

**Real-time all-non-optical method
for complete characterization
of single-shot few-cycle laser pulses**

C D Lin and Zhangjin Chen, Kansas State University

1. Determine

Carrier –envelope-phase

Pulse duration

Peak intensity at focus

of **single-shot** laser pulses from high-energy
photoelectron spectra

2. Determine relative phase and intensity of
2-color measurements

Experiments with few-cycle pulses

Good for:

Waveform dependence in light-matter interactions

Applications—

1. For single attosecond pulse generation
2. Controlling electron emissions
3. Controlling dissociation dynamics of molecules

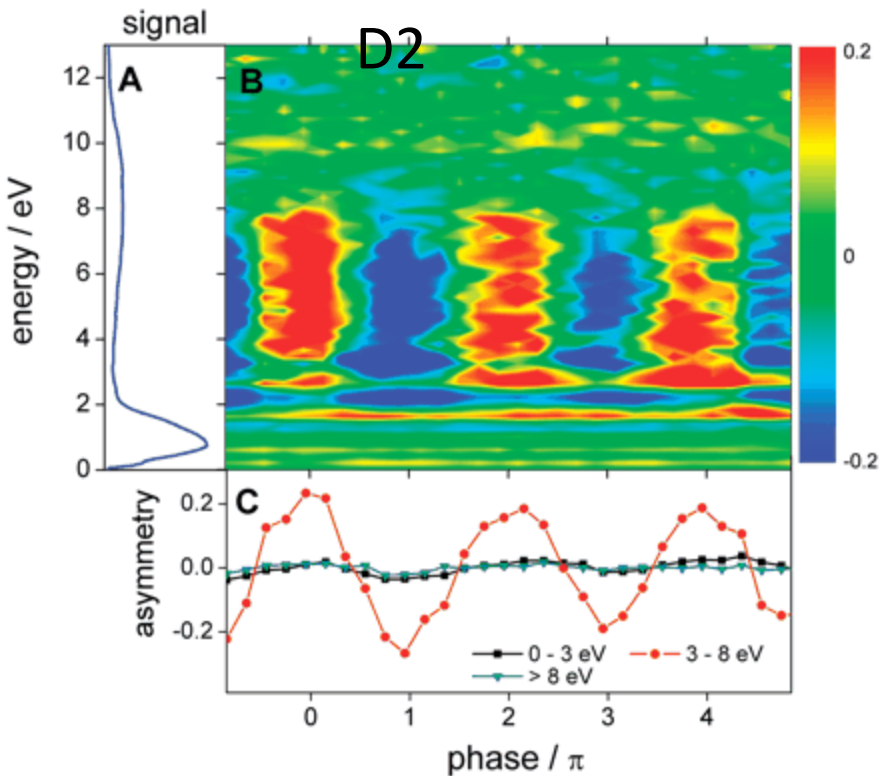
Need phase stabilized pulses

Limitations—

1. CEP stabilization is difficult
2. “Absolute” phase difficult to know -- theory input
3. Pulse duration and peak intensity not known well

Experiments -- CEP-dependent **asymmetric dissociative ionization**

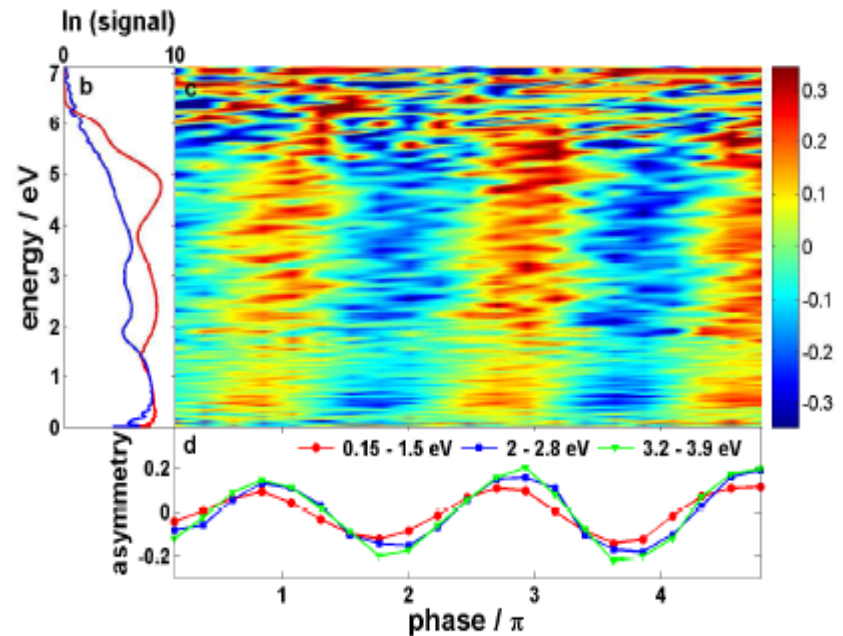
CEP stabilized pulses used



Kling et al Science 2006

Tong and Lin PRL 2007

CO



Znakovskya et al PRL 2009

Need to Know the CEP – requires theory input

Single-shot carrier-envelope phase measurement of few-cycle laser pulses

T. Wittmann¹, B. Horvath¹, W. Helml¹, M. G. Schätzel¹, X. Gu¹, A. L. Cavalieri¹, G. G. Paulus^{2,3}
and R. Kienberger^{1,4}★

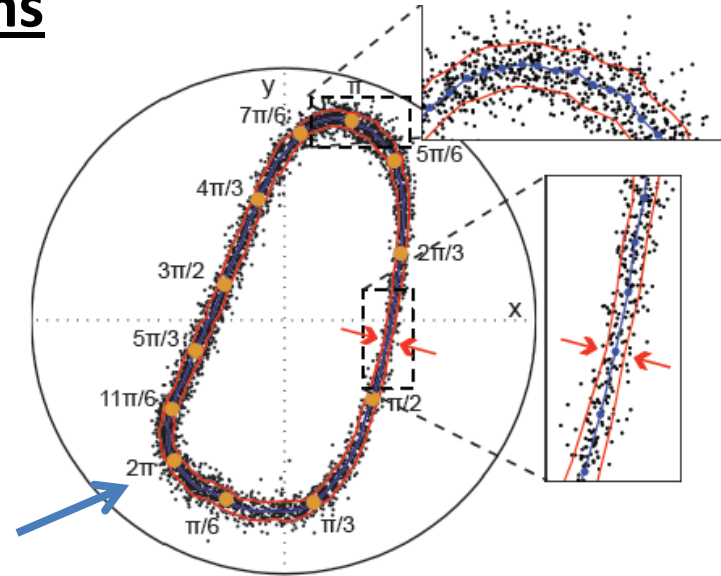
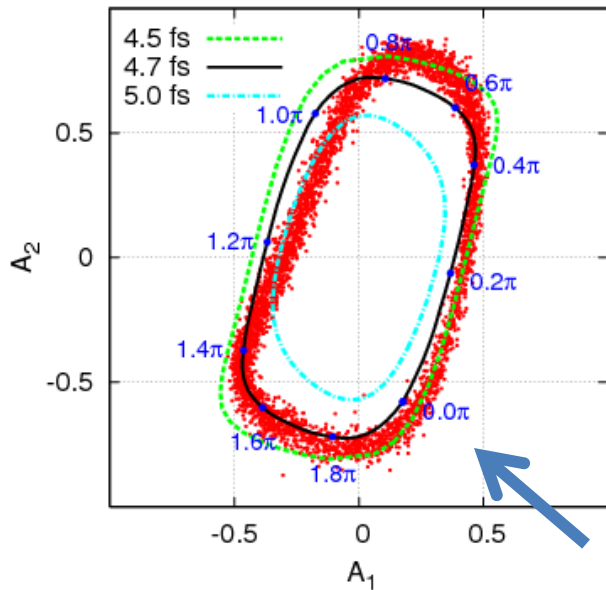
Accomplished:

1. Able to measure left/right electron spectra at **each single shot**
2. Analyze the left/right asymmetry to get the **relative** CEP of each shot

Limitations

1. **Absolute** CEP is not known
2. Pulse duration and Peak intensity determined by optical method (not done usually)

Conclusions



our method:

- Accurate absolute CEP
- Pulse duration
- Pulse peak intensity

Wittmann et al
error of 60 deg
estimated
estimated

- All based on the left/right electron spectra

Our method is fast and accurate— all based on left/right electron spectra — **allow real-time retrieval of laser parameters for single-shot measurements**

Opportunities: To perform waveform dependence studies with

- **CEP-tagging for pulses that are not CEP-stabilized (at the single-shot level)**

Our Method

QRS

fast

accurate

Vs **SFA** and **TDSE**

Quantitative rescattering theory for laser-induced high-energy plateau photoelectron spectra

Zhangjin Chen,¹ Anh-Thu Le,¹ Toru Morishita,² and C. D. Lin¹

Quantitative Rescattering theory (QRS) for high-energy photoelectrons

$$I(p_v, \theta_v) = W(p_r) \sigma(p_r, \theta_r)$$

Features of QRS—

1. Based on the **rescattering concept**
2. Wave packet from strong field approximation— **laser property**
3. Cross section— elastic collisions by free electrons —**target property**
4. Checked against TDSE

Theoretical tools for laser-molecule interactions:

