Probing electron delays in above-threshold ionization: supplementary material

LUCAS J. ZIPP,^{1,2} ADI NATAN,¹ PHILIP H. BUCKSBAUM,^{1,2,3,*}

¹Stanford PULSE Institute, SLAC National Accelerator Laboratory, Menlo Park, CA 94025, USA

²Department of Physics, Stanford University, Stanford, CA 94305, USA

³Department of Applied Physics, Stanford University, Stanford, CA 94305, USA

*Corresponding author: phb@slac.stanford.edu

Published 21 November 2014

This document provides supplementary information to “Probing electron delays in above-threshold ionization,” <http://dx.doi.org/10.1364/optica.1.000361>. Full experimental methods are given as well as details on calculations and simulations performed to support conclusions presented in the primary manuscript. © 2014 Optical Society of America

<http://dx.doi.org/10.1364/optica.1.000361.s001>

A. Experimental Methods

An expanded description of our experimental methods is given here. We produce ~80 fs FWHM laser pulses centered at 800nm from a Ti:Sapphire laser system at a rate of 1 kHz. The pulses are sent through a BBO crystal oriented for Type I phase matching and 400nm light is generated with an orthogonal polarization. This is then sent through a calcite plate with axes oriented so that the group delay introduced between the two colors compensates for that introduced by the rest of the optics. It then passes through a dual order wave plate that rotates the polarization of the 800nm by 90 degrees, so that both colors are nominally linearly polarized in the same direction. They then pass through a wire grid polarizer to ensure a pure polarization, and through glass wedges, one of which was mounted on a motorized delay stage. As the wedge is translated into/out of the laser pulse, the amount of glass the laser travels through changes, leading to a delay in the relative phase of the two colors.

Fig. S1: Schematic of experimental setup.

The pulses pass through an iris that is ~7mm in diameter in order to ensure that the beam diameters of both colors are approximately the same. This ensures that the relative phase between the two colors remains constant as the fields undergo a Gouy phase shift through the focus. The beam then passes through a UV fused silica window into a vacuum chamber with a mu-metal shielded VMI. The beam is then focused by a spherical mirror ($f=15$ cm) into an argon jet. The ionized electrons are focused by electric fields onto an MCP and phosphor screen. The phosphor screen is then imaged by a CCD camera and the images are collected and analyzed on a computer. The images are inverted using an adapted onion peeling technique [1,2], and the signal is integrated over a half plane. The radial signal is then integrated around each ATI peak and sideband peak, excluding any Freeman resonances that appear outside the central part of each peak. At the higher intensity data ($\sim 8 \times 10^{13} \text{ W/cm}^2$), this is done by integrating over the full width at half maximum of each peak. At the lower intensity ($2 \times 10^{13} \text{ W/cm}^2$), this is only necessary for the lowest sideband peak. Freeman resonances are not prominent for the other peaks, and so the whole width of the peak is integrated over. The errors in the phase measurements were estimated as the sum of the fitting error, the standard error over four delay scans, and the uncertainty in the delay stage positioning.

B. Strong Field Approximation for Two Color Fields

The transition amplitude for an electron in the ground state of an atom to ionize with a final momentum $\mathbf{k}$ is given in the Strong Field Approximation (SFA) as [3]:

$$M_{ki} = -i \int_{-\infty}^{\infty} \langle \phi_k^V(t) | H_I(t) | \psi_o(t) \rangle dt, \quad (\text{S1})$$

where $|\phi_k^V(t)\rangle$ is a Volkov state with momentum $\mathbf{k}$, and $H_I(t)$ is the interaction of the atom with the laser field. In the length gauge this probability is given as:

$$M_{ki} = -\frac{i}{(2\pi)^{3/2}} \int_{-\infty}^{\infty} \langle k | e^{-i[k+A(t)] \cdot \mathbf{r}} e^{iS(k,t)} (\mathbf{r} \cdot \mathbf{F}) e^{iU_p t} | \psi_0 \rangle dt, \quad (\text{S2})$$

where $S(\mathbf{k}, t) = \frac{1}{2} \int_{-\infty}^t [\mathbf{k} + \mathbf{A}(t')]^2 dt'$ is the “modified” action of the electron, I_p is the ionization potential of the ground state, and $\mathbf{F}$ is the electric field. If we assume a hydrogenic wavefunction of the ground state, then the spatial integrals can be done analytically, otherwise it can be numerically integrated. The electric field is given by $E_o \sin(\omega t + \phi) + E_p \sin(\frac{\omega}{2} t) \hat{\mathbf{z}}$. Since the interference effect which we wish to observe stems from interferences within a single period of the $\omega/2$ field, we can restrict the integration range to $[0, \frac{4\pi}{\omega}]$ and look at transition rates to ATI peaks whose momentum we know *a priori* from the formula $E_n = n \frac{\omega}{2} - U_p - I_p$. This integral over time is then performed numerically for laser parameters similar to that used in the experiment and the relative phase is varied between the two fields. A hydrogen 1s ground state was used for ease of calculation, but the general oscillation phenomena that we observe should be independent of the exact initial state. The calculation reveals that the oscillation of the peaks in the $+\hat{\mathbf{z}}$ direction follows the pattern:

$$S_{contrast}^{(n)} = \begin{cases} a_n \cos(\phi), & \text{for } n \text{ odd} \\ a_n \cos(\phi - \pi), & \text{for } n \text{ even} \end{cases} \quad (\text{S3})$$

For certain peaks the phase flips by π , but these peaks corresponds to regions of “subcycle” destructive interference [4] from the strong ω field and the ionization rates are low. When SFA rates for a series of intensity values mimicking a Gaussian profile are averaged together, every peak is described by Eq. (S3).

C. Time Dependent Perturbation Theory for a Weak Field in the Presence of a Strong Laser Field

For a two color laser field in the dipole limit, we write our electric field as:

$$\mathbf{E} = -F_1 \sin(\omega t) + F_2 \cos(\frac{\omega}{2} t + \phi') \hat{\mathbf{z}}, \quad (\text{S4})$$

which is equivalent to Eq. 1 in the primary manuscript upon substitution of $\phi' = -2\phi$ and a shift in time $t \rightarrow t' - \frac{2}{\omega}(\phi' + \frac{\pi}{2})$. The Hamiltonian for our system is:

$$H = H_{atom} + H_{\omega}(t) + H_{\omega/2}(t), \quad (\text{S5})$$

where H_{atom} is the atomic Hamiltonian, $H_{\omega}(t) = \mathbf{A}_1(t) \cdot \mathbf{P} = -\frac{F_1}{\omega} \cos(\omega t) \cdot \mathbf{P}$, and $H_{\omega/2}(t) = \mathbf{F}_2(t) \cdot \mathbf{r} = \frac{F_2}{\omega/2} \cos(\frac{\omega}{2} t + \phi') \cdot \mathbf{r}$, where $\mathbf{F}_{1,2}$ are the respective electric field vectors, and $\mathbf{P}$ is the momentum operator. We use this mixed velocity and length gauge for ease of comparison using the Floquet formalism. Note that we are using the form of the velocity gauge (for the ω field) where the $\frac{A^2}{2}$ term has been transformed away. In the case where $H_{\omega/2} \ll (H_{atom} + H_{\omega})$, we can treat H_{ω} exactly and then add in $H_{\omega/2}$ as a perturbation. This perturbation scheme is different from conventional time-dependent perturbation theory in that the “unperturbed” Hamiltonian is now also time-dependent: $H_0 = H_{atom} + H_{\omega}(t)$.

Looking at momenta that can only be reached in the presence of the weak $\omega/2$ field, first order time dependent perturbation theory gives the transition amplitude as:

$$P_{i \rightarrow k_{2N+1}} = \lim_{t_f \rightarrow \infty} \int_{t_o \rightarrow -\infty}^{t_f} \langle \Phi_{k_{2N+1}}^- | U_o(t_f, t') H_{\omega/2}(t') U_o(t', t_o) | \Psi(t_o) \rangle^2 dt', \quad (\text{S6})$$

where $U_o(t, t_o)$ is the time propagator due to $H_{atom} + H_{\omega}(t)$, and $|\Phi_{k_{2N+1}}^- \rangle$ is the Coulomb scattering state corresponding asymptotically to a plane wave and an ingoing spherical wave. Here $k_{2N+1} = \sqrt{2E_{k_{2N+1}}}$ with $E_{k_{2N+1}} = (2N+1) \left(\frac{\omega}{2}\right) + E_i$, where E_i is the energy of the bound state in the strong field and includes the ponderomotive potential and AC stark shift. In what follows we make the assumption that the interacting fields are relatively long pulses with an adiabatic turn on and turn off, allowing us to represent the relevant propagators in the Floquet formalism. We will also represent the states and their propagators in the asymptotic ($r \rightarrow \infty$) limit, since this behavior will dominate the radial integrals between the continuum states.

