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# Optimal quasi-phase-matching for high-order harmonic generation in gases and plasma

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We theoretically demonstrate the feasibility of optimal quasi-phase-matching (QPM) of high-order harmonic generation in gases and plasma with modulated density. QPM optimization, being possible for both tight and loose focusing of the fundamental beam, may increase the conversion efficiency of high-order harmonic generation by several orders of magnitude as compared to the efficiency attainable now.

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High-order harmonic generation (HHG), discovered recently [1], if phase matched, may provide an important and convenient (in principle, table-top) source of short-wavelength coherent radiation. So far, however, HHG phase matching remains poor. It follows from both perturbation theory and nonperturbative models that current experimental conditions correspond to far wings of the phase-matching curve [2]; in fact, under similar conditions, e.g., third-harmonic generation would be characterized as a non-phase-matched process. The only experimental way to improve HHG phase matching has been the weaker focusing of the pumping beam (see, e.g., [3]). An inherent disadvantage of this method is that it lowers the incident intensity, thus decreasing overall power conversion efficiency. Moreover, this technique can have only a limited success in approaching optimum phase matching. Indeed, the $q$th harmonic generation is optimally phase matched if the harmonic wave, while propagating in a medium, remains in phase with nonlinear polarization induced by the fundamental. The phase of this polarization experiences a large shift approximately equal, in the perturbation limit for a Gaussian fundamental beam, to $q \tan^{-1}(2z/b)$, where $b$ is the beam confocal parameter (the distance from the focal plane to the point where beam intensity drops by half), and $z$ is the propagation distance. This so-called geometrical, or diffractive, phase shift should be offset by a large negative dispersion phase mismatch $\Delta k$ (see, e.g., [4(a),5]). At the same time, actual $\Delta k$ is positive: small positive if a medium for HHG is a neutral gas, or, more likely, large positive when this gas becomes ionized. Some other possibilities for improving HHG phase matching discussed recently would either be of no help for very-high-order harmonics (like using resonant refraction; see, e.g., [4(b)]), or would yield phase-matching factors many orders of magnitude lower than optimal ones (like using semi-infinite media; see, e.g., [5,6]).

Recently, Rax and Fisch [7] have suggested plasma density modulation as a method to phase match third-harmonic generation by relativistic plasma electrons. Their idea is essentially a ramification of the well known in nonlinear optics method of quasi-phase-matching (QPM) proposed first in 1962 [8] and extensively studied in the following years (see, e.g., [9] and references therein). Almost all the effort in this area, however, has been concentrated on the second-harmonic generation in solids (see, e.g., [10]) since obviously much more convenient ways to optimize higher-order harmonic generation (until recently—almost exclusively, third-harmonic generation) in gases exist. As a result, to the best of our knowledge, no general consideration of quasi-phase-matching in gases and plasma, in particular for HHG, has yet been published (Ref. [7] is limited to the relativistic third-harmonic generation in plasma in the plane-wave approximation).

In the present paper, we consider quasi-phase-matching of HHG by a focused beam in plasma or a gas whose nonlinear susceptibility and refractive index are spatially modulated in particular through the medium density modulation, and demonstrate that QPM is feasible with the available laser and plasma technology. In accordance with the HHG experimental conditions, we assume that harmonics are generated by bound electrons. Absorption at both fundamental and harmonic frequencies is neglected, as is the pumping beam depletion. We assume that the pumping beam does not change the medium dispersion. Therefore, a plasma medium should be prepared before the pumping pulse comes, and the ionization potential of the plasma ions should be high enough to prevent substantial additional ionization during HHG. In order to obtain analytical results, we rely on the perturbation-theory expressions for the induced nonlinear polarization. Our results remain valid beyond perturbation limits if some general assumptions hold regarding nonlinear polarization induced by a strong laser field [11]. Accurate quantitative estimates of the improvement in HHG due to QPM require nonperturbative calculations of HHG in homogeneous media with large negative $b\Delta k$; to our knowledge, no such calculations have yet been published. On the basis of a nonperturbative model described in Ref. [2], however, one might expect QPM-optimized harmonic intensity to be up to two orders of magnitude larger, with available depth of plasma density modulation, than the output intensity attainable under currently employed loose focusing and poor phase matching. Even more important an advantage of QPM optimization is the opportunity to use tight focusing that has so far been deleterious for HHG; resulting much higher incident intensities would increase power conversion efficiency by many orders of magnitude as compared to the loose-focusing regime.

