Attosecond control of electron emission from atoms

G. Laurent¹,², W. Cao¹, I. Ben-Itzhak¹ and C. L. Cocke¹

¹ James R. Macdonald Laboratory, Physics Department, Kansas State University, Manhattan, KS 66506, USA
² Research Laboratory of Electronics, Massachusetts Institute of Technology, Cambridge, MA 02139, USA

E-mail: glaurent@mit.edu

Abstract. We demonstrate that the electron emission from atoms can be temporally controlled on an attosecond time scale. Electron wave-packets are formed by ionizing an atomic target with an attosecond pulse train composed of both odd and even high-order harmonics in the presence of a relatively weak infrared field. We show that interference between one- and two-photon transitions produces a large asymmetry in the angular distribution of the photoelectrons. The direction of the emission can be controlled on an attosecond time scale by varying the time delay between the two pulses. In addition, we show that such asymmetric emission is also related to the properties of the attosecond pulse train. The temporal analysis of the modulated electron emission, based on an accurate description of the atomic physics of the photoionization process, then provides a way to measure the temporal profile of the attosecond pulse.

1. Introduction

Coherent control of electron dynamics in atoms, molecules and condensed matter is a growing research field in ultrafast science, which has been mainly driven over the last two decades by major advances in laser technology. Recently, the advent of x- or extreme-ultraviolet (XUV-EUV) light pulses in the attosecond time scale has opened up new avenues for experimentalists to manipulate the electron dynamics with unprecedented precision [1–3]. Because the current attosecond sources available in the laboratory are not intense enough to perform “pure” attosecond pump/probe experiments, attosecond control is typically achieved through the exposure of the target to both an attosecond pulse and a phase-locked optical IR field, with a variable time delay between the two. So far, this scheme has been successfully employed to manipulate electron dynamics in various benchmark systems. For instance, in recent works, the dissociative ionization of diatomic molecule was controlled by manipulating the electronic charge distribution within the target [4–8]. Also, Mauritsson et al. reported an electron scattering imaging technique, based on a sequence of attosecond pulses used to release electrons into a sufficiently strong IR field to guide them back to their parent ions [9].

In this work, we demonstrate that an asymmetric electron emission from atomic targets can be generated and controlled by combining an attosecond pulse train (APT) and a much weaker IR field than in the previous study. We show that interference between one- and two-photon transitions produces a large asymmetry in the angular distribution of the photoelectrons though the separate contributions of the two paths have no asymmetry. The direction of the emission can be controlled on an attosecond time scale by varying the time delay between the two pulses. In addition, we show that such asymmetric emission is also related to the properties of the APT.
Figure 1. Principle of the experiment. (a) Energy-degenerate electron wave-packets are formed by ionizing argon atoms with an APT composed of odd and even harmonics in the presence of a weak IR field (\(\sim 10^{11}\) W.cm\(^{-2}\)). (b) Interference between the different quantum paths leads to a modulation of the electron emission as a function of the delay between the APT and IR fields. Such a modulation can be decomposed into three components: a constant term \(I_0\) symmetric along the polarization vector direction, and two oscillating terms \(I_{2\omega}\) and \(I_{\omega}\) symmetric and antisymmetric along the polarization direction, respectively.

The temporal analysis of the modulated electron emission, based on an accurate description of the atomic physics of the photoionization process, then provides a way to measure the temporal profile of the attosecond pulse.

2. Method
The basic principle of our experiment is presented Fig. 1(a). Electron wave-packets are formed by ionizing argon gas with an APT composed of both odd and even high-order harmonics in the presence of a weak IR field (\(\sim 10^{11}\) W.cm\(^{-2}\)). Consequently, a mix of energy-degenerate even \((s,d)\) and odd \((p,f)\) parity states is fed into the continuum by one- and two-photon transitions. These interfere, leading to an asymmetric electron emission along the polarization vector. At some appropriate time delay between the APT and IR fields, the even and odd angular continuum wave-function resulting from one- and two-photon transitions, respectively, add constructively on one side (up) of the polarization vector direction and destructively on the other side (down), thus creating a strong up-down asymmetry in the angular emission of the photoelectrons. This asymmetry will then oscillate as the time delay is varied.

