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Phase-matching optima for high-order multiwave mixing and harmonic generation beyond perturbation limit

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Abstract

We demonstrate that the optimal phase-matching conditions for the high-order optical multiwave mixing and harmonic generation, which are given by the perturbation theory, remain valid in the strong-field limit under quite general assumptions about induced nonlinear polarization.

High-order optical harmonics generation (HHG) has recently attracted much attention as a possible source of short-wavelength coherent radiation [1]; harmonics of the order up to the 135th and wavelengths as short as 7.6 nm have been observed [2,3]. Conversion efficiency of HHG, however, remains too low for applications, largely due to poor phase matching. Phase-matching optimization could substantially increase the output. The actual experimental conditions, however, are, most likely, very far from optimal. Rather, reported HHG experiments correspond to the far wings of the HHG phase-matching factor. Indeed, those experiments are conducted in almost dispersionless (gases) or positively dispersive (plasma) media, whereas phase-matching optimization of HHG, at least according to perturbation theory, requires strong negative dispersion, especially if the pumping beam is tightly focused. To avoid this obstacle, another multiphoton process, high-order difference frequency mixing (HDM)

$$\omega = m\omega_1 - l\omega_2 , \quad (1)$$

where m, l are integers, $m \gg 1$, $m \gg l$, $\omega_1 > \omega_2$, was suggested for large-scale frequency upconversion in ionized gases [5]. It was shown that the phase matching for a given combination m, l is optimal if

$$b\Delta k/2 \approx -(m-l) , \quad (2)$$

where b is the confocal parameter of both beams, Δk is the phase mismatch due to the medium dispersion. For HDM in plasma, Δk is large negative, which may allow one to attain optimal phase matching. This conclusion, however, relied on the perturbation-theory analytical expression for the induced polarization as a function of pumping fields. It is not clear whether Eq. (2) would hold beyond perturbation approximation. Unfortunately, while phase matching for multiwave mixing, including harmonic generation, has been extensively studied within the limits of perturbation theory (see e.g. Refs. [4–6]), the exact dependence of the induced nonlinear polarization on strong pumping fields remains unknown as of now. Moreover, to the best of our knowledge, no model

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calculations of the dipole moment induced by strong biharmonic pumping, have been published yet. In the present paper we theoretically demonstrate, however, that the optimal phase-matching conditions for both HHG and HDM *do not depend*, to a large extent, on a particular form of this dipole moment, as long as some quite general assumptions are valid. As a result, we expect the conditions for phase-matching optimization, Eq. (2), to remain largely intact in strong pumping fields.

We consider multiwave mixing, Eq. (1), of lowest-order Gaussian beams propagating along z-axis and focused at $z=0$ with the same confocal parameters, b (typical situation for most of the experiments), so that the incident field can be written as

$$E=\tilde{E}_1(r,u)\cos(\omega_1 t+\phi_1)+\tilde{E}_2(r,u)\cos(\omega_2 t+\phi_2)\,, \tag{3a}$$

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

$$\tilde{E}_{1,2}(r,u)=E_{1,2}(1+u^2)^{-1/2}\exp[-k_{1,2}r^2/b(1+u^2)]\,, \tag{3b}$$

and

$$\phi_{1,2}=-k_{1,2}z+\tan^{-1}u-k_{1,2}r^2u/b(1+u^2)\,, \tag{3c}$$

$u=2z/b$, $r=(x^2+y^2)^{1/2}$, and $k_{1,2}$ are the wavevectors of the pumping beams. A starting point of our consideration is a particular model suggested in Ref. [7] to explain some features of phase matching of strong-field HHG. For HHG in the weak-field limit (see e.g. Ref. [8]), the qth harmonic of the induced polarization P_q is

$$P_q=A_{\mathrm{weak}}(\tilde{E}_1)\exp[-\mathrm{i}\phi(r,u)],\quad A_{\mathrm{weak}}(r,u)=A_0[\tilde{E}_1(r,u)]^q\,, \tag{4}$$

where A_0 contains the nonlinear susceptibility and other independent of r, u and, therefore here irrelevant factors; we will omit A_0 from now on. The space-dependent part of A_{weak} is real. It is assumed in Ref. [7] that the amplitude A_{weak} is transformed into the strong-field amplitude by replacing q in A_{weak} with some integer number smaller than q; this is 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 slower with the amplitude of the incident field as compared to the weak-field regime. The phase factor $\phi(r,u)$, on the contrary, remains unchanged: $\phi=q\phi_1$. Resulting nonlinear polarization yields closely similar phase matching of harmonics of different orders for actual experimental conditions, that is for a loosely focused pumping beam in dispersionless or positively-dispersive media. In this model, just as in perturbation theory, such conditions correspond to far wings of phase-matching factors.

