Attosecond Pulse Carrier-Envelope-Phase Effects on Ionized Electron Momentum and Energy Distributions

Anthony F. Starace

Department of Physics & Astronomy, University of Nebraska

Collaborators

Liang-You Peng and Evgeny A. Pronin: Dept. of Physics & Astronomy, University of Nebraska

References: *Phys. Rev. A* **76**, 043401 (2007); *New J. Phys.* **10**, 025030 (2008)

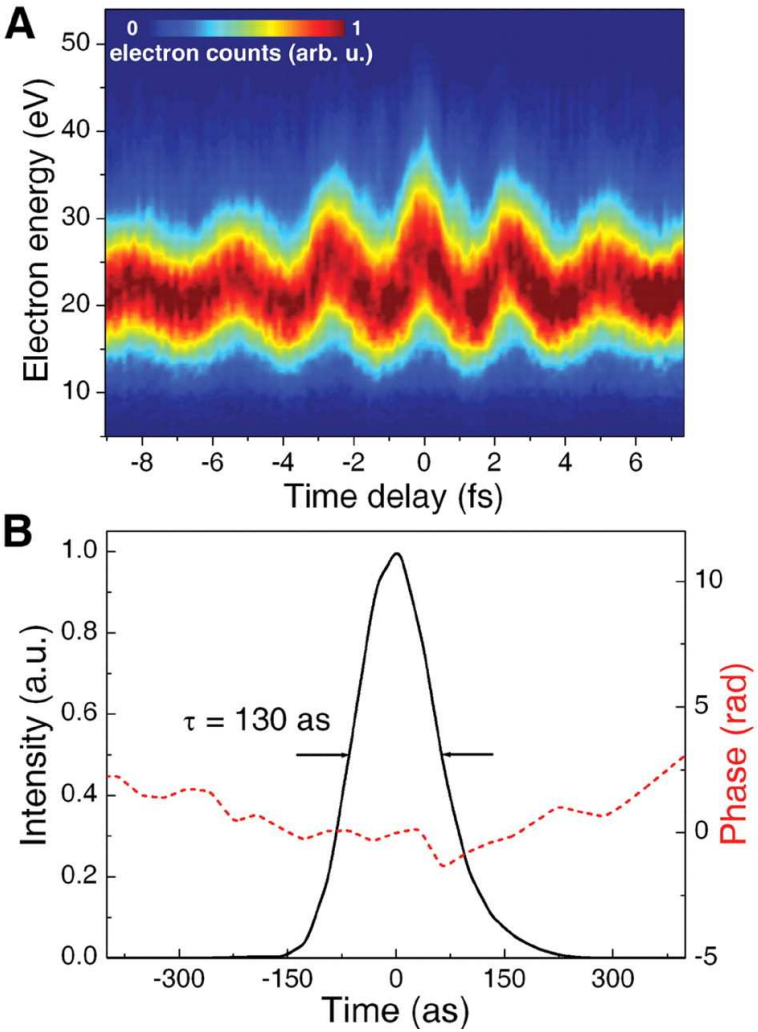
Outline

- *Motivation*
- *Theoretical Method*
- *Results for a Single Few-Cycle Attosecond Pulse*
- *Results for Two Few-Cycle Attosecond Pulses*
- *Effects of an Additional IR Pulse*
- *Effect of Pulse Chirp*
- *Analytic Description of CEP Effects*
- *Concluding Remarks*

Motivation

Motivation

G. Sansone et al., *Science* **314**, 443 (2006).

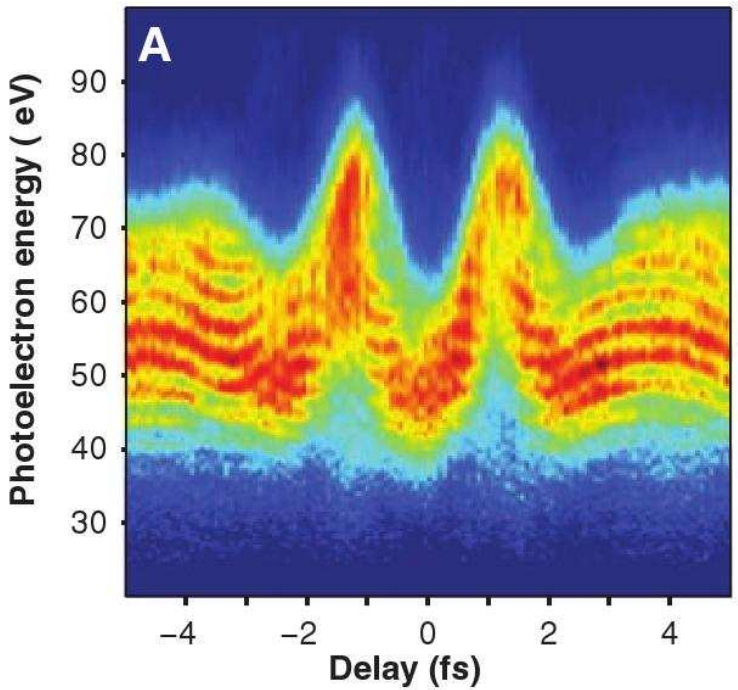


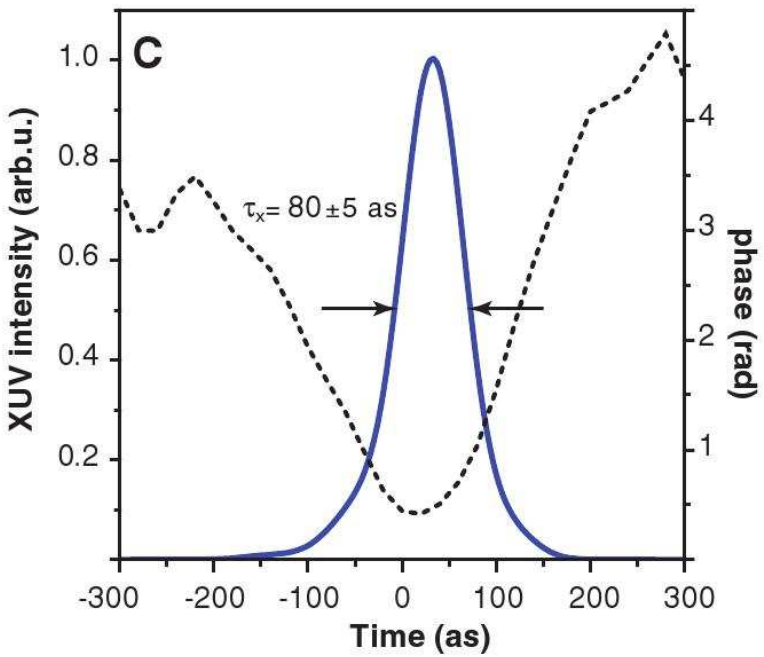
"The availability of **single-cycle isolated attosecond pulses** opens the way to **a new regime in ultrafast physics**, in which the strong-field **electron dynamics** in atoms and molecules **is driven by the electric field** of the attosecond pulses rather than by their intensity profile."

The CEP of the Attosecond Pulse Matters!

Motivation (cont'd)

E. Goulielmakis et al., *Science* **320**, 1614 (2008).





Related Works

- **CEP Effects for a Few-Cycle IR Pulse**
  G.G. Paulus et al., *Nature* **414**, 182 (2001).
- **CEP Effects for a IR Pulse + Attosecond Pulse**
  A.D. Bandrauk et al., *Phys. Rev. Lett.* **89**, 283903 (2002).
- **Low-Energy Electron Wave Packet Produced by Attosecond Pulse Train and Driven by IR Field**
  J. Mauritsson et al., *Phys. Rev. Lett.* **100**, 073003 (2008).

Our work: **CEP Effects for One or Two Few-Cycle Attosecond Pulses with or without an Additional IR Pulse**

Theoretical Method

Time-Dependent Schrödinger Equation

$$i\frac{\partial}{\partial t}\Psi\left(\mathbf{r},t\right) = \left[H_0(\mathbf{r}) + H_I(\mathbf{r},t)\right]\Psi\left(\mathbf{r},t\right)$$

with the atomic and interaction Hamiltonians given by

$$H_0 = -\frac{1}{2}\nabla^2 + V_C(r), \quad H_I(\mathbf{r},t) = -i\mathbf{A}(t)\cdot\nabla$$

respectively, where

$$V_C(r) = \begin{cases} -\frac{1}{r}, & \text{for H,} \\ -\frac{1}{r}\left[1 + (1+\beta r/2)\,e^{-\beta r}\right], & \text{for He,} \end{cases}$$

with $\beta = 27/8$[1]. [1] D.R. Hartree, *The Calculation of Atomic Structures (1957).*

Vector Potential of Two Attosecond Pulses

$$\mathbf{A}(t) \equiv A(t)\hat{\mathbf{z}} = A_1 F_1(t) \sin\left[w_1\left(t + \frac{\tau_1}{2}\right) + \phi_1\right]\hat{\mathbf{z}}$$
$$+A_2 F_2(t) \sin\left[w_2\left(t - T_\mathrm{d} + \frac{\tau_2}{2}\right) + \phi_2\right]\hat{\mathbf{z}},$$

where the envelopes are given by

$$F_1(t) = \begin{cases} \sin^2\left[\pi\left(t + \tau_1/2\right)/\tau_1\right], & |t| \le \tau_1/2; \\ 0, & |t| > \tau_1/2, \end{cases}$$

$$F_2(t) = \begin{cases} \sin^2\left[\pi\left(t - T_\mathrm{d} + \tau_2/2\right)/\tau_2\right], & |t - T_\mathrm{d}| \le \tau_2/2; \\ 0, & |t - T_\mathrm{d}| > \tau_2/2. \end{cases}$$

Electric field strength: $\mathbf{E}(t) = -\frac{1}{c}\frac{\partial}{\partial t}\mathbf{A}(t)$.

Solution of the TDSE

$$\Psi(\mathbf{r},t) \equiv \Psi(r,\theta,\phi,t) = \sum_{l=0}^{L}\sum_{m=-l}^{l} \frac{\varphi_{lm}(r,t)}{r} Y_{lm}(\theta,\phi) \Longrightarrow$$

$$i\frac{\partial}{\partial t}\varphi_{lm}(r,t) = -\frac{1}{2}\frac{d^2}{dr^2}\varphi_{lm}(r,t) + V^l_{\text{eff}}(r)\,\varphi_{lm}(r,t) + [H_I(r,t)]_{lm}$$

where

$$V^l_{\text{eff}}(r) \equiv V_C(r) + \frac{l(l+1)}{2r^2}, \qquad a_{lm} = \sqrt{\frac{(l-m)(l+m)}{(2l-1)(2l+1)}},$$

$$[H_I(r,t)]_{lm} = iA(t)\frac{1}{r}\left[l a_{lm}\varphi_{l-1m}(r,t) - (l+1)\,a_{l+1m}\varphi_{l+1m}(r,t)\right]$$
$$-iA(t)\frac{d}{dr}\left[a_{lm}\varphi_{l-1m}(r,t) + a_{l+1m}\varphi_{l+1m}(r,t)\right].$$

Numerical Methods

- **Radial Coordinate Discretization:**

  Central Finite Difference Method.

- **Time Propagation:**

  Arnoldi Method.

- **Reference for Details:**

  L.-Y. Peng and A.F. Starace, *J. Phys. Chem.* **125**, 154311 (2006).

Ionized Electron Wave Function in Momentum Space

- *Projection onto Field-Free Hamiltonian Eigenstates*:

Project $\Psi\left(\mathbf{r}, t_f\right)$ onto incoming Coulomb waves,

$$\Upsilon(k, \theta', \phi') = \langle \Psi_{\mathbf{k}}^{(-)}\left(\mathbf{r}, t_f\right) | \Psi\left(\mathbf{r}, t_f\right) \rangle,$$

$$\langle \Psi_{\mathbf{k}}^{(-)} | \Psi_{\mathbf{k}'}^{(-)} \rangle = \delta(\mathbf{k} - \mathbf{k}').$$

Calculations of Observables

Without loss of generality, one may set $k_y = 0$. The momentum and energy probability distributions are then calculated according to,

$$P(k_x, k_z) = |\Upsilon(k_x, k_y = 0, k_z)|^2 = P(E, \theta_k),$$

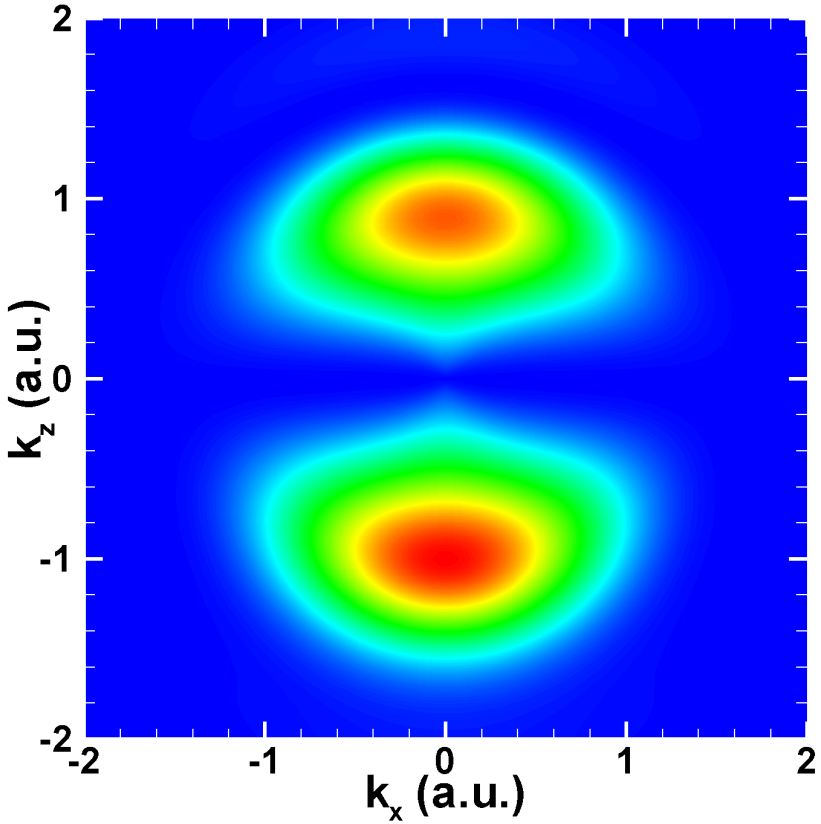
where θ_k is the angle between the laser polarization direction, $\hat{z}$, and the electron momentum vector $\mathbf{k} = (k_x, 0, k_z)$.
The two probabilities are normalized so that

$$\int_{-\infty}^{\infty}\int_{-\infty}^{\infty} P(k_x, k_z) dk_x dk_z \equiv \int_0^{\infty}\int_0^{2\pi} P(E, \theta_k) dE d\theta_k.$$