In our previous studies [10–12], we have shown that the low intensity of both the APT and IR fields allows for a quantum description of the angular electron emission within the framework of second-order perturbation theory and the single active electron approximation. At a time delay \(\tau\) between the attosecond and IR pulses, the probability \(F(\theta,\varepsilon,\tau)\) for emission of an electron in the direction \(\theta\) with respect to the polarization vector and energy \(\varepsilon\) is given by the coherent sum of the amplitudes of the three paths shown in Fig. 1, denoted \(M_q(\theta,\varepsilon)\), \(M_{q-1}(\theta,\varepsilon,\tau)\), and
The general expression for the probability $F(\theta, \varepsilon, \tau)$ obtained by including the final angular momentum of the photoelectrons in the theoretical model is not very transparent. For a sake of clarity, we therefore choose to discuss here the expression which results from the following simplifying assumptions: (i) we consider the initial $m = 0$ state only for the $3p$ electron, (ii) we assume that the matrix element from the ground state to the continuum is approximately independent of the final energy and its phase can be factored out, and (iii) we assume that the continuum to continuum dipole matrix elements are real and equal for single IR photon absorption and emission. Of course, none of these assumptions are justified, and we will relax them later for a proper comparison with our experimental results. Under these simplifications, the probability of emission $F(\theta, \varepsilon, \tau)$ can be written as the sum of three terms:

$$F(\theta, \varepsilon, \tau) \approx E_{q}^{2} f_{c}(\theta)^{2} + (E_{q-1}^{2} + E_{q+1}^{2}) E_{\omega}^{2} f_{o}(\theta)^{2}$$

(1)

$$+ E_{q}[E_{q-1} \sin(\omega \tau + \phi_{q-1} - \phi_{q}) - E_{q+1} \sin(\omega \tau + \phi_{q} - \phi_{q+1})] E_{\omega} f_{c}(\theta) f_{o}(\theta)$$

(2)

$$+ E_{q-1} E_{q+1} \cos(2\omega \tau + \phi_{q-1} - \phi_{q+1}) E_{\omega}^{2} f_{o}(\theta)^{2}$$

where $E_{q}$ and $\phi_{q}$ are the magnitude and phase of the $q^{th}$ component of the APT, $E_{\omega}$ and $\omega$ are the magnitude and angular frequency of the IR field, and $\tau$ the XUV-IR delay. The atomic physics of the photoionization process is absorbed into $f_{c}$ and $f_{o}$. Each of these functions is a weighted sum of spherical harmonics $Y_{\ell,m}(\theta)$ with, respectively, even and odd parity (for an argon target), and both of them are real functions under our assumptions:

$$f_{c}(\theta) = Z_{p-s}^{q} Y_{00}(\theta) + Z_{p-d}^{q} Y_{20}(\theta)$$

$$f_{o}(\theta) = (Z_{p-s}^{q+1} Z_{s-p}^{\omega} + Z_{p-d}^{q+1} Z_{d-p}^{\omega}) Y_{10}(\theta) + Z_{p-d}^{q+1} Z_{d-s}^{\omega} Y_{30}(\theta),$$

(3)

where $(Z_{p-s}^{q+1}, Z_{p-d}^{q+1})$ are the real part of the one-photon matrix elements from the $3p$ ground state to the continuum states, and $(Z_{s-p}^{\omega}, Z_{d-p}^{\omega}, Z_{d-s}^{\omega})$ are the real part of the continuum-continuum matrix elements.

In Eq. (2), the first term ($I_{0}$) is the sum of the angular distributions for the one-photon and two-photon processes individually. This term does not depend on the time delay $\tau$, and the associated angular distribution can be expanded in even order Legendre polynomials, which are symmetric along the polarization vector direction. The second term ($I_{\omega}$) is an interference term coming from the cross product of the one- and two-photon transition amplitudes, a process that we referred to as First order/Second order Interference (FSI). This interference term varies sinusoidally with $\tau$ at the IR frequency $\omega$, and the angular distribution can be expanded in odd-order Legendre polynomials, which are antisymmetric along the polarization vector direction. Finally, the last term ($I_{2\omega}$) comes from the cross product of the two-photon transition amplitudes. This interference term varies sinusoidally with $\tau$ at twice the IR frequency, and is also symmetric along the polarization vector direction. It too can be expanded in even-order Legendre polynomials.

