

Supplementary Information: Effects of ultrashort laser pulses on angular distributions of photoionization spectra

C. H. Raymond Ooi¹, W. L. Ho¹ and A. D. Bandrauk²

¹*Department of Physics, University of Malaya, 50603 Kuala Lumpur, Malaysia*

²*Laboratoire de Chimie Théorique, Faculté des Sciences,
Université de Sherbrooke, Sherbrooke, Québec, Canada, J1K 2R1*

Appendix A: Parabolic pulse

For a parabolic envelope the useful functions take the form

$$g(s) = s(2s_0 - s)/s_0^2 \quad (1)$$

where $s = \omega t$, $s_0 = \omega t_0$ and t_0 can be regarded is the pulse width at the base.

We need to compute the vector potential $\mathbf{A}(t) = -\int_{-\infty}^t \mathbf{E}(t)dt$ and evaluate the expression

$$\hat{a}(s) = \int_0^s g(s') \begin{pmatrix} \hat{z} \cos\left(\frac{\omega}{\omega_0}s' + \varphi\right) \\ \hat{x}\alpha \cos\left(\frac{\omega}{\omega_0}s' + \varphi\right) + \hat{y}\beta \sin\left(\frac{\omega}{\omega_0}s' + \varphi\right) \end{pmatrix} ds' = \begin{pmatrix} \hat{z}\Sigma_c \\ \hat{x}\alpha\Sigma_c + \hat{y}\beta\Sigma_s \end{pmatrix} \quad (2)$$

where we have analytical expressions

$$\Sigma_c(s) = \int g(s') \cos(ws' + \varphi) ds' = \frac{2}{s_0^2 w^3} \left(\left(1 - \frac{1}{2}s^2 w^2 + s w^2 s_0\right) S - w(s - s_0)C \right) \quad (3)$$

$$\Sigma_s(s) = \int g(s') \sin(ws' + \varphi) ds' = -\frac{2}{s_0^2 w^3} \left(\left(1 - \frac{1}{2}s^2 w^2 + s w^2 s_0\right) C + w(s - s_0)S \right) \quad (4)$$

with definitions $w = \frac{\omega}{\omega_0}$, $S = \sin\left(\frac{\omega}{\omega_0}s + \varphi\right)$, $C = \cos\left(\frac{\omega}{\omega_0}s + \varphi\right)$.

Similarly we need

$$\Sigma_c^{(2)} = \int_0^s g^2(s) \cos^2(ws' + \varphi) ds' = \frac{1}{s_0^4} \int s^2(2s_0 - s)^2 \cos^2(ws + \varphi) ds \quad (5)$$

$$\begin{aligned} &= \frac{1}{s_0^4} \left[\frac{1}{30} s^3 (20s_0^2 - 15ss_0 + 3s^2) \right. \\ &\quad \left. + \frac{S_2}{8w^5} \left(3 + 2w^2 \left(s_0^2 - 3(s - s_0)^2 + s^2 w^2 (s - 2s_0)^2 \right) \right) \right. \\ &\quad \left. + \frac{1}{4} \frac{C_2}{w^4} (s - s_0) (2w^2 s(s - 2s_0) - 3) \right] \end{aligned} \quad (6)$$

$$\Sigma_s^{(2)} = \int_0^s g^2(s) \sin^2(ws' + \varphi) ds' = \frac{1}{s_0^4} \int s^2(2s_0 - s)^2 \sin^2(ws + \varphi) ds \quad (7)$$

$$\begin{aligned} &= \frac{1}{s_0^4} \left[\frac{1}{30} s^3 (20s_0^2 - 15ss_0 + 3s^2) \right. \\ &\quad \left. - \frac{S_2}{8w^5} \left(3 + 2w^2 \left(s_0^2 - 3(s - s_0)^2 + s^2 w^2 (s - 2s_0)^2 \right) \right) \right. \\ &\quad \left. - \frac{1}{4} \frac{C_2}{w^4} (s - s_0) (2w^2 s(s - 2s_0) - 3) \right] \end{aligned} \quad (8)$$

where $S_2 = \sin 2\left(\frac{\omega}{\omega_0}s + \varphi\right)$, $C_2 = \cos 2\left(\frac{\omega}{\omega_0}s + \varphi\right)$.