1. Solving the time-dependent Schrödinger Equation numerically (“EXACT”)

$$H = H_{atom} + H_i(t)$$
$$H\Psi(\vec{r}, t) = i\hbar \frac{\partial}{\partial t} \Psi(\vec{r}, t)$$

$$I(p_v, \theta_v) = W(p_r) \sigma(p_r, \theta_r)$$

Limited to one-electron model atomic systems

2. Second-order strong field approximation (Simple)

SFA2:

$$f_2(\vec{k}) = - \int_{-\infty}^{\infty} dt \int_t^{\infty} dt' \int d\vec{p} \langle \chi_{\vec{k}}(t') | V | \chi_{\vec{p}}(t') \rangle \langle \chi_{\vec{p}}(t) | H_i(t) | \Psi_0(t) \rangle$$

$$I_{SFA}(p_v, \theta_v) = W_{SFA}(p_r) \sigma_{Bl}(p_r, \theta_r)$$

3. Quantitative Rescattering Theory (QRS)

$$I_{QRS}(p_v, \theta_v) = W_{SFA}(p_r) \sigma(p_r, \theta_r)$$

$$\mathbf{k} = -\mathbf{A}_r + \mathbf{k}_r$$

$$k_r = 1.26A_r$$

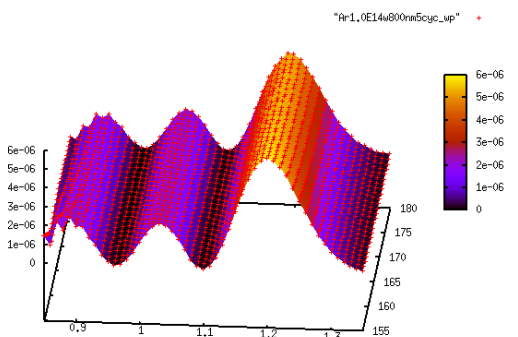
Wave Packets

Th

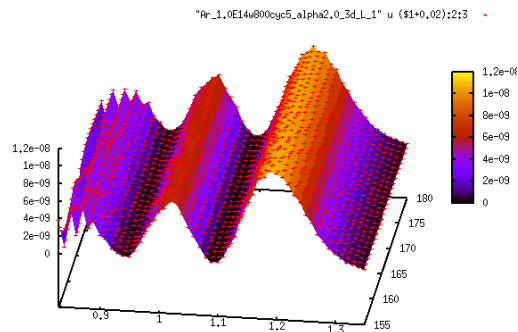
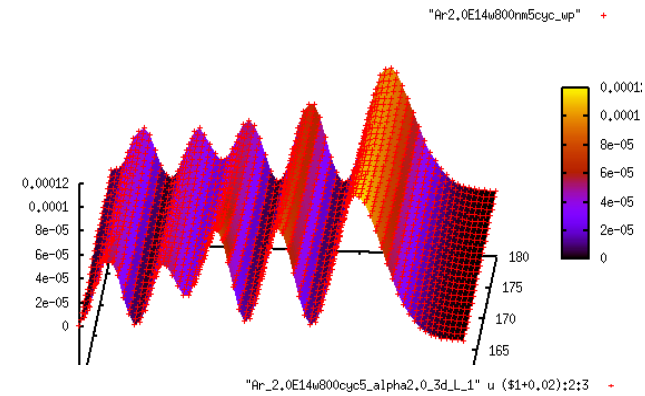
Nearly identical from TDSE and SFA2

$I_0 = 1 \times 10^{14} \text{ W/cm}^2$

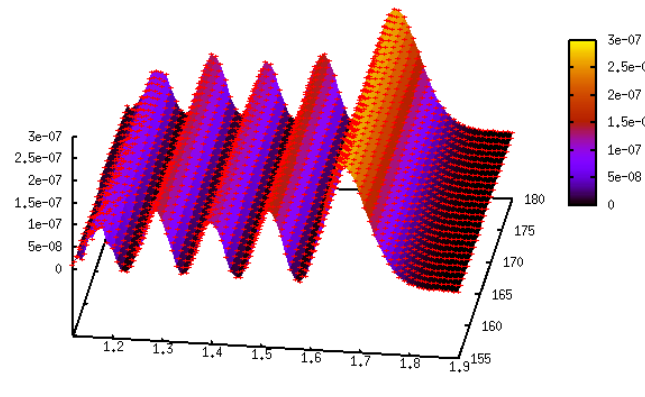
$I_0 = 2 \times 10^{14} \text{ W/cm}^2$



TDSE



SFA2

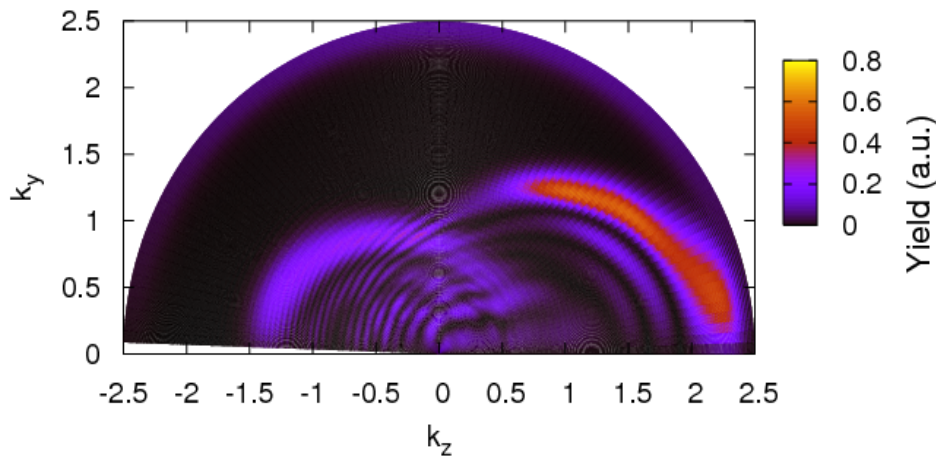


Argon: $\lambda=800 \text{ nm}$, FWHM=5 fs

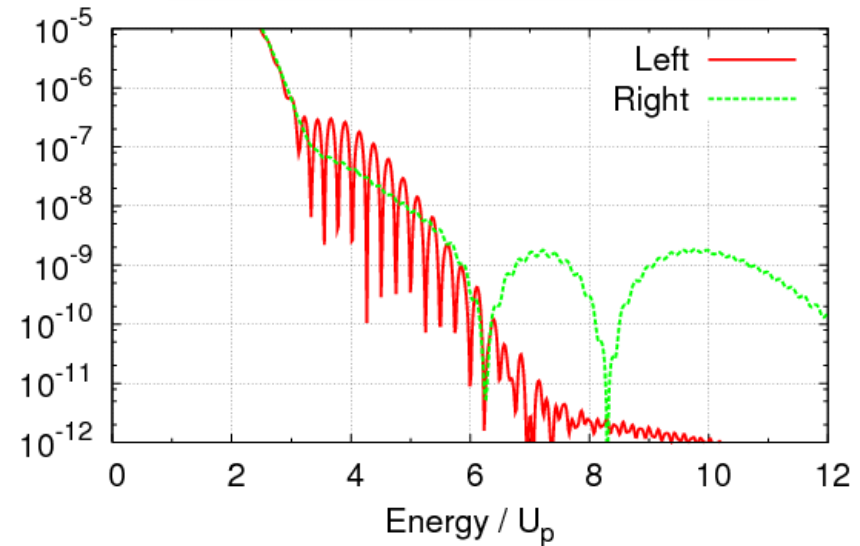
$$I(\mathbf{p}_v) = \sigma(p_r, \theta_r) W(p_r)$$

CEP dependence of high energy photoelectron spectra

High energy 2D momentum distribution



Left and right high energy spectra



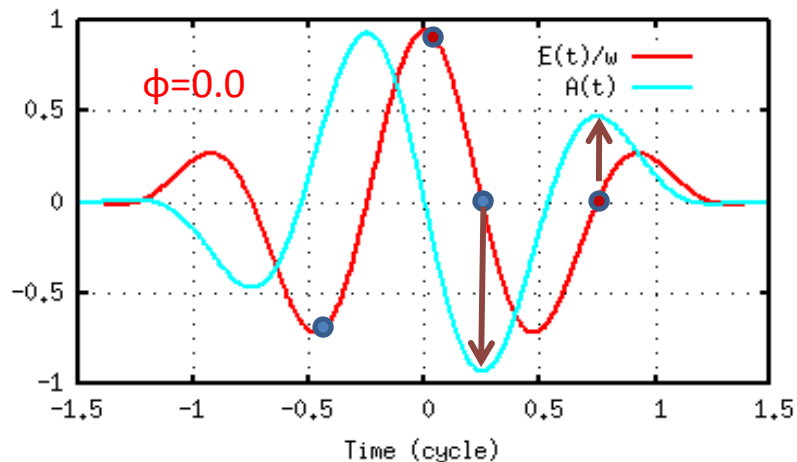
$$E(t) = E_0 a(t) \cos(\omega t + \varphi)$$

Hydrogen:

1.0×10^{14} W/cm²,

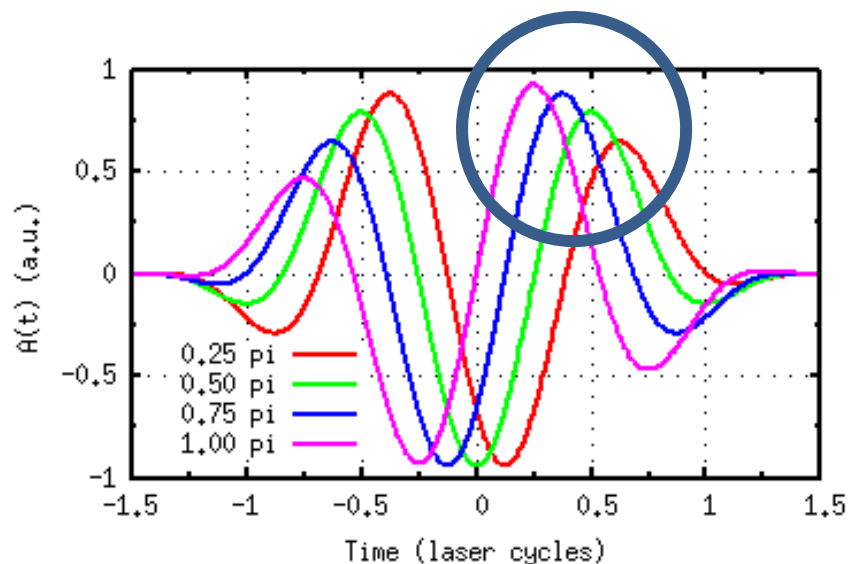
800 nm,

3 optical cycle

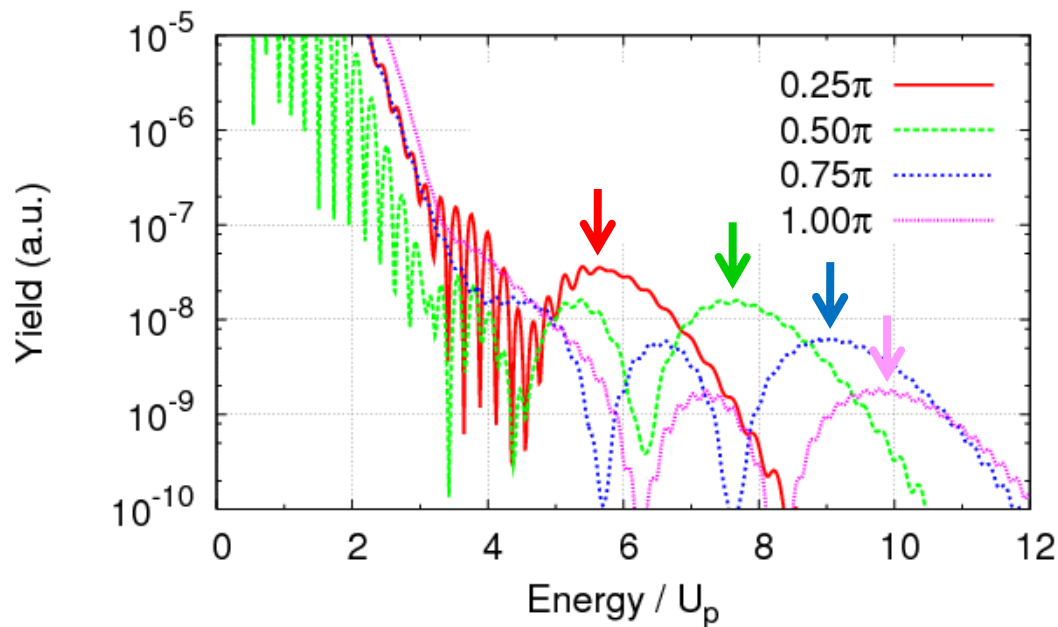


CEP dependence of the high energy photoelectron spectra

A(t) at different CEP



Left-side energy spectra for different CEP



Hydrogen:

$1.0 \times 10^{14} \text{ W/cm}^2$,

800 nm,

3 optical cycle

$$U_p = \frac{A_0^2}{4} = \frac{E_0^2}{4\omega^2}$$

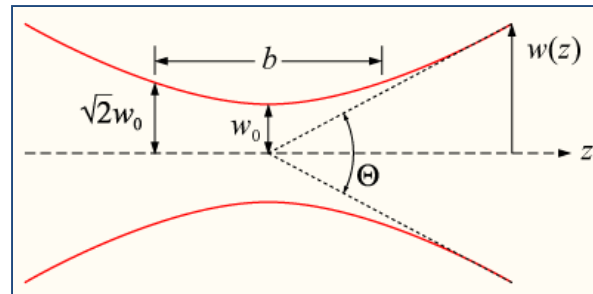
Simplicity and significance of the QRS:

$$I(\mathbf{p}_v) = \sigma(p_r, \theta_r) W(p_r)$$

structure

Lasers

**Photoelectrons are collected from a focused laser beam—
Volume integration is needed**

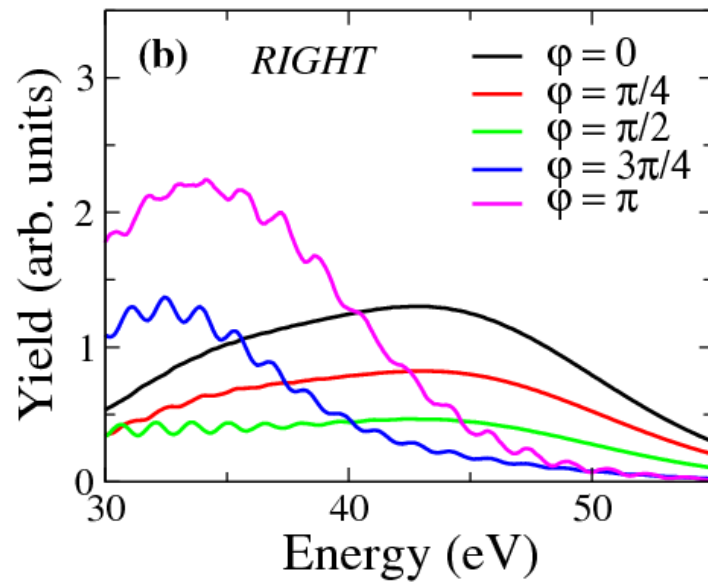
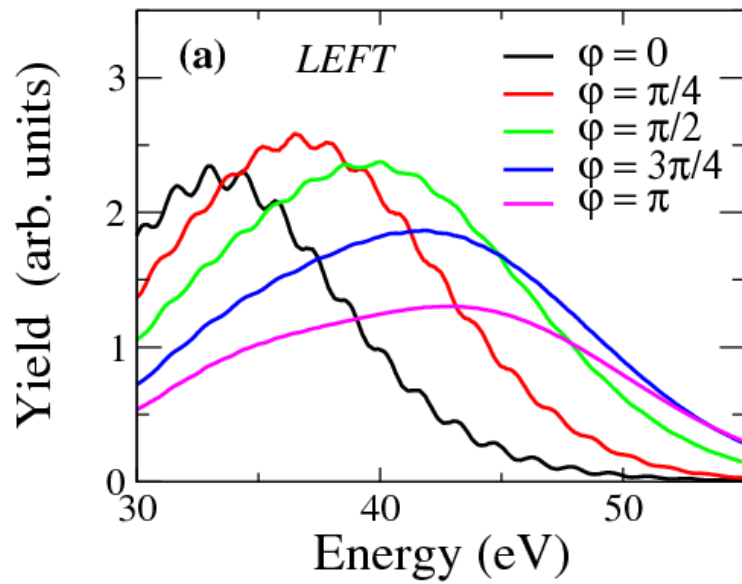


$$\bar{I}(\mathbf{p}_v) = \sigma(p_r, \theta_r) \bar{W}(p_r)$$

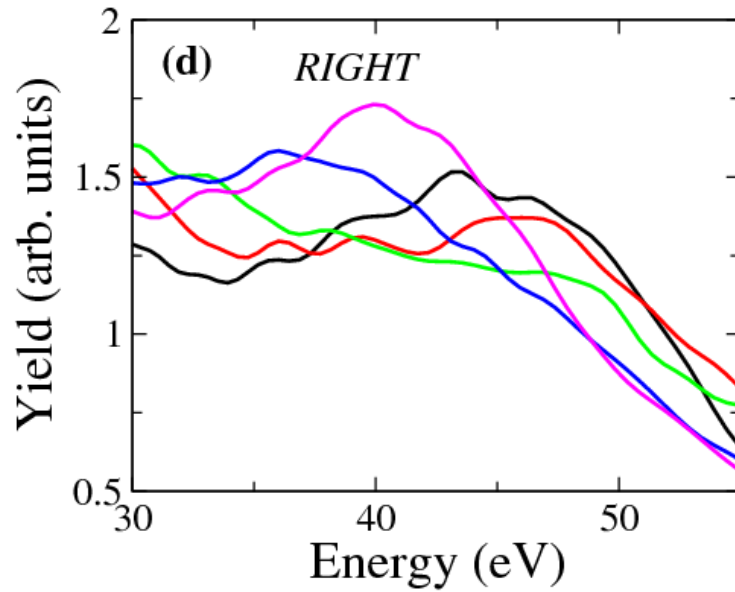
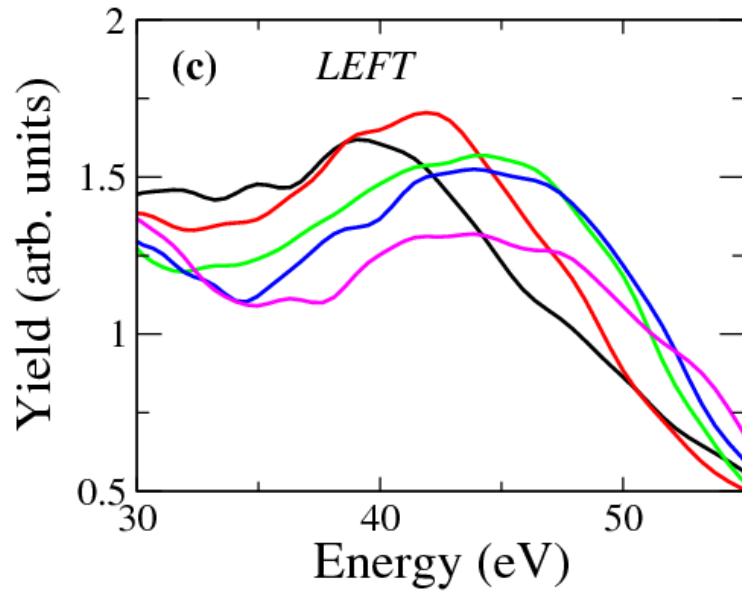
Exp. e-spectra

Volume-integrated w.p.

DCS can be extracted without knowing the laser parameters



theory



Exp.