$\langle \Phi_{k_{2N+1}}^- | U_o(t, t')$ consists of a momentum state scattering in the presence of the strong ω laser field, and may emit or absorb an integer number of photons in the field. $U_o(t', t_o) | \Psi(t_o) \rangle$ is the Floquet state corresponding in the zero field limit to the initial ground state of the atom. The propagators at asymptotically large radial coordinates are:

$$\begin{aligned} \langle \Phi_{k_{2N+1}}^- | U_o(t, t') \sim & e^{-i(k_{2N+1} \cdot \mathbf{r} + \frac{\ln(k_{2N+1} r (1 - \cos \theta_k))}{k_{2N+1}})} e^{i(E_{k_{2N+1}} t' + k_{2N+1} \cdot \frac{F_1}{\omega^2} \sin(\omega t'))} + \\ & \frac{1}{r} e^{i(k_{2N+1} r - \frac{\ln(2k_{2N+1} r)}{k_{2N+1}})} e^{i(E_{k_{2N+1}} t' - k_{2N+1} \cdot \frac{F_1}{\omega^2} \sin(\omega t'))} + \\ & \sum_m \langle \phi_{k_{2m+1}} | f_{2m+1}(\mathbf{k}_{2N+1}; \hat{\mathbf{r}}) e^{i(E_{k_{2m+1}} t' + k_{2m+1} \cdot \frac{F_1}{\omega^2} \sin(\omega t'))} \end{aligned} \quad (\text{S7})$$

and

$$U_o(t', t_o) | \Psi(t_o) \rangle \sim \sum_n e^{-i(E_{k_{2n}} t' + k_{2n} \cdot \frac{F_1}{\omega^2} \sin(\omega t'))} f_{2n}(\hat{\mathbf{r}}) | \phi_{k_{2n}} \rangle \quad (\text{S8})$$

where $f_{2n}(\hat{\mathbf{r}})$ is the angle-dependent transition amplitude to the continuum state $|\phi_{k_{2n}} \rangle$ which asymptotically takes the form of an outgoing spherical wave $|\phi_{k_{2n}} \rangle \sim r^{-1} e^{i(k_{2n} r - \frac{\ln(2k_{2n} r)}{k_{2n}})}$. $f_{2m+1}(\mathbf{k}; \hat{\mathbf{r}})$ is the transition amplitude from the scattering state $|\Phi_{k_{2N+1}}^- \rangle$ to that of $|\Phi_{k_{2m+1}} \rangle$, and θ_k is the angle measured from the $\mathbf{k}_{2N+1}$ axis. In the following, we will neglect the ingoing spherical wave component, which has a very small dipole matrix overlap with the outgoing waves of the ionizing Floquet state, and we will denote the plane wave component as $|\mathbf{k}_{2N+1} \rangle$.

We now have:

$$\begin{aligned} P_{i \rightarrow k_{2N+1}} = & \lim_{t_f \rightarrow \infty} \int_{t_o \rightarrow -\infty}^{t_f} \left| \langle \mathbf{k}_{2N+1} | e^{i(E_{k_{2N+1}} t' + k_{2N+1} \cdot \frac{F_1}{\omega^2} \sin(\omega t'))} + \right. \\ & \left. \sum_m \langle \phi_{k_{2m+1}} | f_{2m+1}(\mathbf{k}_{2N+1}; \hat{\mathbf{r}}) e^{i(E_{k_{2m+1}} t' + k_{2m+1} \cdot \frac{F_1}{\omega^2} \sin(\omega t'))} \right) \times \\ & \left. H_{\omega/2}(t') \sum_n e^{-i(E_{k_{2n}} t' + k_{2n} \cdot \frac{F_1}{\omega^2} \sin(\omega t'))} f_{2n}(\hat{\mathbf{r}}) | \phi_{k_{2n}} \rangle \right|^2 dt'. \end{aligned} \quad (\text{S9})$$

Using the identity $\sum_p J_p(z) e^{ip(\omega t')} = e^{iz \sin(\omega t')}$, where $J_p(z)$ is the Bessel function, we have:

$$P_{i \rightarrow k_{2N+1}} = \lim_{\substack{t_f \rightarrow \infty \\ t_o \rightarrow -\infty}} \int_{t_o}^{t_f} \left| \sum_q \langle \mathbf{k}_{2N+1} | e^{i(E_{k_{2N+1}} t')} J_q \left((\mathbf{k}_{2N+1} - k_{2n} \hat{\mathbf{r}}) \cdot \frac{\mathbf{F}_1}{\omega^2} \right) e^{iq(\omega t')} + \sum_{m,p} \langle \phi_{k_{2m+1}} | f_{2m+1}(\mathbf{k}_{2N+1}; \hat{\mathbf{r}}) e^{i(E_{k_{2m+1}} t')} J_p \left((k_{2m+1} - k_{2n}) \hat{\mathbf{r}} \cdot \frac{\mathbf{F}_1}{\omega^2} \right) e^{ip(\omega t')} \right] \times H_{\frac{\omega}{2}}(t') e^{-iE_{k_{2n}} t'} f_{2n}(\hat{\mathbf{r}}) | \phi_{k_{2n}} \rangle \right|^2 dt', \quad (\text{S10})$$