Results for a Single Few-Cycle Attosecond Pulse

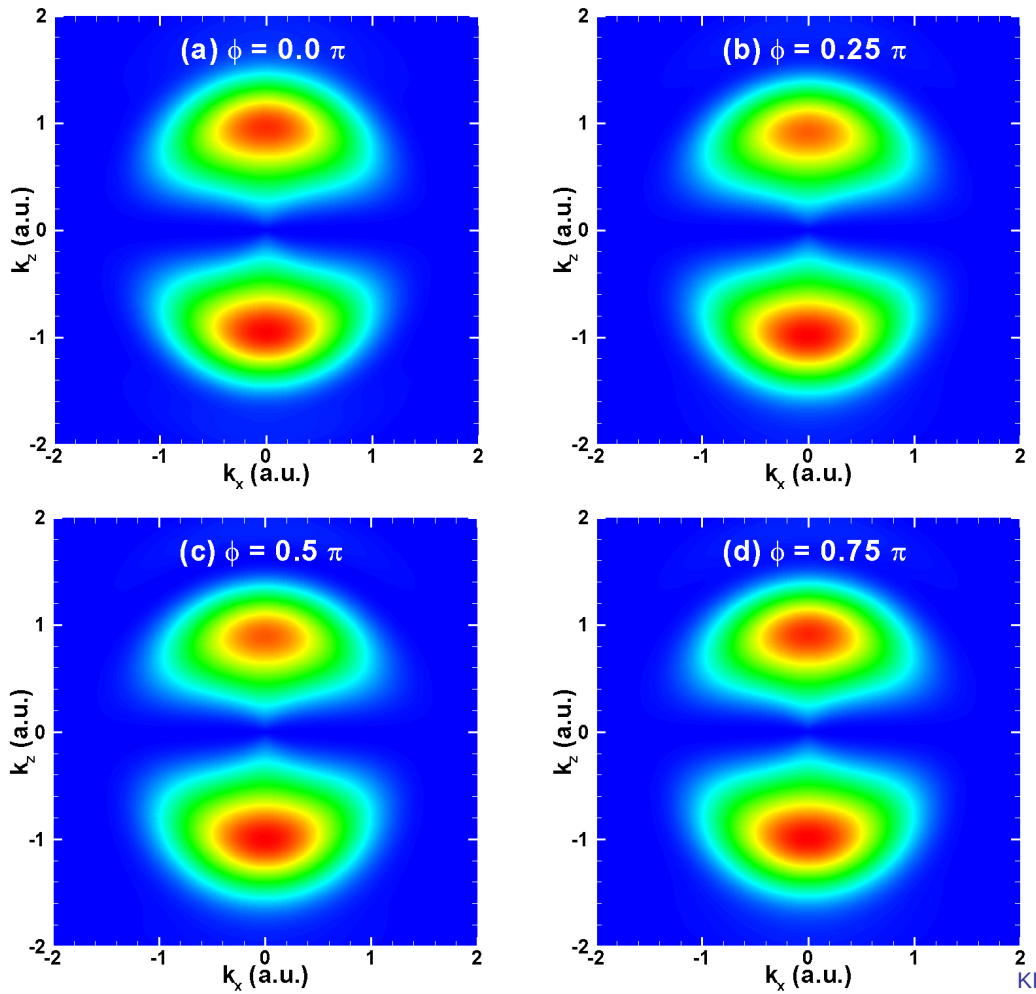
Momentum Distributions of Electrons Ionized from He

$\omega_1 = 36$ eV, $I_1 = 5 \times 10^{15}$ W cm^{-2}, $\tau_1 = 2T_\omega$, $\phi_1 = 0.5\pi$



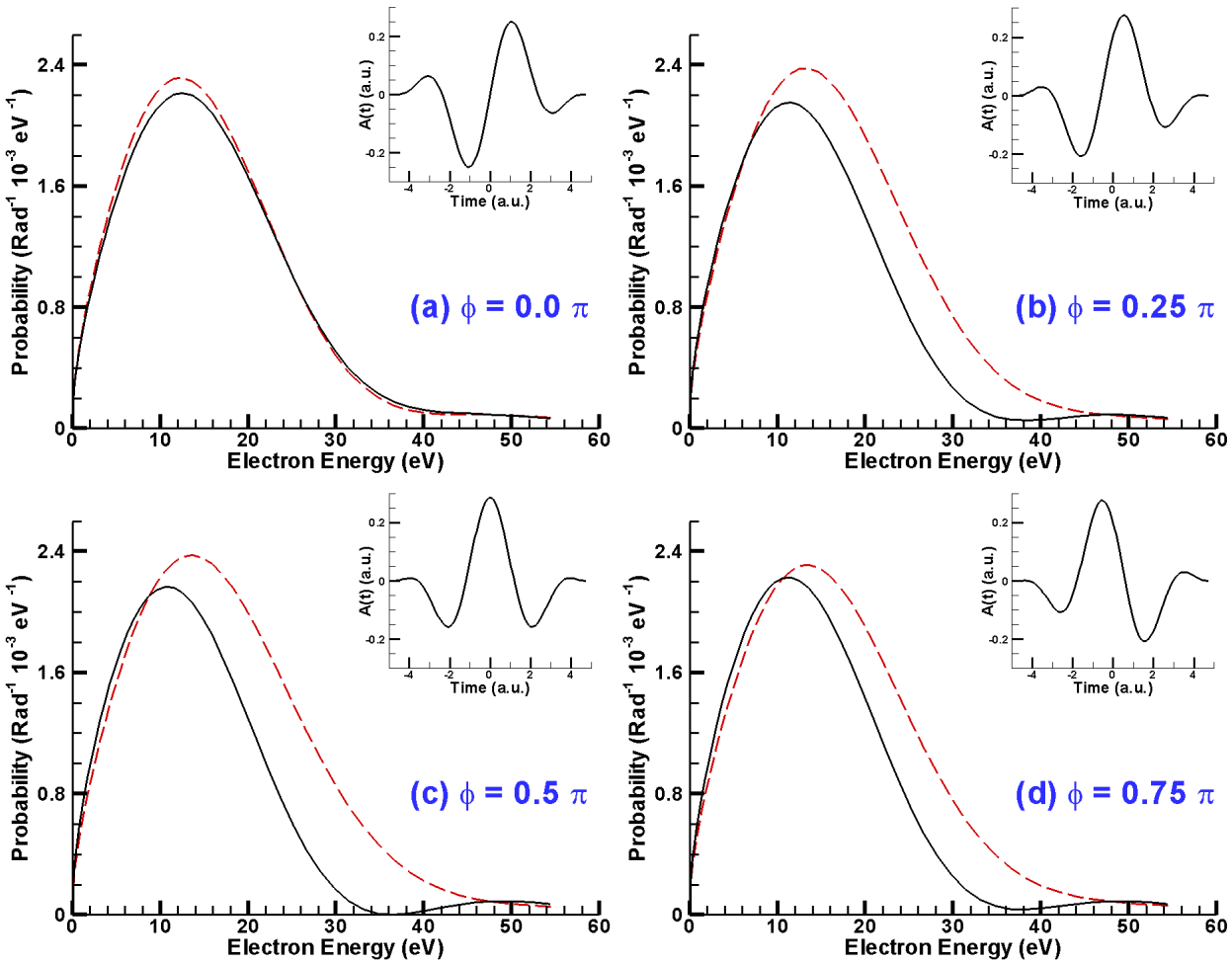
Electron Momentum Distributions vs. CEP for He

$\omega_1 = 36$ eV, $I_1 = 5 \times 10^{15}$ W cm^{-2}, $\tau_1 = 2T_\omega$

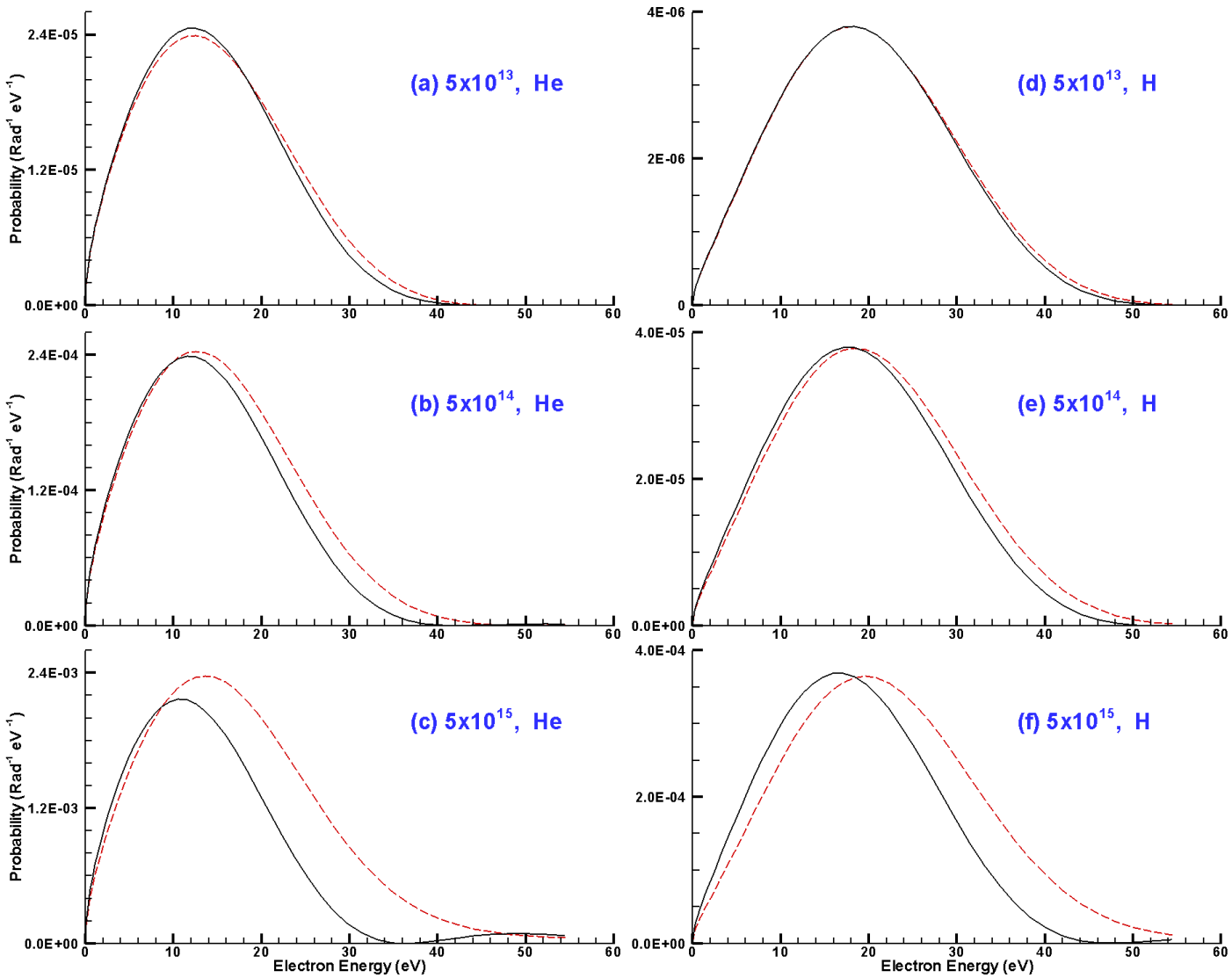


Electron Energy Distributions vs. CEP for He

$\theta_k = 0$ (full lines) or π (dashed lines)