Micheau et al PRL 2009 - not good for single-shot measurements

Energy moment of each single - shot

$$D(\mathbf{k}) = \sigma(k_r, \theta_r) W(k_r)$$

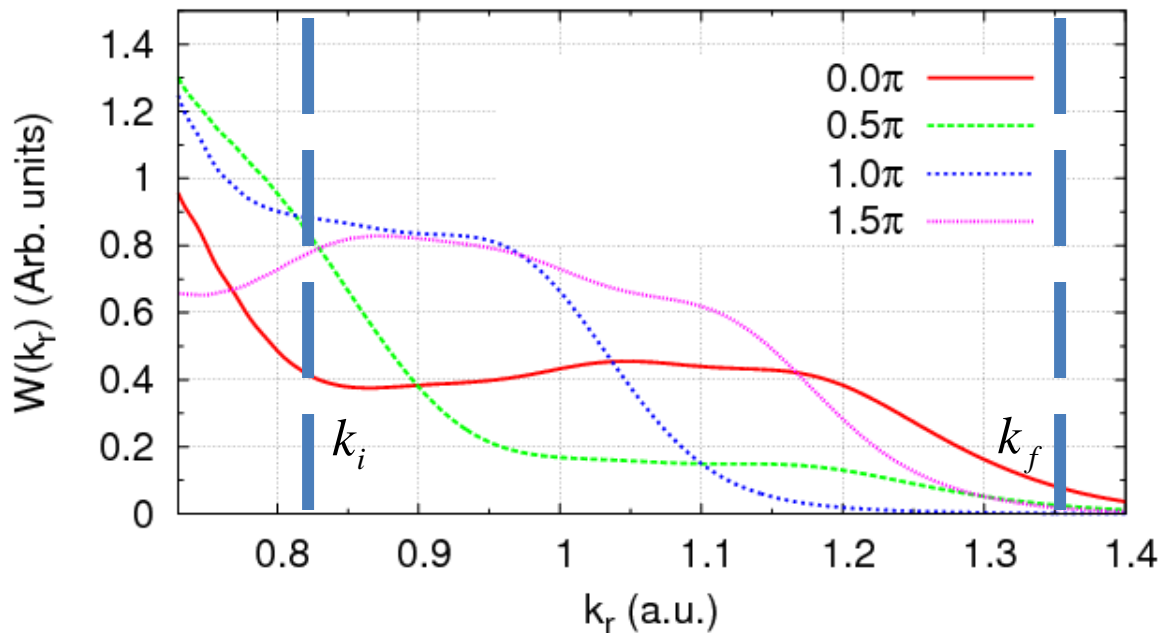
Moment:

$$M = \frac{\int_{k_i}^{k_f} (k_r^2 / 2) W(k_r) dk_r}{\int_{k_i}^{k_f} W(k_r) dk_r}$$

It reflects the cut-off position of the high energy spectra

It does not depend on target

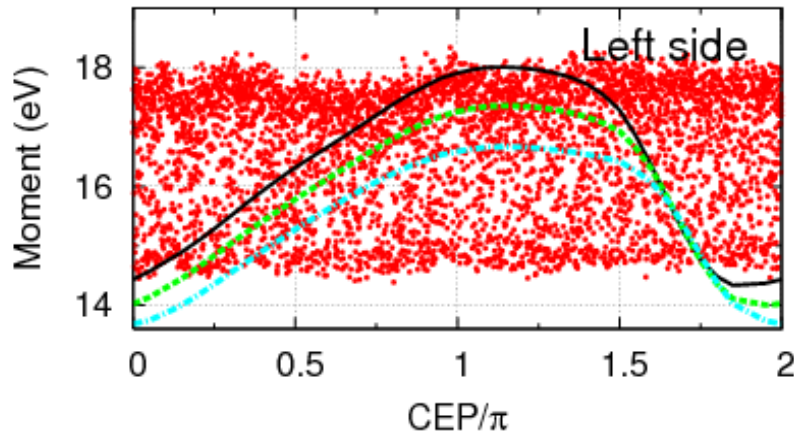
Volume integrated wave packets for different CEP



Retrieve Intensity

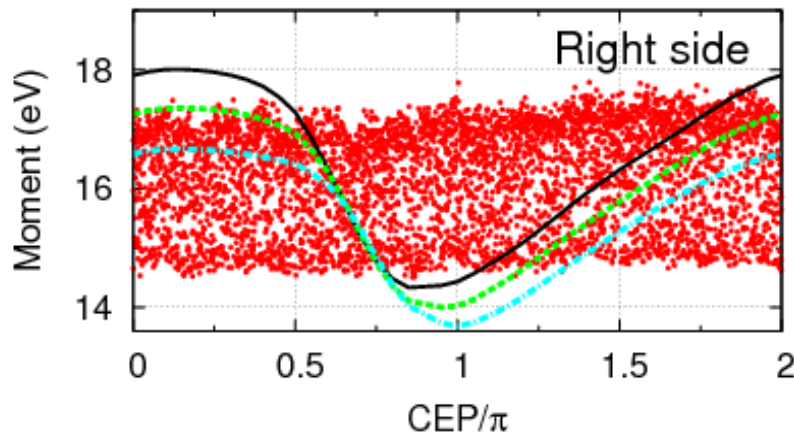
Comparison of the moments from experiment with theory

4500 shots for Xe @
760 nm



Experiment •
 1.4×10^{14} W/cm², 4.5 fs —
 1.3×10^{14} W/cm², 4.5 fs - - -
 1.2×10^{14} W/cm², 4.5 fs - · - ·

Average of moments over one cycle

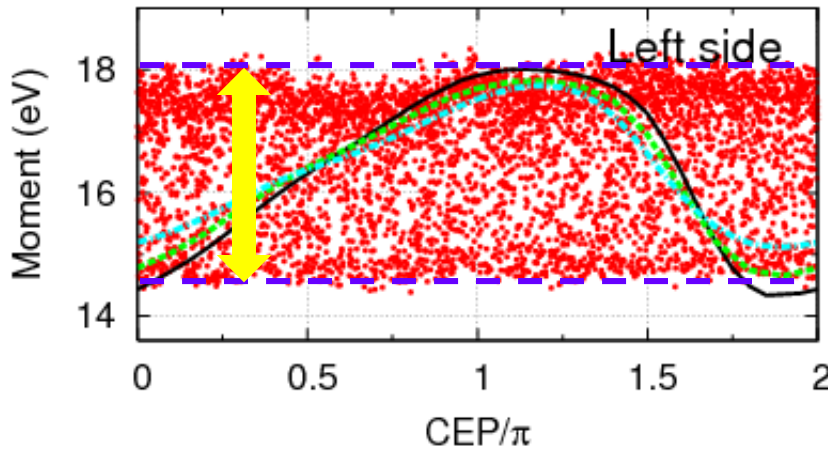


Experiment: 16.60 eV (Left)
16.20 eV (Right)

1.4×10^{14} W/cm²: 16.46 eV
 1.3×10^{14} W/cm²: 16.05 eV
 1.2×10^{14} W/cm²: 15.64 eV

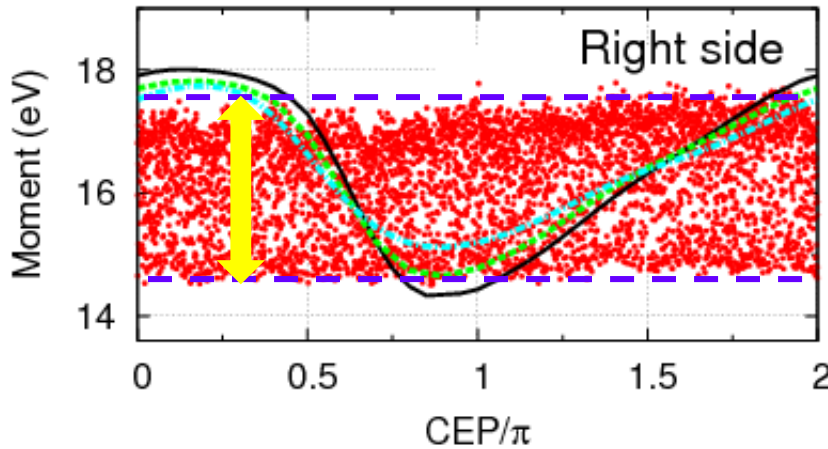
In theoretical simulations, the average of moments over one cycle does not depend on duration !!

Retrieve Duration



Experiment •
 1.4×10^{14} W/cm², 4.5 fs —
 1.4×10^{14} W/cm², 4.7 fs ⋯
 1.4×10^{14} W/cm², 5.0 fs - · - ·

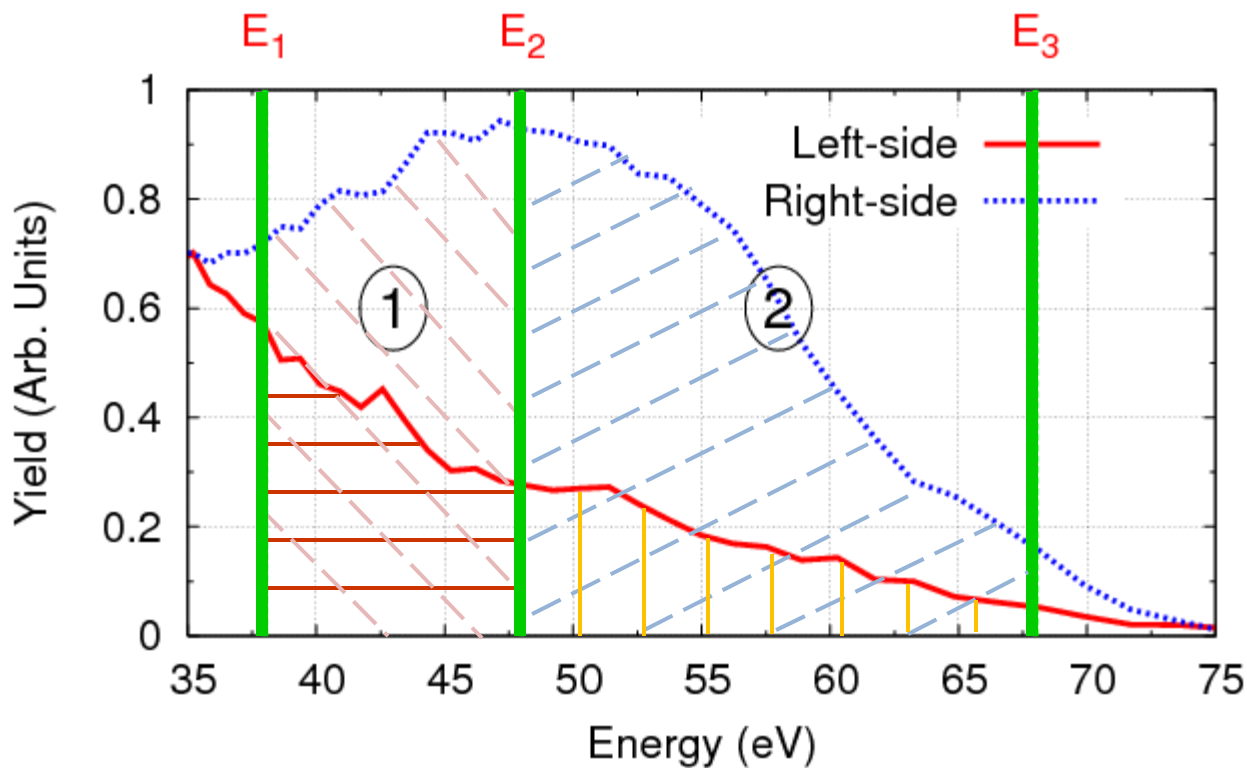
4.6fs



Experiment •
 1.4×10^{14} W/cm², 4.5 fs —
 1.4×10^{14} W/cm², 4.7 fs ⋯
 1.4×10^{14} W/cm², 5.0 fs - · - ·