with $H_{\frac{\omega}{2}}(t') = H_{\frac{\omega}{2}}(e^{-\frac{i\omega}{2}t' - i\phi'} + e^{\frac{i\omega}{2}t' + i\phi'})$, where $H_{\frac{\omega}{2}} = \frac{1}{2} \frac{F_2}{\omega/2} \cdot \mathbf{P}$. Integrating over t' we have:

$$P_{i \rightarrow k_{2N+1}} = \left| \sum_{n,q} \langle \mathbf{k}_{2N+1} | H_{\frac{\omega}{2}} f_{2n}(\hat{\mathbf{r}}) | \phi_{k_{2n}} \rangle J_q \left((\mathbf{k}_{2N+1} - k_{2n} \hat{\mathbf{r}}) \cdot \frac{\mathbf{F}_1}{\omega^2} \right) \times \delta((m-n+p)\omega) e^{-i\phi'} + \delta((m-n+p+1)\omega) e^{i\phi'} + \sum_{m,n,p} \langle \phi_{k_{2m+1}} | f_{2m+1}(\mathbf{k}_{2N+1}; \hat{\mathbf{r}}) H_{\frac{\omega}{2}} f_{2n}(\hat{\mathbf{r}}) | \phi_{k_{2n}} \rangle \times \delta((m-n+p)\omega) e^{-i\phi'} + \delta((m-n+p+1)\omega) \right|^2, \quad (\text{S11})$$

where spatial integration over the Bessel functions is implied. Enforcing the delta function constraint, and transforming the field to replacing the phase of the $\omega/2$ field (ϕ') with that of the ω field ($\phi = 2\phi' - \pi$), we arrive at the result:

$$P_{i \rightarrow k_{2N+1}} = \left| \sum_n \langle \mathbf{k}_{2N+1} | H_{\frac{\omega}{2}} f_{2n}(\hat{\mathbf{r}}) | \phi_{k_{2n}} \rangle \times \{J_{n-N}(\alpha_n) e^{i\phi} + J_{n-N-1}(\alpha_n)\} + \sum_{m,n} \langle \phi_{k_{2m+1}} | f_{2m+1}(\mathbf{k}_{2N+1}; \hat{\mathbf{r}}) H_{\frac{\omega}{2}} f_{2n}(\hat{\mathbf{r}}) | \phi_{k_{2n}} \rangle \times \{J_{n-m}(\beta_{m,n}) e^{i\phi} + J_{n-m-1}(\beta_{m,n})\} \right|^2, \quad (\text{S12})$$

where $\alpha_n = (\mathbf{k}_{2N+1} - k_{2n} \hat{\mathbf{r}}) \cdot \frac{\mathbf{F}_1}{\omega^2}$ and $\beta_{m,n} = (k_{2m+1} - k_{2n}) \hat{\mathbf{r}} \cdot \frac{\mathbf{F}_1}{\omega^2}$. In the weak field limit only the zero order Bessel function contributes, and the result simplifies to

$$P_{i \rightarrow k_{2N+1}} = \left| \left(\langle \mathbf{k}_{2N+1} | H_{\frac{\omega}{2}} f_{2N}(\hat{\mathbf{r}}) | \phi_{k_{2N}} \rangle + \sum_m \langle \phi_{k_{2m+1}} | f_{2m+1}(\mathbf{k}_{2N+1}; \hat{\mathbf{r}}) H_{\frac{\omega}{2}} f_{2m}(\hat{\mathbf{r}}) | \phi_{k_{2m}} \rangle \right) e^{i\phi} + \left(\langle \mathbf{k}_{2N+1} | H_{\frac{\omega}{2}} f_{2N+1}(\hat{\mathbf{r}}) | \phi_{k_{2N+1}} \rangle + \sum_m \langle \phi_{k_{2m+1}} | f_{2m+1}(\mathbf{k}_{2N+1}; \hat{\mathbf{r}}) H_{\frac{\omega}{2}} f_{2m+1}(\hat{\mathbf{r}}) | \phi_{k_{2m+1}} \rangle \right) \right|^2. \quad (\text{S13})$$

The sum over m ensures that all multiphoton pathways are included, ie. those in which the IR probe photon is absorbed or emitted after first absorbing $N_o, N_o + 1, N_o + 2$, etc. ω photons.