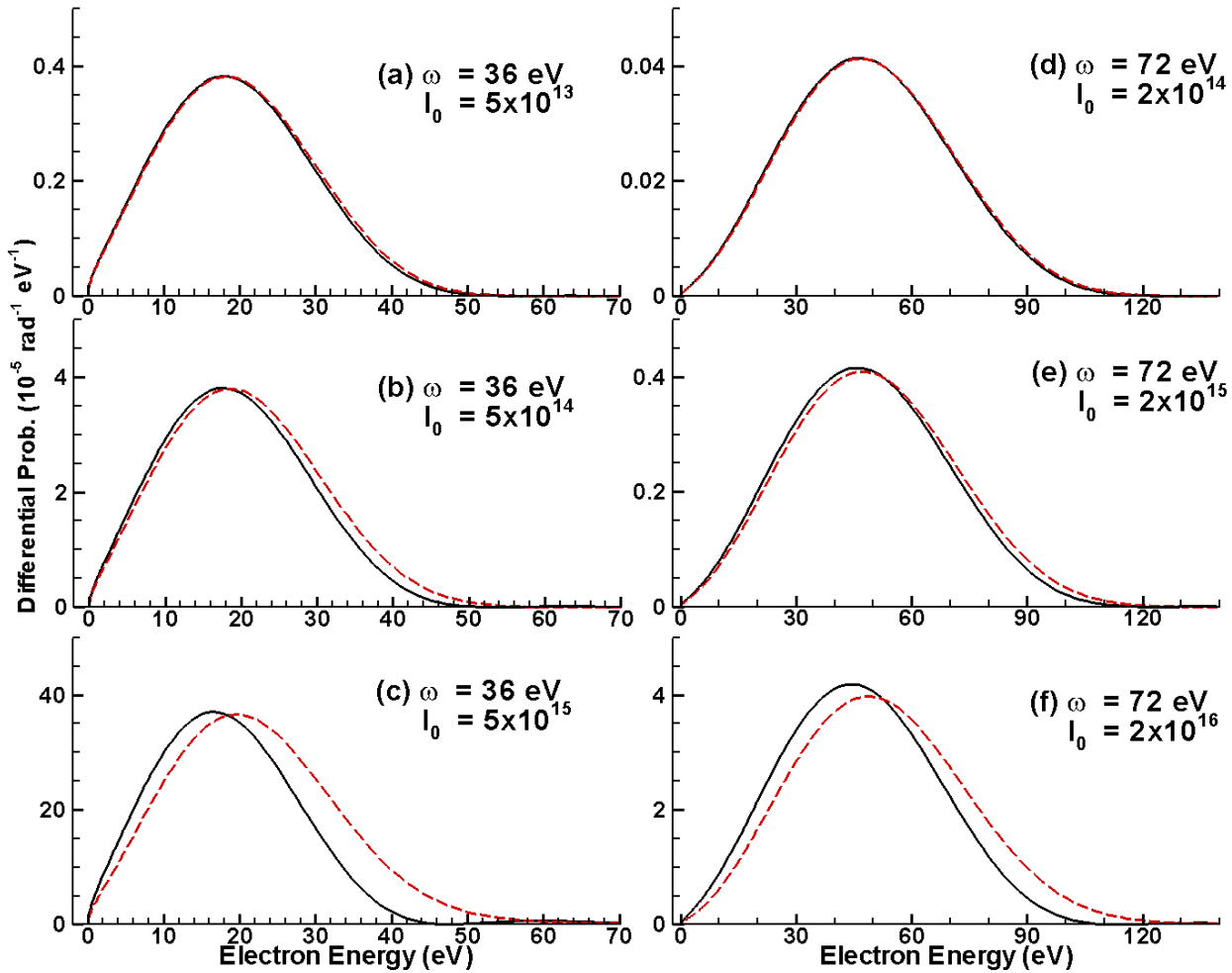


Comparison of He and H at different I_1 for $\phi_1 = 0.5\pi$



Scaling Law for CEP Effects for H

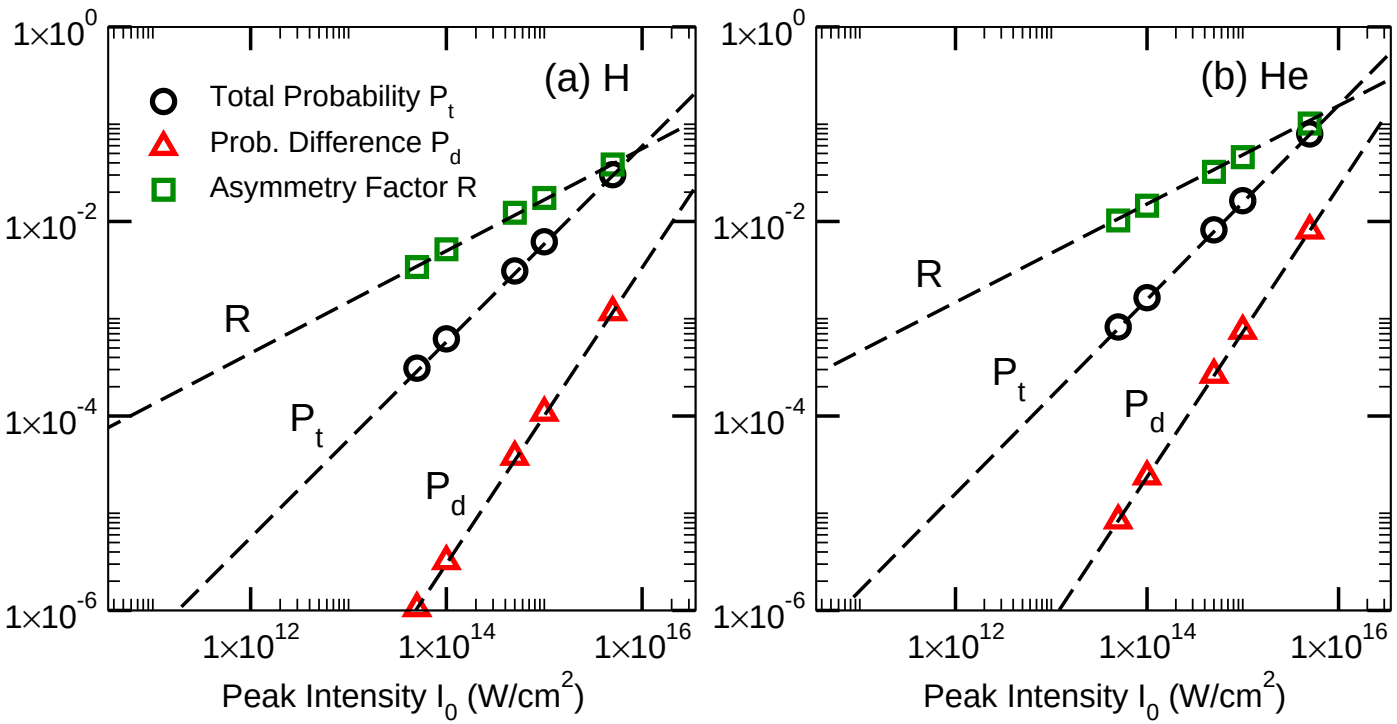
$$\sqrt{I_0(1)}/\omega_1 = \sqrt{I_0(2)}/\omega_2$$



Intensity Dependence of the CEP-Induced Asymmetries

$$P_t \equiv P_- + P_+ \propto I^{1.0}; \quad P_d \equiv P_- - P_+ \propto I^{1.5};$$

$$R \equiv P_d/P_t \propto I^{0.5}$$

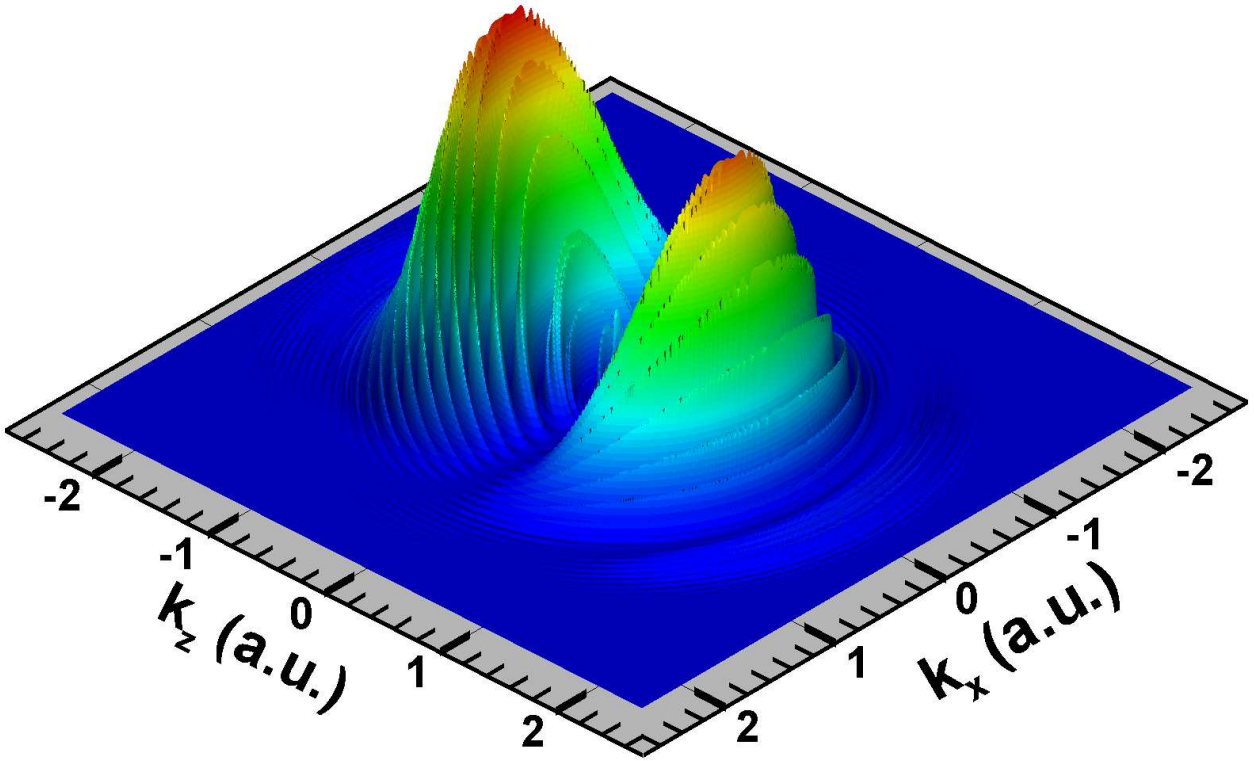


Results for Two Few-Cycle Attosecond Pulses

3D Momentum Distribution of Electrons for He

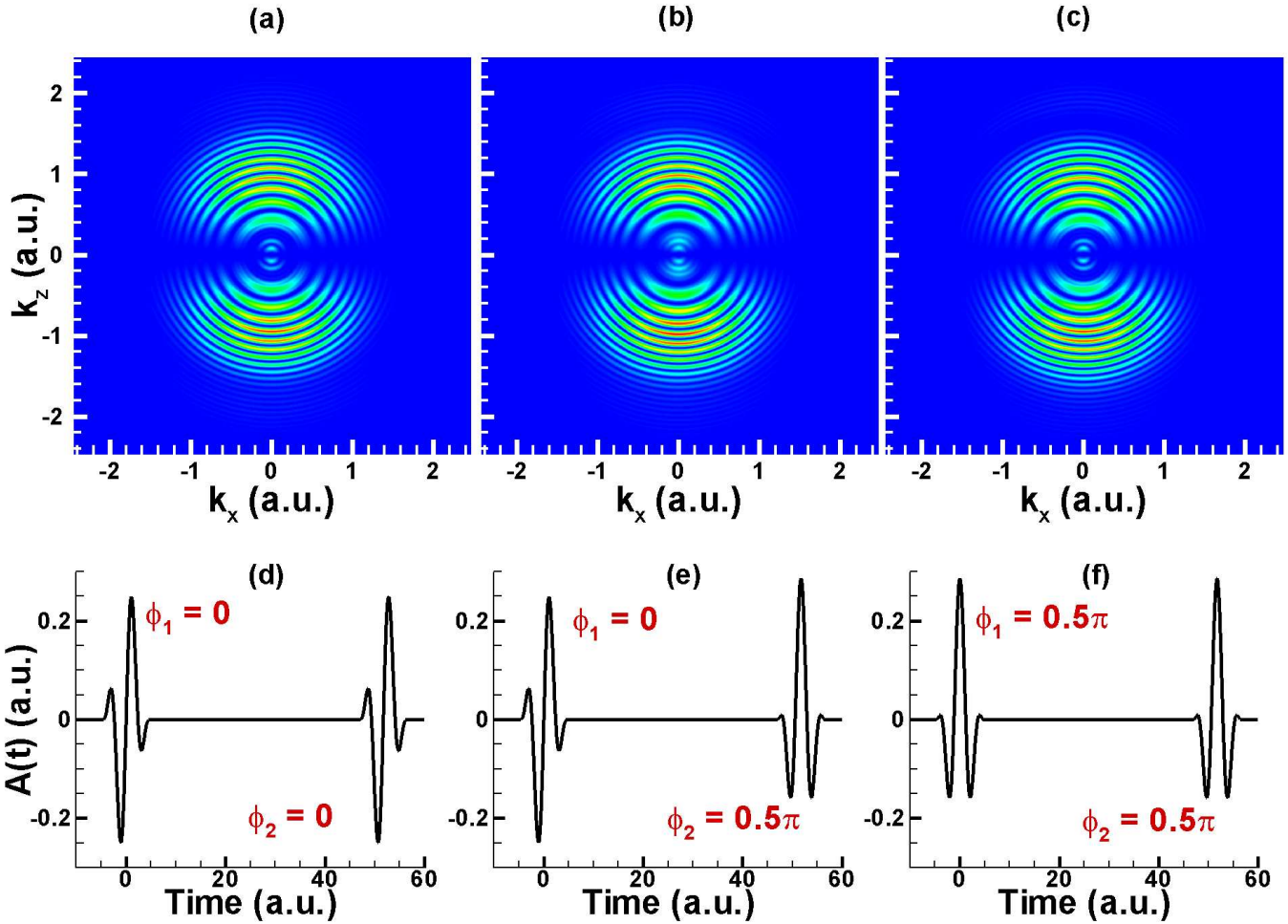
$$\omega_1 = \omega_2 = 36 \text{ eV}, I_1 = I_2 = 5 \times 10^{15} \text{ W cm}^{-2}, \tau_1 = \tau_2 = 2T_\omega$$

$$\phi_1 = \phi_2 = 0.5\pi$$



Momentum Distribution of Electrons Ionized from He

$\omega_1 = \omega_2 = 36$ eV, $I_1 = I_2 = 5 \times 10^{15}$ W cm^{-2}, $\tau_1 = \tau_2 = 2T_\omega$



