHG is strongly enhanced because of a two-photon resonance; another possibility, coherent control (that is, control of the phase of the induced dipole moment by reshaping of the incident pulse) is suggested in Ref. 26. The relative mathematical simplicity of this case allows us to obtain optimal QPM conditions that are then shown to hold in density-modulated media as well.

More specifically, we assume that the nonlinear susceptibility responsible for the qth-harmonic generation, $\chi^{(q)}$, is spatially modulated as

$$\chi^{(q)}(u) = \chi_0^{(q)}[1 + A\cos(au)], \qquad a = \pi b/\lambda_m\,, \tag{7}$$

where λ_m and $A < 1$ are the modulation wavelength and the amplitude, respectively, and $\chi_0^{(q)}$ is the ambient (unperturbed) nonlinear susceptibility. Equation (7) provides maximum nonlinearity at $z = 0$, where the pumping field is maximal. In principle, the amplitude A may depend on q, but we omit this dependence from A for brevity.

Our goal is to determine what values of the modulation parameter a make it possible to generate harmonics efficiently in nearly dispersionless (rare-gas) or positively dispersive (plasma) media in which, because the parameter p is positive, $F_0 = 0$. The nonlinear susceptibility modulation, Eq. (7), results in the new factor, $[1 + A^{(q)}\cos(au)]$, that appears in the integrand of Eq. (6), so that F_0 is replaced by F_m:

$$F_m = I(p) + (A/2)[I(p + a) + I(p - a)]\,. \tag{8}$$

If $p \geq 0$, then $I(p) = I(p + a) = 0$, and the phase-matching factor $|F_m|^2 = (A^2/4)|I(p - a)|^2$ reaches its maximum, for a given p, at

$$a = a_{\text{opt}} = p + q - 2,$$
$$\lambda_m{}^{\text{opt}} \approx \pi b[(q - 2)(1 + B)]^{-1},$$
$$B = b\Delta k/[2(q - 2)]\,, \tag{9}$$

where $\lambda_m{}^{\text{opt}}$ is the optimal modulation wavelength. The dispersion of neutral noble gases, which are the media of choice for HHG experiments, is so low[27] that we can neglect B in $\lambda_m{}^{\text{opt}}$, and HHG will be optimally quasi-phase matched by adjustment of the modulation wavelength to only the geometry of the process:

$$\lambda_m{}^{\text{opt}} \approx \lambda_m{}^{\text{geom}} = \pi b/(q - 2)\,. \tag{10}$$

In plasma, in the absence of close ionic resonances to the fundamental or to the qth-harmonic frequencies, phase mismatch is determined largely by free-electron dispersion [relation (4)]. Two distinct QPM regimes in plasma can then be considered. The first is characterized by $B \ll 1$, which means that the geometrical contribution dominates the phase mismatch, as in rare gases. This is the case if a beam is focused to a small confocal parameter in a plasma that is not too dense. Optimal QPM conditions in this low-dispersion regime are determined by relation (10), so that the optimal modulation wavelength for a given q depends only on the confocal parameter, and not on the fundamental frequency or the plasma density. If, on the contrary, dispersive phase mismatch dominates, $B \gg 1$ (the high-dispersion regime), then

$$\lambda_m{}^{\text{opt}} = \lambda_m{}^{\text{disp}} \approx 2\pi/\Delta k \ll \lambda_m{}^{\text{geom}}\,. \tag{11}$$

As an illustration, consider QPM for the 51st harmonic of a Ti:sapphire laser ($\lambda = 0.8\ \mu$m), which is near the middle of the harmonic plateau in recent HHG experiments,[6,7] and assume that $b = 100\ \mu$m (instead of the ~1-mm value commonly used). In a plasma with $N_e \sim 10^{18}\ \text{cm}^{-3}$, $B \sim 0.1$; for this low-dispersion regime, relation (10) yields $\lambda_m{}^{\text{opt}} \approx 6.2\ \mu$m. In contrast, in a plasma with $N_e \sim 10^{20}\ \text{cm}^{-3}$, $B \sim 10$ and $\lambda_m{}^{\text{opt}} \approx 0.54\ \mu$m.

Relations (10) and (11) yield, generally speaking, a different optimal modulation wavelength for harmonics of different orders. One might, however, expect phase-matching curves (i.e., phase-matching factors versus $p = b\Delta k/2$) to be relatively broad for high-order harmonics. For instance, one can calculate, using Ref. 12, that the perturbative phase-matching factor for the 31st harmonic is only ≈10% smaller than its maximal value at p, which is optimal for the 29th harmonic. As a result, a given λ_m would yield a substantial phase-matching factor for a number of harmonics simultaneously, and, importantly, without the need for precise control of the modulation wavelength.

B. Density Modulation

Now let us assume that the nonlinear susceptibility modulation [relation (10)] is due only to the medium density modulation and is therefore accompanied by the refractive-index modulation:

$$n(u) = 1 + \tilde{n}[1 + A\cos(au)] = n_0 + \tilde{n}A\cos(au)\,, \tag{12}$$

where $n_0 = 1 + \tilde{n}$ is the ambient (unperturbed) refractive index and $\tilde{n}$ is proportional to the ambient medium density. Both A and a are obviously the same in Eqs. (7) and (12). In gases and in plasma, $\tilde{n}$ is usually very small ($\tilde{n} \ll 1$), so that modulation of the refractive index is very weak (smaller than $\tilde{n}$). If $\lambda \ll \lambda_m$ also, which is commonly the case for $\lambda_m{}^{\text{opt}}$ in low-dispersion regimes as well as in some high-dispersion regimes, then radiation propagation in the medium can be described by the approximation of geometrical optics[28]: propagation in an inhomogeneous medium is assumed to be approximately the same as in a homogeneous medium with a variable dielectric permittivity. This approximation is characterized, in particular, by the absence of Bragg reflection, which would substantially complicate the consideration (similar to the complications resulting from Fresnel reflection in QPM in solids[29]). As we do not intend to investigate fully HHG in density-modulated media but rather wish to identify some conditions favorable for QPM, we limit our consideration to this approximation, in which the situation is both the simplest and common. Then, as we show now, the refractive-index modulation [Eq. (12)] does not substantially affect the optimal QPM conditions [relations (11)]. Indeed, if (1) the lowest-order Gaussian (outside the nonlinear medium) beam is focused at the point $z = 0$ inside a nonabsorbing medium that begins at $z = -L$; (2) the refractive index of the medium is modified, $n = n(z)$; and (3) the approximation of geometrical optics is valid, then the laser field inside the medium can be expressed as