The dot products which we define in the text may be expressed as

$$\hat{n} \cdot \hat{a} = \begin{pmatrix} \Sigma_c \cos\left(\frac{\omega}{\omega_0}s + \varphi\right) \\ \alpha^2 \Sigma_c \cos\left(\frac{\omega}{\omega_0}s + \varphi\right) + \beta^2 \Sigma_s \sin\left(\frac{\omega}{\omega_0}s + \varphi\right) \end{pmatrix} \quad (9)$$

$$\hat{a} \cdot \hat{a} = \begin{pmatrix} \Sigma_c^2 \\ \alpha^2 \Sigma_c^2 + \beta^2 \Sigma_s^2 \end{pmatrix} \quad (10)$$

$$\vec{\rho} \cdot \hat{a} = \begin{pmatrix} \rho_z \Sigma_c \\ \rho_x \alpha \Sigma_c + \rho_y \beta \Sigma_s \end{pmatrix} \quad (11)$$

The next level of integrations are

$$\int_0^s \hat{a}(s) ds = \begin{pmatrix} \hat{z} \Xi_c \\ \hat{x} \alpha \Xi_c + \hat{y} \beta \Xi_s \end{pmatrix}, \quad (12)$$

$$\int_0^s \hat{a} \cdot \hat{a} ds = \int_0^s \begin{pmatrix} \Sigma_c^2 \\ \alpha^2 \Sigma_c^2 + \beta^2 \Sigma_s^2 \end{pmatrix} ds = \begin{pmatrix} \Xi_c^{(2)} \\ \alpha^2 \Xi_c^{(2)} + \beta^2 \Xi_s^{(2)} \end{pmatrix}, \quad (13)$$

where

$$\Xi_{c(s)}(s) = \int_0^s \Sigma_{c(s)}(s') ds'.$$

Appendix B: Spherical Coordinates in Generalized Momentum Space

The full hydrogenic wavefunction (dimension $\text{m}^{-3/2}$) is

$$\begin{aligned} \psi_{nlm}(\mathbf{r}) &= \sigma \frac{1}{(2\pi)^{1/2}} e^{im\phi} \sqrt{\frac{(2l+1)(l-|m|)!}{2(l+|m|)!}} P_l^m(y) \left[(2Y)^{l+1} \sqrt{\frac{Y(n-l-1)!}{n[(n+1)!]^3}} e^{-Yr} r^l L_{n-l-1}^{2l+1}(2Yr) \right] \\ &= \sigma \sqrt{\frac{(2l+1)(l-|m|)!}{4\pi(l+|m|)!}} \sqrt{\left(\frac{2}{na_0}\right)^3 \frac{(n-l-1)!}{2n[(n+1)!]^3}} \left[e^{-x/2} x^l L_{n-l-1}^{2l+1}(x) \right] P_l^m(y) e^{im\phi} \end{aligned} \quad (15)$$

and its Fourier transform (dimension $(\text{kgms}^{-1})^{-3/2}$) that was obtained analytically by Pauling and Podolsky [?]]

$$\begin{aligned} \bar{\psi}_s(\Pi) &= \frac{1}{\sqrt{(2\pi\hbar)^3}} \int \psi_s(\mathbf{r}) \exp(-\frac{i}{\hbar} \Pi(s) \cdot \mathbf{r}) d^3r = \frac{1}{\sqrt{(2\pi\hbar Y)^3}} FGH \\ &= \frac{1}{\sqrt{(2\pi\hbar Y)^3}} \left\{ \frac{1}{(2\pi)^{1/2}} e^{\pm im\Phi'} \right\} \left\{ \left(\frac{(2l+1)(l-m)!}{2(l+m)!} \right)^{\frac{1}{2}} P_l^m(\cos \Theta') \right\} \times \\ &\quad \left\{ -(-1)^{2l+1} (-i)^l \pi 2^{2l+4} l! \left(\frac{n(n-l-1)!}{(n+l)!} \right)^{\frac{1}{2}} \frac{\zeta^l}{(1+\zeta^2)^{l+2}} C_{n-l-1}^{l+1} \left(\frac{1-\zeta^2}{1+\zeta^2} \right) \right\} \end{aligned} \quad (16)$$

The generalized momentum vector and the corresponding variables in spherical coordinates are

$$= \mathbf{P} + q \frac{\mathcal{E}_0}{\omega_0} \hat{a}(s) = \begin{pmatrix} P \sin \Theta \cos \Phi + q \frac{\mathcal{E}_0}{\omega_0} \begin{pmatrix} 0 \\ \alpha \Sigma_c \end{pmatrix} \\ P \sin \Theta \sin \Phi + q \frac{\mathcal{E}_0}{\omega_0} \begin{pmatrix} 0 \\ \beta \Sigma_s \end{pmatrix} \\ P \cos \Theta + q \frac{\mathcal{E}_0}{\omega_0} \begin{pmatrix} \Sigma_c \\ 0 \end{pmatrix} \end{pmatrix} = \begin{pmatrix} \Pi \sin \Theta' \cos \Phi' \\ \Pi \sin \Theta' \sin \Phi' \\ \Pi \cos \Theta' \end{pmatrix} \quad (17)$$

