Tailoring isolated attosecond pulses using quantum path interferences

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Abstract. By performing calculations on the generation of isolated attosecond pulses by intense and few-cycle laser fields, we demonstrate that the pulse temporal width is sensitive to the angle of emission. We show that this effect results from the interference of the short and long electronic trajectory contributions, responsible for high-order harmonic generation. In particular, we find that the shortest pulses are emitted off-axis, which are substantially shorter than those emitted on-axis, where single trajectory contributions dominate.

1. Introduction
There is an astonishing simplicity in some processes of intense laser-matter interactions. At high field amplitudes, there is a large probability for the electron to tunnel out of the atom. Afterwards, the field interaction dominates and the electrons evolve almost freely in the continuum. High-order harmonics are emitted when these fast electrons are driven back to rescatter the ion, converting their kinetic energy into high-frequency radiation, in a process known as high-order harmonic generation (HHG) [1, 2]. The simplicity of this process is mapped into the quantum mechanical wavefunction, described by a small set of semiclassical orbits, or Feynman's paths [3]. The energy conservation demands that each orbit contributes to a single harmonic, with a phase given by its action. However, as different paths may have the same rescattering energy, the final harmonic signal is the result of the interference of the degenerated quantum path contributions.

One of the most exciting perspectives of HHG by intense lasers is the possibility of synthesizing XUV pulses of sub-femtosecond duration [4, 5, 6]. An attosecond pulse train (1 as = 10^{-18} s) is obtained by the selection of the higher frequency part of the harmonic spectrum which conform the plateau region. For the correct synthesis, the spectrum should approach two conditions: on one side its structure should approach that of a frequency comb, in which the harmonic intensities are similar; on the other side, the relative phase between the harmonics should be nearly constant (phase locking) [4]. Fortunately, these two conditions are approached in the typical harmonic spectra generated during the interaction of intense fields with matter.

However, the HHG mechanism itself prevents the harmonics to be emitted in the Fourier limit. Since the different electronic trajectories rescatter at different times [7], a chirp is imprinted in the emitted radiation. The emission from the short trajectories is positively chirped (the lower harmonics are emitted before the higher), while the emission from the long trajectories exhibits...
a negative chirp (the higher harmonics are emitted before the lower) [8, 9]. This chirp is referred in the literature as attochirp.

The first experimental evidence of an attosecond pulse train was obtained by selecting five consecutive harmonics generated in argon, obtaining 250 as pulses [10]. Almost simultaneously, isolated pulses with durations of 650 as were generated by spectrally filtering a few cut-off harmonics produced by an ultrashort laser pulse [11]. At present, after postcompression of the attosecond pulses, isolated pulses of temporal durations < 100 attoseconds have been measured experimentally [12, 13]. Recent works proposed the synthesis of X-ray waveforms in the zeptosecond regime [14], using mid-infrared laser sources to drive ultra-high order harmonics generation [15].

In this contribution we explore the relevance of macroscopic phase-matching conditions for the final phase-locking of the harmonics. In particular, we show that at certain angles of detection, where the short and the long trajectories are relevant, the interference of these contributions can lead to a final attosecond burst with smaller duration than the corresponding to a single trajectory. This effect was already observed in [16] for longer pulses, and the interference between short and long paths was identified to occur between paths from two consecutive ionization maxima. Here we explore this effect over isolated attosecond pulses driven by few-cycle pulses.

2. Methods and setup
Our computations are carried on using the extended strong field approximation (SFA+) [17] for the single atom response, combined with a discrete dipole approach for computing the harmonic propagation [18]. Our approach is based in the integral solution of the wave equation, instead of resorting to its numerical integration [19]. Every point in the target is treated as source of an elementary wave, which is then propagated to the detector. The final field at the detector is, thus, the coherent addition of these elementary contributions. We take into account propagation effects in the fundamental field such as the free charges and neutrals. More information on this method and the SFA+ model used to compute the single-atom harmonic generation may be found in [20, 16].

Figure 1. Scheme of the off-axis detection geometry used to demonstrate the angular chirping compensation of the attosecond bursts. A Gaussian beam propagating along the z direction is focused into an argon gas jet. The gas jet profile is constant in the x direction, while Gaussian in y and z directions, with 500 μm full width at half maximum (FWHM), and peak density 10^{18} atoms/cm^3. For the angular resolved detection, we consider a window of 0.5 mrad in the far field profile.

We have performed our computations assuming a fundamental field modeled by a sin^2 envelope at 800 nm, with intensity at focus ≃ 2.45 × 10^{14} W/cm^2 and polarized along the
The beam is assumed Gaussian, propagating along the z direction. The beam waist at focus is 22.5 µm and the confocal parameter results \( b = 3.98 \) mm. The target is modeled as an argon gas jet flowing perpendicularly to the Gaussian beam. The gas jet profile is constant in the \( x \) direction, while Gaussian in \( y \) and \( z \) directions, with 500 µm full width at half maximum (FWHM), and peak density \( 10^{18} \) atoms/cm\(^3\). For the angular resolved detection, we consider a window of 0.5 mrad in the far field profile, that would correspond experimentally to an annular gate. Figure 1 illustrates the scheme of this setup.