$$E(R, z) = E_0[n(z)D]^{-1} \exp[-k_0R^2/b_0D] \times \exp\left[i \int_{-L}^{z} k(z')\mathrm{d}z'\right],$$
$$D = 1 + 2ib_0{}^{-1}\left[\int_{-L}^{z} n^{-1}(z)\mathrm{d}z - L\right], \qquad (13)$$

where $R^2 = x^2 + y^2$ and E_0, b_0, and k_0 are the beam amplitude, the confocal parameter, and the wave number, respectively, outside the medium. Equations (13) are straightforwardly derived from a more general expression given in Ref. 30. The very weak dependence of the refractive index on z can be neglected almost everywhere in Eqs. (13), so that, e.g., $D \approx 1 + iu$. However, we retain the z' dependence in the final exponential term in $E(R, z)$ because this exponential term will eventually include Δk, whose dependence on z is as strong as that of $\chi^{(q)}$. Thus we arrive at an expression for the fundamental field in a modulated medium that differs from the ordinary lowest-order Gaussian beam only in that $\exp(ikz)$ is replaced with $\exp[i \int_{-L}^{z} k(z')\mathrm{d}z']$. Such an expression first appeared in Ref. 31 [relation (4)] and was used in numerous publications on HHG since then (see, e.g., Ref. 9), without, however, containing any references to the approximation of the geometrical optics implied. With this modified input, the procedure developed in Ref. 31 yields the phase-matching factor $|F|^2$:

$$F = [\chi_0{}^{(q)}]^{-1} \int_{-\infty}^{\infty} \mathrm{d}u\chi^{(q)}(u)\exp[-i(z\Delta k)]f_q(u),$$
$$\{z\Delta k\} \equiv \int_{-\infty}^{z} \mathrm{d}z'\Delta k(z') = p_0[u + A \sin(au)/a], \qquad (14)$$

where $p_0 = b\Delta k_0/2$ and Δk_0 is the phase mismatch resulting from the ambient dispersion. It follows from Eqs. (14), (12), and (7) that, to account for the modulated index of refraction, it is enough to replace the term pu in each integral I in Eq. (8) [see also Eq. (6)] with $p_0[u + A \sin(au)/a]$. All three integrals that are taken in symmetrical limits are real, so $F \approx 2(I_0 + I_{+1} + I_{-1})$, and

$$I_j = \int_0^{\infty} \mathrm{d}u \cos[(p_0 + ja)u + (q - 1) \times \tan^{-1} u + p_0A \sin(au)/a]f_q(u),$$
$$j = 0, \pm 1. \qquad (15)$$

For large q the factor $f_q(u)$ differs substantially from zero only for such a small value of u that $\tan^{-1} u \approx u$, so that

$$I_j \approx \int_0^{\infty} \mathrm{d}u\alpha_j f_q(u),$$
$$\alpha_j = \cos\{u(p_0 + ja + q - 1)[1 + \delta_j \sin(au)/(au)]\},$$
$$\delta_j = p_0\beta/(p_0 + ja + q - 1). \qquad (16)$$

Because $|\sin(au)/(au)| \le 1$ and $\delta_{+1} < \delta_0 < 1$, the coefficients at u in both α_0 and α_{+1} are positive, so that I_0 and I_{+1} disappear under the conditions of interest: $p_0 > 0$. The modulus of the remaining integral I_{-1} is approximately maximized at $a = a_{\text{opt}}$ [relations (9)], with p being replaced by p_0. Indeed, for $a' = p_0 + q - 1$,

$$I_{-1} \approx I_{-1}{}^{\text{opt}} \approx \int_0^{\infty} \mathrm{d}u \cos[(p_0A/a')\sin(a'u)]f_q(u). \qquad (17)$$

Because $p_0/a' < 1$ and $|\sin(a'u)| \le 1$, the argument of the cosine function in relation (17) is smaller than A. If the modulation is not too deep, e.g., if $A < 0.3$, which is quite realistic, the integral $I_{-1}{}^{\text{opt}}$ is equal, within a 4% error, to $\int_0^{\infty} \mathrm{d}uf_q(u)$, which in turn provides the upper limit for $|I_{-1}|$. Because $a' \approx a_{\text{opt}}$ for large q, one may conclude that optimal QPM conditions are almost independent of the modulation of the refractive index [Eq. (12)].

So far we have used perturbation-theory expressions for the phase-matching integral. In fact, however, it can be seen that we have utilized only the particular form of the phase factor in Eq. (6) and a fast decrease of the function $f_q(u)$ with increasing $|u|$. If the same conditions hold beyond the perturbation limits, which indeed seems to be the case (see Section 4 below), relations (9) are also valid for strong pumping. Then our consideration has been limited to tight focusing. It can be shown, however, that, for large q, relations (9) are an equally good approximation for the opposite limit of loose focusing, $b \gg L$, if λ_m, which is much smaller than b, is also much smaller than L. (In this regard, it is worth noting that the HHG phase-matching optima in the perturbation limit correspond to almost the same $b\Delta k \approx -2q$ (see, e.g., Ref. 12).

The most obvious and important advantage of QPM optimization is that it permits tight focusing otherwise detrimental for HHG in rare gases and plasma. The incident intensity could then easily be increased by 2 orders of magnitude for the same pumping power by, e.g., a simple change in the confocal parameter from the value of ~1 mm used now to the readily attainable 100 μm. Without a general theory of phase matching beyond the perturbation limits, or at least numerical simulations for a particular laser and a medium, it is impossible to calculate accurately the resulting increase in harmonic intensity. It is commonly assumed, however, that the intensity of high-order harmonics is approximately proportional to the 12th power of the incident intensity (see, e.g., Ref. 9). Moreover, QPM optimization may significantly increase the HHG conversion efficiency even for the incident intensity currently used. Indeed, a model (see Fig. 31 in Ref. 9) developed for HHG of a loosely focused incident beam allows one to assume a high-order harmonic conversion efficiency under the current experimental conditions to be 5 orders of magnitude lower than the conversion efficiency that might be optimized in a conventional way (that is, by the provision of $b\Delta k \approx -2q$ in a homogeneous medium). In contrast, the harmonic intensity at the QPM optimum, under otherwise equal conditions, differs by a factor of $A^2/4$ from conventionally optimized output, as one can see by comparing the first and the third integrals in Eq. (8). As a result, with the recently reported[32] plasma density modulation done by irradiation of a grating with a ruby laser ($A \approx 0.08$, $\lambda_m \approx 2$–$6\ \mu$m in a plasma with $N_e \sim 10^{18}\ \mathrm{cm}^{-3}$), QPM optimization might raise the harmonic intensity by a factor of $(A^2/4) \times 10^5 \approx 160$ for the same pumping intensity.