In contrast to Ref. [7], we are interested in phase-matching optima, and for much broader variety of processes. Generalizing the model, we assume that the Fourier component of the induced nonlinear polarization responsible for high-order difference-frequency mixing, Eq. (1), of strong fields is:

$$P_{m,l}=p(\tilde{E}_1,\tilde{E}_2)\exp(-\mathrm{i}\phi),\quad p^*=p,\quad \phi=-k'z+(m-l)\tan^{-1}u-uk'r^2/(1+u^2),\quad k'=mk_1-lk_2\,, \tag{5}$$

(compare with Ref. [6]). The space-dependent amplitude p which is a real quantity $[\tilde{E}_1(r,u)]^m[\tilde{E}_2(r,u)]^l$ for weak fields, is again real; in other words, we assume that the *space-dependent* phase of the induced dipole moment is the same, ϕ, for both weak and strong fields.. (It is worth noting that a similar suggestion was made in Ref. [9] on the basis of a simple 1D model of HHG, as well as on the basis of a quantum-mechanical model which treats atomic field as small perturbation.) In contrast to Ref. [7], however, we do not presume any particular expression for the amplitude p; in fact, our conclusions do not depend on such an expression. Instead, it is enough for the real amplitude p, Eq. (5), to be 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 will neglect contributions to the generated field from the points of a medium at which the amplitude of the first (or the only, for HHG) incident field is smaller than 85% of its maximal value $E_1=\tilde{E}_1(0,0)$; since the intensity of high-order harmonics inside the plateau is approximately proportional to the 12th degree of the incident intensity [7], one expects only a small fraction ($\approx 2\%$) of the harmonic intensity to originate at these points. From Eqs. (3), the pumping amplitude drops below $\approx 85\%$ of its maximal value if

$$\text{either } |u|>0.6 \quad \text{for any } r, \quad \text{or } r^2>0.16\, w_2^2>0.16\, w_1^2 \quad \text{for any } u\,, \tag{6}$$

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

Regardless of the intensity of the incident fields, the electric field $\mathscr{E}(r', z')$ generated in a homogeneous medium and observed at a point (r', z') outside of it, can be written as (see e.g. Ref. [7]):

$$\mathscr{E}(r', z') \sim \int \frac{\tilde{P}(r,z)\exp(-\mathrm{i}z\Delta k)}{z'-z} \exp\left(\frac{\mathrm{i}k(r^2+r'^2)}{z'-z}\right) J_0\left(\frac{kr'r}{z'-z}\right) r\,\mathrm{d}r\,\mathrm{d}z, \tag{7}$$

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 $\exp(\mathrm{i}k'z)$, $\Delta k = k-k'$, $k'=mk_1-lk_2$, and k is the wavevector of the generated field; the integral is taken over the entire nonlinear medium. Substituting the polarization from Eq. (5), we obtain:

$$\mathscr{E}(r', u') \sim \int \exp\left[-\mathrm{i}\left(b\Delta k u/2 + (m-l)\tan^{-1} u - \frac{k'r^2u}{b(1+u^2)} - 2\frac{k(r^2+r'^2)}{b(u'-u)}\right)\right]$$
$$\times \frac{p(r,u)}{u'-u} J_0\left(2\frac{kr'r}{b(u'-u)}\right) r\,\mathrm{d}r\,\mathrm{d}u\,, \tag{8}$$