4.8 fs

Retrieve the CEP

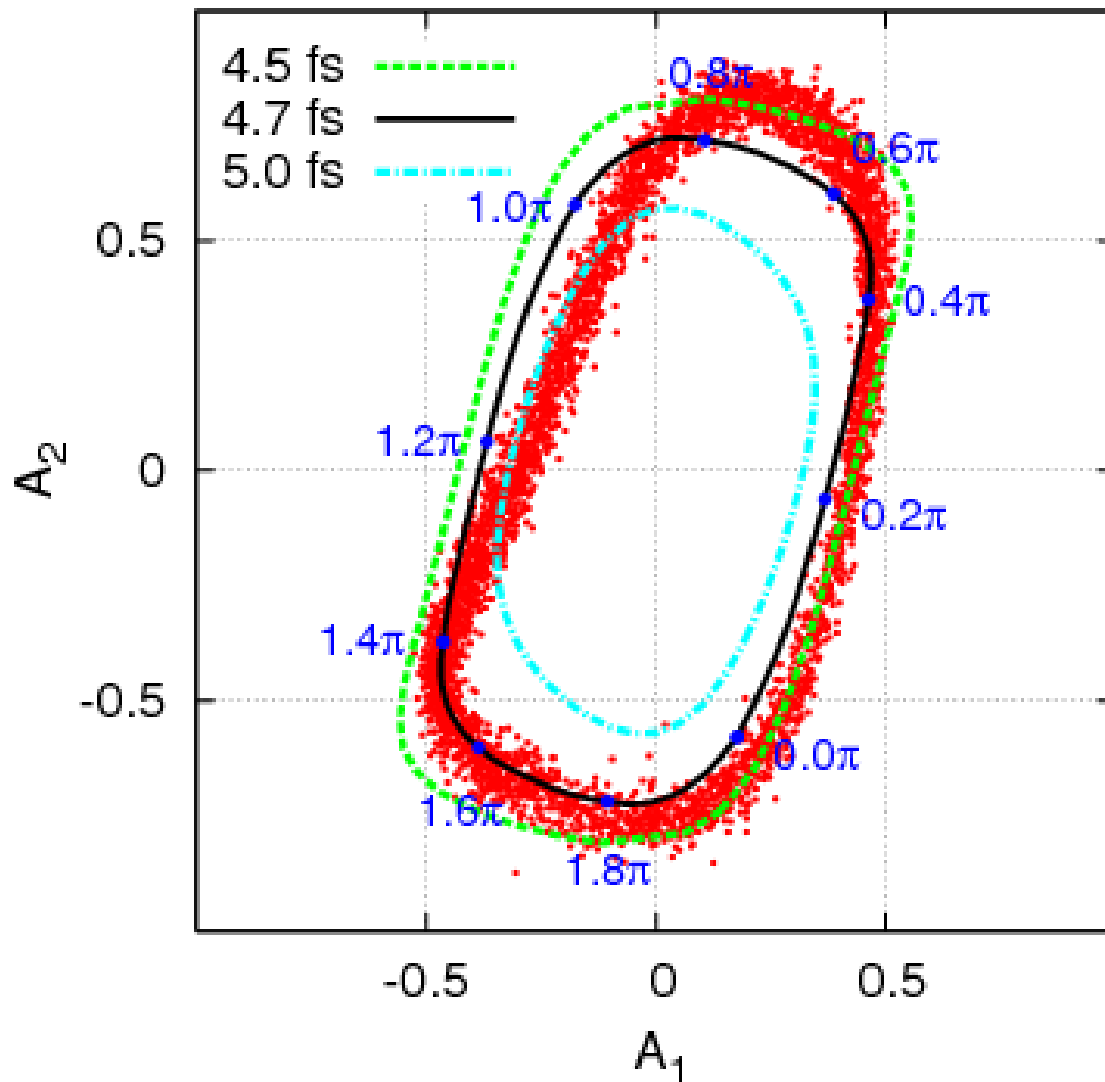


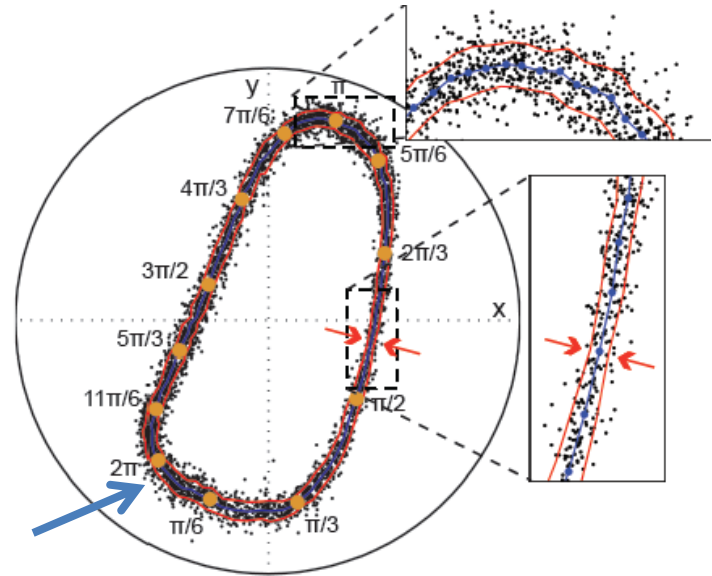
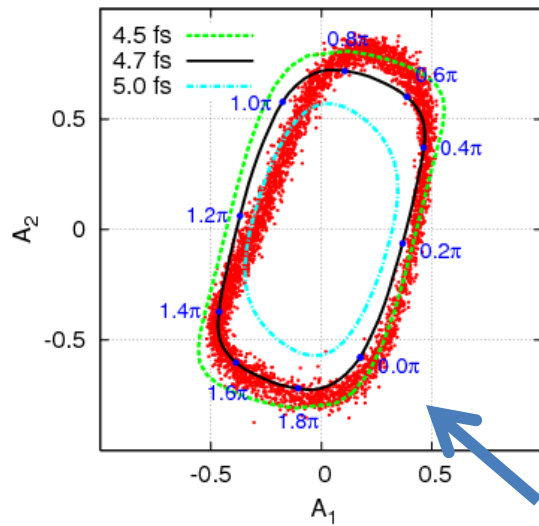
Phase asymmetry parameters:

$$A_1 = \frac{Y_L^1 - Y_R^1}{Y_L^1 + Y_R^1}, \quad A_2 = \frac{Y_L^2 - Y_R^2}{Y_L^2 + Y_R^2}$$

Retrieve the CEP

4500 shots





our method:

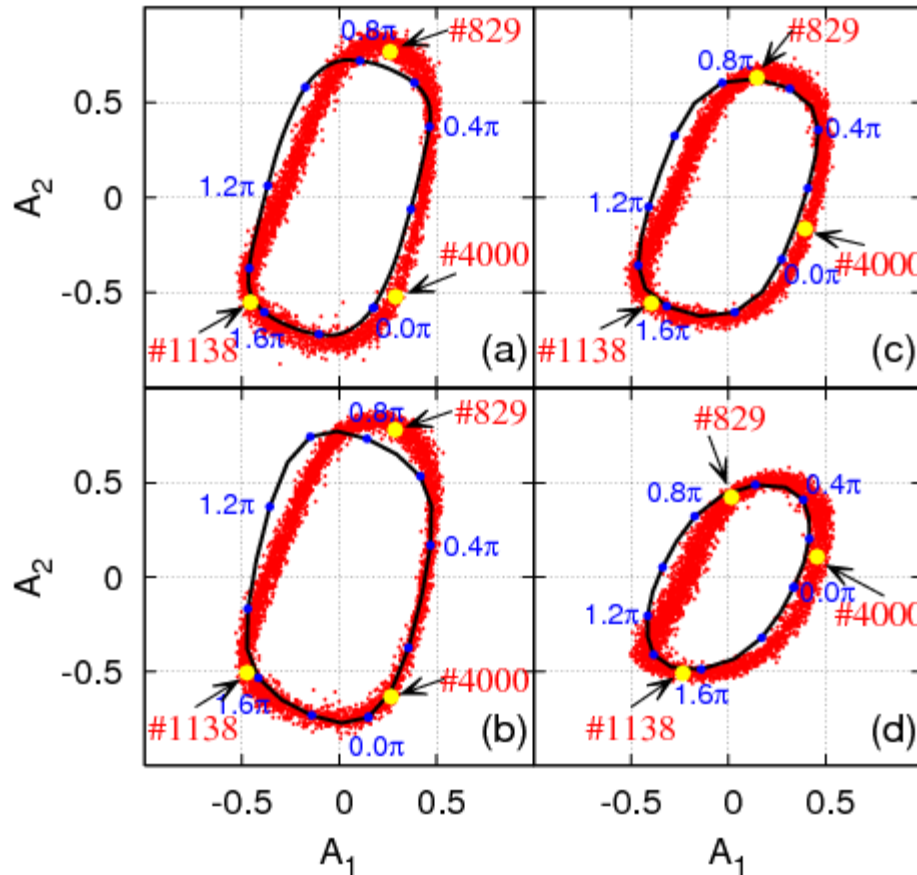
- **Accurate absolute CEP**
- **Pulse duration (4.7fs)**
- **Pulse peak intensity $1.4 \times 10^{14} \text{ W / cm}^2$**

