Quantum-path control in high-order harmonic generation at high photon energies

Xiaoshi Zhang1, Amy L Lytle, Oren Cohen, Margaret M Murnane and Henry C Kapteyn
JILA and Department of Physics, University of Colorado at Boulder, Boulder, CO 80309, USA
E-mail: xiaoshi.zhang@colorado.edu

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Abstract. We show through experiment and calculations how all-optical quasi-phase-matching of high-order harmonic generation can be used to selectively enhance emission from distinct quantum trajectories at high photon energies. Electrons rescattered in a strong field can traverse short and long quantum trajectories that exhibit differing coherence lengths as a result of variations in intensity of the driving laser along the direction of propagation. By varying the separation of the pulses in a counterpropagating pulse train, we selectively enhance either the long or the short quantum trajectory, and observe distinct spectral signatures in each case. This demonstrates a new type of coupling between the coherence of high-order harmonic beams and the attosecond time-scale quantum dynamics inherent in the process.

1 Author to whom any correspondence should be addressed.
High-order harmonic generation (HHG) of intense femtosecond laser pulses results in a tabletop source of ultrashort-pulse short-wavelength radiation with unique characteristics [1]. The emerging short wavelength beams can exhibit full spatial coherence [2, 3] making them useful for applications such as time resolved holography [2, 4], lensless imaging [5], or as a coherent seed pulse for further amplification in x-ray free electron lasers [6]. Furthermore, the coherent superposition of several harmonic orders results in a sequence of attosecond-duration bursts [7], which in some conditions can be limited to a single isolated attosecond pulse [8]–[10]. The short pulse duration has been used for a variety of experiments in high-energy atomic and molecular dynamics [11]–[13].

However, the spatial and temporal coherence of the visible laser beam that drives the HHG process is not directly imprinted on the high-harmonic beam, as is the case in conventional perturbative nonlinear optical processes. HHG results from a ‘rescattering’ process, where an electron is ionized from the atom, and a high-harmonic photon is emitted if the electron re-encounters the atomic core ∼3/4 cycle later. The phase of an emitted high-harmonic photon depends on the quantum phase advance that the electron accumulates during its free trajectory in the strong laser field [14]–[16]. An analysis of the possible trajectories of these electrons shows that for a particular harmonic order below the cutoff region, there exist two different electron quantum paths that contribute to the emission [17]. The so-called long quantum trajectory harmonics result from electrons that are ionized immediately following the peak of the laser field in a particular half-cycle, that re-encounter the ion core after a relatively long excursion (up to a full cycle of the laser field). The short quantum trajectory results from electrons that are ionized shortly after the peak in the laser field, and propagate in the continuum for approximately 1/2 laser cycle.

Quantum interference between the two paths, and their very different response to variations in the intensity of the driving laser, are major sources of potential spectral and temporal decoherence in HHG [18]. Hence, to obtain a highly coherent beam, or the shortest possible attosecond duration pulses, typically it is best to find operating conditions where only one of the two electron trajectories contributes to the coherent buildup of the signal. This can be done using two general approaches. First, for photon energies near the high-harmonic cutoff, the two trajectories lose their distinction. This near-cutoff region is useful for the generation of isolated attosecond pulse bursts. However, this ultimately limits the obtainable pulse duration, since it limits the useful spectral bandwidth. To produce shorter and more intense attosecond pulses, a single pulse comprising coherent light spanning the entire plateau region would be ideal [8]–[10], [19].

The second approach relies on the fact that the predominance of harmonic emission from either the short or long quantum trajectories depends on phase matching conditions in a non-trivial way. The two trajectories respond differently to variations in laser intensity, leading to differences in the ideal phase matching conditions [20, 21]. In a simple picture, the relative phase between the driving laser and the emitted harmonic light changes proportionally to the intensity of the driving laser field, with a proportionality factor much larger for the long quantum path than for the short [22]. As a result, the two quantum path components lead to generated fields with differing center frequencies and divergence angles. In its simplest manifestation, the short (long) trajectory emission is efficiently generated when the driving laser is diverging (converging) in the nonlinear medium. By varying the focusing conditions and geometry, the two contributions can be distinguished to some extent [18]. For example, the use of a diverging driving laser beam in a free focus, or phase-matched generation in a
hollow waveguide, preferentially selects harmonic emission originating from short electron trajectories [23]. However, this approach works best in regimes where relatively good phase matching can be achieved; i.e. where a balance of geometrical, material and plasma dispersions results in a long coherence length. This is possible only at relatively low levels of ionization of the nonlinear medium, which restricts this type of control to photon energies $\lesssim 130$ eV. Higher photon energies are generated at higher laser intensities and ionization levels. This clearly imposes a limit on the shortest possible attosecond pulse that can be generated, since, for example, the duration of a two-cycle pulse at 130 eV is 63 attoseconds. Such a pulse would require tens of electron volts of coherent bandwidth in the plateau and cutoff region.

