Quasi-periodic and random quasi-phase matching of high harmonic generation

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Quasi-phase matching schemes employing quasi-periodic or random spatial modulations, previously applied to perturbative nonlinear optics, are demonstrated theoretically for the extreme nonlinear optical process of high harmonic generation. We show that quasi-periodic quasi-phase matching of high harmonic generation can be used for simultaneous enhancement of arbitrarily chosen spectral regions. We also demonstrate enhancement of a single extremely wide bandwidth using random quasi-phase matching.

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The yield of a nonlinear optical process in which new frequency components are generated is often limited by dispersion [1]. Different frequency components propagate with different phase velocities: thus, in general the direction of energy transfer between the fundamental pump beam and generated light is reversed every coherence length, when the accumulated phase difference is equal to π. This dispersion problem exists both for perturbative nonlinear optics, where the radiating electron stays bound in an anharmonic potential, and for the extreme nonlinear optical process of high-order harmonic generation (HHG), which involves transitions between bound and continuum states. In spite of the two different physical generation mechanisms, in both cases a dispersion-induced macroscopic phase mismatch can be formulated as a microscopic momentum imbalance between the interacting photons. Thus, a common remedy, known as quasi-phase matching (QPM) [1–3], changes the required momentum conservation condition by employing an ordered spatial modulation of the nonlinear medium. In momentum space, the momentum imbalance is compensated by a quasi-momentum component related to a reciprocal lattice vector of the modulated structure [2]. The interpretation in the spatial domain is a sequential correction of the phase mismatch.

The most basic QPM scheme uses a one-dimensional periodic modulation structure and is typically used for selective enhancement of a single nonlinear process using the shortest available reciprocal lattice vector. Simultaneous enhancement of several nonlinear processes (with different coherence lengths) typically requires more complex QPM structures that, for example, involve random or quasi-periodic modulations. Quasi-periodic QPM [4–6] uses an ordered spatial modulation lacking translational symmetry, giving rise to a wealth of reciprocal lattice vectors that are able to phase match processes with incommensurate phase mismatch values. Random QPM [7,8] can phase match a broad bandwidth for a given nonlinear process with no need for prior knowledge about the exact phase matching conditions (or the spatiotemporal dynamics) or for strictly controlling the optical properties of the media. Introducing randomness into a modulated structure corresponds to adding a semi-continuous set of reciprocal lattice vectors—each able to compensate a nonlinear process with a slightly different phase mismatch value.

Quasi-phase matching schemes developed for perturbative nonlinear optics have already been adapted to extreme nonlinear optics. Past work demonstrated that periodic QPM schemes could enhance HHG using modulated waveguides to induce amplitude modulations in the driving laser intensity [9–11], by using interference patterns created by a sequence of counterpropagating light pulses [12–14] or by using a sequence of nonlinear sources [15]. Periodic self-induced QPM for enhancing HHG emission was suggested by using periodic refocusing of an intense laser pulse in an ionizing medium [16,17] and by using mode beating [18]. Finally, QPM can be extended to very high (keV) photon energies by using a counterpropagating quasi-cw field [19]. Aperiodic chirped and target function driven amplitude modulations were also proposed to enhance harmonics near the cutoff region over narrow bandwidths of up to approximately 30 eV [20–22]. However, quasi-periodic or random QPM schemes have not been proposed or used to date for extreme nonlinear optics.

In this work we demonstrate for the first time to our knowledge that quasi-periodic and random QPM schemes can be used to selectively enhance HHG. We show that quasi-periodic QPM can result in the simultaneous spectral enhancement of several arbitrarily chosen spectral regions, while random QPM can enhance a single extremely wide bandwidth (over hundreds of eV). Both of these capabilities are important for applications of the new fields of coherent, nonlinear, and attosecond x-ray science. The ability to selectively enhance a broad spectral bandwidth could enable bright attosecond pulses at higher photon energies than are now possible. Moreover, the ability to spectrally shape the high harmonic spectrum such that two different wavelengths are efficiently generated will be useful for nonlinear optics using high harmonic beams, or for high contrast spectroscopy of molecules and materials [23,24].