Approximate Formula for the Interference Minima

The total energy distribution of electrons ionized by two attosecond pulses is given by

$$P(E,\theta) = \left[f_1(E)e^{-i\Delta\Phi} + f_2(E)\right]\left[f_1(E)e^{i\Delta\Phi} + f_2(E)\right]$$
$$= |f_1(E)|^2 + |f_2(E)|^2 + 2\,|f_1(E)f_2(E)|\cos\Delta\Phi,$$

where the relative phase is

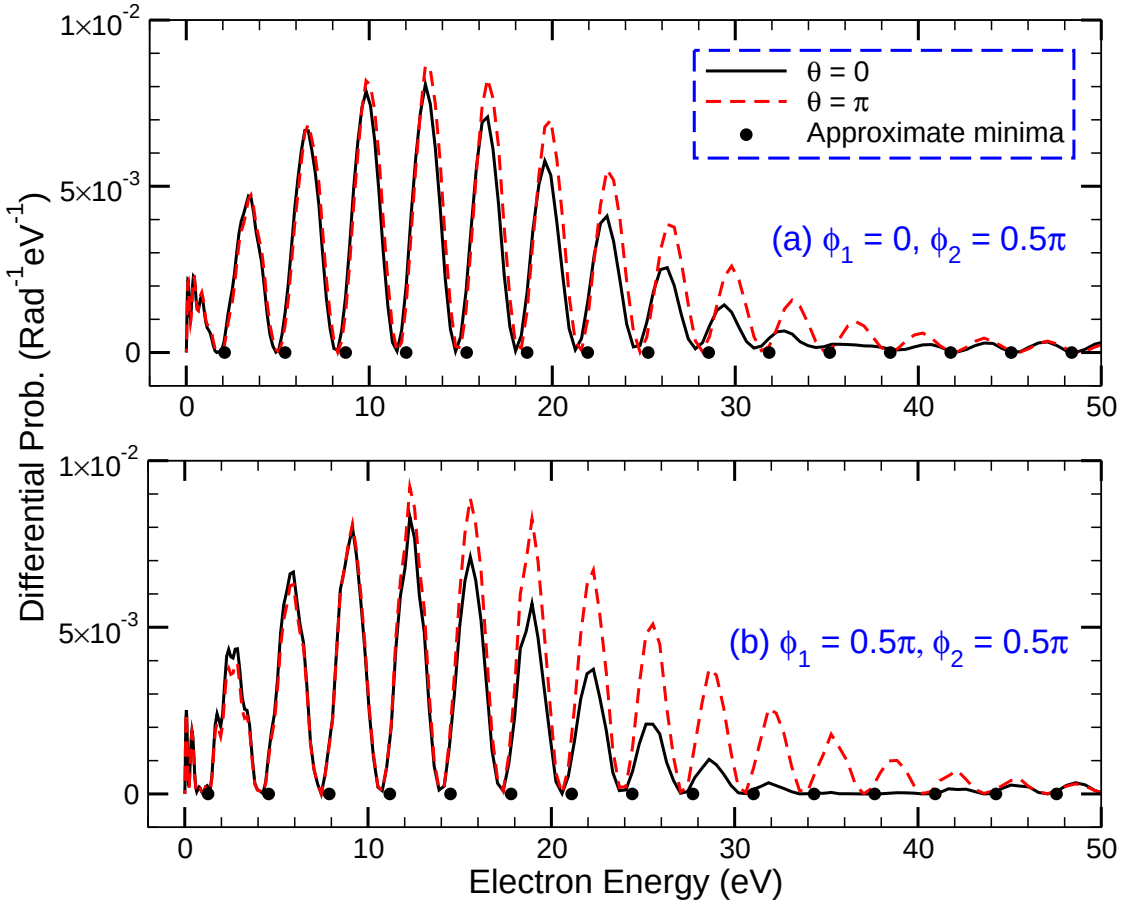
$$\Delta\Phi = (E + E_\mathrm{b})T_\mathrm{d} + (\phi_1 - \phi_2).$$

The interference minima in the energy spectra occur when $\Delta\Phi = (2n+1)\pi$, which gives

$$E_n^{\mathrm{min}} = -E_\mathrm{b} + \frac{\pi}{T_\mathrm{d}}\left(2n + 1 - \frac{\phi_1 - \phi_2}{\pi}\right).$$

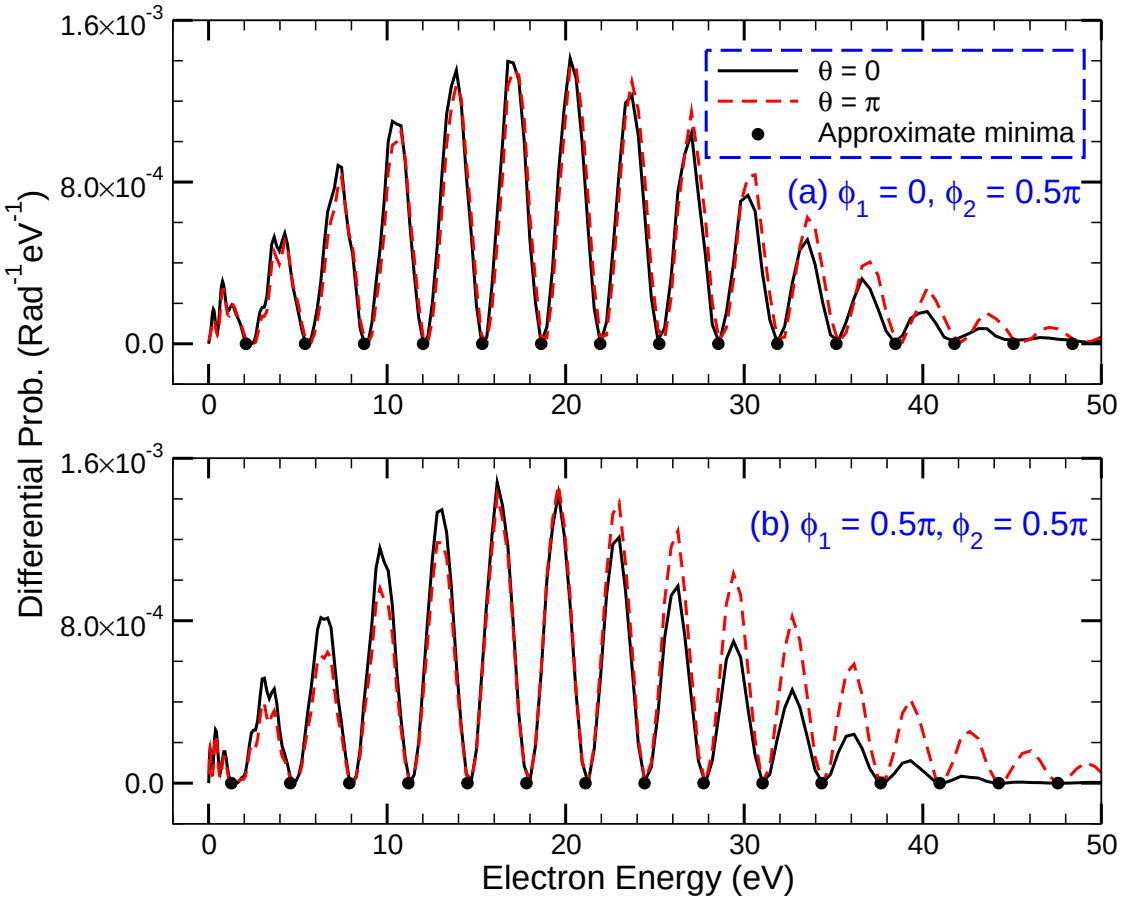
Energy Distribution of Electrons Ionized from He

$\omega_1 = \omega_2 = 36$ eV, $I_1 = I_2 = 5 \times 10^{15}$ W cm^{-2}, $\tau_1 = \tau_2 = 2T_\omega$



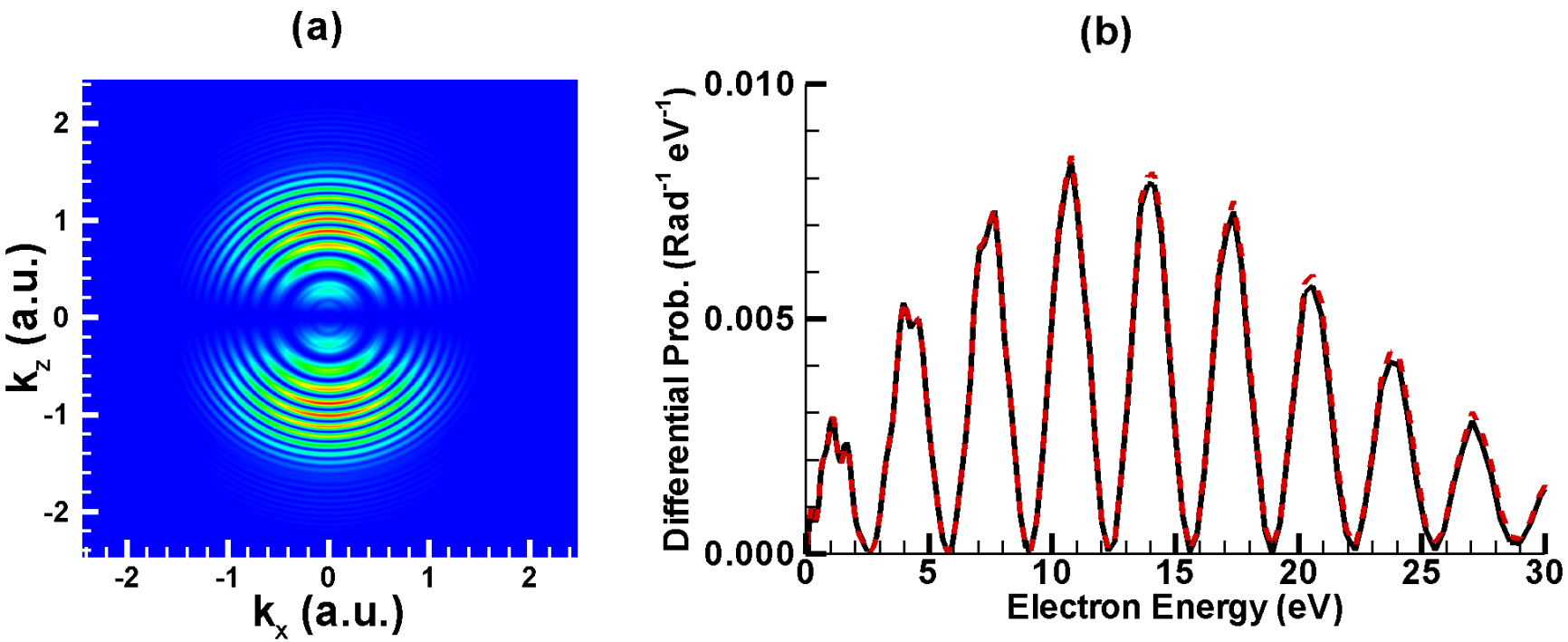
Energy Distribution of Electrons Ionized from H

$\omega_1 = \omega_2 = 36$ eV, $I_1 = I_2 = 5 \times 10^{15}$ W cm^{-2}, $\tau_1 = \tau_2 = 2T_\omega$



Distributions of Electrons Ionized from He by Two Pulses with a Phase Difference of π