Note that QPM can also be applied to x-ray THG. Indeed, for a typical XRL wavelength $\lambda_{\mathrm{XRL}} \sim 200$ Å, a now feasible $b \sim 100\ \mu$m, and a reasonable plasma density of $< 10^{19}\ \mathrm{cm}^{-3}$, one can obtain $\lambda_m{}^{\mathrm{opt}} = \lambda_m{}^{\mathrm{geom}} \approx \pi b \sim 300\ \mu$m. With such an easily attainable modulation wavelength, QPM may appear as an attractive method to optimize XRL frequency tripling in plasma.

3. LARGE-SCALE NONLINEAR FREQUENCY UPCONVERSION BY HIGH-ORDER DIFFERENCE-FREQUENCY MIXING

As we pointed out in Section 2, even QPM, which is potentially the most promising method of HHG phase matching, would yield a phase-matching factor 2 orders of magnitude smaller than the factor attainable in media with negative dispersion. In other words, plasma remains an inherently hostile medium for harmonic generation as far as phase matching is concerned. At the same time, the substantial presence of free electrons in any experiment with nonlinear laser–matter interactions at high intensities is practically unavoidable. Moreover, theoretical models and recent experimental results[19] suggest that HHG in ions would yield a substantial output at a much shorter wavelength than does HHG in neutral atoms. It would therefore be much more advantageous to use plasma dispersion as an ally rather than to fight an uphill battle with it. The need for such an ally is also obvious from the necessity to compensate for the large geometrical mismatch by large dispersion. Plasma dispersion is large; unfortunately for HHG its sign is wrong. What we suggest is that, instead of HHG, another nonlinear effect, high-order difference-frequency mixing (HDM) in plasma, be used for large-scale nonlinear upconversion.[17] HDM is the process of generating coherent radiation at the frequency ω:

$$\omega = m\omega_1 - l\omega_2 , \tag{18}$$

where m and l are integers, $m \gg 1$, by laser beams with two substantially different frequencies ω_1 and ω_2, interacting in a nonlinear medium. We assume that $\omega_2 \ll \omega_1$ and $m > l$, so that shorter wavelengths can be attained by HDM of a given overall order $(m + l)$. In this section we demonstrate that HDM presents a much better potential for large-scale nonlinear upconversion of the frequency ω_1 than does HHG, in that HDM permits optimal phase matching in ionized media. Indeed, in the absence of close resonances to both incident and generated radiation, Δk for HDM is determined only by free-electron dispersion and can be written for collinear beams as

$$\Delta k \approx r_e N_e m[\lambda_1 - (l/m)\lambda_2 - \lambda/m] ,$$
$$\lambda = \lambda_1\lambda_2/(m\lambda_2 - l\lambda_1) . \tag{19}$$

Obviously, by choosing (or tuning) the second laser and/or changing the plasma density and the confocal parameters, one can in principle adjust the phase matching [relation (19)] to any sign or size of the optimal $b\Delta k$.

To transform these qualitative remarks into quantitative estimations of optimal media and laser parameters, we need the theory of phase matching, which would generalize to arbitrary orders the theory developed by Bjorklund[11] for third-order mixing. As with QPM for high-order processes, there was little interest in such a theory before HHG was discovered. (Reference 12 contains only some general suggestions regarding the locations of phase-matching optima, whereas an extensive general analysis in Ref. 31 yields complicated integrals in the complex plane.) In this section we derive analytical expressions for the phase-matching factor that determines the HDM output for tightly focused collinear beams in homogeneous plasma as well as analytical approximations of optimal phase-matching conditions.[17] The present results confirm that HDM can be optimally phase matched in ionized media. The experimental feasibility of HDM is obvious from numerous recent experiments with strong biharmonic pumping[33]; those experiments, however, have not addressed the problem of HDM phase matching.

A. High-Order Difference-Frequency Mixing Phase-Matching Factor

We consider general multiphoton mixing:

$$\omega = \sum_{j=1}^{m} \omega_j - \sum_{j=1}^{l} \omega_j , \tag{20}$$

where all the beams are the lowest-order Gaussian beams propagating along the z axis with coinciding waist locations, so their electric-field amplitudes can be written as[11]

$$E_j(x, y, z) = E_{j0} \exp(izk_j)(1 + iu_j)^{-1} \times \exp[-k_j R^2/b_j(1 + iu_j)] . \tag{21}$$

Here $R^2 \equiv x^2 + y^2$, $u_j = 2(z - f)/b_j$, and f is the z coordinate of the focus ($z = 0$ at the input window). For the sake of simplicity, and in a manner similar to that reported in Ref. 11, we first assume equal confocal parameters for all the beams; a more general situation is briefly addressed below. 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:

$$P(x, y, z) = S\chi N_i \prod_{j=1}^{m} E_j \prod_{j=1}^{l} \tilde{E}_j^{\,*}$$
$$= S\chi N_i \prod_{j=1}^{m} E_{j0} \prod_{j=1}^{m} \tilde{E}_{j0}^{\,*} \exp(ik'z)$$
$$\times \exp\left[\frac{R^2(iuk' - k'')}{b(1+u^2)}\right](1+iu)^{-m}(1-iu)^{-1}, \tag{22}$$

where the $\tilde{E}$'s correspond to the frequencies $\tilde{\omega}$; $\Delta k = k - k'$; k_0 and k are the wave vectors of the generated radiation in vacuum and in a nonlinear medium, respectively; and

$$k' = \sum_{j=1}^{m} k_j - \sum_{j=1}^{l} \tilde{k}_j, \qquad k'' = \sum_{j=1}^{m} k_j + \sum_{j=1}^{l} k_j. \tag{23}$$

The numerical factor S is determined by m and l and depends on the way the susceptibility χ is defined. Because neither S nor χ contributes to phase matching, we do not specify them further. Equation (22) represents a straightforward generalization of Eqs. (10) and (12) from Ref. 11. Following the procedure developed in Ref. 11 further, one arrives at the general expression for the amplitude of the electric field generated at the frequency ω:

$$E(x, y, z)$$
$$= S\chi N_i \prod_{j=1}^{m} E_{j0} \prod_{j=1}^{m} \tilde{E}_{j0}^{\,*}(ik_0^{\,2}k^{-1}b)\exp(izk')$$
$$\times \int_{-\zeta}^{u} du' \frac{\exp[-i(b\Delta k/2)(u-u')]}{(1+iu')^{m-1}(1-iu')^{l-1}(k''-iu'k')H}$$
$$\times \exp\left(\frac{-R^2}{bH}\right),$$
$$H = (1+u'^2)(k''-iu')^{-1} - i(u'-u)k'^{-1}. \tag{24}$$

