Simultaneous ultrabroadband quasi-phase-matching for high-order harmonic generation

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We propose a simple quasi-phase-matching (QPM) scheme in high-order harmonic generation (HHG) based on the approximately linear dependence of the phase mismatch on harmonic order. With this scheme, essentially all the harmonic orders are phase matched simultaneously, with harmonic order q experiencing a qth-order QPM. We validate this proposal by simulations using a semiclassical numerical model of HHG.

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I. INTRODUCTION

High-order harmonic generation (HHG) is a nonlinear optical up-conversion process creating extremely broadband radiation emitted in the form of (tens of) attosecond-duration pulses [1,2]. At the single-atom level, the HHG process can be described by a three-step model [3]: tunnel ionization, at which an intense laser field ionizes an electron out of its parent atom; oscillating acceleration; and, finally, recombination of the accelerated electron with its parent ion, where excess kinetic energy is delivered to an emitted high-energy photon with a frequency lying in the soft x-ray part of the spectrum. Such a process suffers in many cases from a mismatch between the phase velocities of the pump pulse and the HHG emission, hindering efficient up-conversion.

Quasi-phase-matching (QPM) is a well-known, widely explored, and commonly used technique for phase matching optical nonlinear conversion processes [4,5]. In QPM, a modulation of a parameter relevant to the interaction replaces the strict requirement of momentum matching for the photons involved in the process with a quasimomentum balance [6]. The most common modulation geometry of QPM used for HHG is a periodic modulation, which is useful for efficiently phase matching a specific harmonic order [7–19], although the use of random and quasiperiodic modulations was also suggested to phase match several or a group of harmonic orders [20], while a sophisticated, hard to realize, spatiotemporal accelerating QPM modulation was proposed to phase match an extremely broadband HHG emission [21]. Other QPM techniques for HHG, such as the use of multiple plasma jets [22], might also be relevant provided the features of the spatial modulation can be varied fast enough.

Here we propose and validate through numerical simulations that a judicious choice of the period in a simple periodic QPM scheme can phase match simultaneously essentially all the HHG emission.

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II. THEORY

Consider an up-conversion of the fundamental harmonic to a qth harmonic. The phase mismatch for this process is $\Delta k_q = qk_0 - k_q$, with k_0 and k_q being the wave vectors of the fundamental and the qth harmonics, respectively. The phase mismatch also defines the coherence length $l_q \triangleq \pi/\Delta k_q$ over which the qth harmonic would be built constructively (in phase). QPM with a periodic modulation of period $2l_q$ (that is, with a frequency Δk_q) would phase match the generation of the qth harmonic. Using the regular dispersion relation $k(\omega) = \omega n(\omega)/c$, we set

$$l_q = \frac{1}{q}\frac{\pi c}{\omega_0[n(\omega_0) - n(\omega_q)]} = \frac{1}{q}l_0(q), \tag{1}$$

where ω_0 is the fundamental frequency. As the index of refraction of the high harmonics depends only weakly on the frequency, $n(\omega_q) \approx 1$ (this approximation gets better as q gets higher), we can safely approximate $l_0(q) \approx l_0 = \frac{\pi c}{\omega_0[n(\omega_0)-1]}$. With $\Delta k_0 = \pi/l_0$ we also get $\Delta k_q \approx q\Delta k_0$. We call l_0 and Δk_0 the fundamental coherence length and the fundamental phase mismatch, respectively.

Now, using a QPM modulation at the fundamental phase mismatch frequency (with a period $2l_0$), it is clear that harmonic order q would experience a qth-order QPM [23] provided that the modulation contains a component with a frequency $q\Delta k_0$ in its spectrum. Clearly, a perfect sinusoidal modulation with a frequency Δk_0 would not serve for this purpose, but a rectangular modulation with a period $2l_0$ and a duty cycle of 50% (giving a square wave) would be able to phase match all odd-order harmonics simultaneously. We define the relative enhancement η_q in a medium of length L for each harmonic as the intensity of that harmonic at the end of the medium with QPM modulation divided by the same without the modulation. Without modulation the intensity is proportional to $I_q \propto |\int_0^L \exp(i\Delta k_q z)\exp(-\alpha_q z/2)dz|^2$, with α_q being the absorption coefficient of a harmonic order q. With QPM of order q the intensity at the end of the medium is proportional to $I_q \propto |\int_0^L c_q \exp(-\alpha_q z/2)dz|^2$, where $c_q = 2/\pi q$ is the Fourier coefficient of the qth harmonic of the rectangular modulation [23] (this last expression is accurate only for long enough interaction lengths: $L/l_q \gg 1$). Dividing the latter by the former, we obtain the relative enhancement:

$$\eta_q = \left[\left(\frac{2}{\pi q}\right)^2 + \left(\frac{4}{\alpha_q l_0}\right)^2\right] \frac{1 - 2e^{-\alpha_q L/2} + e^{-\alpha_q L}}{1 - 2e^{-\alpha_q L/2}\cos\Delta k_q L + e^{-\alpha_q L}}. \tag{2}$$

Remarkably, if we neglect absorption altogether and calculate the ratio of the intensity at the end of the medium to just after one coherence length, we get a value which is independent of the harmonic order:

$$\left.\frac{I_q(L \gg l_q)}{I_q(l_q)}\right|_{\text{no absorption}} = |L\Delta k_0/\pi|^2 = |L/l_0|^2. \tag{3}$$

We can easily understand this result: each harmonic order is phase matched with a qth-order QPM, and as such, its intensity is lower by q^2 than that with a first-order QPM dedicated to phase match solely that harmonic. However, with no QPM the maximum intensity of harmonic order q also scales as $1/q^2$, which causes Eq. (3) to be constant.

Of course, a perfect rectangular modulation is impossible as its spectrum is unbounded, but we can assume that a finite-bandwidth QPM modulation would be possible and practical as we need to phase match only the harmonic orders up to the cutoff. Thus, this method can achieve a simultaneous QPM of practically all the HHG spectrum.

In passing, we mention several possible experimental approaches to realize a QPM scheme for HHG which is relatively sharp enough, that is, composed of several Fourier components. The first is an all-optical scheme in which the interference of several spatial optical modes realizes a spatial intensity grating which translates to a phase grating of the harmonic emission. Such a QPM approach for HHG was recently demonstrated in a gaseous medium for a small number of spatial modes [19]; however, in principle, it can easily be extended to the superposition of several or many modes. This is especially relevant to scenarios where the required fundamental spatial frequency is small (say, of the order of $\sim$1 mm^{-1}), as is the case considered in the simulations in the current work. Another approach, which was demonstrated experimentally a few years ago, uses a periodically poled lithium niobate waveguide for HHG in a solid-state medium [24], where the electric field poling technique which can be applied at a submicron poling resolution [25] can produce a rather sharp modulation. Another approach might be using an ultrasonic transducer driven with an electronic wave-form generator to modulate the gas pressure in the medium, a method which was suggested for third-harmonic generation [26] but needs to be modified for HHG as, generally, it is inefficient for invariant conditions along the propagation length [27].

III. SIMULATIONS

We test our hypothesis using a semiclassical one-dimensional numerical model of high-order harmonic generation. The propagation of the fundamental harmonic along the optical axis z is calculated using a known extreme nonlinear propagation equation [28] (which includes the effects of plasma dispersion). At each propagation step we solve a one-dimensional (1D) time-dependent Schrödinger equation with absorbing boundary conditions using a Crank-Nicolson scheme. The Coulombic potential was modeled with a modified 1D cusp potential [29] of the form $-2I_p/(1+\sqrt{2I_p}|x|)$ (in atomic units), with I_p being the ionization potential. The polarization in each step is calculated using the expectation value of the electron's position along the direction of polarization of the electric field x multiplied by the electron's charge e and the atomic density of the medium N: $P = Ne\langle\psi|x|\psi\rangle$. Finally, the emitted field is calculated by integrating the propagation equation [28]

$$\frac{\partial E(z,\tau)}{\partial z} = -\frac{2\pi}{c}\frac{\partial P}{\partial \tau} \tag{4}$$