We first investigate quasi-periodic QPM in HHG. We designed the quasi-periodic spatial modulation using the dual grid method, which has been shown to be capable of solving the general problem of phase matching of multiple perturbative nonlinear processes [4,25–27]. This method is able to create an ordered QPM structure containing an arbitrary set of predefined discrete spectral components. Each such component, located at a reciprocal lattice vector of the ordered QPM structure, can phase match the generation of a relatively narrow bandwidth of harmonics, as expected from a simple periodic modulation. However, the utility of the dual grid method is that it permits several different arbitrary narrow bandwidths to be phase matched simultaneously.

We study numerically the effect of such a phase matching scheme for the case of HHG in a preformed plasma waveguide [28]. We consider a 20 fs, hyperbolic secant, driving laser pulse at $\lambda = 0.8\ \mu$m with a peak intensity of $20 \times 10^{14}\ \mathrm{W/cm^2}$ propagating in a medium of singly ionized argon ions at a pressure of 3 Torr within a circular waveguide with a diameter of 150 μm and length of 6 mm. The generated harmonic field is calculated using a 1-D generalized Lewenstein model [29]. For these parameters the transmission of the generated harmonics is higher than 0.75. Thus, in our simulations absorption was neglected completely in order to emphasize propagation effects related to the phase mismatch. We used the dual grid method [4] to design a quasi-periodic structure that can enhance simultaneously two (arbitrarily chosen) spectral regions—the first in the plateau region around the 183rd harmonic with associated coherence length of 66 μm, and the second in the cutoff region around the 283rd harmonic where the coherence length is 46 μm (the coherence lengths were calculated while taking into account waveguide, plasma, and neutral atom dispersion terms [13]). This in turn yields a quasi-periodic QPM pattern (shown as an inset in Fig. 1d) consisting of elements that are on the scale of the coherence lengths associated with the chosen harmonics. The application of this pattern was carried out by eliminating the harmonic emission from the dark (blue online) zones of the pattern. This was shown to be experimentally possible using a counterpropagating pulse train [12,13]. However, our conclusions also apply qualitatively to cases where harmonic emission is modulated rather than eliminated.

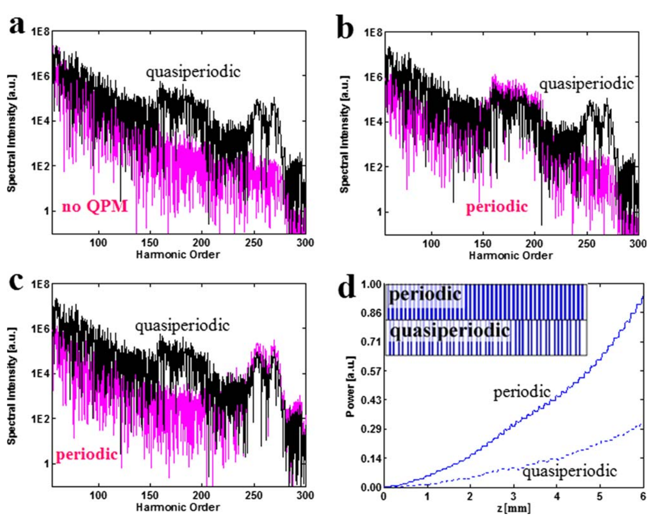

Fig. 1. (Color online) Quasi-periodic QPM. a, Spectral intensity of the emitted harmonic emission with quasi-periodic QPM for enhancing the emission around the 183rd and the 283rd harmonics (quasi-periodic) compared with the emission with no QPM. This quasi-periodic scheme is also compared with periodic QPM for enhancement around b, the 183rd harmonic and around c, the 283rd harmonics. d, Power buildup of the 183rd harmonic as a function of propagation length for a periodic QPM (solid line) and for the quasi-periodic QPM (dotted line). The inset shows the spatial modulation pattern for periodic and quasi-periodic cases.