We assume that the plasma is contained in a cell of length L, so that $z = L$, $\zeta = 2f/b$, and $u = 2(L-f)/b$ in the right-hand side of Eqs. (24). For the processes of interest for large-scale frequency transformations, $\sum_{j=1}^{m} k_j \gg \sum_{j=1}^{l} \tilde{k}_j$, so that

$$k' \approx k''. \tag{25}$$

As a result, $H \approx (1+iu)k'^{-1}$, and $E(x, y, z)$ corresponds, consistently with Ref. 11, to a lowest-order Gaussian beam. The intensity of this beam, obtained by evaluation of an integral $\int_0^\infty 2\pi R|E(R, z)|^2 \mathrm{d}R$, is proportional to the phase-matching factor $|F_{m,l}|^2$:

$$F_{m,l} = \int_{-\zeta}^{u} \mathrm{d}u' \frac{\exp[-ib\Delta k u'/2]}{(1+iu')^{m-1}(1-iu')^{l}}. \tag{26}$$

$|F_{m,l}|^2$ as defined by Eq. (26), reduces to the corresponding phase-matching factors given in Ref. 11 if $m + l = 3$.

Of main interest to us are beams that are focused tightly ($b \ll L$) inside the cell, so that we can assume that $u, \zeta \to \infty$ in Eq. (26). The resulting integral can be solved in closed form with Eq. 3.384.9 from Ref. 34 to yield

$$F_{m,l} = [\pi 2^{(3-m-l)}/(m-2)!](b|\Delta k|)^{(m+l-3)/2}$$
$$\times W_{(m-l-1)/2,-(m+l-2)/2}(b|\Delta k|), \qquad \Delta k < 0, \tag{27}$$

$$F_{m,l} = -[\pi 2^{(3-m-l)}/(l-1)!](b\Delta k)^{(m+l-3)/2}$$
$$\times W_{(l-m+1)/2,-(m+l-2)/2}(b\Delta k), \qquad \Delta k \geq 0, \tag{28}$$

where W stands for the Whittaker function. It is convenient to express W through the Laguerre polynomials[35]:

$$W_{a+d+1/2,d}(x) = (-1)^a x^{d+1/2} \exp(-x/2) a! L_a^{\,2d}(x). \tag{29}$$

(Please note that the corresponding Eq. 9.237.2 in Ref. 34 contains a misprint: $a!$ was omitted.) Using the following definition of the Laguerre polynomials (Ref. 34, Eq. 8.970.1):

$$L_n^{\,\alpha}(x) = \sum_{j=0}^{n} (-1)^j \binom{n+\alpha}{n-j} \frac{x^j}{j!}, \tag{30}$$

with the binomial coefficients

$$\binom{n+\alpha}{n-j}$$

being defined as[36]

$$\binom{j}{n} = \begin{cases} \dfrac{j(j-1)\ldots(j-n+1)}{n!} & n > 0 \\ 1 & n = 0 \\ 0 & n < 0 \end{cases}, \tag{31}$$

we finally arrive at the relatively simple polynomial expressions for the phase-matching factor for HDM of two tightly focused beams with equal confocal parameters b:

$$|F_{r,s}|^2 \begin{aligned} &= U_{r,s}^{\,2}(b\Delta k), \quad \text{if } \Delta k > 0, \\ &= U_{s,r}^{\,2}(b|\Delta k|), \quad \text{if } \Delta k < 0, \end{aligned} \tag{32a}$$

where

$$U_{r,s}(x) \equiv \frac{\pi \exp(-x/2)}{r!2^{r+s}} \sum_{k=0}^{s} \frac{(r+s-k)!}{(s-k)!k!} x^k,$$
$$r = m - 2, \qquad s \equiv l - 1. \tag{32b}$$

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

$$|F_{r,0}|^2 = \pi^2 2^{-2r} \exp(-b\Delta k), \qquad \Delta k \geq 0,$$
$$= \pi^2 2^{-2r} \exp(-b|\Delta k|)\left[\sum_{k=0}^{r} (b|\Delta k|)^k k!\right]^2,$$
$$\Delta k < 0. \tag{33}$$

B. Phase-Matching Optimization

Now we are ready to find the optimal phase-matching conditions for HDM, which correspond to the maxima of $|F_{r,s}|^2$. We shall do it for $s > 0$; the particular case of $s =$

0 yields the same conclusions in a similar way. First, let us show that there are no maxima at $\Delta k > 0$. Indeed, the condition of the maximum for $|F_{r,s}|^2$, $\mathrm{d}|F_{r,s}|^2/\mathrm{d}(b\Delta k) = 0$, leads to the equation

$$\tilde{f}_{>}(x) \equiv \sum_{k=0}^{s} \frac{(r+s-k)!}{(s-k)!}(r-s+k)\frac{x^k}{k!} = 0, \quad x = b\Delta k > 0. \tag{34}$$

It is easy to see that all the coefficients of the polynomial $\tilde{f}_{>}$ are positive for $r > s$; therefore Eq. (34) has no real roots, and the phase-matching factor has no maxima for $b\Delta k > 0$. In contrast, for $\Delta k < 0$, the condition of the maximum leads to

$$\tilde{f}_{<}(x) \equiv \sum_{k=0}^{r} \frac{(r+s-k)!}{(r-k)!}(s-r+k)\frac{x^k}{k!} = 0, \quad x = b|\Delta k| > 0. \tag{35}$$

The polynomial $\tilde{f}_{<}(x)$ contains only one change of sign and therefore, according to Descartes' rule,[37] has only one real root. The easiest way to approximate this root or, in other words, to find the optimal $b\Delta k$ is to use Eq. (26) directly. For infinite integration limits the integral

$$F_{m,l} = \int_{-\infty}^{\infty} \mathrm{d}u' \frac{\exp[-ib\Delta k u'/2]}{(1+iu')^{m-1}(1-iu')^{l}} \tag{36}$$

is real and can be simplified to

$$F_{m,l}(b\Delta k) = 2\int_{0}^{\infty} \mathrm{d}u \frac{\cos[b\Delta k u/2 - (r-s)\tan^{-1}(u)]}{(1+u^2)^{(r+s+2)/2}}, \tag{37}$$

where we replaced u' with u for convenience. Because $r + s \gg 1$ for HDM, the integral relation (38) is largely determined by contributions from $u < a \ll 1$, such that

$$\tan^{-1}(u) \approx u, \tag{38}$$

where a can be assumed to be as large as 0.4; then the error in Eq. (37) is less than 5%. In such an approximation,

$$F_{m,l} \approx 2\int_{0}^{a} \mathrm{d}u \frac{\cos[u(b\Delta k/2 - r + s)]}{(1+u^2)^{(r+s+2)/2}}. \tag{39}$$