using a fourth-order Runge-Kutta method. We chose to simulate a medium of a preformed plasma waveguide [30,31] made of argon ions. Such a medium is characterized by relatively short coherence lengths such that a pump pulse propagating over the length of dozens of QPM modulation periods at moderate pressures (tens of torr) would hardly be distorted due to dispersion. Such a medium was already suggested to serve in several macroscopic manipulation schemes of HHG [21,32]. Explicitly, we simulated HHG in 50 torr of a singly ionized argon ($I_p = 27.629$ eV) at a temperature of 24 °C; absorption in the medium was modeled according to the work by Reilman and Manson [33]; the pump pulse was a transform-limited Gaussian with a 20-fs FWHM duration, an 800-nm center wavelength, and a peak intensity of 4×10^{18} W/m^2 (leading to a maximal ionization level of $\sim$2% above the already preionized medium); the total propagation length was 5 mm.

Note that for the conditions simulated in this work, where there are no diffraction effects providing geometric phase terms and the intrinsic atomic phase [34] can be neglected, the phase mismatch of any specific harmonic order is proportional to the pressure [19,35,36]. The HHG cutoff harmonic order is calculated to be 56. For these parameters the value of the fundamental coherence length is $l_0 = 0.857$ mm. We simulated QPM with a fundamental frequency of Δk_0 by modulating the polarization through multiplication with a rectangular periodic function $S_1(z) = \text{sgn}[\sin(\Delta k_0 z)]$. The results of the simulation are summarized in Fig. 1, where we see the evolution of the intensity of various harmonic orders as well as the spectrum at the end of the interaction with and without the applied QPM scheme. It is evident that all harmonic orders are indeed simultaneously phase matched, with each harmonic order experiencing a qth-order QPM. The relative enhancement [Eq. (2)] is shown in Fig. 1(d). We note that the prediction of the simplified theoretical model [Eq. (2)] is not constant in the harmonic order due to absorption, but the variations in our case are very small on a log scale.

It is also interesting to consider a more elaborate QPM modulation geometry where we add to the previous modulation another frequency component which is set to phase match a particular harmonic order q'. In this case the QPM modulation takes the form $S_2(z) = \{\text{sgn}[\sin(\Delta k_0 z)] + \sin(q'\Delta k_0 z)\}/2$. This way we shift the efficiencies of the generation of different harmonic orders to better favor the generation of harmonic order q', while all harmonic orders are still simultaneously phase matched. The results of simulations with this modulation are shown in Fig. 2

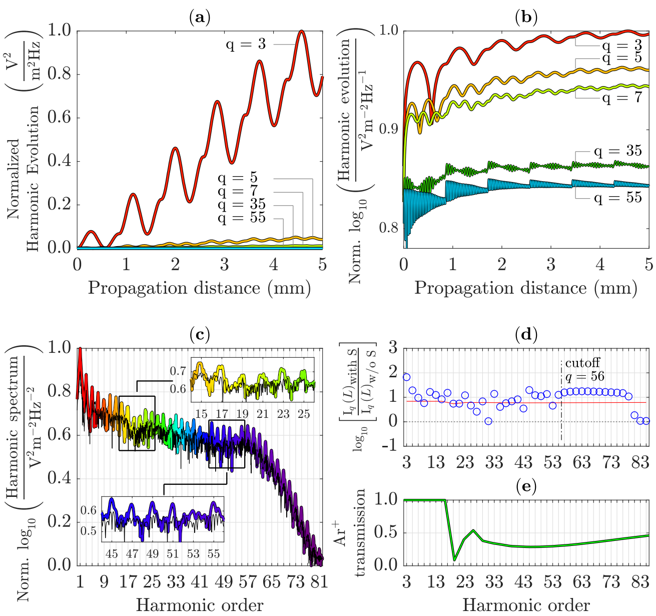

FIG. 1. HHG with simultaneous QPM of all harmonic orders using modulation of the polarization vector by the function $S_1(z)$ (see text). (a) Evolution of the intensity I_q of various harmonic orders, (b) the same evolution in logarithmic scale, (c) the HHG spectrum at the end of the interaction without (black) and with (colored line) the application of $S_1(z)$ modulation at the fundamental phase mismatch frequency, (d) relative enhancement of each harmonic order (blue circles) in a logarithmic scale, where the solid red line shows the theoretical prediction according to Eq. (2) and the vertical dash-dotted line indicates the cutoff location, and (e) the Ar^+ transmission [33] for a 5-mm propagation.