$$\phi_1 = 0.5\pi,\ \phi_2 = 1.5\pi$$

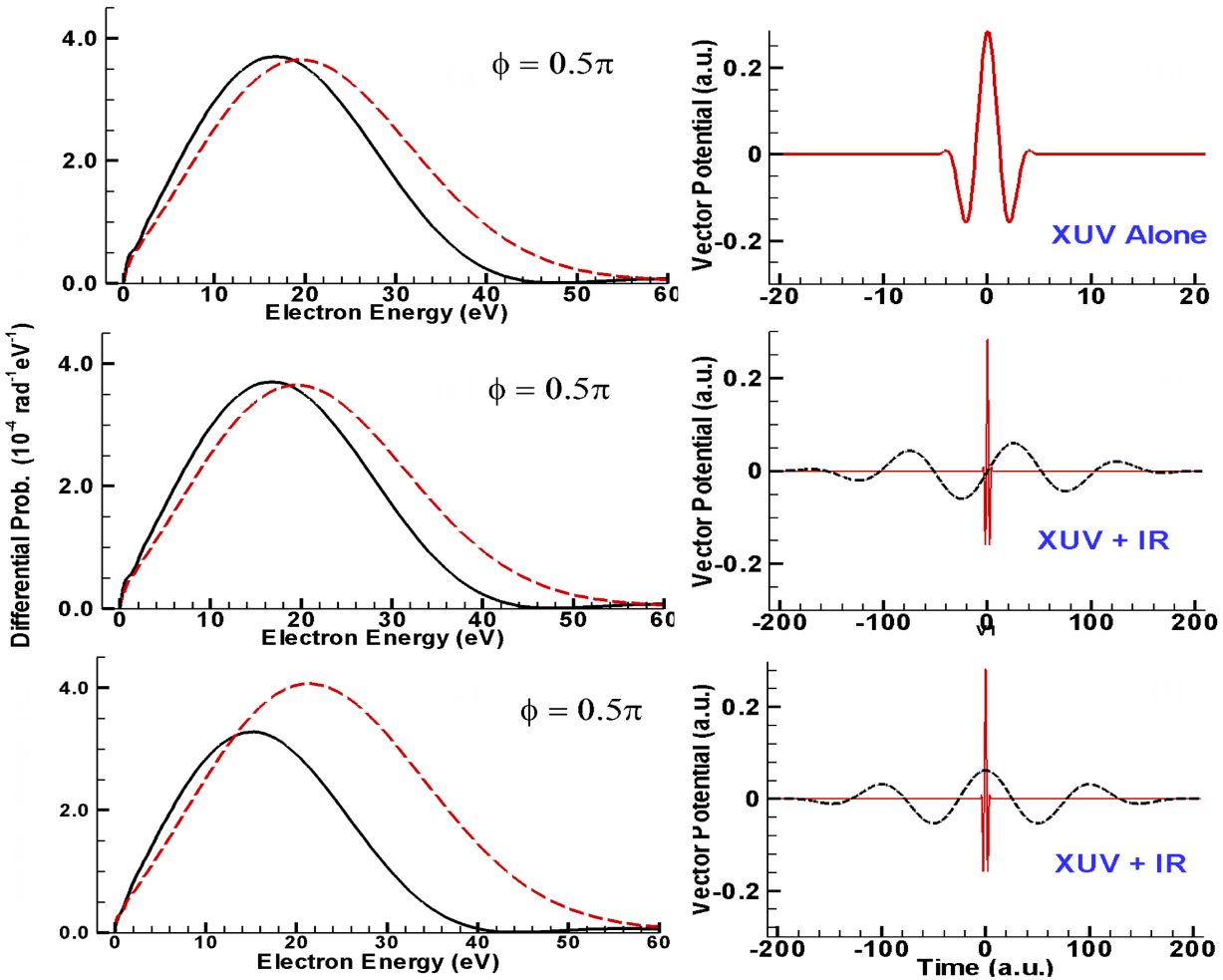


Effects of an Additional IR Pulse

Results with Additional IR Pulse

$P(E, \theta_k)$ for H with an Additional IR Pulse for $\phi_1 = 0.5\pi$

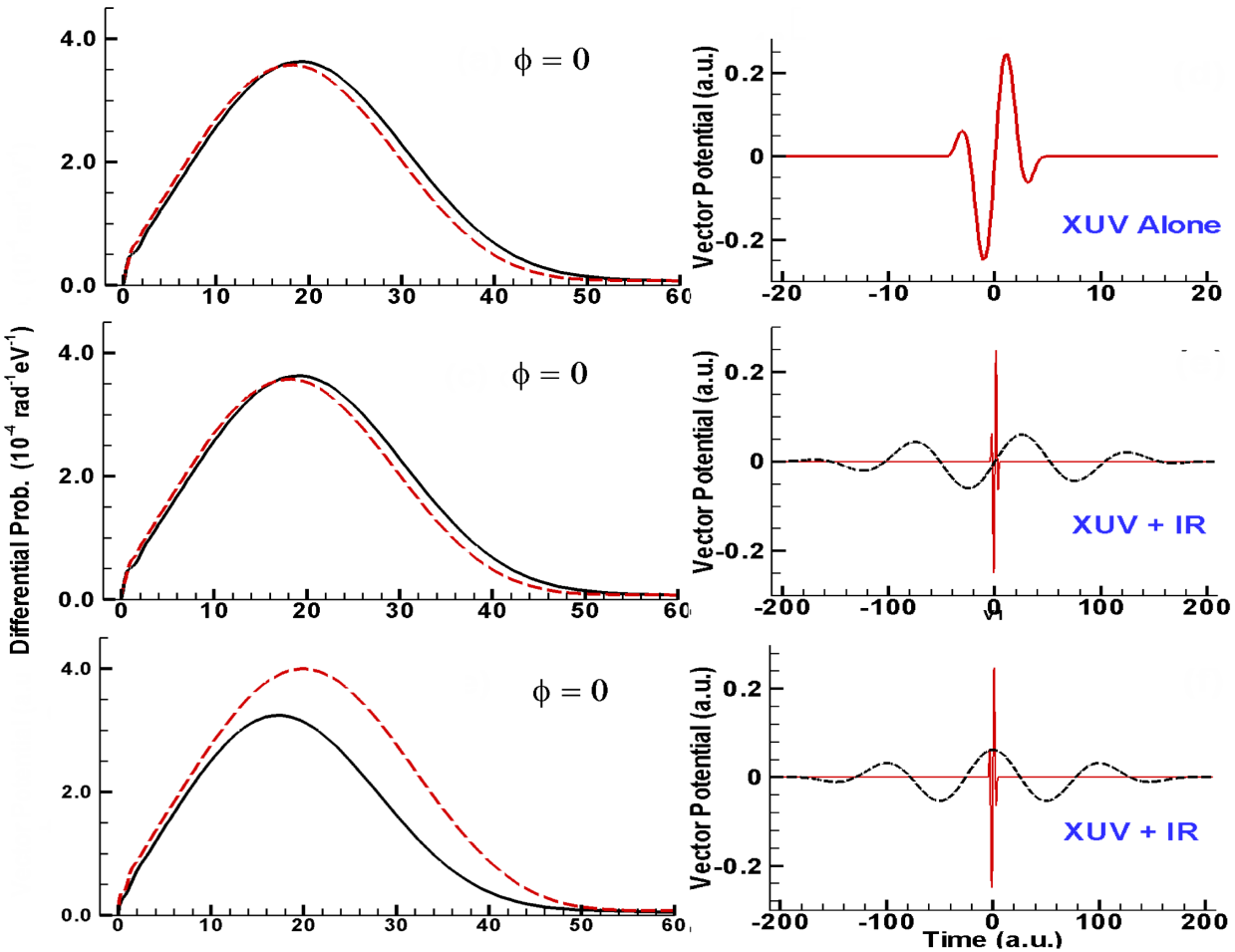
$\lambda_{\rm IR} = 750$ nm, $I_{\rm IR} = 5 \times 10^{11}$ W cm^{-2}, $\tau_{\rm IR} = 4T_{\rm IR}$



Results with Additional IR Pulse

$P(E, \theta_k)$ for H with an Additional IR Pulse for $\phi_1 = 0$

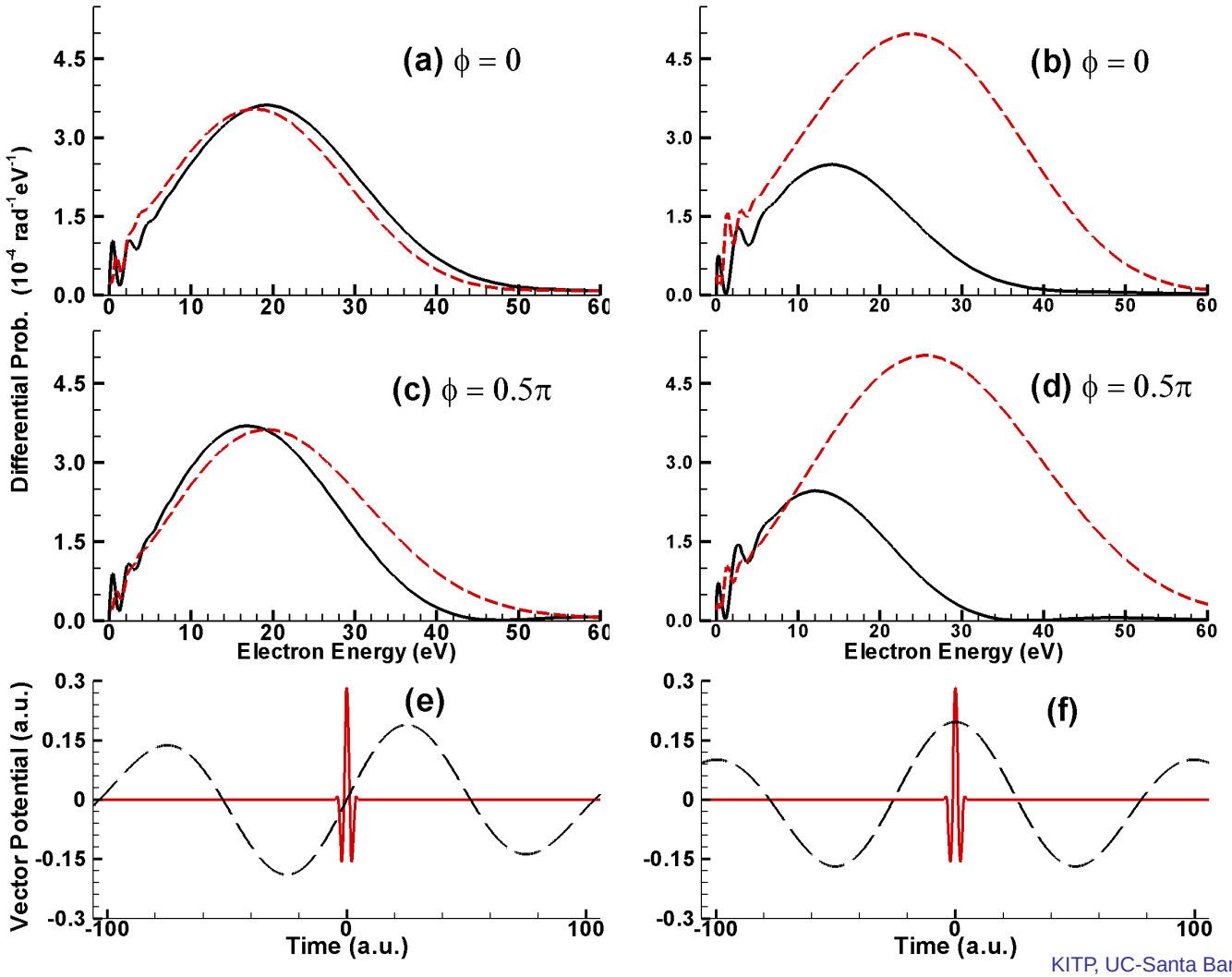
$\lambda_{\rm IR} = 750$ nm, $I_{\rm IR} = 5 \times 10^{11}$ W cm^{-2}, $\tau_{\rm IR} = 4T_{\rm IR}$



Results with Additional IR Pulse

$P(E, \theta_k)$ for H with an Additional IR Pulse for $\phi_1 = \frac{\pi}{2}$

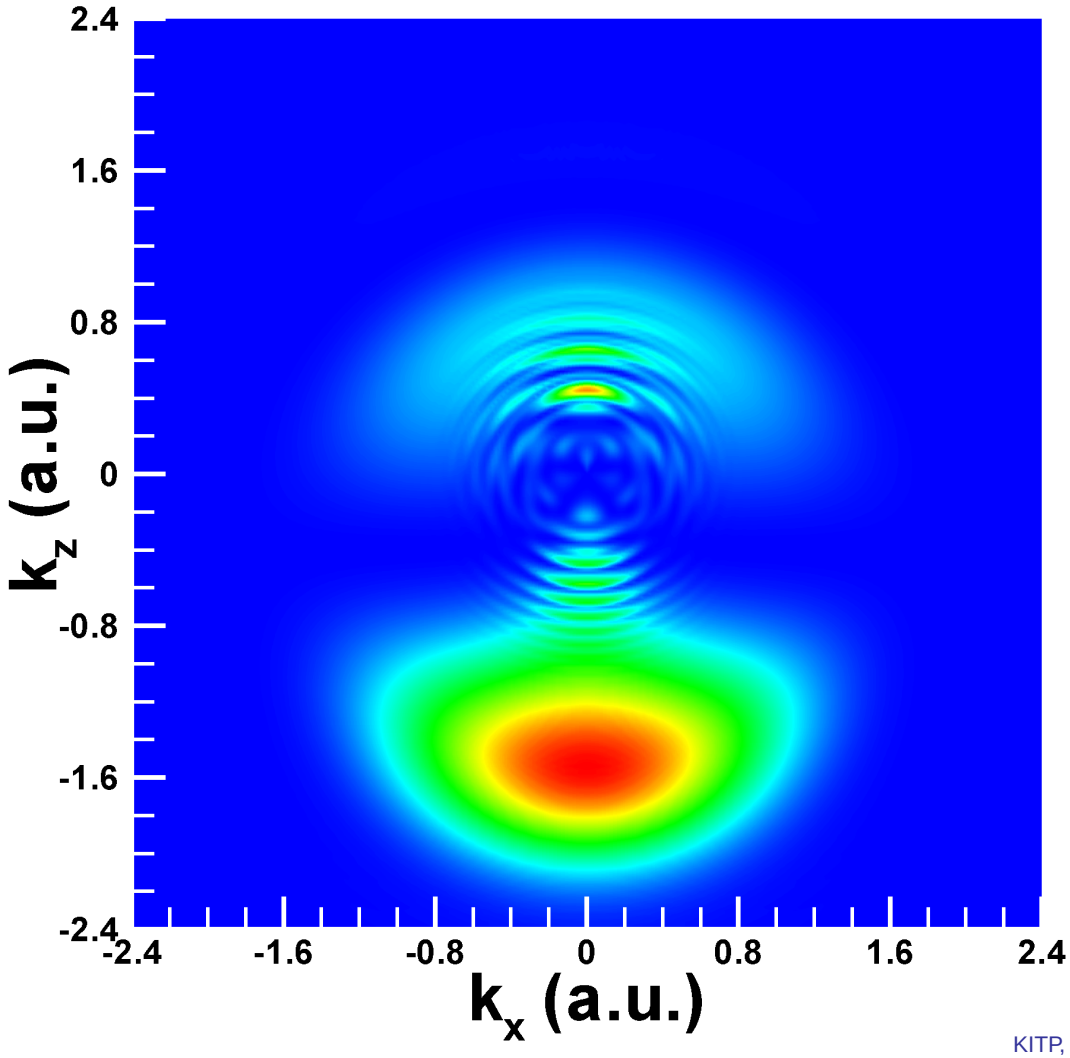
$\lambda_{\rm IR} = 750$ nm, $I_{\rm IR} = 5 \times 10^{12}$ W cm^{-2}, $\tau_{\rm IR} = 4T_{\rm IR}$



Results with Additional IR Pulse

$P(E, \theta_k)$ **for H with an Additional IR Pulse for** $\phi_1 = \frac{\pi}{2}$