Now, because

$$\cos(0) \geq \cos[u(b\Delta k/2 - r + s)] \tag{40}$$

for any $b\Delta k/2$ different from $r - s$, the integrand in relation (39) is maximal at each point in u for $b\Delta k = 2(r - s)$. Therefore the approximate condition of maximum for $f_{m,l}$ and the HDM phase-matching integral is

$$(b\Delta k)_{\mathrm{opt}} \approx -2(r-s). \tag{41}$$

Although approximation (41) was obtained for HDM, it is obvious from the outlined procedure that, for $s = 0$ and $r = q$, this relation is a good approximation for HHG, in full agreement with known results.[12] [This is not true, however, for Eqs. (32): no choice of r, s makes Eqs. (32) yield a HHG phase-matching factor.]

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 readily be obtained in a similar, although more cumbersome, way. The result is

$$\begin{aligned} |\tilde{F}_{r,s}|^2 &= (b_2/b_1)^{s+1}\tilde{U}_{r,s}{}^2, \quad \text{if } \Delta k > 0, \\ &= (b_2/b_1)^{s+1}\tilde{U}_{s,r}{}^2, \quad \text{if } \Delta k < 0, \end{aligned} \tag{42a}$$

where

$$\tilde{U}_{r,s} \equiv \left(\frac{b_1+b_2}{2b_1}\right)^{-(r+s+1)} \frac{\pi \exp(-b_2\Delta k/2)}{r!2^{r+s}} \sum_{k=0}^{s} \frac{(r+s-k)!}{(s-k)!k!} \times [(b_1+b_2)|\Delta k|]^k. \tag{42b}$$

For $m + l = 3$, more extensive results including a discussion of the influence of variation of the confocal parameters on the conversion efficiency can be found in Ref. 38.

Given relations (19) and (41), phase matching of a particular HDM process can be optimized as follows. For given values of m, l, the optimal $b\Delta k$ is calculated with Relation (41). Substituting the result into relation (19), one obtains the relation between the confocal parameter and the electron density required for optimal phase matching. Moreover, with phase-matching curves being quite broad, any given combination of N_e and b provides nearly optimal phase matching, not just for one, but for a number of m, l sets determined by relations (19) and (41) for large m, as follows:

$$l/m \approx (1+\beta)/(1+h\beta), \qquad \beta = (1/2)r_e N_e \lambda_1 \approx 1.4\tilde{b}\tilde{N}_e\tilde{\lambda}_1, \tag{43}$$

where $h = \lambda_2/\lambda_1$ and $\tilde{b}$, $\tilde{N}_e$, and $\tilde{\lambda}_1$ are in units of millimeters, $10^{18}\ \mathrm{cm}^{-3}$, and micrometers, respectively. One should then apply the ratio l/m, together with relation (19), keeping in mind that $l + m$ should be an odd integer. It is obvious from relation (43) that, for a given overall order of the process, the lowest l and therefore the shortest λ is attainable with the highest ratio h. In other words, from this point of view, the two pumping frequencies should be as far apart as possible.

In practical terms, the proof-of-principle experiments on HDM could utilize the fundamental and a harmonic of the same laser, although it may not be the best conceivable combination. To illustrate the numbers involved, let us consider HDM of the fourth ($\lambda_1 \approx 0.266\ \mu$m) and the fundamental ($\lambda_2 \approx 1.064\ \mu$m) harmonics of a Nd:YAG laser, using equal confocal parameters of 1 mm. Then, e.g., HDM with $l = 5$, $m = 18$ ($\lambda \approx 14.9$ nm) will be optimally phase matched in a plasma with $N_e \approx 9 \times 10^{18}\ \mathrm{cm}^{-3}$. For $l = 12$, $m = 43$ ($\lambda = 7.8$ nm), the optimal plasma density is $\approx 1.6 \times 10^{19}\ \mathrm{cm}^{-3}$. A more promising, and more difficult, proof-of-principle experiment could be on HDM of a harmonic and a Stokes wave of the same laser. In addition, it is worthy of note that noncollinear phase matching is also theoretically possible for HDM.

4. PHASE MATCHING OPTIMA BEYOND THE PERTURBATION LIMIT

Our conclusions on the feasibility and usefulness of HDM and QPM are based on relation (41) for the process of phase-matching optima. This relation, however, eventually relied on the perturbation-theory analytical expression for induced polarization as a function of the pumping fields [of the kind described by Eq. (22)]. It is not clear, therefore, whether relation (41) holds beyond the perturbation approximation. Unfortunately, the exact dependence of the induced harmonic polarization on strong pumping fields remains unknown. Moreover, although numerous models of the atomic response in strong-field HHG have appeared in recent years (see, e.g., Ref. 9 and references therein, as well as Ref. 39), no model calculations of the dipole moment induced by strong biharmonic pumping have been published yet. It may seem, therefore, that there is no way of knowing to what extent, if any, relation (41) is applicable beyond the perturbation limit. However, in this section we theoretically demonstrate that the optimal phase-matching conditions for both HHG and HDM do not depend, to a large extent, on a particular form of the induced dipole moment, as long as some very general assumptions are valid.[25] As a result, we expect the conditions for phase-matching optimization [relation (41)] to remain largely intact in strong pumping fields.