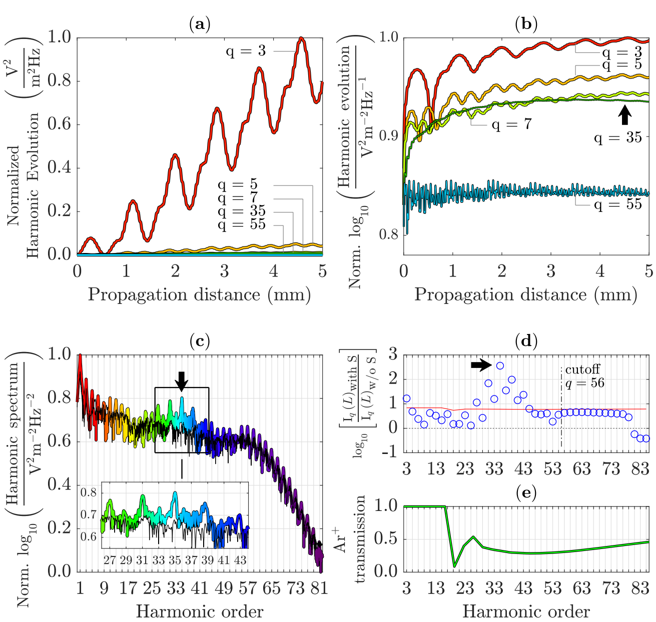

FIG. 2. Same as Fig. 1, but the QPM modulation is of the form of $S_2(z)$ (see text) set to phase match all harmonic orders with a preference to harmonic order $q\prime = 35$. In this case, $l_0 = \pi/\Delta k_0 = 857\ \mu$m, and $l_{ph} = \pi/q\prime\Delta k_0 = 24.5\ \mu$m is the coherence length of the 35th harmonic. The thick black arrows denote the values related to harmonic order $q\prime = 35$.

for $q\prime = 35$. The preference given to harmonic $q\prime$ (and its neighbor harmonics) is obvious in this case. The relative enhancement of this harmonic order [Fig. 2(d)] is clearly shifted above that of the other harmonic orders which are shifted slightly below the value of the previous square-wave modulation [$S_1(z)$] case. We note that a longer modulation would better isolate the preferred harmonic from its neighbor harmonics.

We would like to note that, as indicated above, we modeled the QPM modulation as an amplitude perturbation of the polarization excited in the medium. Other types of modulations would change only the Fourier components associated with the spatial frequencies of the QPM grating, while the overall effect would be the same in all cases (up to overall scaling of the conversion efficiency). The exact modeling of the QPM modulation should depend on the specific physical realization of the modulation. For example, with pump intensity modulations the grating is essentially a phase modulation applied to the polarization [19], while for a polarization beating modulation [14] or a density (pressure) modulation [27] the grating is applied as both an amplitude and a phase modulation.

IV. CONCLUSIONS

In conclusion, we have proposed a simple periodic QPM modulation to simultaneously phase match essentially all harmonic orders. The low spatial frequency of this modulation compared to regular QPM schemes necessitates the use of relatively long interaction lengths with a relatively constant pump. Thus, this method requires a condition in which the pump pulse does not vary much during several coherence lengths for all relevant harmonic orders, while absorption should not be too dominant (generally, the absorption length for any relevant harmonic order should be longer than a few coherence lengths). Thus, conditions that can be relevant are guiding the pump beam within a preionized medium as considered in this work or, alternatively, using a neutral gas with a long interaction length at low pressures and loose focusing (or using long waveguides) while keeping ionization to a low level to prevent pulse distortion. Such conditions would also keep the integrity of the QPM modulation along the propagation length in the case it is applied all optically [19]. Another option, which was already mentioned, is to use HHG in long, nonlinear crystal waveguides [24]. We numerically demonstrated that this technique can be successfully combined with a traditional single-order harmonic QPM, in which a particular, prechosen harmonic order is favorably quasi-phase-matched. This work is also relevant in the context of creating attosecond pulses by employing macroscopic effects [37], which can benefit from ultrabroadband enhancement, as well as for probing electronic and nuclear dynamics through HHG [38,39].

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