$$\Pi = \left(\frac{\sqrt{P^2 \sin^2 \Theta + (P \cos \Theta + q \Sigma_c \frac{\mathcal{E}_0}{\omega_0})^2}}{\sqrt{(P \sin \Theta \cos \Phi + q \frac{\mathcal{E}_0}{\omega_0} \alpha \Sigma_c)^2 + (P \sin \Theta \sin \Phi + q \frac{\mathcal{E}_0}{\omega_0} \beta \Sigma_s)^2 + (P \cos \Theta)^2}} \right) \quad (18)$$

$$\tan \Phi' = \frac{P \sin \Theta \sin \Phi + q \frac{\mathcal{E}_0}{\omega_0} \begin{pmatrix} 0 \\ \beta \Sigma_s \end{pmatrix}}{P \sin \Theta \cos \Phi + q \frac{\mathcal{E}_0}{\omega_0} \begin{pmatrix} 0 \\ \alpha \Sigma_c \end{pmatrix}} = \begin{pmatrix} \tan \Phi \\ \frac{P \sin \Theta \sin \Phi + q \frac{\mathcal{E}_0}{\omega_0} \beta \Sigma_s}{P \sin \Theta \cos \Phi + q \frac{\mathcal{E}_0}{\omega_0} \alpha \Sigma_c} \end{pmatrix} \quad (19)$$

$$\tan \Theta' = \left(\frac{\frac{P \sin \Theta}{\sqrt{(P \cos \Theta + q \Sigma_c \frac{\mathcal{E}_0}{\omega_0})^2}}}{\frac{\sqrt{(P \sin \Theta \cos \Phi + q \frac{\mathcal{E}_0}{\omega_0} \alpha \Sigma_c)^2 + (P \sin \Theta \sin \Phi + q \frac{\mathcal{E}_0}{\omega_0} \beta \Sigma_s)^2}}{P \cos \Theta}} \right) \quad (20)$$

Appendix C: Derivatives of the Wavefunctions in Generalized Momentum Space

The derivatives of the separated functions in $\bar{\psi}_s(\Pi)$ are

$$\frac{\partial \bar{\psi}_s}{\partial \Phi'} = im \bar{\psi}_s, \quad \frac{\partial F}{\partial \Phi'} = im F, \quad (21)$$

$$\begin{aligned} \frac{\partial G(\Theta')}{\partial \Theta'} &= \frac{\partial}{\partial \Theta'} \left\{ \left(\frac{(2l+1)(l-m)!}{2(l+m)!} \right)^{\frac{1}{2}} P_l^m(\cos \Theta') \right\} \\ &= \left(\frac{(2l+1)(l-m)!}{2(l+m)!} \right)^{\frac{1}{2}} \frac{l(\cos \theta) P_l^m(\cos \Theta') - (l+m) P_{l-1}^m(\cos \Theta')}{\sin^2 \Theta'}. \end{aligned} \quad (22)$$

$$\begin{aligned} \frac{\partial H}{\partial \zeta} &= -(-i)^l \pi 2^{2l+4} l! \left(\frac{n(n-l-1)!}{(n+l)!} \right)^{\frac{1}{2}} \times \\ &\quad \left\{ \frac{8(l+1)\zeta^{l+1}}{(\zeta^2+1)^{l+4}} C_{n-l-2}^{l+2} \left(\frac{\zeta^2-1}{\zeta^2+1} \right) + C_{n-l-1}^{l+1} \left(\frac{\zeta^2-1}{\zeta^2+1} \right) \left[\frac{l\zeta^{l-1}}{(\zeta^2+1)^{(l+2)}} - 2(l+2) \frac{\zeta^{l+1}}{(\zeta^2+1)^{(l+3)}} \right] \right\}. \end{aligned} \quad (23)$$

where we have used the identity $\frac{\partial}{\partial \Theta'} P_l^m(\cos \Theta') = \frac{l(\cos \Theta') P_l^m(\cos \Theta') - (l+m) P_{l-1}^m(\cos \Theta')}{\sin^2 \Theta'}$ for the associated Legendre polynomials and evaluated the derivative

$$\frac{\partial H(\Pi)}{\partial \Pi} = -(-i)^l \pi 2^{2l+4} l! \left(\frac{n(n-l-1)!}{(n+l)!} \right)^{\frac{1}{2}} \frac{\partial}{\partial \Pi} \left[\frac{\zeta^l}{(\zeta^2 + 1)^{l+2}} C_{n-l-1}^{l+1} \left(\frac{\zeta^2 - 1}{\zeta^2 + 1} \right) \right]. \quad (24)$$