In this work, we demonstrate that by controlling the quasi-phase matching (QPM) conditions using counterpropagating light fields in a gas-filled waveguide, we can selectively enhance high-harmonic emission from either the short or long quantum trajectories. This approach allows us to regain quantum path selectivity in HHG at high photon energies, while also enhancing the output using all-optical QPM [24]–[27]. (Note: just before this paper was submitted, the theoretical paper in [27] was published that predicts selective enhancement of the trajectories using QPM. Our analysis was developed independently.) We first show that the coherence lengths of HHG generated by the short and long trajectories, even in the same spectral regions, are quite different because of ionization dynamics in the medium as the laser propagates. Then, we use a counterpropagating pulse train to implement all-optical QPM, selectively enhancing high-harmonic emission for either the short or long electron trajectories at photon energies around $\sim 150$ eV. This is accomplished by tuning the separation between two counterpropagating pulses. This ability to enhance and select different quantum trajectories has important implications for attosecond pulse generation since isolated attosecond pulses are generated from a single electron recollision with an ion. Thus, by selectively optimizing a single electron quantum trajectory, it should be possible to generate tunable, sub-femtosecond, pulses at high photon energies with adjustable spectral bandwidth. Such pulses will have broad application for probing ultrafast electron dynamics in atoms and molecules.

HHG is a coherent nonlinear-optical process. Thus, if the process is not phase-matched, the buildup of a coherent signal occurs only over a distance corresponding to the coherence length $L_c = \pi / \Delta k$, where $\Delta k$ is the phase mismatch between the polarization and the harmonic waves. In HHG, the total phase mismatch has both extrinsic and intrinsic contributions, i.e. $\Delta k = \Delta k_{\text{ext}} + \Delta k_{\text{int}}$. The extrinsic phase mismatch, which results from different phase velocities of the pump and harmonic fields, is a sum of the contributions from neutral atoms, free electrons and geometric dispersions (e.g. free focusing or wave-guiding with or without Gouy phase shift) [23]. In our experiment, we used a gas-filled hollow fiber geometry, where the driving laser is guided in the fiber, and therefore it does not suffer from any Gouy phase shift. The intrinsic phase mismatch contribution, $\Delta k_{\text{int}}$, results from the intensity-dependent phase of the rescattering electron. At high ionization levels, where the free electron dispersion is dominant, the intrinsic phase mismatch has a sign opposite to the extrinsic term, and therefore will increase the coherence length. The intrinsic phase shift of the long trajectory harmonic emission is much larger than that of the short trajectory, and is thus more sensitive to variations in the driving laser pulse. Therefore, as will be demonstrated below, the coherence length is typically longer for the long trajectory.

First, we model the generated harmonic signal by solving the propagation equations for the driving and harmonic fields in time and along the propagation direction. The ionization rate is calculated using ADK theory [28]. The atomic dipole moment, which is the source for the
Figure 1. Calculated laser field at the input ($z = 0$ cm) and output ($z = 1$ cm) of the medium near the peak of the laser pulse ($T = 0$ fs), with (a) and without (b) the effects of time-dependent ionization by the laser field. The times within the optical cycle when an electron is ionized and then recombines to produce the 95th harmonic are also indicated for both the short (blue) and the long (red) trajectories. (c) Phase of long and short trajectories for the 95th harmonic as a function of $z$, both considering time-dependent ionization and preformed plasma contributions to dispersion, and excluding time-dependent ionization but including a preformed plasma contribution such that the plasma densities are the same at the peak of the laser field. (d) Coherence length at the peak of the laser pulse as a function of harmonic order for both cases.