- **All based on the left/right electron spectra**

Wittmann et al

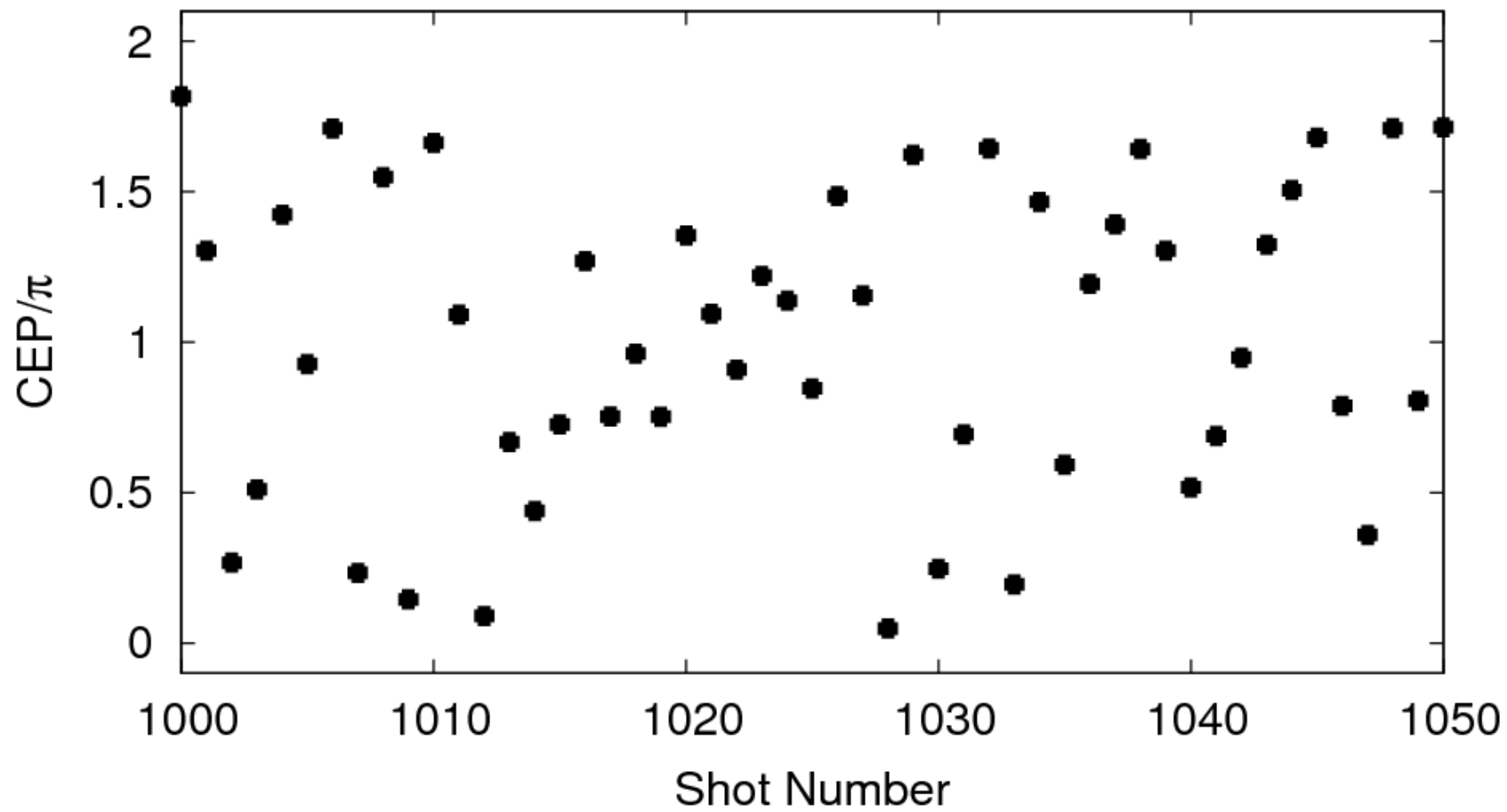
**error of 60 deg
estimated
estimated**

Results – Get CEP



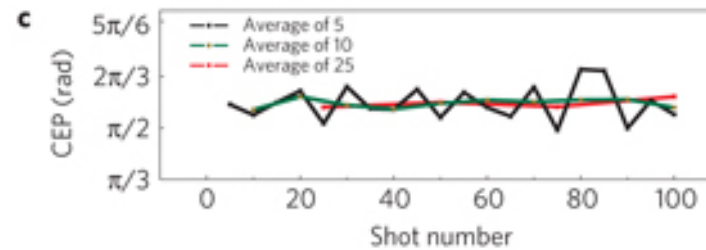
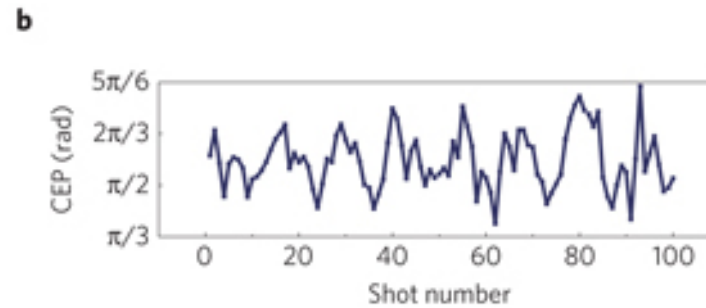
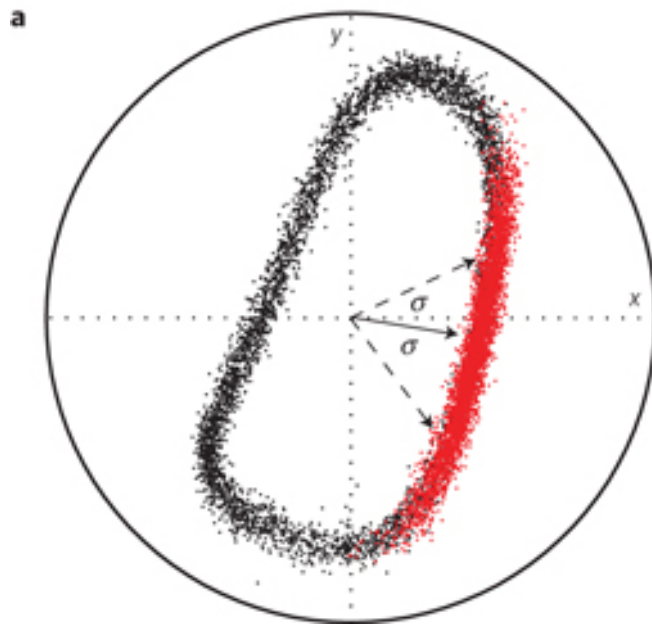
Shot#	1138	829	4000	2500
(a)	275 deg	129 deg	11 deg	196 deg
(b)	279 deg	126 deg	17 deg	199 deg
(c)	281 deg	131 deg	17 deg	196 deg
(d)	279 deg	121 deg	20 deg	200 deg