It is important to note, though, that the FSI process is not reflected in the total photoionization probability, since the continuum states populated by the one- and two-photon quantum paths are orthogonal to each other. Experimental observation of the FSI modulation thus requires a photoelectron momentum spectroscopy measurement.

$$M_{q+1}(\theta, \varepsilon, \tau),$$

respectively:

$$F(\theta, \varepsilon, \tau) = |M_{q}(\theta, \varepsilon) + M_{q-1}(\theta, \varepsilon, \tau) + M_{q+1}(\theta, \varepsilon, \tau)|^{2},$$

where $\varepsilon = \omega_{q} - \varepsilon_{i}$, and $\omega_{q} = \omega_{q-1} + \omega = \omega_{q+1} - \omega$. The general expression for the probability $F(\theta, \varepsilon, \tau)$ obtained by including the final angular momentum of the photoelectrons in the theoretical model is not very transparent. For a sake of clarity, we therefore choose to discuss here the expression which results from the following simplifying assumptions: (i) we consider the initial $m = 0$ state only for the $3p$ electron, (ii) we assume that the matrix element from the ground state to the continuum is approximately independent of the final energy and its phase can be factored out, and (iii) we assume that the continuum to continuum dipole matrix elements are real and equal for single IR photon absorption and emission. Of course, none of these assumptions are justified, and we will relax them later for a proper comparison with our experimental results. Under these simplifications, the probability of emission $F(\theta, \varepsilon, \tau)$ can be written as the sum of three terms:

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$$+ E_{q-1} E_{q+1} \cos(2\omega \tau + \phi_{q-1} - \phi_{q+1}) E_{\omega}^{2} f_{o}(\theta)^{2}$$

where $E_{q}$ and $\phi_{q}$ are the magnitude and phase of the $q^{th}$ component of the APT, $E_{\omega}$ and $\omega$ are the magnitude and angular frequency of the IR field, and $\tau$ the XUV-IR delay. The atomic physics of the photoionization process is absorbed into $f_{c}$ and $f_{o}$. Each of these functions is a weighted sum of spherical harmonics $Y_{\ell,m}(\theta)$ with, respectively, even and odd parity (for an argon target), and both of them are real functions under our assumptions:

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where $(Z_{p-s}^{q+1}, Z_{p-d}^{q+1})$ are the real part of the one-photon matrix elements from the $3p$ ground state to the continuum states, and $(Z_{s-p}^{\omega}, Z_{d-p}^{\omega}, Z_{d-s}^{\omega})$ are the real part of the continuum-continuum matrix elements.

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It is important to note, though, that the FSI process is not reflected in the total photoionization probability, since the continuum states populated by the one- and two-photon quantum paths are orthogonal to each other. Experimental observation of the FSI modulation thus requires a photoelectron momentum spectroscopy measurement.
3. Experimental setup
An overview of the experimental setup is shown in Fig. 2. It combines a Ti:Sapphire laser delivering 40-fs 800-nm pulses at a repetition rate of 1 kHz, a XUV-IR Mach-Zehnder interferometer, and a velocity map imaging system. Part of the incoming linearly polarized IR beam was sent into the first arm of the interferometer where the APT was generated via high harmonic generation with a two-color (800- and 400-nm) field [13]. The IR beam was first focused by a lens of 50-cm focal length, and then frequency doubled in a 140-µm-thick β-barium borate (BBO) crystal. The resulting orthogonally polarized two-color field focused into a 1.5 mm windowless gas cell filled with 10 Torr of argon. At the focal point, the intensity of the 400-nm component was typically 0.5% of that of the 800-nm field. By combining 800 and 400 nm for the harmonics generation, odd and even harmonic orders were produced [14, 15]. The APT was then filtered by using a spatial aperture (2 mm diameter at 0.5 m) and a 200 nm thick Al thin film to remove harmonics below the 11th order. A replica of the IR (without the 400 nm) was sent into the second arm of the interferometer, whose total length could be changed to vary the time delay \( \tau \) between the APT and the IR pulse. Both beams were focused, collinearly recombined with parallel polarization (the direction of polarization of the APT was adjusted by rotating the polarization of the IR driving field with a half-wave plate before the high harmonic generation), and finally sent into a vacuum chamber containing an effusive argon gas jet. At the focal point, the IR intensity is estimated to be below \( 10^{11} \) W cm\(^{-2} \). A velocity map imaging system was used to measure the photoelectrons momenta [16].