It is possible to obtain an isolated attosecond pulse using few-cycle driving pulses [6, 11], since high order harmonics are then generated in a single rescattering event. For that purpose we have performed our computations assuming a fundamental few-cycle laser pulse of 1.4 cycles (3.8 fs) FWHM. Other techniques have been applied for isolating a single attosecond pulse [5, 21, 22], whereas the extraction of the different attosecond pulses from the train has also been reported recently [23].

### 3. Compensating the attochirp of isolated pulses

We plot in Fig. 2 the attosecond pulses obtained with our numerical simulation after Fourier transforming the detected harmonic spectra. The low energy part of the harmonic spectra has been filtered with an Al plate 100 µm width. We have considered two different situations: target placed before the laser focus (left column) and after the focus (right column). The first row in Fig. 2—plots (a) and (b)—corresponds to the attosecond pulses detected on-axis, while the second row shows the off-axis detection—plots (c) and (d)—, where the spatial window was centered at 3.3 mrad. First, we observe that the CEP choice and the parameters of the propagation led to an isolated attosecond pulse when the gas jet is placed before the focus, whereas a two-pulse train structure after the focus. This behavior can be modified by changing the CEP of the incident laser pulse.

![Figure 2](image_url)

**Figure 2.** Temporal distribution of the attosecond pulses when the high-order harmonics are detected in a window (a) on-axis and (b) centered at 3.3 mrad, when the gas jet is placed 1 mm before the focus position. Plots (c) and (d) represent the same situation as (a) and (b), but when the gas jet is placed 1 mm after the focus position. The driving laser pulse is 1.4 cycles (3.8 fs) FWHM sin\(^2\), 800 nm, with intensity at focus \( \simeq 2.45 \times 10^{14} \) W/cm\(^2\).
More interesting is the comparison between the attosecond pulse width at different detection angles. From plots (a) and (c) we can observe how the attosecond pulse width has been reduced from 311 as on-axis to 183 as off-axis. This reduction has a physical explanation in the interference between the radiation bursts associated with the rescattering of short and long paths, which is more acute off-axis, where both trajectories have comparable weights, rather than on-axis, where short trajectories dominate. For the same reason, this effect is more acute when the gas jet is placed after the focus. In that situation, short trajectories are phase-matched on-axis, whereas as we increase the detection angle, long trajectories become dominant. In an intermediate situation, both quantum paths can have similar weights and interfere. In the temporal domain, this interference has been identified to occur between the short and long trajectories originated respectively, at two consecutive ionization maxima [16]. Counterintuitively, even with few-cycle pulses, the weight of quantum paths from two consecutive ionization maxima in each half-cycle is high enough to interfere with similar weight.

On the other hand, if we place the gas jet after the focus, phase-matching favors only short trajectories at any angle. As a consequence, in plots (b) and (d) we do not observe any major change in the attosecond pulse width when increasing the detection angle.

Let us now study in detail how the isolated attosecond pulse changes with the detection angle for a gas jet placed before the focus. In Fig. we present the isolated attosecond pulse and a time-frequency analysis of the spectra when detected (a-b) on-axis, and at (c-d) 0.7, (e-f) 1.3, (g-h) 2.0, (i-j) 2.6, (k-l) 3.3 and (m-n) 4.0 mrad off-axis. If we look at the time-frequency analysis on-axis –plot (b)– we observe that the electron’s recollision energy shows a roughly linear dependence on time, in which the lower harmonics are emitted before the higher ones, giving rise to a positive chirp typical from the harmonic spectrum emitted by short trajectories. However, as we increase the detection angle, we observe how the time-frequency structure changes leading to a vertical structure when detected above 3.3 mrad –plots (l) and (n)–. In those cases, the destructive interference between paths tends to confine the harmonic radiation into a narrower window, reducing drastically the positive chirp associated to short trajectories alone.

In the left column of Fig. 3 we observe how one can tailor the attosecond pulse waveform by changing the detection angle. Although the efficiency of the signal decreases when for off-axis detection, the pulse width is reduced to nearly half of the width obtained on-axis.

4. Conclusions
We have proposed a route to directly compensate the chirp of isolated attosecond pulses by detecting them at a certain angle from the propagation axis. Performing a time-frequency analysis, we have identified the interference between different quantum paths as the underlying mechanism for this compensation. We have presented the detection angle as a parameter for controlling the macroscopic signatures of these single-atom processes.

Acknowledgments
We acknowledge support from Junta de Castilla y León (Consejería de Educación and Fondo Social Europeo), Spanish MINECO (FIS2009-09522, Consolider Program SAUUL CSD2007-00013), and Centro de Láseres Pulsados, CLPU.