A. Model of High-Order Difference-Frequency Mixing Phase Matching for Strong Fields

We consider multiwave mixing [Eq. (18)] of the lowest-order Gaussian beams propagating along the z axis and focused at $z = 0$ to the same confocal parameters b (the typical situation for most of the experiments), so that the incident field can be written as

$$E = \tilde{E}_1(R, u)\cos(\omega_1 t + \phi_1) + \tilde{E}_2(R, u)\cos(\omega_2 t + \phi_2), \tag{44a}$$

where $\tilde{E}_{1,2}$ are the Gaussian profiles:

$$\tilde{E}_{1,2}(R, u) = E_{1,2}(1 + u^2)^{-1/2} \exp[-k_{1,2}R^2/b(1 + u^2)], \tag{44b}$$

and where

$$\phi_{1,2} = -k_{1,2}z + \tan^{-1} u - k_{1,2}R^2 u/b(1 + u^2), \tag{44c}$$

with $u = 2z/b$, $R = (x^2 + y^2)^{1/2}$, and $k_{1,2}$ being the wave vectors of the pump beams. A starting point of our consideration is a particular model suggested in Ref. 9 to explain some features of phase matching for strong-field HHG. For HHG in the weak-field limit [see, e.g., Eq. (22) with $l - 0$], the qth harmonic of the induced polarization P_q is

$$P_q = A_{\text{weak}}(R, u)\exp[-i\phi(R, u)],$$
$$A_{\text{weak}}(R, u) = A_0[\tilde{E}_1(R, u)]^q, \tag{45}$$

where A_0 contains the nonlinear susceptibility and other factors independent of R, u and is therefore irrelevant here. We thus omit A_0 from now on. The space-dependent part of A_{weak} is real. It is assumed in Ref. 9 that A_{weak} is transformed into the strong-field amplitude when q in A_{weak} is replaced with a smaller integer number. This substitution is performed to reflect the fact that the nonlinearity reaches some kind of saturation for strong pumping, and therefore the amplitude of the induced dipole moment varies more slowly with the amplitude of the incident field, as compared with the weak-field regime. The phase factor $\phi(r, u)$, on the contrary, is assumed (implicitly) to remain the same as the one for weak fields: $\phi = q\phi_1$. The resulting nonlinear polarization yields closely similar phase-matching factors of the harmonics of different orders for the actual experimental conditions, that is, for a loosely focused pump beam in dispersionless or positively dispersive media. In this model, just as in perturbation theory, such conditions correspond to the remote wings of the phase-matching factors.

In contrast to the study reported in Ref. 9, we are interested in phase-matching optimas for both HHG and HDM. Generalizing the model, we assume that the Fourier component of the induced nonlinear polarization responsible for HDM [Eq. (18)] of strong field is

$$P_{m,l} = P_0(\tilde{E}_1, \tilde{E}_2)\exp(-i\phi),$$
$$P_0{}^* = P_0,$$
$$\phi = -k'z + (m - l)\tan^{-1} u - uk'R^2/(1 + u^2),$$
$$k' = mk_1 - lk_2. \tag{46}$$

The space-dependent amplitude P_0, which is a real quantity $[E_1(R, u)]^m[\tilde{E}_2(R, u)]^l$ for weak fields [see Eq. (22)] is still real in Eqs. (45). This, in turn, means that we assume (as is done implicitly in Ref. 9) that the space-dependent phase of the induced dipole moment is the same, ϕ, for both weak and strong fields. (A similar suggestion was also made in Ref. 40 on the basis of a simple one-dimensional model of HHG, as well as on the basis of a quantum-mechanical model that treats the atomic field as a small perturbation.) In contrast to the authors of Ref. 9, however, we do not presume any particular expression for the amplitude P_0; in fact, our consideration does not require such an expression. Instead, it is enough for our conclusions that the real amplitude P_0 [Eqs. (46)] is a positive, monotonic, rapidly increasing function of $\tilde{E}_1$, $\tilde{E}_2$ (and therefore a rapidly decreasing function of R, u). A physical ground for such an assumption is the fact that the intensity of high-order harmonics rapidly increases, on average, with the intensity of the pumping. To be more specific, we neglect contributions to the generated field from the points of a medium at which the amplitude of the first (or, for HHG, the only) incident field is smaller than 85% of its maximal value $E_1 = \tilde{E}_1(0, 0)$; because the intensities of high-order harmonics inside the plateau are approximately proportional to the twelfth degree of the incident intensity,[9] we expect only a small fraction (≈2%) of the harmonic intensity to originate at these points. From Eqs. (44) we can see that the pumping amplitude drops below ≈85% of its maximal value if

$$\text{either } |u| > 0.6 \quad \text{for any } R$$
$$\text{or } R^2 > 0.16{w_2}^2 > 0.16{w_1}^2 \quad \text{for any } u, \tag{47}$$

where $w_1(w_2)$ is the spot size of the first (second) beam, ${w_{1,2}}^2 = b/k_{1,2}$. Also, because $\tilde{E}_1$, $\tilde{E}_2$ are symmetrical in u, so is p.

Regardless of the intensity of the incident fields, we can write the electric field $\mathscr{E}(R', z')$ generated in a homogeneous medium and observed at a point (r', z') outside it as[9]

$$\mathscr{E}(R', z') \sim \int \frac{\tilde{P}(R, z)\exp(-iz\Delta k)}{z' - z} \exp\left[\frac{ik(R^2 + R'^2)}{z' - z}\right] \times J_0\left(\frac{kR'R}{z' - z}\right) R\mathrm{d}R\mathrm{d}z\,, \tag{48}$$

where J_0 is the ordinary Bessel function of the zeroth order, $\tilde{P}(r, z)$ stands for the Fourier component $P_{m,l}$ without the phase factor $\exp(ik'z)$, $\Delta k = k - k'$, $k' = mk_1 - lk_2$, and k is the wave vector of the generated field; the integral is taken over the entire nonlinear medium. Substituting the polarization from Eqs. (46), we obtain

$$\mathscr{E}(R', u') \sim \int \exp\left\{-i\left[b\Delta ku/2 + (m - l) \times \tan^{-1} u - \frac{k'R^2u}{b(1 + u^2)} - 2\frac{k(R^2 + R'^2)}{b(u' - u)}\right]\right\} \times \frac{p(R, u)}{u' - u} J_0\left[2\frac{kR'R}{b(u' - u)}\right] R\mathrm{d}R\mathrm{d}u\,, \tag{49}$$

where $u' = 2z'/b$. Seeking the solution in the far-field area,

$$u' \gg 1, \qquad u' \gg u\,, \tag{50}$$

we may neglect u as compared with u' in Eq. (8) and move $\exp(2ikR'^2/bu')$ out of the integrand. The remainder of the fourth term in the exponent, $2kR^2/bu'$ in relation (49), is much smaller than 1 for a sufficiently large u'. However, it cannot simply be ignored because for very small u ($u < 1/u'$) it is larger than the third term; thus we retain this remainder, but in the form $\exp(2ikR^2/bu') \approx 1 + 2ikR^2/bu'$. Because $k' < mk_1$ and $u/(1 + u^2) < u$, it follows from conditions (47) that the third term in the exponent, relation (49), is smaller than 12% of the second term, and we neglect the former. Furthermore, as the integrand in relation (49) is now symmetrical in u, we may replace the exponent, relation (49), with the cosine function. Finally, note that for $|u| < 0.6$, $\tan^{-1} u \approx u$ to within 5% accuracy. All these approximations yield