Random CEP in each single shot



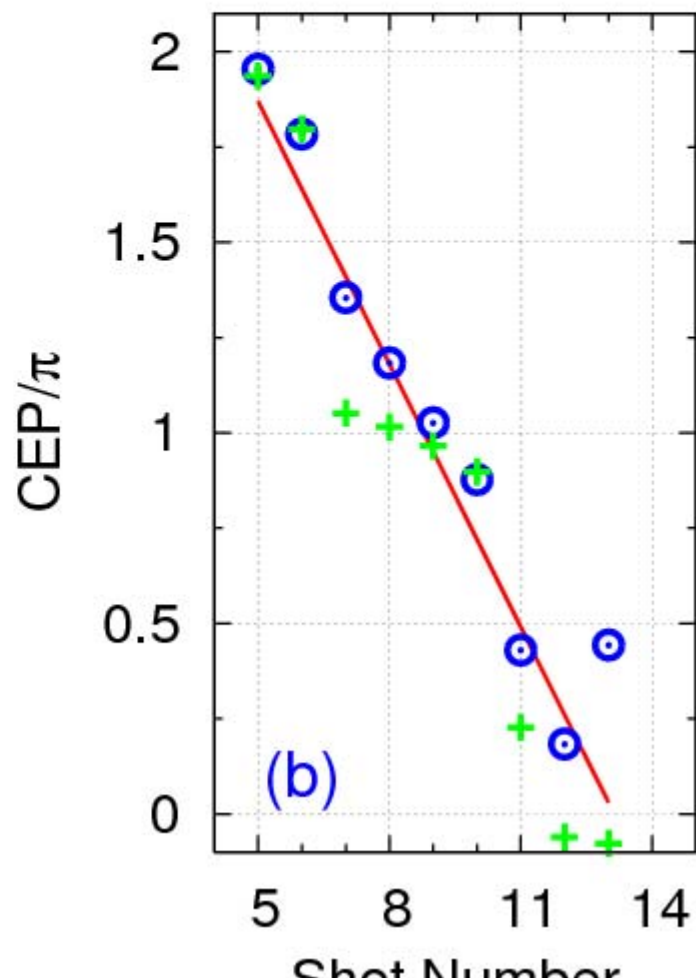
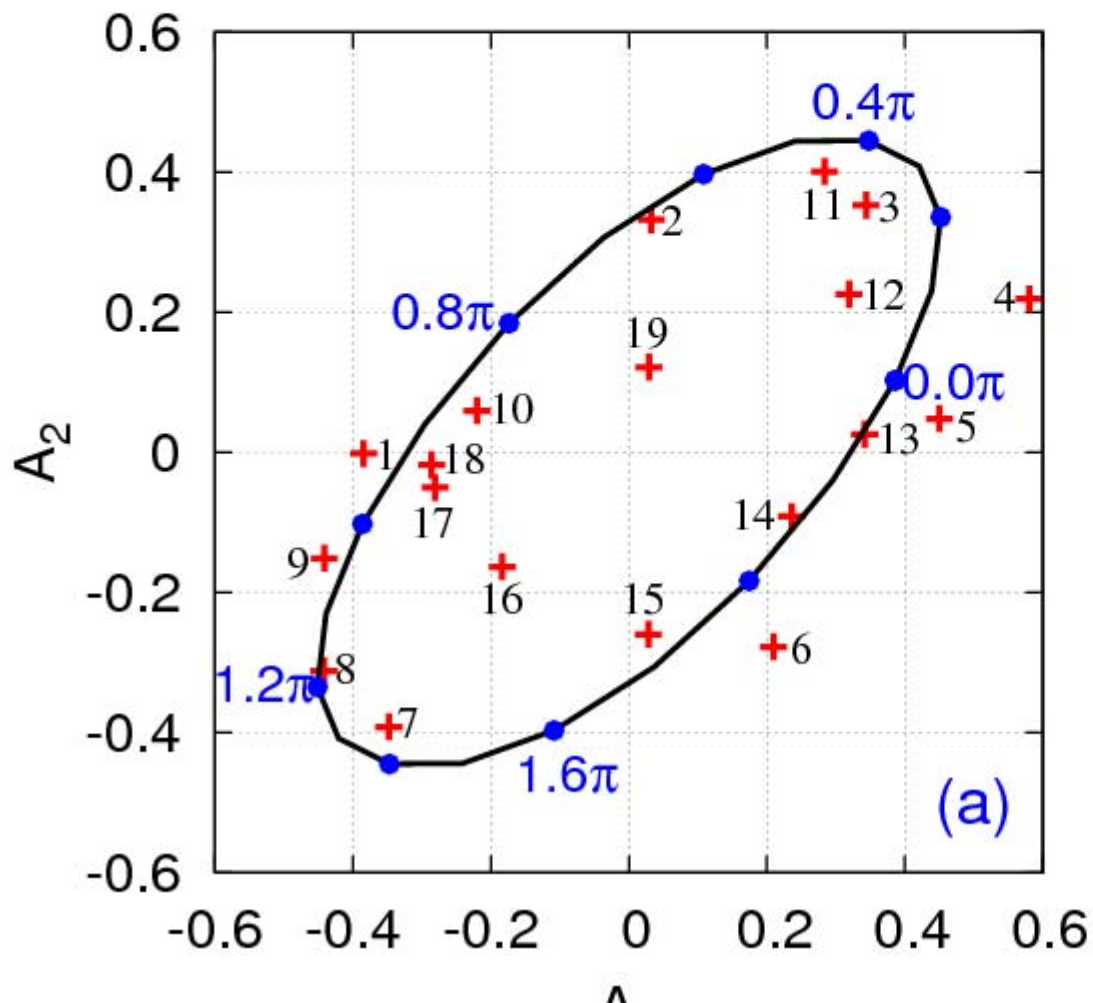
“CEP” of phase-stabilized laser pulses – from Wittmann et al

About 20 degrees



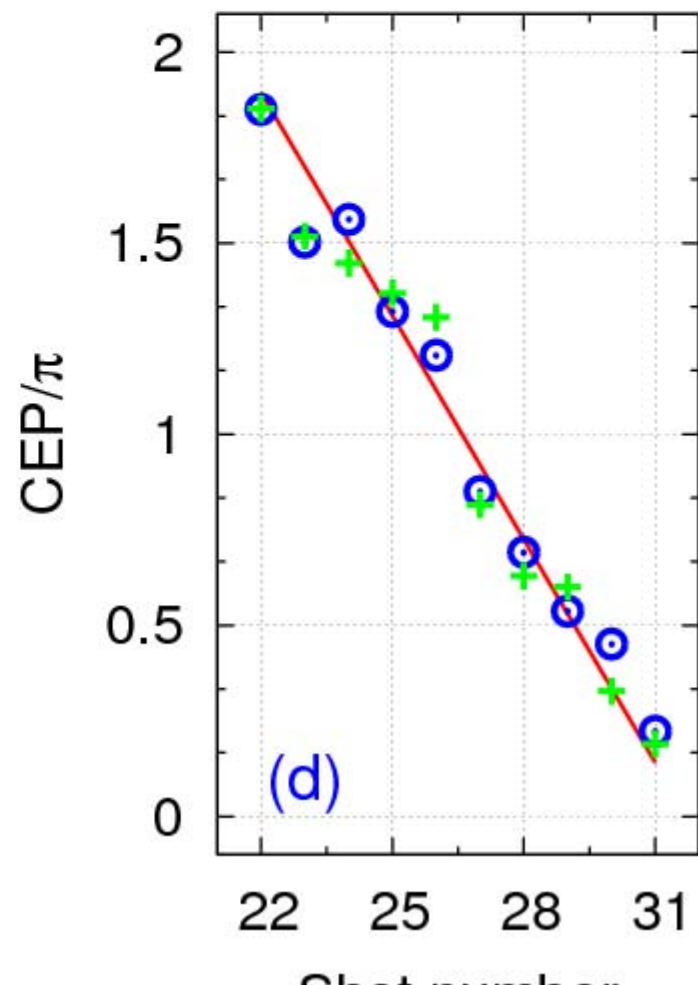
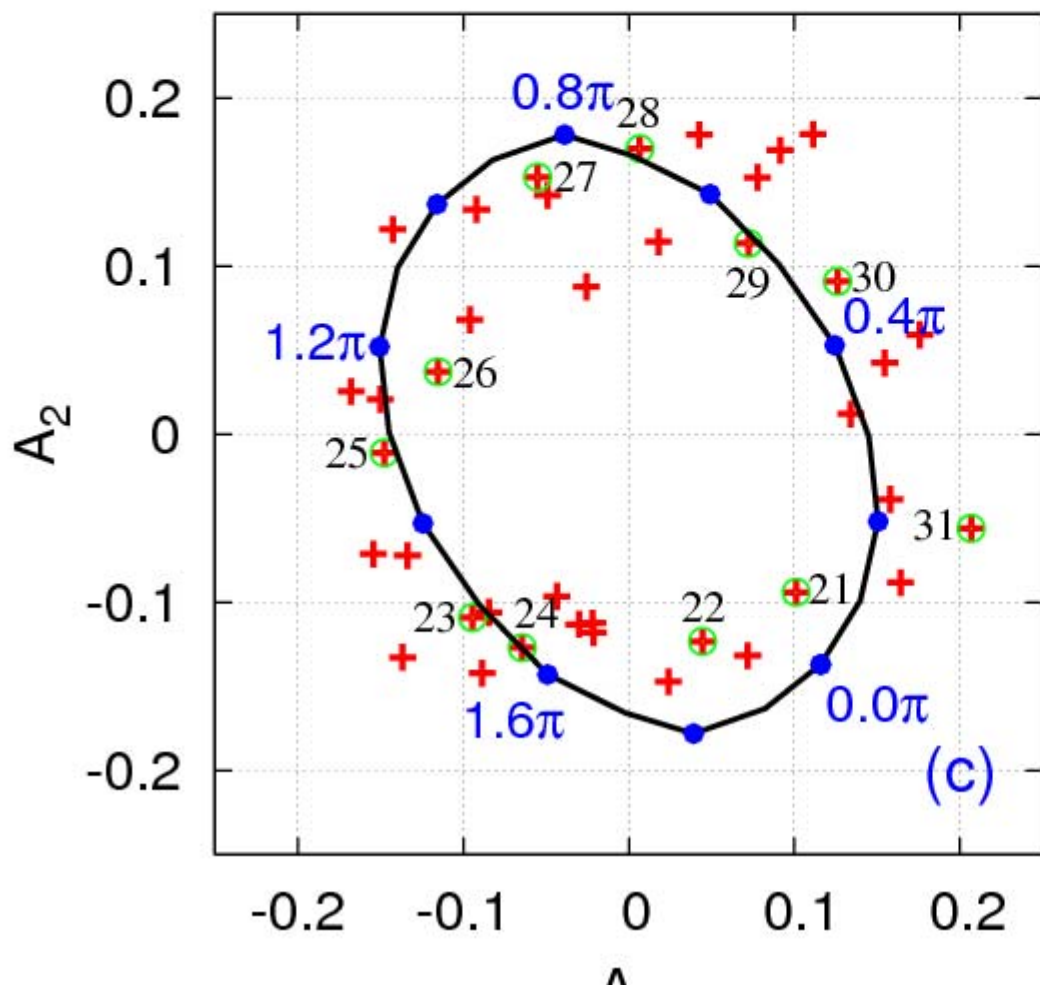
Applications— retrieving phases of CEP-stabilized pulses from multi-shot measurements

Znakovskya et al PRL 2009



For longer pulses, the ellipse is smaller

Data from Kling et al (2008) 7fs (instead of 6fs)