The power of the $q$th harmonic generated by the lowest-order Gaussian fundamental beam, which propagates along

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the $z$ axes in a homogeneous medium and is tightly focused with the conformal parameter $b$ at $z=0$, is proportional to the phase-matching factor $|F_0|^2$, where (see, e.g., [4])

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

$\Delta k=k_q-qk$ is the dispersive phase mismatch, $k$ is the wave vector of the incident beam, and $k_q$ is the harmonic wave vector. Integral $F_0$ corresponds to direct generation of the $q$th harmonic by the fundamental beam, $\omega_q=q\omega$. Generally speaking, so-called cascade mixings could also generate the $q$th harmonic; e.g., $\omega_{q-2}=(q-2)\omega$ and then $\omega_q=\omega+\omega+\omega_{q-2}$. Beyond perturbation limits, however, the distinction between direct and cascade processes becomes meaningless; then, if some general assumptions hold, the phase-matching factor could again be written, similarly to Eq. (1), in such a way that the optimal parameter $p$ is approximately the same as for perturbation HHG [11].

## MODULATION OF NONLINEAR SUSCEPTIBILITY

We begin with the case where only nonlinear susceptibility is modulated. Experimentally, it could be achieved, e.g., by shifting the resonant level if HHG were to be strongly enhanced due to a two-photon resonance; another possibility, "coherent control" (that is, control of the phase of the induced dipole moment by changing parameters of the incident pulse), is suggested in Ref. [13]. The relative mathematical simplicity of this case will allow us to obtain optimal quasi-phase-matching conditions which then will be shown to be valid in density modulated media as well. More specifically, we assume the nonlinear susceptibility that is responsible for the $q$th harmonic generation $\chi^{(q)}$ to be spatially modulated as

$$\chi^{(q)}(u)=\chi_0^{(q)}[1+A\ \cos(au)], \quad a=\pi b/\lambda_m, \tag{2}$$

where $\lambda_m$ and $A<1$ are the modulation wavelength and amplitude, respectively, $\chi_0^{(q)}$ is the ambient (unperturbed) nonlinear susceptibility. Equation (2) provides maximum nonlinearity at $z=0$, where the pumping field is maximal. (If $\chi^{(q)}$ is modulated by either of the two methods just mentioned, the amplitude $A$ may depend on $q$.) Now our goal is to determine the magnitudes of the modulation parameter $a$ which make it possible to generate harmonics efficiently in almost dispersionless (rare gas) or positively dispersive (plasma) media where, due to the parameter $p$ being positive, $F_0=0$. The nonlinear susceptibility modulation, Eq. (2), results in the new factor $[1+A^{(q)}\cos(au)]$ appearing in the integrand of Eq. (1), so that $F_0$ is replaced by $F_m$,

$$F_m=I(p)+(A/2)[I(p+a)+I(p-a)]. \tag{3}$$

It is well known that $I(p\geqslant 0)=0$, and $|I(p)|^2$ is maximal at $p=-(q-2)$ (see, e.g., [5]). In the media of interest to us, $p\geqslant 0$, so that $I(p)=I(p+a)=0$, and the phase-matching factor $|F_m|^2=(A^2/4)|I(p-a)|^2$ reaches its maximum for a given $p$ at

$$a=a_{\text{opt}}=p+q-2, \quad \lambda_m^{\text{opt}}\approx \pi b[(q-2)(1+B)]^{-1},$$
$$B=b\Delta k/2(q-2), \tag{4}$$

where $\lambda_m^{\text{opt}}$ is the optimal modulation wavelength. The dispersion of neutral noble gases, which are the media of choice for the experiments of HHG, is so small (see, e.g., [14]) that we can neglect $B$ in Eq. (4), and HHG will be optimally quasi-phase-matched at

$$\lambda_m^{\text{opt}}\approx\lambda_m^{\text{geom}}=\pi b/(q-2). \tag{5}$$

In plasma, in the absence of resonances close to either the fundamental or the $q$th harmonic frequencies, $\Delta k\approx r_eN_eq\lambda$, where $r_e$ is the classical electron radius, $N_e$ is the plasma electron density, and $\lambda$ is the wavelength of the incident beam. Two distinct QPM regimes in plasma can be considered. The first is characterized by $B\ll 1$, which means that the geometrical phase mismatch dominates the dispersive one (such a relation holds in rare gases). It is the case if a beam is focused on a small confocal parameter in a not too dense plasma. Optimal QPM conditions in this "low-dispersion" regime are determined by Eq. (5), so that the optimal modulation wavelength for a given $q$ depends only on the conformal parameter, and not on the fundamental frequency or plasma density. If, on the contrary, dispersive phase mismatch dominates, $B\gg 1$ ("high-dispersion" regime); then

$$\lambda_m^{\text{opt}}=\lambda_m^{\text{disp}}\approx 2\pi/\Delta k\ll\lambda_m^{\text{geom}}. \tag{6}$$

As an illustration, consider QPM for the 51st harmonic of a Ti:sapphire laser ($\lambda=0.8\ \mu$m), which was near the middle of the harmonic plateau in the recent HHG experiments [15], and assume $b=100\ \mu$m. In plasma with $N_e\sim 10^{18}$ cm$^{-3}$, $B\sim 0.1$; for this low-dispersion regime, Eq. (5) yields $\lambda_m^{\text{opt}}\approx 6.2\ \mu$m. On the other hand, in a plasma with $N_e\sim 10^{20}$ cm$^{-3}$, $B\sim 10$ and $\lambda_m^{\text{opt}}\approx 0.54\ \mu$m.