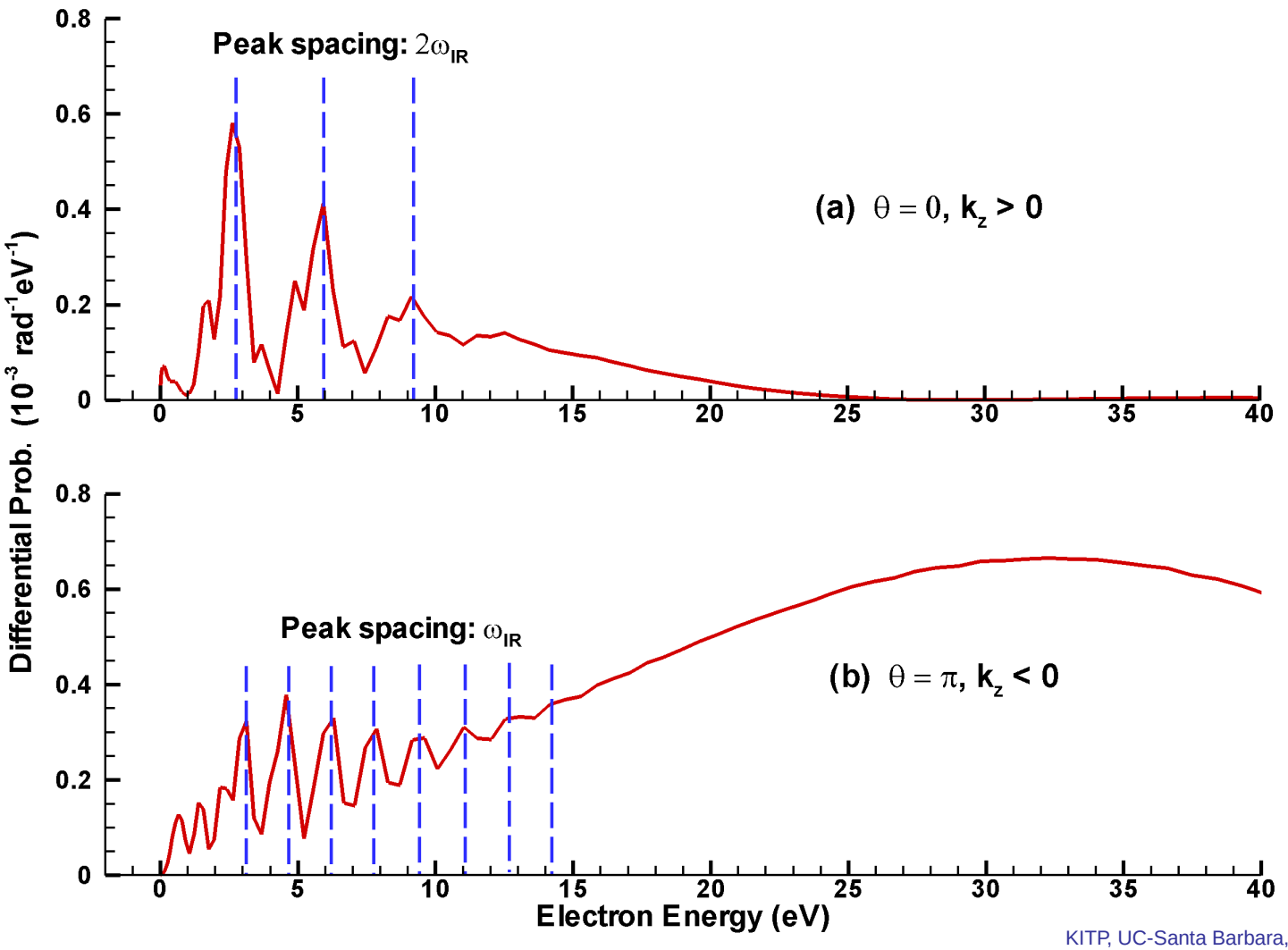
$\lambda_{\rm IR} = 750$ nm, $I_{\rm IR} = 2 \times 10^{13}$ W cm^{-2}, $\tau_{\rm IR} = 8T_{\rm IR}$



Results with Additional IR Pulse

$P(E, \theta_k)$ for H with an Additional IR Pulse for $\phi_1 = \frac{\pi}{2}$

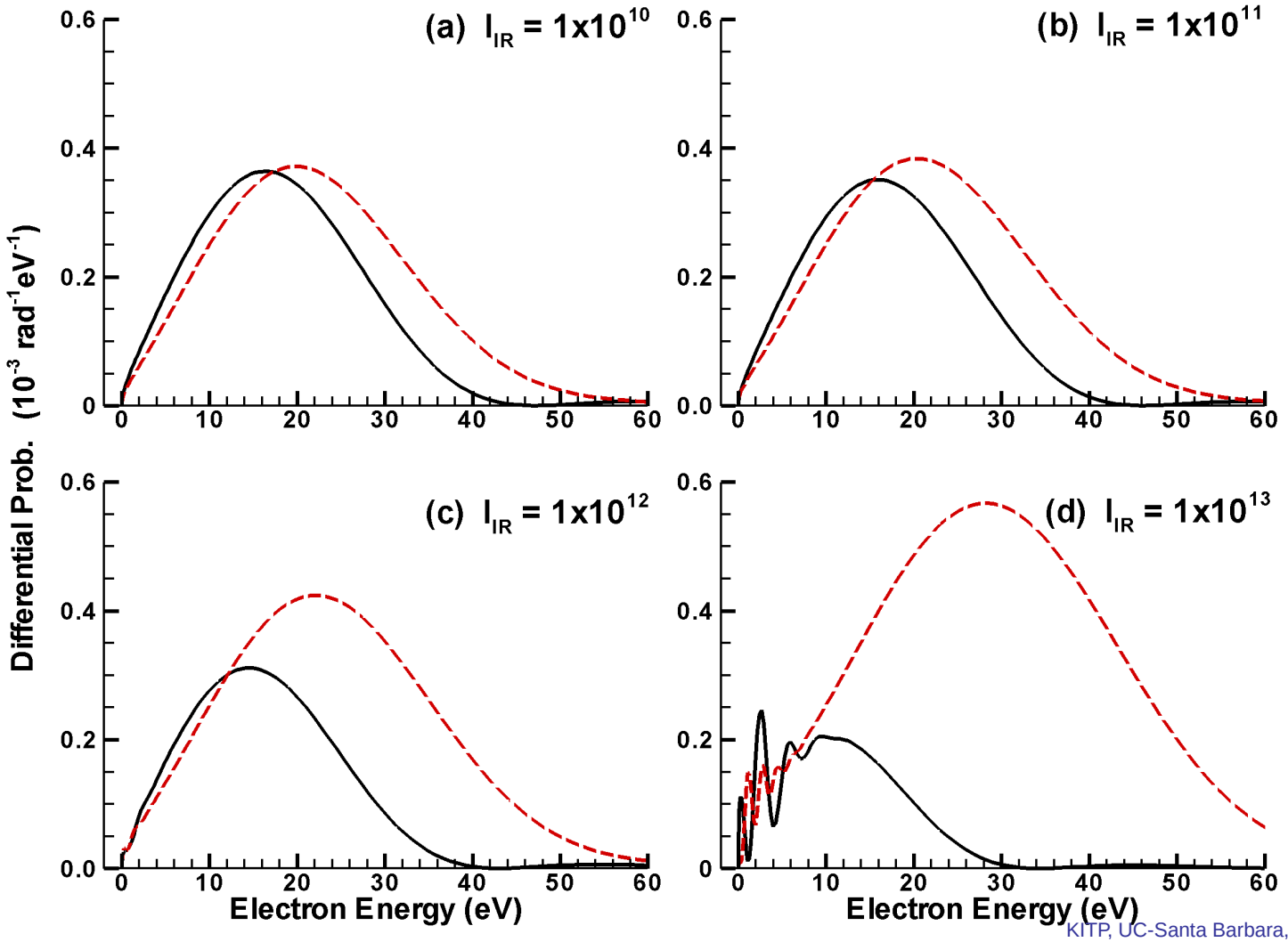
$\lambda_{\rm IR} = 750$ nm, $I_{\rm IR} = 2 \times 10^{13}$ W cm^{-2}, $\tau_{\rm IR} = 8T_{\rm IR}$



Dependence on IR Pulse Intensity

$P(E, \theta_k)$ for H with an Additional IR Pulse for $\phi_1 = \frac{\pi}{2}$

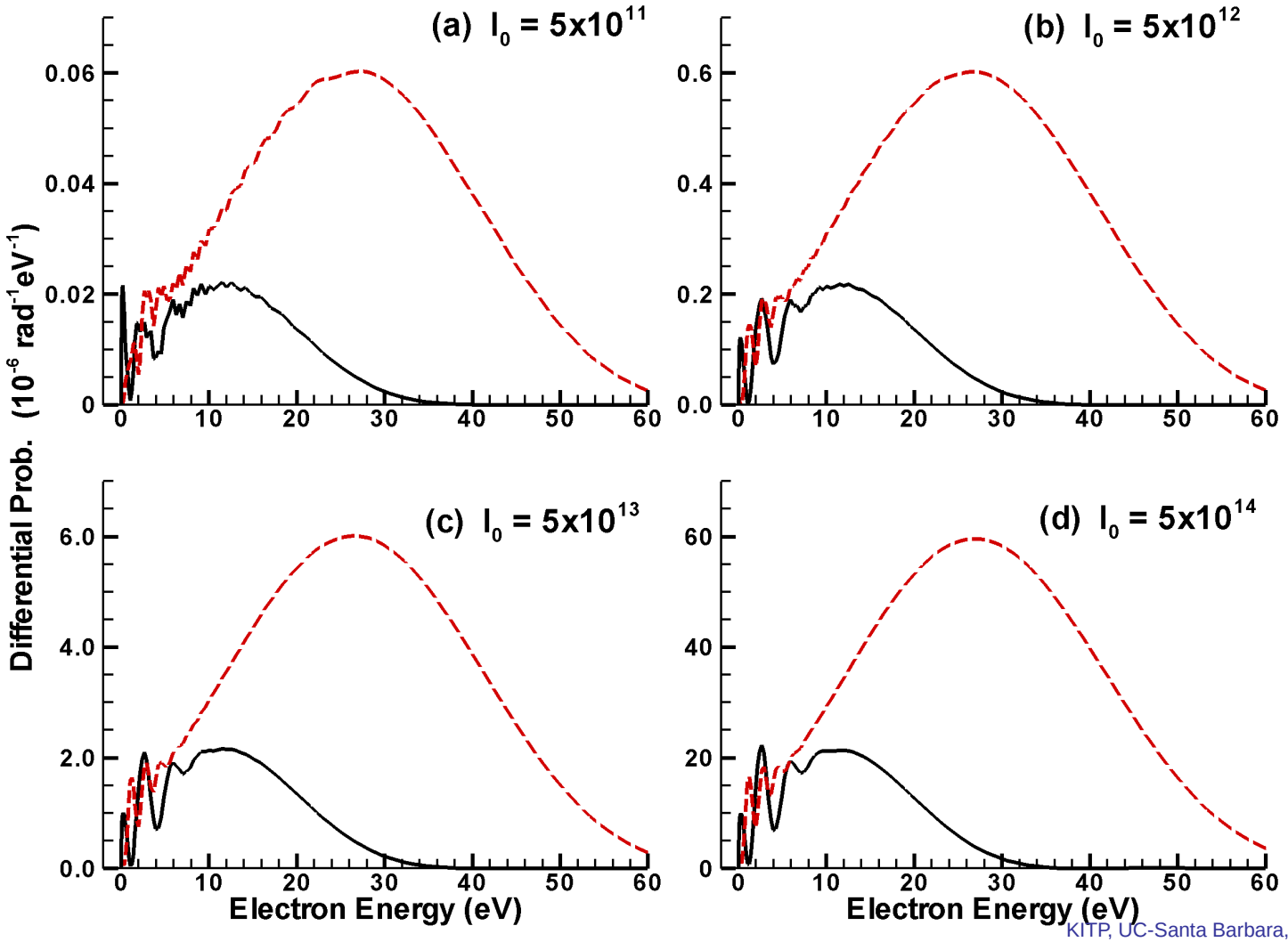
$\lambda_{\rm IR} = 750$ nm, $\tau_{\rm IR} = 4T_{\rm IR}$



Dependence on XUV Pulse Intensity

$P(E, \theta_k)$ for H with an Additional IR Pulse for $\phi_1 = \frac{\pi}{2}$

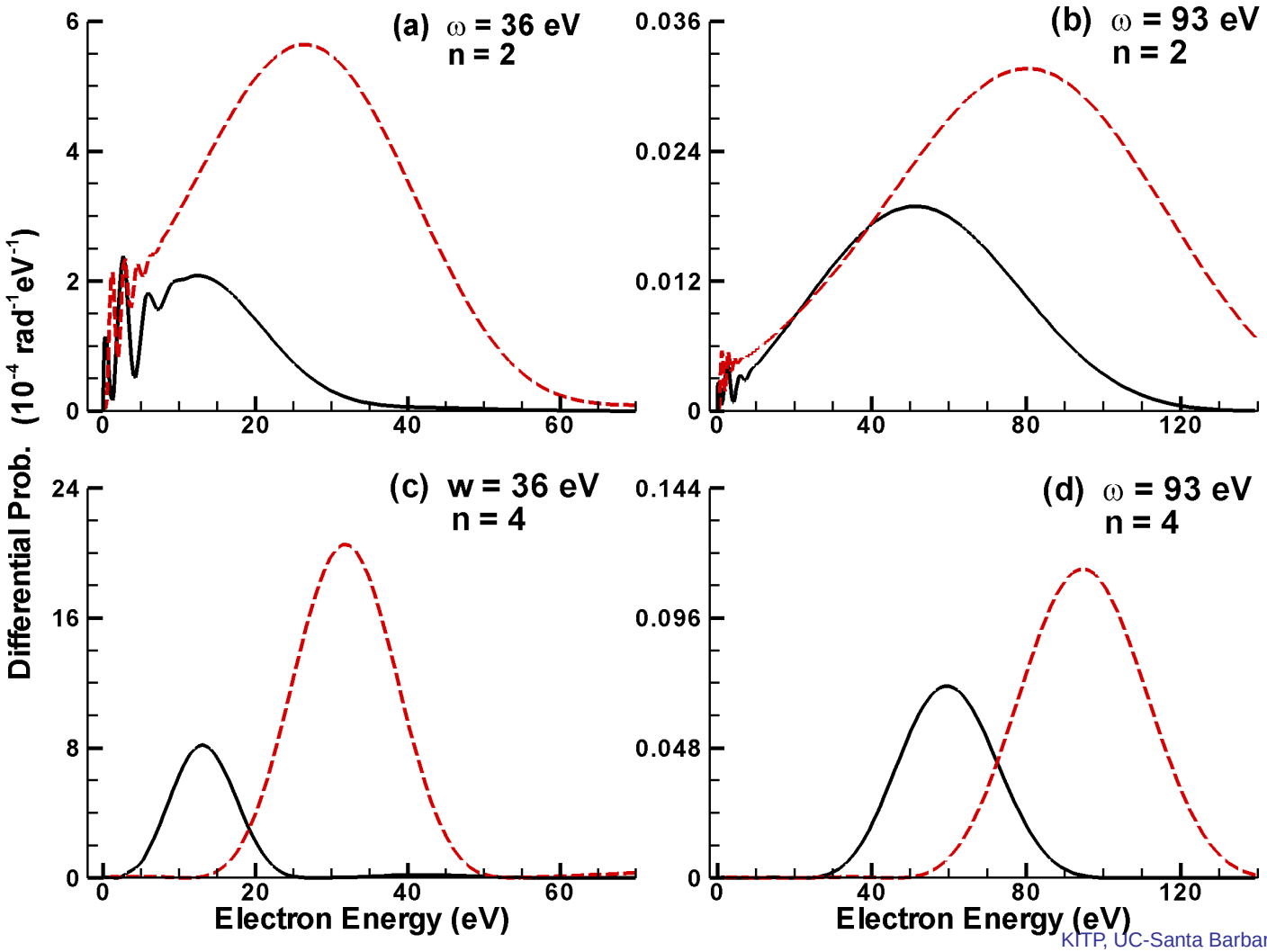
$\lambda_{\rm IR} = 750$ nm, $I_{\rm IR} = 1 \times 10^{13}$ W cm^{-2}, $\tau_{\rm IR} = 4T_{\rm IR}$