References
[1] Corkum P B 1993 Phys. Rev. Lett. 71, 1994
[2] Schader K, Yang B, DiMauro L F and Kulander K C 1993 Phys. Rev. Lett. 70, 1599
[3] Salières P, Carré B, Le Déoff L, Grashov F, Paulus G, Walther H, Kopold R, Becker W, Milosovic D, Sanpera A and Lewenstein M 2001 Science, 292, 902
[4] Farkas G and Toth C 1992 Phys. Lett. A 168, 447
[5] Corkum P B, Burnett N H and Ivanov M Y 1994 Opt. Lett. 19, 1870
[6] Christov I P, Murnane M M and Kapteyn H C 1997 Phys. Rev. Lett 78, 1251
Figure 3. Temporal distribution of the attosecond train of pulses (left column) and time-frequency structure (right column) when the high-order harmonics are detected in a window (a-b) on-axis in the first row, and centered at (c-d) 0.7, (e-f) 1.3, (g-h) 2.0, (i-j) 2.6, (k-l) 3.3 and (m-n) 4.0 mrad off-axis in the following rows. The gas jet is placed 1 mm before the focus position, and the laser pulse is 1.4 cycles (3.8 fs) FWHM $\sin^2$, 800 nm, with intensity at focus $\simeq 2.45 \times 10^{14}$ W/cm$^2$. 
[7] Antoine P, L’Huillier A and Lewenstein M, 1996 Phys. Rev. Lett. 77, 1234
[8] Mairesse Y, de Bohan A, Frasinski L J, Merdji H, Dinh L C, Monchicourt P, Breger P, Kovacev M, Taieb R, Carré B, Muller H G, Agostini P and Salières P 2003 Science, 302, 1540
[9] Varjú K, Mairesse Y, Carré B, Gaarde M B, Johnson P, Kazamias S, López-Martens R, Mauritsson J, Schafer K J, Balcou Ph, L’Huillier A and Salières P 2005 J. of Mod. Opt., 52, 379
[10] P. M. Paul, E. S. Toma, P. Breger, G. Mullot, F. Augé, Ph. Balcou, H. G. Muller and P. Agostini Science 292, 1689 (2001).
[11] Hentschel M, Kienberger R, Spielmann C, Reider G A, Milosevic N, Heinzmann U, Drescher M and Krausz F 2001 Nature 414, 509
[12] Goulielmakis E, Schultze M, Hofstetter M, Yakovlev V S, Gagnon J, Uiberacker M, Aquila A L, Gullikson E M, Attwood D T, Kienberger R, Krausz F and Kleineberg U 2008 Science, 320, 1614
[13] Zhao K, Zhang Q, Chini M, Wu Y, Wang X and Chang Z 2012 Opt. Lett. 37, 389
[14] Hernández-García C, Pérez-Hernández J A, Popmintchev T, Murnane M M, Kapteyn H C, Jaron-Becker A, Becker A and Plaja L 2012 Zeptosecond keV X-ray waveforms driven by mid-infrared laser pulses, submitted
[15] Popmintchev T, Chen M-C, Popmintchev D, Arpin P, Brown S, Alisaukas S, Andriukaitis G, Balciunas T, Mücke O, Pugzlys A, Baltuška A, Shim B, Schrauth S E, Gaeta A, Hernández-García C, Plaja L, Becker A, Jaron-Becker A, Murnane M M and Kapteyn H C 2012 Science 336, 1287
[16] Hernández-García C and Plaja L 2012 Journal of Physics B: At. Mol. Opt. Phys. 45, 074021
[17] Pérez-Hernández J A, Roso L and Plaja L 2009 Opt. Exp. 17, 9891
[18] Hernández-García C, Pérez-Hernández J A, Ramos J, Conejero Jarque E, Roso L and Plaja L, 2010 Phys. Rev. A 82, 033432
[19] L’Huillier A, Li X F and Louprière L A 1999 J. Opt. Soc. Am. B. 7, 527
[20] Pérez-Hernández J A, Hernández-García C, Ramos J, Conejero E, Plaja L and Roso L 2011, New Methods For Computing High-OrderHarmonic Generation and Propagation (Progress in Ultrafast Intense Laser Science VII), (pp 145-162), Springer chapter 7 pp 145-162
Kuhn T 1998 Density matrix theory of coherent ultrafast dynamics Theory of Transport Properties of Semiconductor Nanostructures (Electronic Materials vol 4) ed E Scholl (London: Chapman and Hall) chapter 6 pp 173214
[21] Sola I J, Mével E, Elouga L, Constant E, Strekalov V, Poletto L, Villoresi P, Benedetti E, Caumes J-P, Stagira S, Vozzi C, Sansone G and Nisoli M, 2006 Nature Phys. 2, 319
[22] Sansone G, Benedetti E, Calegari F, Vozzi C, Avaldi L, Flammini R, Poletto L, Villoresi P, Altucci C, Velotta R, Stagira S, De Silvestri S and Nisoli M 2006 Science 314, 443
[23] Vincenti H and Quéré F 2012 Phys. Rev. Lett. 108, 113904