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

$$u' \gg 1, \qquad u' \gg u\,, \tag{9}$$

we may neglect u as compared to u' in Eq. (8) and move $\exp(2\mathrm{i}kr'^2/bu')$ out of the integrand. The remainder of the fourth term in the exponent, $2kr^2/bu'$, is much smaller than 1 (for sufficiently large u'). It, however, cannot be simply ignored since for very small u, $u<1/u'$, it is larger then the third term; thus, we retain this remainder, but in the form $\exp(2\mathrm{i}kr^2/bu') \approx 1+2\mathrm{i}kr^2/bu'$. Since $k'<mk_1$ and $u/(1+u^2)<u$, it follows from Eq. (6) that the third term in the exponent, Eq. (8), is smaller than 12% of the second one, and we neglect the former. Furthermore, since the integrand in Eq. (8) is now symmetrical in u, we may replace the exponent, Eq. (8), with the cosine function. Finally we note that for $|u|<0.6$, $\tan^{-1}u \approx u$. All these approximations yield:

$$\mathscr{E}(r', u', M) \sim \int_0^{0.6} \cos(Mu)\,[F_1(u)+\mathrm{i}F_2(u)]\,\mathrm{d}u, \quad M=b\Delta k/2+(m-l)\,, \tag{10}$$

where

$$F_1(u) \approx \int \frac{p(r,u)}{u'} J_0\left(2\frac{kr'r}{bu'}\right) r\,\mathrm{d}r, \quad F_2(u) \approx \int \frac{2kr^2p(r,u)}{bu'^2} J_0\left(2\frac{kr'r}{bu'}\right) r\,\mathrm{d}r\,.$$

It can be shown that for any positive, monotonically decreasing function $\tilde{f}(r)$,

$$\int_0^{r_0} \tilde{f}(r)\, J_0(ar)\,\mathrm{d}r > 0\,, \tag{11}$$

for $a>0$, $r_0>0$; therefore, $F_{1,2}(u)>0$ for any u. Now, since $\cos(0) \geq \cos(Mu)$ for any u and $M\neq 0$, both $|\mathrm{Re}\mathscr{E}|$ and $|\mathrm{Im}\mathscr{E}|$ and, therefore, the generated field amplitude $|\mathscr{E}|$, are maximal if $M=0$. Hence, the optimal product of the confocal parameter and the dispersion phase mismatch is close to

$$(b\Delta k)_{\mathrm{opt}} \approx -2(m-l)\,, \tag{12}$$

which corresponds to the optimal phase matching of HDM in perturbation theory, Eq. (2).

We can roughly estimate an additional error that may be brought in by the approximations we have used to implement our assumption regarding the function p. Neglecting u as compared to u' should not lead to a substantial distortion in the far-field area. Then, replacing $\tan^{-1}u$ with u, we might overestimate $(b\Delta k)_{\rm opt}$ by up to 10% – the largest difference between u and $\tan^{-1}u$ at the interval 0–0.6. On the other hand, neglecting the third term in the exponent of Eq. (8) may introduce an error of the same magnitude but in the opposite direction. Therefore, we expect Eq. (12) to reflect optimal phase-matching conditions with about 10% accuracy. Since at least in the weak-field limit, the phase-matching factor is quite broad for higher orders of interaction (see e.g. Ref. [8]), Eq. (12) may be a good approximation provided our general assumptions are justified.

In the loose-focusing limit ($b \gg L$), one can drop any special restrictions of u. Indeed, in such a limit, $|u| \ll 1$ within the entire medium, so that the integral (10) could be extended to the full medium length, again with Eq. (12) as the outcome. Note that in the weak-field regime, phase-matching maxima for HHG with tight and loose focusing also correspond to almost the same $(b\Delta k)_{\rm opt}$ [8].

Now, the question is under what conditions the assumption, Eq. (5), together with the assumed monotonic behavior of p, can be justified. First of all, Eq. (5) is valid if the induced nonlinear polarization $P(x, y, z, t)$ is a (continuous) function of the incident field:

$$P(x,y,z,t)=f[E(x,y,z,t)] \,. \tag{13}$$

In other words, we presume here spatial and temporal locality of the induced polarization, as well as non-depletion of the pumping. One may expect Eq. (13) to hold for the pumping pulses being much longer that the relaxation times of the nonlinear media; pumping frequencies being far from any resonances, including intensity-induced resonances [10]; and the generated field being so weak that the induced polarization is largely determined by the pumping field. In the simpler case of the qth harmonic generation ($l=0$, $m=q$) with a monochromatic pumping field, $f=f(E_1)$ is periodical in time with the period $2\pi/\omega_1$, so that

$$P_q=\frac{\omega_1}{\pi}\int\limits_{-\pi/\omega_1}^{\pi/\omega_1} \mathrm{d}t\, f[\tilde{E}_1\cos(\omega_1 t+\phi_1)]\exp(\mathrm{i}q\omega_1 t)=p\exp(-\mathrm{i}q\phi_1) \,, \tag{14}$$

where

$$p=\frac{\omega_1}{\pi}\int\limits_{-\pi/\omega_1}^{\pi/\omega_1} \mathrm{d}t\, f[\tilde{E}_1\cos(\omega_1 t)]\cos(q\omega_1 t)$$

is real, and the phase factor $\exp(-\mathrm{i}q\phi_1)$ is the same as that for weak fields.