Equations (5) and (6), generally speaking, require different optimal modulation wavelengths for harmonics of different orders. One, however, might expect phase-matching curves (i.e., phase-matching factors vs $b\Delta k$) to be relatively broad for high-order harmonics. (For instance, one can calculate, using Ref. [5], that the perturbative phase-matching factor for the 31st harmonic is only ~10% smaller than its maximal value, at $b\Delta k$, that corresponds to the maximum of the 29th harmonic.) As a result, a given $\lambda_m$ would yield a substantial phase-matching factor simultaneously for a number of harmonics, and moreover without precise control of the modulation wavelength.

## DENSITY MODULATION

Now, let us assume that the nonlinear susceptibility modulation, Eq. (2), is due to medium density modulation only, and is therefore accompanied by the refractive-index modulation:

$$n(u)=1+\tilde{n}[1+A\ \cos(au)]=n_0+\tilde{n}A\ \cos(au), \tag{7}$$

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. (2) and (7). In gases and in plasma, $\tilde{n}$ is usually very small, so that modulation of the refractive index is very weak. If also $\lambda \ll \lambda_m$, which is commonly the case for $\lambda_m^{\rm opt}$ 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 [16]: propagation in an inhomogeneous medium is assumed to be approximately the same as in a homogeneous medium with variable dielectric permittivity. This approximation is characterized, in particular, by the absence of Bragg reflection, which would substantially complicate the consideration (similar to the way in which complications result from Fresnel reflection in QPM in solids; see, e.g., [10]). Since we do not intend to fully investigate HHG in density-modulated media but rather want to identify some conditions favorable to quasi-phase-matching, we limit our consideration to this approximation as the simplest, while most common, situation. In this case, as we will now show, the refractive-index modulation, Eq. (7), does not substantially change the optimal QPM conditions, Eq. (4). Indeed, if (i) the lowest-order Gaussian beam is focused at the point $z=0$ inside a nonabsorbing medium that begins at $z=-L$, (ii) the refractive index of the medium is modified, $n=n(z)$, and (iii) the approximation of geometrical optics is valid, then the laser field inside the medium can be expressed as

$$E(r,z)=E_0[n(z)D]^{-1}\exp[-k_0r^2/b_0D]\times\exp\left(i\int_{-L}^{z}k(z')dz'\right),$$

$$D=1+2ib_0^{-1}\left(\int_{-L}^{z}n^{-1}(z)dz-L\right), \tag{8}$$

where $r^2=x^2+y^2$ and $E_0$, $b_0$, and $k_0$ are the beam amplitude, confocal parameter, and wave number, respectively, outside the medium. Equation (8) is straightforwardly derived from a more general expression given in Ref. [17]. Very weak dependence of refractive index on $z$ can be neglected almost everywhere in Eq. (8), so that, e.g., $D\approx 1+iu$. We, however, retain $z'$ dependence in the last exponential term in $E(r,z)$ since this exponential term will eventually include $\Delta k$, whose dependence on $z$ is as strong as that of $\chi^{(q)}$. Thus, we arrive at the expression for the fundamental field in a modulated medium which differs from the ordinary lowest-order Gaussian beam only in that $\exp(ikz)$ is replaced by $\exp[i\int_{-L}^{z}k(z')dz']$. [Such an expression first appeared in Ref. [12] and has been used in numerous publications on HHG since then (see, e.g., [2]), without, however, any reference to the approximation of geometrical optics.] Then, the procedure developed in Ref. [12] yields the phase-matching factor $|F|^2$,

$$F=[\chi_0^{(q)}]^{-1}\int_{-\infty}^{\infty}du\ \chi^{(q)}(u)e^{-i\{z\,\Delta k\}}f_q(u),$$

$$\{z\Delta k\}\equiv\int_{-\infty}^{z}dz'\Delta k(z')=p_0[u+A\ \sin(au)/a], \tag{9}$$

where $p_0=b\Delta k_0/2$, and $\Delta k_0$ is the phase mismatch due to the ambient dispersion. It follows from Eqs. (9), (7), and (2) that, to account for the modulated refraction, it is enough to replace the term $pu$ in each integral in Eq. (3) with $p_0[u+A\ \sin(au)/a]$. All three integrals taken in symmetrical limits are real, so that $F\approx 2(I_0+I_{+1}+I_{-1})$,

$$I_j=\int_0^{\infty}du\ \cos[(p_0+ja)u+(q-1)\tan^{-1}u+p_0A\ \sin(au)/a]f_q(u),\quad j=0,\pm 1. \tag{10}$$