4. Results and discussions
First, we discuss the results of the well-know RABBITT experiment (Reconstruction of Attosecond Beating By Interference of Two-photon Transitions) [2,3,17]. In such an experiment, a comb of odd-order harmonics only was used to ionize argon in the presence of a weak IR field. Figures 3(a) and 3(b) present the up and down photoelectron energy spectra as a function of the delay \( \tau \) between the APT and the IR field. Electrons are observed at energies corresponding
Figure 3. Density plot of the photoelectron emission probability from argon measured in both direction (up and down) along the polarization vector ($\pm 30$ deg.) as a function of the electron energy (in units of harmonic order) and the time delay between the APT and IR fields: (a,b) APT composed of odd harmonics only, (c,d) APT composed of odd and even harmonics. The corresponding experimental XUV harmonic spectrum is shown in the right panel of each plot.

To one-photon absorption of the odd harmonics, and, located in between, sideband peaks due to two-photon transitions (absorption of one XUV photon plus absorption or emission of one IR photon). Two different quantum paths involving two consecutive harmonics contribute to the same sideband quantum state, and thus interfere. This interference leads to a modulation of the intensity of each sideband peak, which oscillates with $\tau$ at twice the frequency of the IR field, as predicted by second-order perturbation theory [18]. By comparing Figs. 3(a) and 3(b), we also observe that the oscillations of the sidebands associated with electrons emitted in the up and down directions are in phase. This means that the electron emission is symmetric along the polarization vector direction at any time delay between the APT and the IR pulse.

When the APT is composed of both even and odd harmonics, the situation becomes slightly more complicated. Now, three transitions contribute to the generation of photoelectrons at energy corresponding to each harmonic: two transitions involving absorption of one XUV photon plus absorption or emission of one IR photon, and the direct transition induced by absorption of a single XUV photon. Interference between the one- and two-photon transitions adds new features in the electron emission compared to the RABBITT experiment. The up and down energy spectra as a function of the delay $\tau$ when even and odd harmonics are present are shown in Fig. 3(c) and 3(d). First, we see that oscillations with periods of both 1.35 and 2.7 fs are present in the yields. The oscillation with the shorter period is due to the interference between the two-photon transitions, as in the RABBITT experiment, while the 2.7 fs oscillation is the result of the interference between the one- and two-photon transitions, the FSI process. Second,
the yields of electrons emitted in the up and down directions oscillate out-of-phase with each other, indicating that the emission is now asymmetric along the polarization vector direction.

To unravel the symmetric and asymmetric components of the electron emission, we have expanded the full angular-resolved photoelectron distribution measured as a sum of odd- and even-Legendre polynomials:

\[ F(\theta, \varepsilon, \tau) = 2L_{\text{max}} \sum_{J=0}^{2L_{\text{max}}} \beta_J(\varepsilon, \tau) P_J[\cos(\theta)]. \tag{4} \]

The expansion coefficients \( \beta_J \) associated with the first four polynomials \( P_J \) as a function of the delay \( \tau \) between the APT and the IR field are plotted in Fig. 4. The asymmetric component of the electron emission represented by the coefficients \( \beta_1 \) and \( \beta_3 \) oscillates at the IR frequency, while the symmetric component represented by the coefficients \( \beta_0 \) and \( \beta_2 \) oscillates at twice the IR frequency, as predicted by Eq. 2.