The results of our HHG simulations with the quasi-periodic QPM structure are shown in Fig. 1. The normalized harmonic spectrum after a propagation distance of 6 mm is shown in Fig. 1a both without (no QPM) and with (quasi-periodic) this QPM scheme. Quasi-periodic QPM shapes the HHG emission to enhance harmonic emission by about two orders of magnitude in the two designated spectral windows corresponding to the 183rd and 283rd harmonics. Figures 1b and 1c compare the quasi-periodic modulation with a periodic QPM pattern designed to phase match a single narrow bandwidth. The enhancements observed due to quasi-periodic QPM and periodic QPM are of the same order of magnitude. The power growth as a function of propagation distance for a narrow spectral window of 3 harmonics around the 183rd harmonic is shown for the periodic and quasi-periodic QPM schemes on a normalized scale in Fig. 1d. Both periodic and quasi-periodic QPM exhibit quadratic growth, which is characteristic of a coherent, in-phase, buildup of the harmonic field.

Next, we investigate HHG under random QPM modulation. We use the same physical system as in the quasi-periodic case. We simulated HHG from regions that are one coherence length long (appropriate for phase matching around the 183rd harmonic) and that are spaced randomly with a mean separation (between the centers of adjacent regions) of twice the coherence length and with a variance separation of 0.3 coherence lengths (a regular deterministic periodic modulation corresponds to zero variance). We averaged the results over an ensemble of 40 different such random modulations. The HHG spectral intensity without and with the ensemble-averaged random QPM scheme is shown in Fig. 2a. Clearly a wide bandwidth spanning approximately 150 harmonics ($\sim$230 eV) is significantly enhanced. Increasing the separation variance would result in a broader enhanced bandwidth at the expense of a lower enhancement factor. The buildup of HHG power as a function of distance in a narrow spectral window (3 harmonics) around the 183rd harmonic is shown in Fig. 2b. The averaged random evolution shows a linear buildup, characteristic of a random-walk type of incoherent summation of fields.

In conclusion, we have shown that quasi-periodic and random QPM schemes that were demonstrated

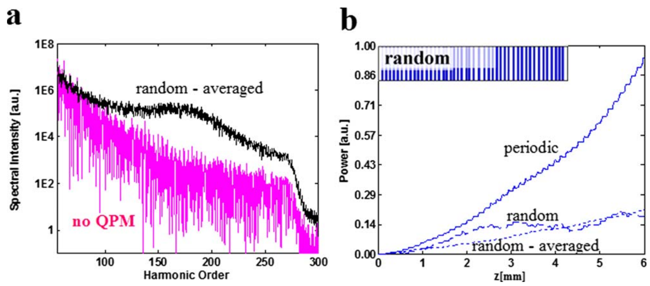

Fig. 2. (Color online) Random QPM. a, Spectral intensity of the emitted harmonic emission with random QPM for enhancing the emission in a wide spectral window around the 183rd harmonic (black), compared with the emission with no QPM. The intensity with random QPM is averaged over an ensemble of 40 different random modulation patterns. b, Power buildup of the 183rd harmonic as a function of propagation length for a periodic QPM (solid line), for a typical random pattern (dashed line), and for the average over the ensemble (dotted line). The inset shows a typical spatial modulation pattern.

to be very useful in perturbative nonlinear optics are also applicable to the extreme nonlinear optics of HHG. We used quasi-periodic QPM to enhance two distinct spectral regions, and our method can be extended for simultaneous enhancement of arbitrarily chosen spectral regions. This control over the shape of the spectrum will be useful for pump–probe and imaging applications in atoms, molecules, and materials. Another possible application is simultaneous phase matching of the long and short electronic trajectories associated with the emission of harmonics in the plateau region, which possess different coherence lengths [30,31]. We also demonstrated wideband enhancement using random QPM structures, which could be useful for bright attosecond pulse generation [32]. Another important advantage of random QPM for HHG is that it is insensitive to small temporal and spatial variations of the phase matching conditions [14]. Finally, in contrast to HHG using periodic modulations, no exact knowledge of the coherence length, or its spatiotemporal dynamics, is required for applying random QPM to HHG.

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