For large $q$ the factor $f_q(u)$ differs substantially from 0 only for such a small $u$ that $\tan^{-1}u\approx u$, so that

$$I_j=\int_0^{\infty}du\ \alpha_j\ f(u),$$

$$\alpha_j=\cos\{u(p_0+ja+q-1)[1+\delta_j\sin(au)/(au)]\}, \tag{11}$$

$$\delta_j=p_0\beta/(p_0+ja+q-1).$$

Since $|\sin(au)/(au)|\leq 1$ and $\delta_{+1}<\delta_0<1$, the coefficients at $u$ in both $\alpha_0$ and $\alpha_{+1}$ 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 $a=a_{\rm opt}$, Eq. (4), with $p$ replaced by $p_0$. Indeed, for $a'=p_0+q-1$,

$$I_{-1}\approx I_{-1}^{\rm opt}=\int_0^{\infty}du\ \cos[(p_0A/a')\sin(a'u)]f(u). \tag{12}$$

Since $p_0/a'<1$ and $|\sin(a'u)|\leq 1$, the argument of the cosine function in Eq. (12) is smaller than $A$. If the modulation is not too deep; e.g, if $A<0.3$, which is quite realistic, the integral $I_{-1}^{\rm opt}$ is equal, within 4% error, to $\int_0^{\infty}du\ f(u)$, which in turn provides the upper limit for $|I_{-1}|$. Since for large $q$, $a'\approx a_{\rm opt}$, one may conclude that optimal QPM conditions are almost independent of the modulation of the refractive index, Eq. (7).

So far, we have used perturbation-theory expressions for the phase-matching integral. In fact, however, one may see that we have only utilized the particular form of the phase factor in Eq. (1), as well as the fast decrease of the function $f(u)$ with increasing $|u|$. If the same holds beyond perturbation limits, as seems indeed to be the case [11], Eq. (4) is also valid for strong pumping. Our consideration has been limited to tight focusing. It can be shown, however, that for large $q$ Eq. (4) is an equally good approximation for the opposite limit of loose focusing, $b\gg L$ if $\lambda_m$, which is much smaller than $b$, is also much smaller than $L$.

The most obvious advantage of QPM optimization is that it allows tight focusing, which is otherwise detrimental to HHG in rare gases and plasma. The incident intensity can then be easily increased by two orders of magnitude for the same pumping power by, e.g., simply changing the confocal parameter from the $\sim$1 mm now used, to the readily attainable 100 $\mu$m. Without a general theory of phase matching beyond 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., [2]). Moreover, even for the same currently used incident intensity, QPM optimization may significantly increase HHG conversion efficiency. Indeed, a model ([2], Fig. 31) developed for HHG of a loosely focused incident beam, allows one to assume the high-order harmonic conversion efficiency under current experimental conditions to be five orders of magnitude lower than the conversion efficiency that might be optimized in a conventional way (that is, by providing $b\Delta k \approx -2q$ in a homogeneous medium). On the other hand, harmonic intensity at the QPM optimum, under otherwise equal conditions, differs by a factor of $A^2/4$ from conventionally optimized output, as one may see by comparing the third and the first integrals in Eq. (3). As a result, with, e.g., recently reported [18] plasma density modulation achieved by irradiating a grating with a ruby laser ($A \approx 0.08$, $\lambda_m \approx 2-6$ $\mu$m in a plasma with $N_e \sim 10^{18}$ cm$^{-3}$), QPM optimization might increase the harmonic intensity by a factor of $(A^2/4)\times 10^5 \approx 160$ for the same pumping intensity.

In conclusion, we demonstrate theoretically the feasibility of optimal quasi-phase-matching of HHG in gases and plasmas with modulated density. This technique may increase high-order harmonic conversion efficiency by several orders of magnitude. QPM can also be applied to x-ray third-harmonic generation [19]. Indeed, for a typical x-ray laser wavelength $\lambda_{\mathrm{XRL}} \sim 200$ Å, $b \sim 100$ $\mu$m now being attainable, and reasonable plasma density $<10^{19}$ cm$^{-3}$, $\lambda_m^{\mathrm{opt}} = \lambda_m^{\mathrm{geom}} \approx \pi b \sim 300$ $\mu$m. With such an easily attainable modulation wavelength, QPM may appear to be an attractive method to optimize XRL frequency tripling in plasma.

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