harmonic field, is calculated using a generalized Lewenstein model that includes nonadiabatic effects [29, 30]. This method allows us to directly calculate the phase of a single harmonic-order from a distinct electronic trajectory under the ideal conditions. A typical example is presented in figure 1, where a Gaussian shape driving pulse with peak intensity $I = 7.4 \times 10^{14}$ W cm$^{-2}$ and pulse width $\tau = 27$ fs, propagates in 200 Torr of helium. Figure 1(a) plots a few optical cycles near the peak of the driving laser pulse, at the input ($z = 0$ cm) and output ($z = 1$ cm) of the medium. The times within the optical cycle when an electron that generates the 95th harmonic is ionized and recombines are also indicated, for both the short (blue) and the long (red) trajectories. Clearly, the shape of the pulse changes along the propagation direction due...
to time-dependent ionization-induced self-phase modulation as well as energy loss. This leads to a change in the intrinsic phase of the high-harmonic emission. To illustrate the effect of the ionization dynamics, figure 1(b) shows the laser pulse after traversing a distance of 1 cm, in the case where the laser propagates through a preformed plasma, with a free electron density fixed at the same electron density generated at the peak of the laser pulse in the case of figure 1(a). Figure 1(b) shows that the quantum trajectories are changed due to ionization dynamics in the medium, and not simply due to linear propagation of the laser pulse. Comparing figures 1(a) and (b), we note that any extrinsic phase mismatch contribution is identical for both cases, while the intrinsic contribution to the phase mismatch is significant only in the case of figure 1(a).

The influence of the intrinsic phase shift on the phase matching conditions is shown in figures 1(c) and (d). Figure 1(c) shows the phase of the long and short trajectory emissions of the 95th harmonic, along the propagation direction, for the two cases shown in figures 1(a) and (b). The calculated coherence lengths for several harmonic orders are plotted in figure 1(d). The effect of the intrinsic phase mismatch, which has opposite sign to the plasma-induced mismatch, is to increase the coherence length. Moreover, $L_c$ significantly differentiates between the long and short trajectories, allowing for high selectivity for QPM from a single trajectory.

We should note that in the calculations and experiments performed here, the laser intensity along the propagation direction gently decreases due to ionization/absorption. In experiments performed at higher laser intensities, the laser intensity distribution is often very complex, for example, due to ionization-induced periodic defocusing [31]–[33] or mode beating [34, 35]. Moreover, the coherence length of the long trajectory emission could be shorter than that corresponding to short trajectory emission, in those waveguide regions where the laser intensity increases significantly along the direction of propagation. Thus, direct in situ measurement of the different coherence lengths is very useful in understanding the ionization dynamics in the medium [36].

Using counterpropagating pulses to manipulate the HHG process, we can both directly measure and selectively enhance the emission originating from the two quantum paths. The experimental set-up is shown in figure 2(a). The driving laser and counterpropagating pulses are coupled into the opposite sides of a hollow waveguide by lens with 50 cm focal length. The coupling efficiencies for the driving laser and counterpropagating beams are 50 and 20%, respectively. We used a low-energy counterpropagating beam to directly measure the coherence length of harmonics that are generated under relatively benign conditions of low ionization levels (where plasma defocusing and mode beating are not strong and where good comparison of theory and experiment is possible). In this technique, a single weak counterpropagating pulse suppresses the generation process in regions where this pulse intersects with the driving laser pulse [37]. Consequently, a scan of the intersection point between the pulses results in oscillations in the harmonic output with a period corresponding to $2L_c$ [36]. In the few-optical-cycle regime of HHG, emission from the short and the long trajectories also experiences blueshifts of different magnitude due to the time-dependence of the intrinsic phase, which allows us to spectrally distinguish the two quantum path contributions [17]. Thus, we can separately measure $L_c$ for the short and long trajectories. Figure 2(b) shows a scan of the counterpropagating pulse intersection point for the 19th harmonic generated in an argon-filled hollow waveguide pressurized at 5 Torr. The harmonic emission is not phase matched at such low pressures, and both short and long trajectory emissions are observable. The measured coherence lengths of the long and short trajectory harmonics are 1.55 and 1.25 mm, respectively. As predicted by theory, $L_c$ is longer for the long trajectory than for the short trajectory.
Figure 2. (a) Experimental set-up for selecting emission from either the long or the short trajectory. (b) Experimentally measurement of coherence length of long and short trajectories of 19th harmonic. The driving and counterpropagating pulse energy and duration are 0.13 mJ and 25 fs, and 0.2 mJ and 1.6 ps, respectively. The black and red curves show 19th harmonic intensity as a function of position of intersection of the counterpropagating and driving laser pulses, for both long and short trajectories. Coherence lengths are measured as 1.55 and 1.25 mm for the long and short trajectories, respectively, near $z = 5$ mm. Harmonics are produced in an argon-filled hollow waveguide with 150 $\mu$m inner diameter and 5 Torr pressure.