Going back to the nonperturbative regime in Eq. (S12), we now ignore transition matrix elements between non-adjacent continuum states, because they are exponentially decreasing functions of the difference in momentum $(k_{2m+1} - k_{2n})$ [5]. We see that the transition amplitude becomes:

$$P_{i \rightarrow k_{2N+1}} \approx \left| \left[G_{2N+1,2N} J_0(\alpha_N) + G_{2N+1,2N+2} J_1(\alpha_{N+1}) + \sum_m T_{2m+1,2m} J_0(\beta_{m,m}) + T_{2m+1,2m+2} J_1(\beta_{m,m+1}) \right] e^{i\phi} + \left[G_{2N+1,2N+2} J_0(\alpha_{N+1}) + G_{2N+1,2N} J_{-1}(\alpha_N) + \sum_m T_{2m+1,2m+2} J_0(\beta_{m,m+1}) + T_{2m+1,2m} J_{-1}(\beta_{m,m}) \right] \right|^2, \quad (\text{S14})$$

where $G_{2N+1,2N}$ denotes the transition matrix term $\langle \mathbf{k}_{2N+1} | H_{\frac{\omega}{2}} f_{2N}(\hat{\mathbf{r}}) | \phi_{k_{2N}} \rangle$, while $T_{2m+1,2m}$ is equal to $\langle \phi_{k_{2m+1}} | f_{2m+1}(\mathbf{k}_{2N+1}; \hat{\mathbf{r}}) H_{\frac{\omega}{2}} f_{2m}(\hat{\mathbf{r}}) | \phi_{k_{2m}} \rangle$. We see that the term with $e^{i\phi}$ which we associate with an absorption of an IR photon includes a transition matrix element $T_{2m+1,2m+2}$ from an upper state while the emission term includes a transition from a lower state. This contribution is limited however if J_1 is sufficiently small. At our highest intensity and lowest order ATI peak, both $J_1(\beta_{m,m+1})$ and $J_1(\alpha_n) \sim 0.2$, while the J_0 terms are ~ 1 . To see the relative magnitude of this contribution in the resulting phase, we perform the sum over m and replace them with "effective" values to estimate the error. Upon expanding the square we see that the interference term is:

$$S_{\text{contrast}} \sim |T_{\uparrow}^{J_0} T_{\downarrow}^{J_0}| \cos(\phi - (\phi_{\downarrow} - \phi_{\uparrow})) + \left(|T_{\uparrow}^{J_0 J_1}|^2 + |T_{\downarrow}^{J_0 J_1}|^2 \right) \cos \phi, \quad (\text{S15})$$

where for example $T_{\uparrow}^{J_0}$ denotes an "effective" value for the J_0 weighted transitions from the lower state, and $\phi_{\uparrow}$ is the argument of $T_{\uparrow}^{J_0}$. We have neglected a contribution from a term that is of the order of J_1^2 . The $\cos \phi$ term causes a deviation of the experimentally measured delay δ_n in $S_{\text{contrast}}^{(n)} = a_n \cos(\phi - \delta_n)$ away from the desired value $\phi_{\downarrow} - \phi_{\uparrow}$. We can estimate the ratio between the second and first terms as being $J_1(\beta_{m,m+1})$ which gives a maximum deviation in phase of $\sim 25\%$ for the lowest order sideband in the experiment, and significantly lower deviation at higher momenta.

References

1. C. Bordas, F. Paulig, H. Helm, and D. L. Huestis, "Photoelectron imaging spectrometry: Principle and inversion method," *Review of Scientific Instruments* **67**, 2257–2268 (1996).
2. G. M. Roberts, J. L. Nixon, J. Lecointre, E. Wrede, and J. R. R. Verlet, "Toward real-time charged-particle image reconstruction using polar onion-peeling," *Review of Scientific Instruments* **80**, 053104 (2009).
3. Z. Chen, A.-T. Le, T. Morishita, and C. D. Lin, "Quantitative rescattering theory for laser-induced high-energy plateau photoelectron spectra," *Phys. Rev. A* **79**, 033409 (2009).
4. D. G. Arbó, K. L. Ishikawa, K. Schiessl, E. Persson, and J. Burgdörfer, "Intracycle and intercycle interferences in above-threshold ionization: The time grating," *Phys. Rev. A* **81**, 021403 (2010).
5. J. M. Dahlström, D. Guénot, K. Klünder, M. Gisselbrecht, J. Mauritsson, A. L'Huillier, A. Maquet, and R. Taïeb, "Theory of attosecond delays in laser-assisted photoionization," *Chemical Physics* **414**, 53–64 (2013).