Dependence on XUV Frequency, ω, and Number of Cycles, n

$P(E, \theta_k)$ for H with an Additional IR Pulse for $\phi_1 = \frac{\pi}{2}$

$$I_{\mathrm{IR}} = 1 \times 10^{13}\ \mathrm{W\ cm^{-2}}$$



Chirped, Few-Cycle Attosecond Pulses

Collaborators

Liang-You Peng, Fang Tan and Qihuang Gong

Peking University, Beijing

Evgeny A. Pronin and Anthony F. Starace

University of Nebraska, Lincoln

Reference: *Phys. Rev. A* (submitted, 2009)

Chirped Pulses

$$\mathbf{A}(t) = AF(t)\sin\left[\omega(t)(t-t_0)+\phi_0\right]\mathbf{e}_z, \tag{1}$$

$$A = \frac{E}{\omega_0} = \frac{1}{\omega_0}\sqrt{\frac{I_0/I_{\text{au}}}{\sqrt{1+\xi^2}}}, \tag{2}$$

$$\omega(t) = \omega_0 + 4\ln 2\frac{\xi}{1+\xi^2}\frac{(t-t_0)}{\tau_0^2}, \tag{3}$$

$$F(t) = \exp\left[-4\ln 2\frac{1}{1+\xi^2}\frac{(t-t_0)^2}{\tau_0^2}\right], \tag{4}$$

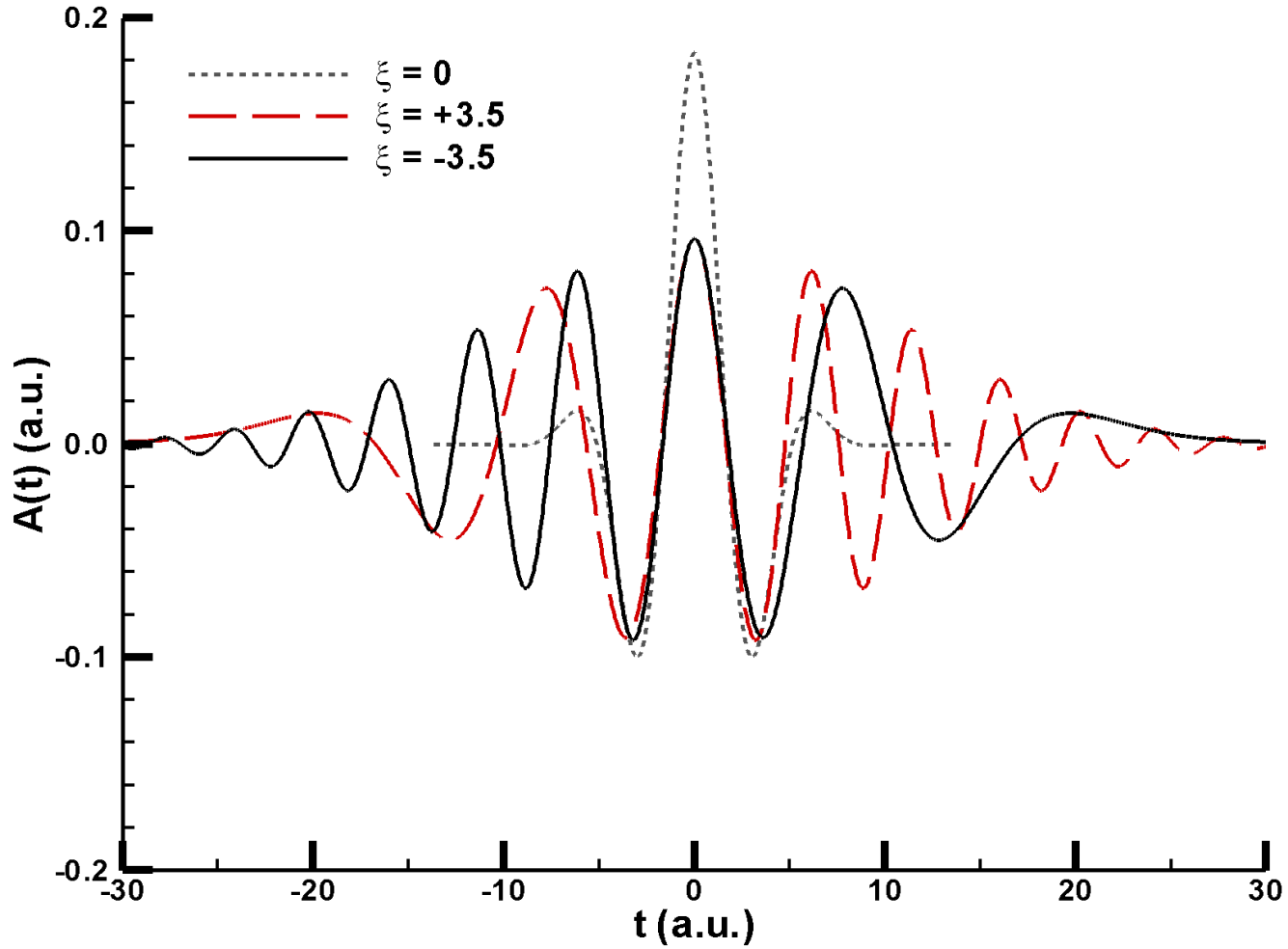
Key features:

- Pulse BANDWIDTH does NOT depend on chirp ξ
- Pulse ENERGY does NOT depend on chirp ξ
- Chirped pulse duration $\tau = \tau_0\sqrt{1+\xi^2}$, peak intensity $I = I_0/\sqrt{1+\xi^2}$

Differential Probabilities

Laser pulse vector potential. The laser pulse for $\xi = 0$ has

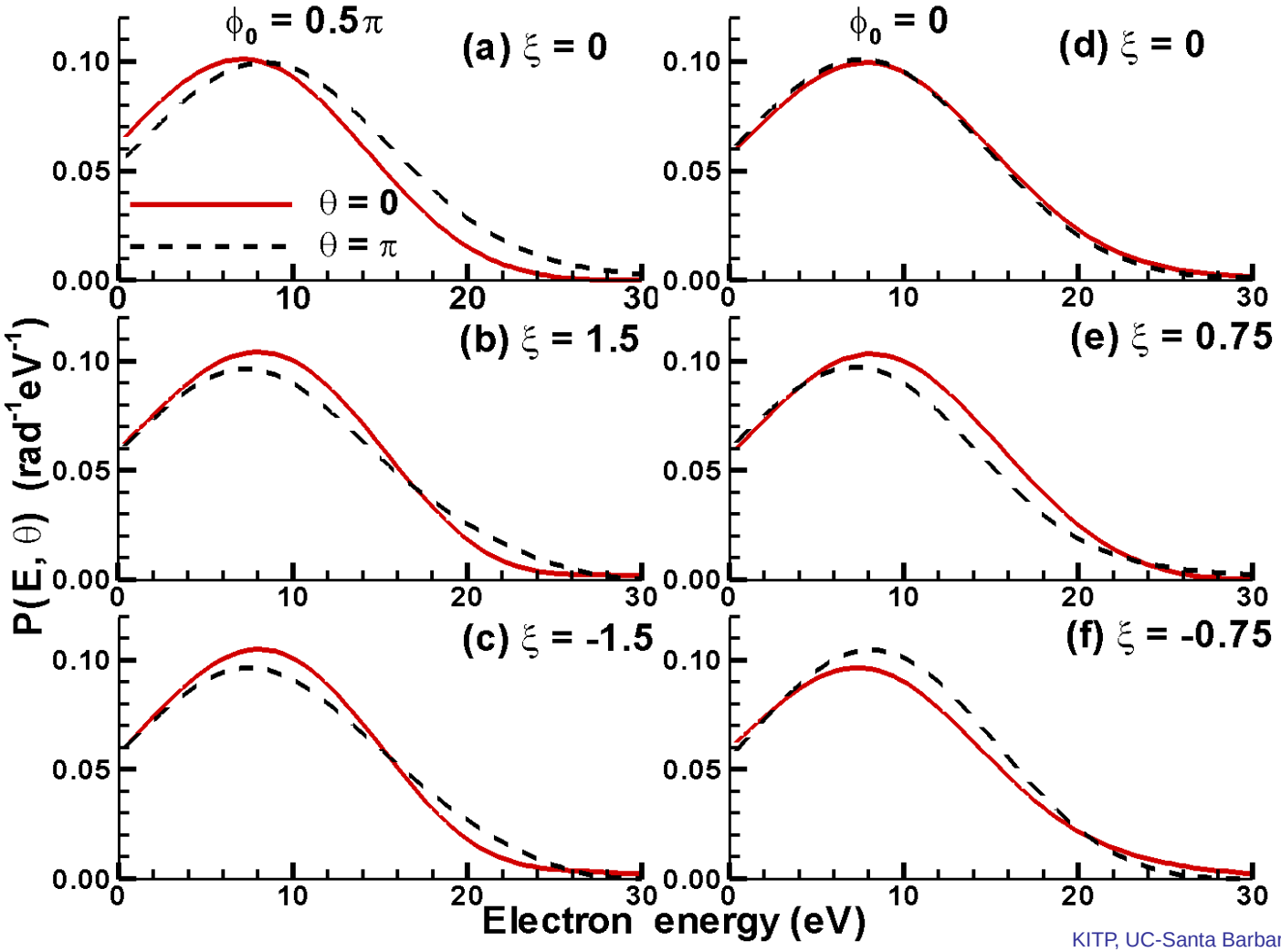
$\omega_0 = 25$ eV, $I_0 = 10^{15}$ W/cm^2 , $\tau_0 = T_0$ (1 cycle), $\phi_0 = \pi/2$



Differential Probabilities

$P(E, \theta_k)$ **for Hydrogen. The laser pulse for** $\xi = 0$ **has**

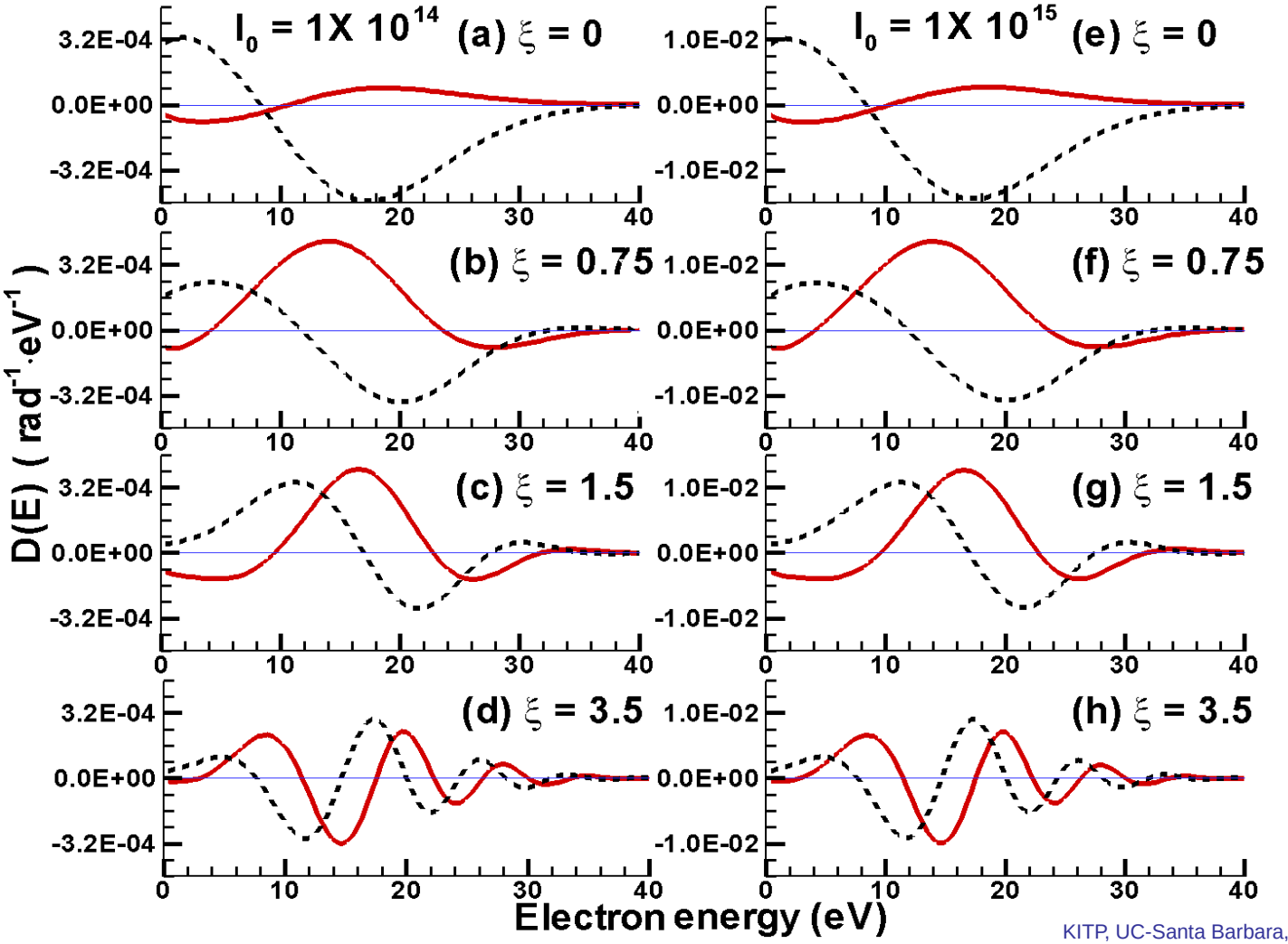
$\omega_0 = 25$ eV, $I_0 = 10^{15}$ W/cm^2 , and $\tau_0 = T_0$ (1 cycle)