An unexpected feature is observed in the maps for the coefficient \( \beta_1 \) and \( \beta_3 \), though. Both maps resemble a checkerboard. This pattern indicates that, at a given time delay \( \tau \), the asymmetric emission of photoelectrons associated with odd harmonics points in a direction along the polarization vector opposite to that associated with the even harmonics. A closer look at Eq. (2) reveals that the electron emission depends on both the amplitude \( E_q \) and the phase \( \phi_q \) of the harmonics. Our measurements have shown that this pattern is insensitive to substantial changes in the relative intensities of the harmonics in the comb. We therefore believe that the origin of this effect lies in the intrinsic relative phases of the consecutive harmonic orders. In
order to explore this characteristic, we have carried out a calculation of the expected values of the $\beta$ coefficients without the simplifying assumptions underlying Eq. (2). To describe the one- and two-photon ionization process, we have used the results developed by Dahlstr"om et al. within the framework of first and second-order perturbation theory and the single active electron approximation [19]. The calculations were performed for the three possible initial $m$ values for the $3p$ state and added incoherently. We have used the harmonic intensities and linewidths consistent with the experimental XUV spectrum shown in Fig. 3. The phase between consecutive odd and even harmonics, on the other hand, was used as a parameter in our model (the phase for each harmonic was kept constant across the harmonic itself). The resulting maps for the coefficient $\beta_1$ are shown in Figs. 5(a) and 5(b) for a phase shift between odd and even harmonics of $0.35\pi$ and $0.5\pi$, respectively. Comparison of Fig. 5(b) with the experimental result of Fig. 4(b) strongly suggests that the actual even-odd phase shift is near $\pi/2$. On the contrary, the calculation performed for an even-odd phase shift of $0.35\pi$ is significantly different from the measurement. We note that the result for $0.65\pi$ (not shown) is nearly the same as that for $0.35\pi$.

We do not yet know of a physical explanation for these phase shifts. But, we have recently observed that the quantitative value of the even-odd phase shift is specific to the procedure used to generate the APT. In a recent study, where a parallel polarized two-color field has been used to generate the APT, we have found that the phase shift is not constant over the whole harmonic spectrum [12]. It decreases (or increases) from $\pi/2$ for the lower harmonics in the plateau region to close to 0 (or $\pi$) for the higher ones close to the cut-off (our experiment cannot distinguish between these two values). We note that the previous theoretical studies of Ivanov and Zuo also predicted, for harmonics near the cutoff, very large phase shifts, nearly $\pi$ [20,21]. Further investigation of this question would be a separate, and very worthy, endeavor.

Finally, it is interesting to notice the implications this phase-shift has for the physical picture of the electric field of the APT. It is often assumed that the combination of even and odd harmonics in the pulse train creates a series of pulses which occur only once per optical cycle of the fundamental field, rather than half when only odd harmonics are present in the pulse, and thus offer the possibility to strobe an atomic system only once during each IR cycle. The temporal profile of the APT shown in Fig. 6 contradicts this nice physical picture. The picture holds only if there is no phase shift between even and odd harmonics as shown in Fig. 6(a). If the even-odd phase shift is different from zero, as our measurement suggests it can be, the resulting APT has a more complex structure not resembling a single AP once per IR period.

Figure 6. Temporal profile of the attosecond pulse train retrieved for an even-odd phase shift of 0 (a), and $\pi/2$ (b).
It is still correct that the repetition time of the pulse is a full optical cycle, but the waveform becomes a more complex and extended pulse.

5. Conclusions
In conclusion, we have demonstrated that an asymmetric electron emission from atomic targets can be generated and controlled by combining an attosecond pulse train composed of both odd and even harmonics and a weak IR field. The direction of the emission can be controlled by varying the time delay between the two pulses. In addition, we show that such asymmetric emission is also related to the properties of the APT, allowing the measurement of the phase difference between consecutive odd and even harmonics. We have found the previously unreported result that the relative phase shift between consecutive odd and even harmonics appears to be near $\pi/2$. It is interesting to notice that the resulting APT has a complex structure not resembling a single AP once per IR period.

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