Figure 3 plots our results obtained by using a pair of pulses to selectively enhance either the short or the long trajectory, in the photon energy range around the 95th harmonic order ($\approx 150$ eV) in helium. The driving and counterpropagating pulse energies and durations are 0.13 mJ and 25 fs, and 0.2 mJ and 1.6 ps, respectively. The gray curve shows the harmonic spectrum in the absence of any counterpropagating pulses, which, under our experimental
Figure 3. Selective enhancement of either the long or short quantum trajectories at high photon energy using counterpropagating pulse trains containing two pulses with different separations, in He at 200 Torr. The dashed lines mark the position of the harmonic peaks for the short electron trajectories. The gray, red and blue curves show harmonic spectra with no counterpropagating pulses (gray); with two counterpropagating pulses at pulse separations of either 356 µm (blue) and 840 µm (red), respectively.

Conditions of 200 Torr He, is dominated by the short trajectory because the coherence length associated with the short trajectory is longer than the coherence length associated with the long trajectory. The blue curve is the spectrum obtained for an interpulse spacing of 356 µm between the two counterpropagating pulses. In this case, the emission is enhanced in the range of harmonic orders 85–97, while the peak positions for the harmonics 83–95 are significantly blueshifted compared with the positions of the short trajectory harmonics—as is expected if the emission is now predominantly originating from the long trajectory. Here, the 356 µm interpulse spacing corresponds to first-order QPM and optimizes harmonic orders with coherence length ~183 µm. Also, the positions of the peaks for harmonics 97–99 show little or no observable shift. This is as expected, since the distinction between the long and short trajectories diminishes near the cutoff photon energy. Interestingly, for harmonic orders around 81, no spectral shift is observed. This is because the coherence length varies with harmonic order according to $L_c \propto 1/q$. Therefore, using a fixed pulse separation of 356 µm, only a limited numbers of harmonics (83–95) of long trajectories are enhanced.

In both present and past work, harmonics originating from the long quantum trajectory are most easily enhanced using all-optical QPM, since this trajectory spends a longer time in the continuum and is thus influenced more easily by the field of the counterpropagating pulse. However, the harmonics originating from the short quantum trajectory can also be enhanced—most effectively by using a larger interpulse spacing that allows for more selectivity. Higher order QPM can better differentiate harmonic emission corresponding to different coherence lengths, because of the larger phase slip over longer distances containing more coherent zones. The red curve plots the measured harmonic emission in the presence of a pair of
counterpropagating pulses separated by 840 µm. In this case, the positions of the spectral peaks are not shifted from their position without counterpropagating pulses present—i.e. the short trajectory emission is enhanced. The interpulse spacing for this spectrum corresponds to second-order QPM for the 95th order, or $4L_c$.

In conclusion, we demonstrate both theoretically and experimentally that the intrinsic phase shift as a result of ionization dynamics in the medium induces different coherence lengths for harmonic emissions from the long and short electron quantum trajectories. These differing coherence lengths allow for selective enhancement of the emission from either trajectory alone, even at high-photon energies around 150 eV. Detailed control over harmonic emission and phase matching conditions suggests a number of possibilities in shaping short wavelength light pulses, in generating fully coherent beams from high harmonics at high-photon energies, and in generating very short attosecond pulses using both the plateau and cutoff harmonics.

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