With two pumping frequencies, ω_1 and ω_2, there is no common "time scale", and the previous procedure with the 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 a sum $S_n=\sum_{k=1}^{n} a_k\psi_k(x)$, where ψ_k are orthonormal polynomials [11]. 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 then be transformed into the expression:

$$S_n=\sum_{j1j2} s_{j1j2}=\sum_{j1j2} A_{j1j2}\cos(j_1\Phi_1-j_2\Phi_2) \,, \tag{15}$$

where $\Phi_{1,2}=\omega_{1,2}t+\phi_{1,2}$, real amplitudes $A_{j1,j2}$ 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_{j1j2} which contains a given combination of frequencies, $\omega_{ml}=m\omega_1-l\omega_2$, contains also the phase $\phi_{ml}=m\phi_1-l\phi_2$: $s_{j1,j2}=A_{ml}\cos(\omega_{ml}+\phi_{ml})$, so that the Fourier transform of $s_{j1,j2}$ is

$$P_{m,l}=p\exp[-\mathrm{i}(m\phi_1-l\phi_2)], \quad p=A_{ml}(\tilde{E}_1,\tilde{E}_2) \,, \tag{16}$$

the validity of Eq. (5) follows.

In real experiments, however, one can hardly expect Eq. (13) to be strictly correct. Consequently, any deviation from Eq. (13) would be reflected in p being a complex, and not 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 Eq. (8) if the change of Ψ within the interval of interest, Eq. (6), is much smaller than the respective change of the geometrical phase $(m-l)u$. If also $|p|$ drops with $|\tilde{E}_1|$ rapidly and monotonically, Eq. (12) can still hold. Due to the lack of calculations of the single-atom response to strong biharmonic pumping, we may only try to verify these assumptions for HHG. The three models of HHG [12,13,10] that have been most successfully compared with experimental data, seem to show sufficiently small variation of the phase Ψ. Indeed, the intensity-dependent phase of the dipole moment of the 51st harmonic, calculated in Ref. [3] according to the model [12], changes only by $\sim 4\pi$ 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 developed in LLNL/Saclay (see e.g. Ref. [13]) do not include Ψ explicitly, in all the theoretical considerations this phase is assumed to be constant (see e.g. Ref. [14]), or at least not to interfere strongly with phase matching [7]. Finally, the non-perturbative two-level model of HHG [10] 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 Eq. (8). (We already mentioned yet another model [9] that essentially presumed $\Psi = 0$.)

As to the modulus of the dipole moment, $|p|$, in Ref. [3], as well as in the simplified quantum-mechanical model published recently [15], it behaves in a desirable manner up to the cutoff intensity. After that, in the both LLNL/Saclay [13] and the two-level [10] models, $|p|$ experiences sharp resonance-like 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 experimental dependence of the output on the incident field. Moreover, in all the experiments on HHG and HDM, the pumping field is in a form of a pulse, so that 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 where the pumping intensity is lower.

To the best of our knowledge, no calculations of the amplitude or phase of the induced dipole moment in HDM have been published yet. If our general assumptions hold also for this process, the optimal phase-matching conditions, Eq. (2), calculated in Ref. [5] within the perturbation-theory limit, would present a quite accurate approximation for nonperturbative HDM as well. One should bear in mind, though, that the same optimal phase matching determined by Eq. (2) may imply very different laser and media parameters (such as confocal parameters and media density) for low and high intensities, e.g. due to potentially different dispersion properties of media in weak and strong laser fields.

In conclusion, we have demonstrated that optimal phase-matching conditions for high-order harmonic generation and high-order difference-frequency mixing of weak fields remain largely intact beyond the perturbation limit under quite broad assumptions regarding nonlinear polarization. It allows one to use safely perturbative phase-matching conditions, Eq. (2), for both HHG and HDM in strong fields.

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