Differential Probability Differences

$$D(E) = P(E,0) - P(E,\pi).$$

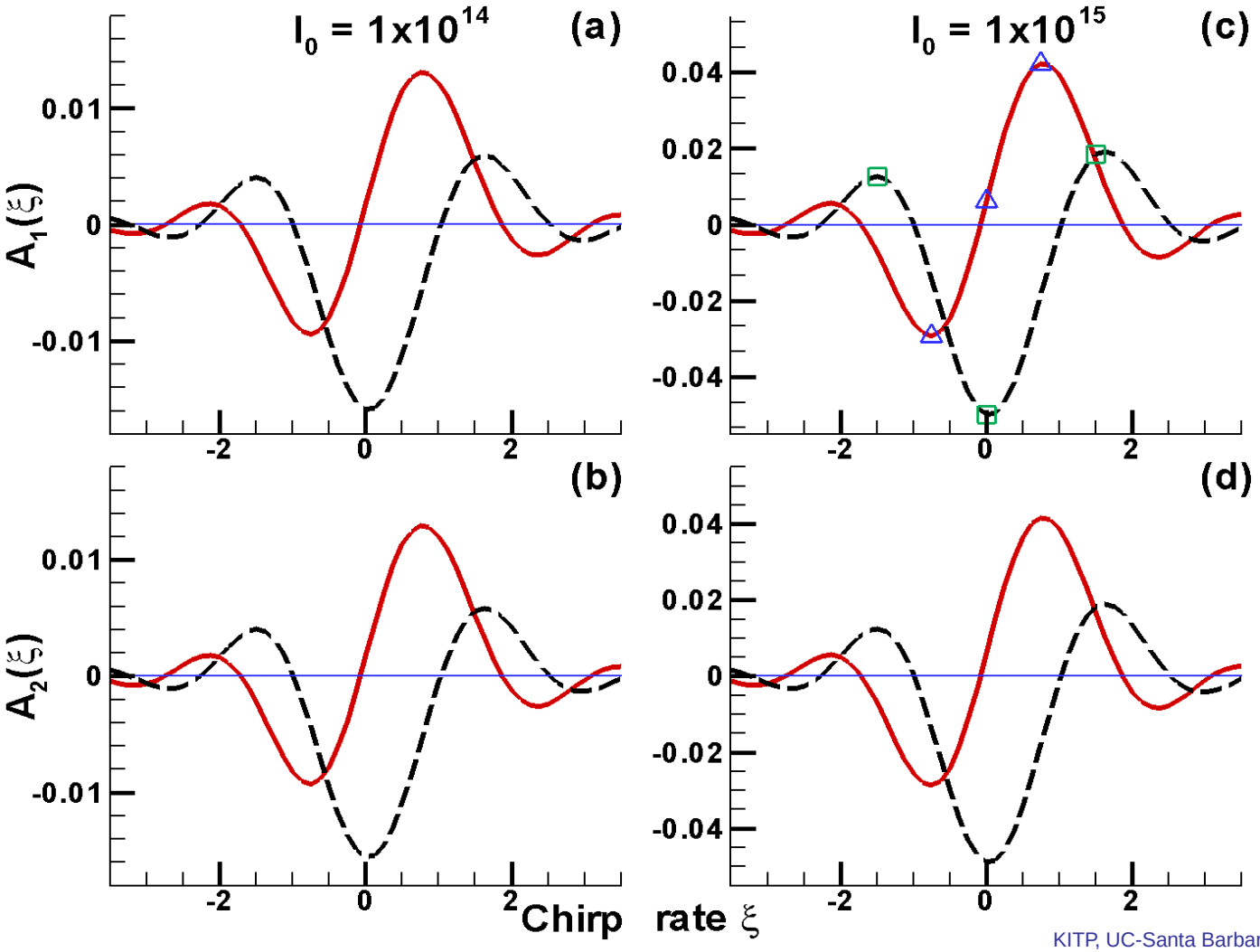
Solid lines, $\phi_0 = 0$; dashed lines, $\phi_0 = \pi/2$.



Asymmetry Dependence on Chirp ξ

$A_1 = \frac{P_0^+ - P_0^-}{P_0^+ + P_0^-}$, $A_2 = \frac{P_{10^\circ}^+ - P_{10^\circ}^-}{P_{10^\circ}^+ + P_{10^\circ}^-}$ **(integrated over energy).**

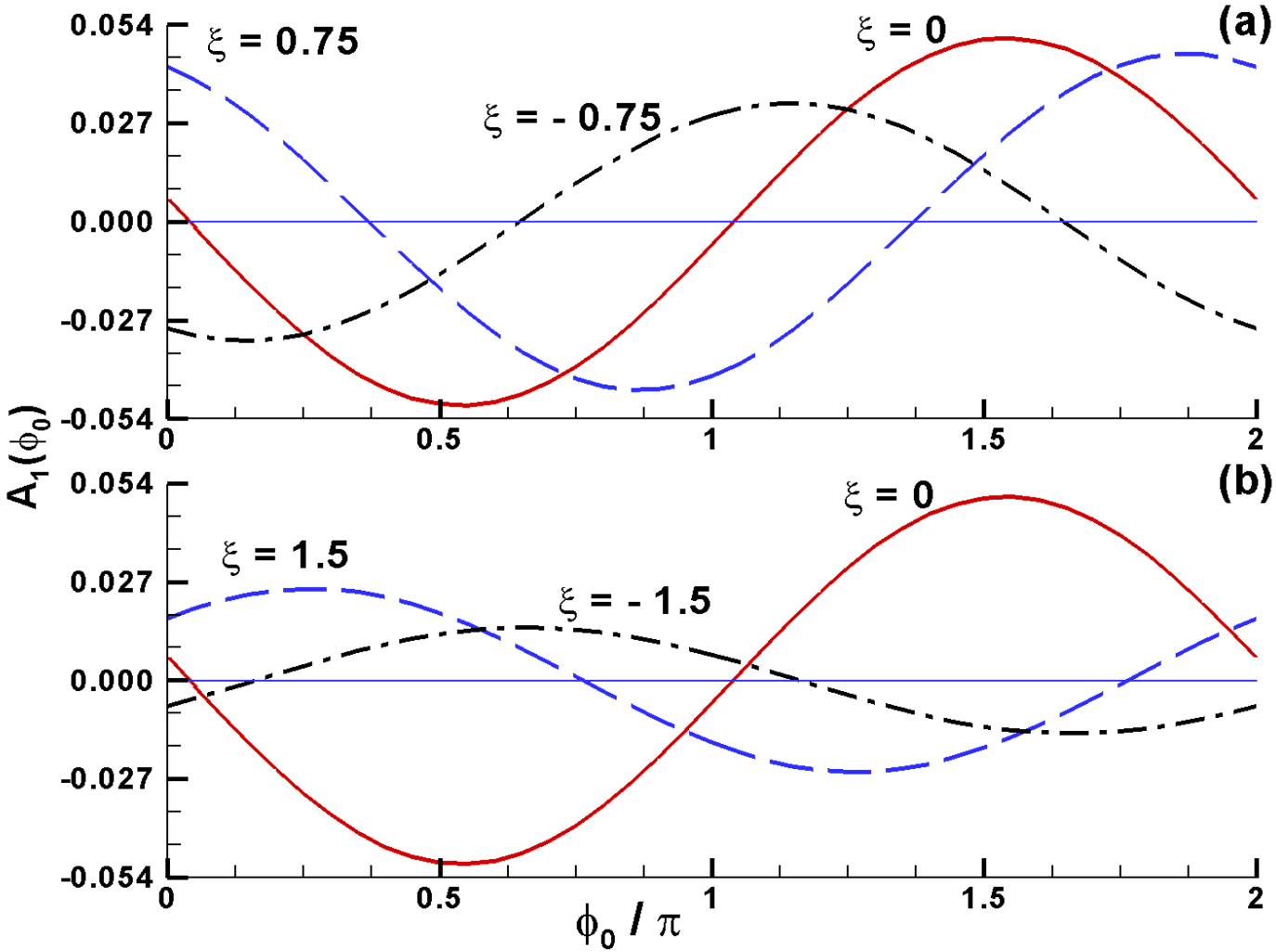
Solid lines, $\phi_0 = 0$; dashed lines, $\phi_0 = \pi/2$.



Asymmetry Dependence on CEP ϕ_0

$A_1 = \frac{P_0^+ - P_0^-}{P_0^+ + P_0^-}$ **(integrated over energy).**

$I_0 = 10^{15}$ W/cm^2.



Concluding Remarks

Summary and Conclusions

- The **CEPs of few-cycle attosecond pulses produce asymmetries** in ionized electron momentum and energy distributions that become significant at attosecond pulse intensities $\gtrsim 10^{14}$ W/cm^2 **(within range of current experimental capabilities)**.
- Even **a weak IR pulse can augment the CEP effects** of a few-cycle attosecond pulse.
- For short attosecond pulses having ionized electron spectra with significant numbers of low-energy electrons, **an IR pulse may allow one to explore rescattering of ionized electrons from the ionic core.**

Concluding Remarks (cont'd)

- **The chirp** of a few-cycle attosecond pulse can **affect significantly the asymmetry** in the ionized electron distributions, which remain sensitive to the CEP.

Outlook:

- The CEP and chirp of a few-cycle attosecond pulse provide **additional tools for controlling electron dynamics** in AMO processes initiated by a few-cycle attosecond pulse.

References:

Phys. Rev. A **76**, 043401 (2007);

New J. Phys. **10**, 025030 (2008);

Phys. Rev. A (submitted).

Beam Parameters and Gouy Phase

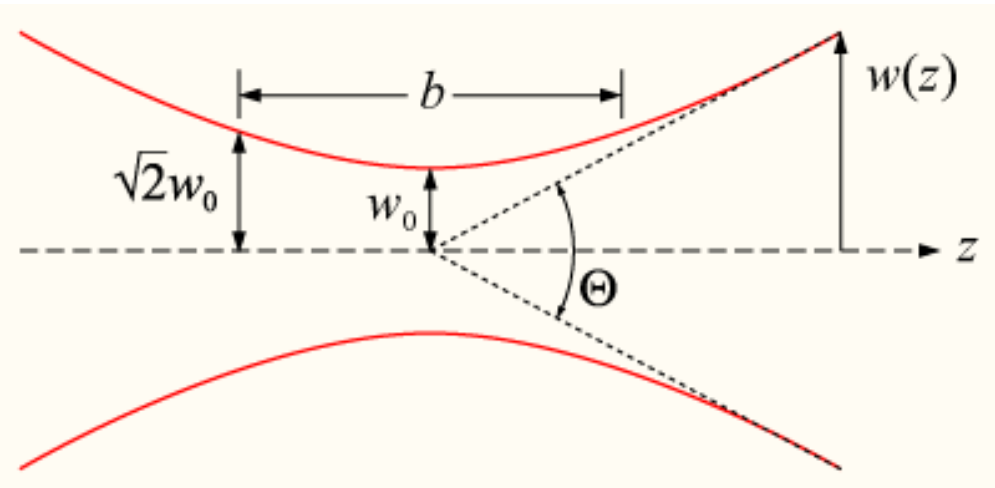
Beam Waist: w_0 **Rayleigh Length:** $z_0 = \pi w_0^2/\lambda$

Spot Size: $w(z) = w_0\sqrt{1+(z/z_0)^2}$

Confocal Parameter: $b = 2z_0$

Beam Divergence: $\theta = \Theta/2 \approx \lambda/\pi w_0$

Ref.: F. Lindner et al., *PRL* **92**, 113001 (2004)



Gouy Phase: $\zeta(z) = \arctan(z/z_0)$

Estimate of Gouy Phase Variation

Beam Waist: $w_0 \gtrsim 1\mu\mathrm{m}$ **Wave Length:** $\lambda \sim 3.45 \times 10^{-2}\mu\mathrm{m}$

Rayleigh Length: $z_0 = \pi w_0^2/\lambda \gtrsim 100\mu\mathrm{m}$

Gouy Phase: $\zeta(z) = \arctan(z/100)$