$$|\mathscr{E}(R', u', M)| \sim [\mathscr{E}_1{}^2 + \mathscr{E}_2{}^2]^{1/2},$$
$$\mathscr{E}_1 = \int_0^{0.6} \cos(Mu)F_1(u)\mathrm{d}u,$$
$$\mathscr{E}_2 = \int_0^{0.6} \cos(Mu)F_2(u)\mathrm{d}u,$$
$$M = b\Delta k/2 + (m - l)\,, \tag{51}$$

where

$$F_1(u) \approx \int \frac{P_0(R, u)}{u'} J_0\left(2\frac{kR'R}{bu'}\right) R\mathrm{d}R,$$
$$F_2(u) \approx \int \frac{2kR^2P_0(R, u)}{bu'^2} J_0\left(2\frac{kR'R}{bu'}\right) R\mathrm{d}R\,.$$

Because of conditions (47), $kR^2/b \sim (m - l)$, so that $F_2 \approx [(m - l)/u']F_1$, and we can neglect F_2 in $|\mathscr{E}(R', u', M)|$, relation (51), for $z' \gg (m - l)b$.

To proceed further, we now show that, if a positive function $f(x)$ does not increase in the interval $[0, C]$, then

$$\int_0^C f(x)J_j(ax)\mathrm{d}x > 0, \qquad j = 0, 1, 2, \ldots\,. \tag{52}$$

for $a > 0$, $C > 0$. Indeed, it is known that the graph of $J_j(x)$ for $j = 0, 1, 2, \ldots$ and for $x \geq 0$ resembles the graph of dumped oscillators. The successive areas of half-waves that lie above and below the x axes form a decreasing sequence [Ref. 41, Chap. 7.9]. In other words,

$$\int_0^{x_1} J_j(x)\mathrm{d}x > \int_{x_1}^{x_2} |J_j(x)|\mathrm{d}x > \int_{x_2}^{x_3} J_j(x)\mathrm{d}x > \int_{x_3}^{x_4} |J_j(x)|\mathrm{d}x \ldots\,, \tag{53}$$

where $x_1, x_2, x_3, \ldots$ are consecutive zeros of $J_j(x)$. Let us first make a replacement $x/a \to x$ in relation (52). It follows from relations (51) and the assumed properties of $f(x)$ that

$$\int_0^{x_1} f(x/a)J_j(x)\mathrm{d}x > \int_{x_1}^{x_2} f(x/a)|J_j(x)|\mathrm{d}x > \int_{x_2}^{x_3} f(x/a)J_j(x)\mathrm{d}x > \int_{x_3}^{x_4} f(x/a)|J_j(x)|\mathrm{d}x \ldots\,,$$

so that

$$\int_0^{x_1} f(x/a)J_j(x)\mathrm{d}x - \int_{x_1}^{x_2} f(x/a)|J_j(x)|\mathrm{d}x + \int_{x_2}^{x_3} f(x/a)|J_j(x)|\mathrm{d}x - \ldots \int_{x_m}^{aC} f(x/a)J_j(x)|\mathrm{d}x = \int_0^{aC} f(x/a)|J_j(x)|\mathrm{d}x > 0\,.$$

The validity of relation (52) is thus obvious.

It follows then from relation (52) and from the assumed properties of P_0 that $F_1(u) > 0$ for any u. Now, because $\cos(0) \geq \cos(Mu)$ for any u nonzero M, both $|\mathrm{Re}\mathscr{E}|$ and $|\mathrm{Im}\mathscr{E}|$, and therefore the generated field amplitude $|\mathscr{E}|$, are maximal at $M = 0$. Hence the optimal product of the confocal parameter and the dispersion-phase mismatch is close to

$$(b\Delta k)_{\mathrm{opt}} = -2(m - l)\,, \tag{53}$$

which corresponds to the optimal phase matching of HDM in perturbation theory, relation (41).

We can roughly estimate an additional error that may be brought in by the approximations that we used to implement our assumption regarding the function P_0. Neglecting u as compared with u' should not lead to a substantial distortion in the far-field area. Then, replacing $\tan^{-1} u$ with u, we might overestimate $(b\Delta k)_{\mathrm{opt}}$ by as much as 10%, the largest difference between u and $\tan^{-1} u$ at the interval 0–0.6. At the same time, neglecting the third term in the exponent of relations (50) may introduce an error of the same magnitude but in the opposite direction. Therefore we expect relations (50) to reflect optimal phase-matching conditions with an accuracy of ≈10%. Because, at least in the weak-field limit, the phase-matching factor is quite broad for higher orders of interaction,[12] relation (53) may be a good approximation, provided that our general assumptions are justified.

In the loose-focusing limit, $b \gg L$, we can drop any special restrictions of u. Indeed, in such a limit $|u| \ll 1$ within the entire medium, so that the integral in relations (51) could be extended to the full medium length, with relation (53) again being the outcome.

B. Limits of the Model's Validity

Now the question is, Under what conditions can our basic assumptions [Eqs. (46) with the assumed monotonic behavior of P_0] be justified? First, Eqs. (46) are valid if the induced nonlinear polarization $P(x, y, z, t)$ is a (continuous) function of the incident field:

$$P(x, y, z, t) = f[E(x, y, z, t)], \tag{54}$$

in other words, for spatial and temporal localities of the induced polarization and for nondepletion of the pumping. One may expect Eq. (54) to hold for pump pulses that are much longer than the relaxation times of the nonlinear media; pumping frequencies that are far from any resonances, including intensity-induced resonances[39]; and the generated field that is so weak that the induced polarization is largely determined by the pumping field. Then, in the simpler case of the qth-harmonic generation ($l = 0$ and $m = q$) with a monochromatic pumping field, $f = f(\mathbf{E}_1)$ is periodical in time with the period $2\pi/\omega_1$, so that

$$\begin{aligned} P_q &= \frac{\omega_1}{\pi} \int_{-\pi/\omega_1}^{\pi/\omega_1} \mathrm{d}t f[\tilde{E}_1 \cos(\omega_1 t + \phi_1)]\exp(iq\omega_1 t) \\ &= P_0 \exp(-iq\phi_1), \end{aligned} \tag{55}$$

where $P_0 = (\omega_1/\pi) \int_{-\pi/\omega_1}^{\pi/\omega_1} \mathrm{d}t f[\tilde{E}_1 \cos(\omega_1 t)]\cos(q\omega_1 t)$ is real, and the phase factor $\exp(-iq\phi_1)$ is the same as that for weak fields. The last equality in Eq. (54) is obtained by the time shift $t \rightarrow t - \phi/\omega_1$.