Retrieval of the Relative Phase in Two-Color Pulses

Zhangjin Chen & C D Lin
Dipanwita Ray & C L Cocke

Waveform of the two-color pulse

800 nm + 400nm

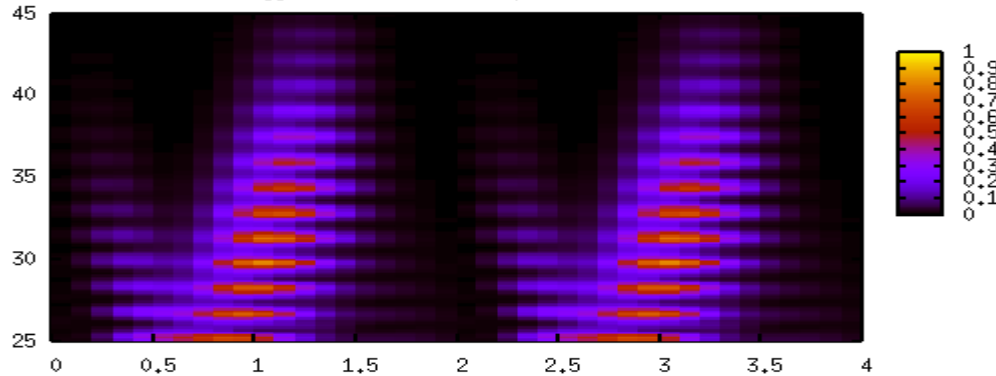
$$E(t) = E_0 \cos^2\left(\frac{\pi}{\tau}t\right) \left[\cos(\omega t) + \alpha \cos(2\omega t + \phi) \right]$$

$$U_P = \frac{E_0^2}{4\omega^2} \left[1 + \frac{\alpha^2}{4} \right]$$

Ratio

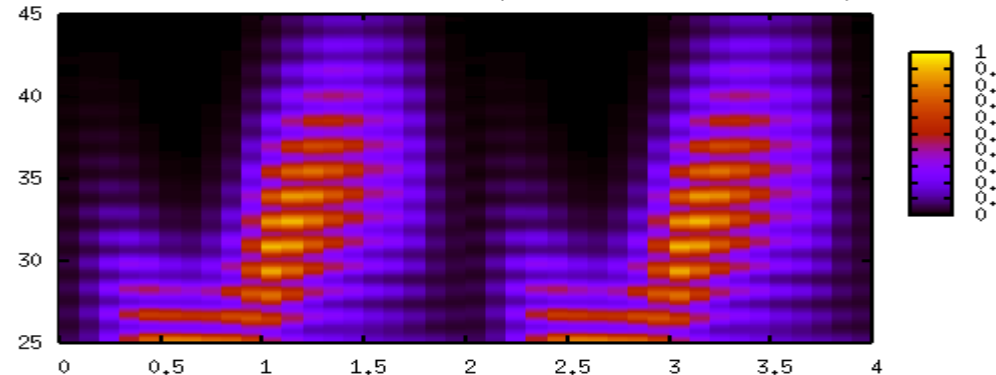
Relative phase

"Energy_0.50E14w800nm10fs_plot.dat" u 1:(\$2*27.2):(\$3*1.3e5)



QRS

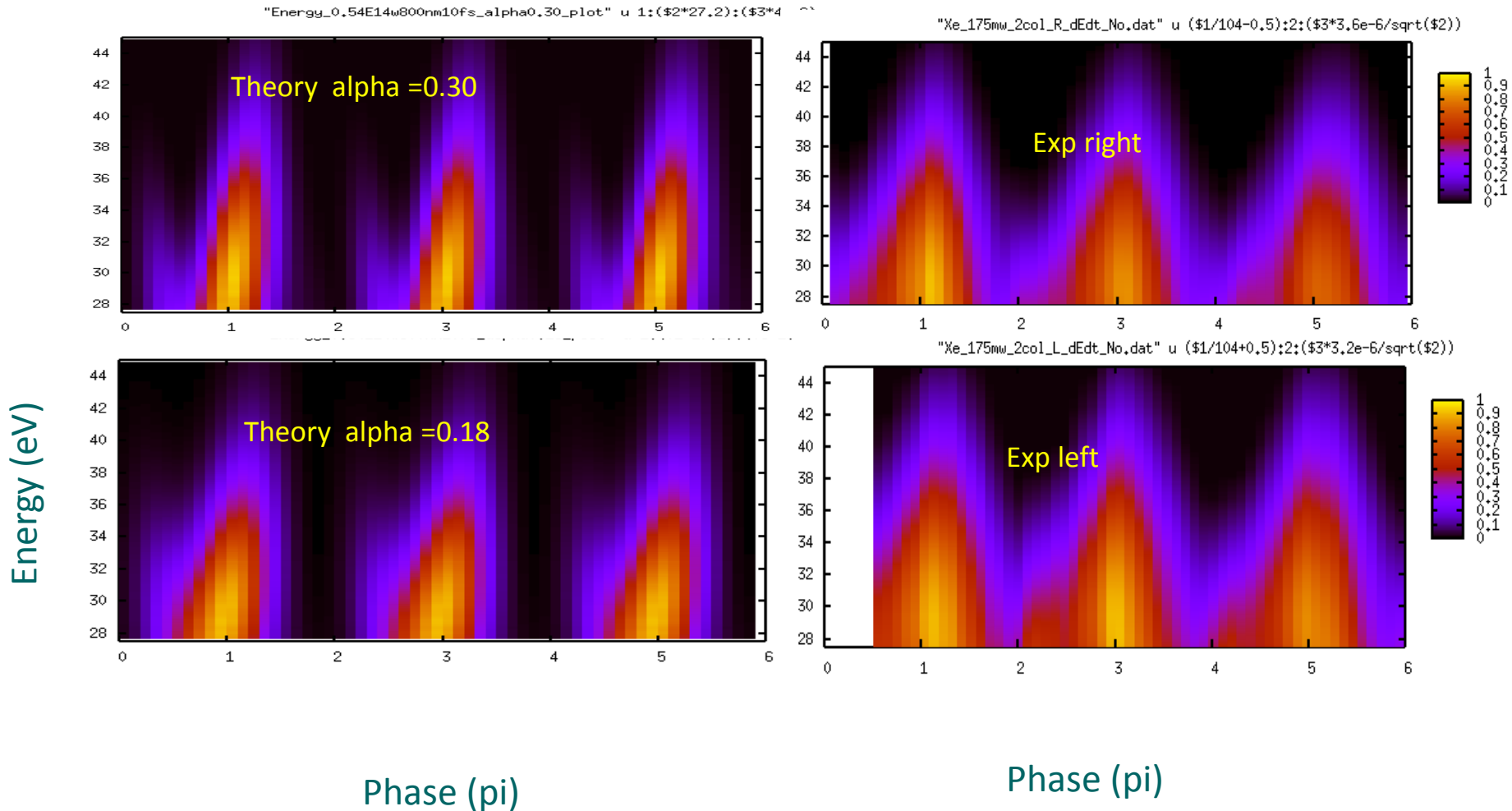
"IR10fs800nm0.50E14ratio0.3_2d_s_plot" u 1:(\$2*27.2):(\$3*20/sqrt(\$2))



TDSE

Single
intensity

Compare the QRS with experiment –Single ratio



Relative phase and relative intensity retrieved

Summary--

Based on the **wave packet** from the **QRS** theory—

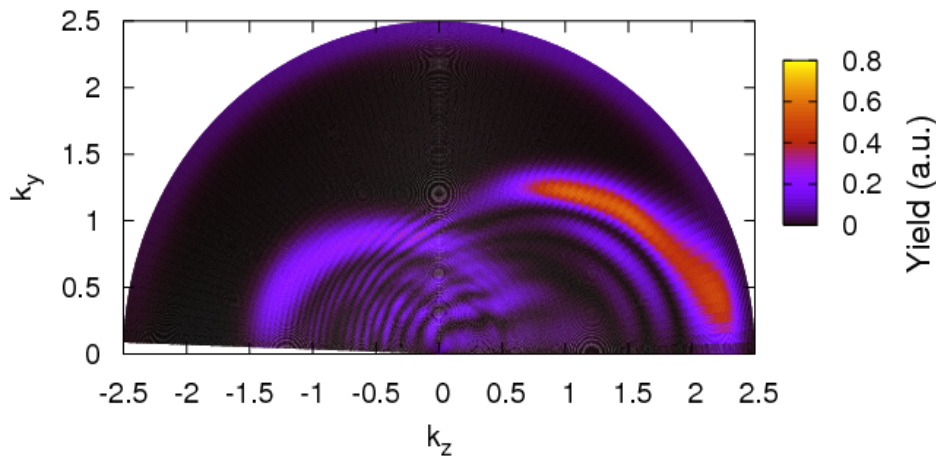
1. Can retrieve CEP, pulse duration and peak intensity quickly and accurately from high-energy ATI electron spectra
2. Can retrieve relative phase and relative peak intensity in two-color measurements

Not discussed— the **elastic scattering cross sections** (independent of lasers) derived from QRS can be used for structural imaging of the target

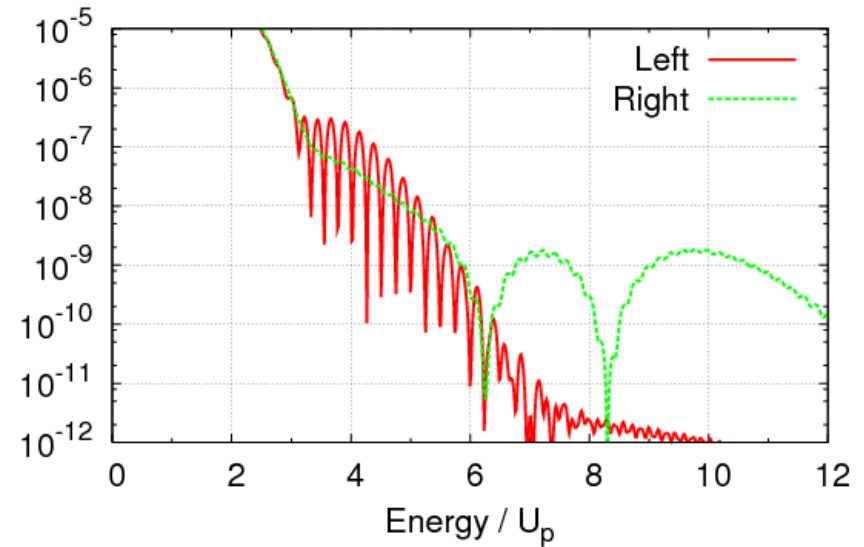
QRS for **HHG** from molecules— A. T. Le 's talk

CEP dependence of the high energy photoelectron spectra

High energy 2D momentum distribution



Left and right high energy spectra



$$E(t) = E_0 a(t) \cos(\omega t + \varphi)$$

Hydrogen:

$1.0 \times 10^{14} \text{ W/cm}^2$,

800 nm,

3 optical cycle