With two pumping frequencies present, there is no common time scale, and a time shift cannot be applied. Instead, we use a well-known mathematical result: any continuous on some finite interval function can be approximated on this interval, to any required accuracy, by the sum $S_n = \sum_{k=1}^{n} a_k \Psi_k(x)$, where Ψ_k are orthonormal polynomials (Ref. 41, Chap. 10). Therefore $f(E)$, $E = \tilde{E}_1 \cos(\omega_1 t + \phi_1) + \tilde{E}_2 \cos(\omega_2 t + \phi_2)$, can be approximated by such a sum and can then be transformed into the expression

$$S_n = \sum_{j_1 j_2} s_{j_1 j_2} = \sum_{j_1 j_2} A_{j_1 j_2} \cos(j_1\Phi_1 - j_2\Phi_2), \tag{56}$$

where $\Phi_{1,2} = \omega_{1,2}t + \phi_{1,2}$, the real amplitudes A_{j_1,j_2} are sums of products of various degrees of $\tilde{E}_1$ and $\tilde{E}_2$, and j_1, j_2 are integers (positive or negative). Therefore any term $s_{j_1 j_2}$ that contains a given combination of frequencies, $\omega_{ml} = m\omega_1 - l\omega_2$, also contains the phase $\phi_{ml} = m\phi_1 - l\phi_2$: $s_{j_1,j_2} = A_{ml} \cos(\omega_{ml} + \phi_{ml})$, so that the Fourier transform of s_{j_1,j_2} is

$$P_{m,l} = P_0 \exp[-i(m\phi_1 - l\phi_2)], \quad P_0 = A_{ml}(\tilde{E}_1, \tilde{E}_2), \tag{57}$$

from which the validity of Eqs. (46) follows.

In real experiments, however, one can hardly expect Eq. (54) to be strictly correct. Consequently, any deviation from Eq. (54) would be reflected in P_0's being a complex, and not a real, function with an amplitude-dependent (and therefore coordinate-dependent) phase $\Psi(\tilde{E}_1, \tilde{E}_2)$. Nevertheless, this phase would not substantially affect relation (49) if the change of Ψ within the interval of interest [conditions (47)] is much smaller than the respective change of the geometrical phase $(m - l)u$. If $|P_0|$ also drops rapidly and monotonically with $|\tilde{E}_1|$, relation (53) can still hold. Because of the lack of calculations for the single-atom response to strong biharmonic pumping, we can try to verify these assumptions only for HHG. The three models of HHG[39,42,43] that have been most successfully compared with experimental data seem to show a sufficiently small variation of the phase Ψ. Indeed, e.g., the intensity-dependent phase of the dipole moment of the 51st harmonic, calculated in Ref. 5 according to the model described in Ref. 12, changes by only $\sim 4\pi$ within the interval of interest, $|u| = 0-0.6$, whereas the change of the geometrical phase qu is $0.6q$, which is much larger. Although published results of a numerical model[43] do not include Ψ explicitly, in all the theoretical considerations this phase is assumed to be constant (see, e.g., Ref. 44), or at least not to interfere strongly with phase matching.[9] Finally, the nonperturbative two-level model of HHG[39] predicts the intensity-dependent phase, which is almost constant between the intensity-induced resonances, and experiences a jump by π at such resonances; obviously, such behavior allows one to neglect this phase in relation (49). (We have already mentioned yet another model 40 that essentially presumed $\Psi = 0$.) One therefore may conclude that the phase Ψ could be compensated for by an additional relatively [as compared with $(m - l)u$] small change in the dispersive phase mismatch, which leaves relation (53) still valid as a reasonable approximation.

As for the modulus of the dipole moment $|P_0|$ in Ref. 5, as well as in the simplified quantum-mechanical model published recently,[45] it behaves in a desirable manner up to the cut-off intensity. After that, in both the numerical model[43,45] and the two-level[39] model, $|P_0|$ experiences sharp resonancelike jumps, so that within some intervals of the incident intensity it may even be a decreasing function. Such intervals, however, are relatively small, so that for most values of the pumping intensity and on average the amplitude of the dipole moment *is* a monotonic and rapidly increasing function of the pumping intensity, in accordance with the experimental dependence of the output on the incident field. Moreover, in all the experiments on HHG and HDM, the pumping field is in the form of a pulse, so its intensity at any given point changes in time. Obviously, for most of the pulse duration we should expect p to be smaller at the points in space at which the pumping intensity is lower.

To the best of our knowledge, no calculations of the amplitude or the phase of the induced dipole moment in HDM have been published yet. If our general assumptions also hold for this process (some preliminary calculations for a one-dimensional model of the atom response to biharmonic pumping do show small Ψ variations[46]), the optimal phase-matching conditions [Eq. (18)] estimated within the perturbation-theory limits would present a highly accurate approximation for nonperturbative HDM as well. One should keep in mind, though, that the same optimal phase matching determined by relation (2) may imply very different laser and media parameters, such as confocal parameters and media density, for low and high intensities, e.g., those due to potentially different dispersion properties of media in weak and strong laser fields.

5. CONCLUSION

In the present paper we discuss the reasons underlying poor phase matching in experiments on high-order harmonic generation and suggest two techniques to improve radically the efficiency of large-scale frequency transformation: quasi-phase matching of high-order harmonic generation in density-modulated plasma, and high-order difference-frequency mixing in plasma. We derive, in the perturbation limit, analytical expressions for phase-matching factors and optimal phase-matching conditions for both processes. We demonstrate theoretically that both techniques permit optimal phase matching for nonlinear transformations of tightly focused beams in neutral and especially ionized gases, which is detrimental to current approaches to frequency upconversion, with a potential increase in the conversion efficiency by several orders of magnitude. Some suggestions are made for proof-of-principle experiments with available laser and plasma technologies. Optimal phase-matching conditions that we obtained in the perturbation limit are shown to be likely to hold for strong pumping fields as well.

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*Permanent address, Depto de Engenharia Mecanica, Universidado Federal de Santa Catarina, Florianapolis, SC 88040-900, Brazil.

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