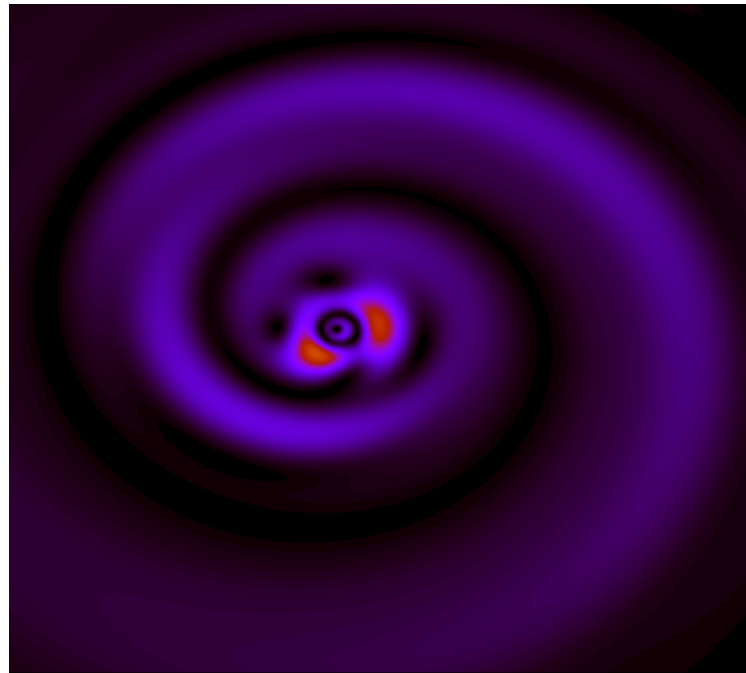


Coherent Control and Attosecond Dynamics with Pulsed XUV and IR Radiation

Missouri University of Science & Technology,
March 21, 2019

Klaus Bartschat

Drake University, Des Moines, IA 50311, USA



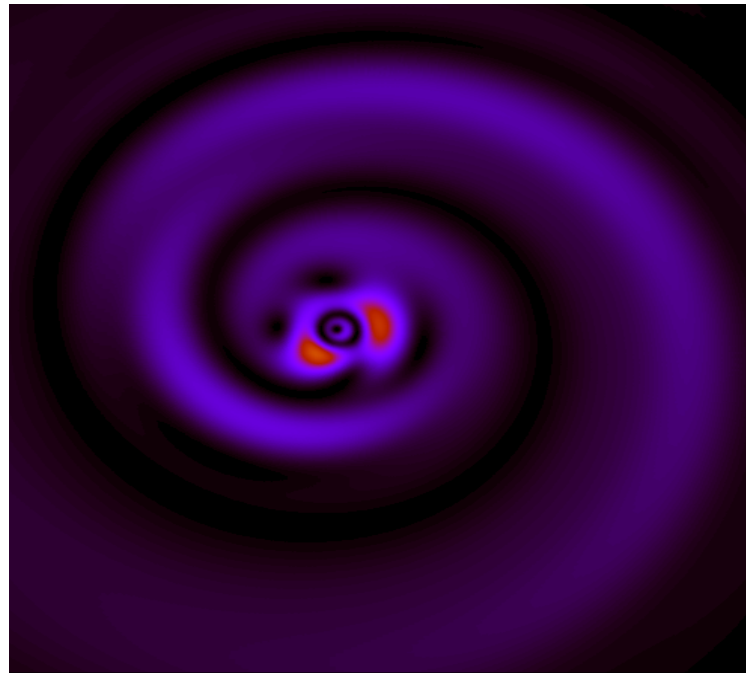
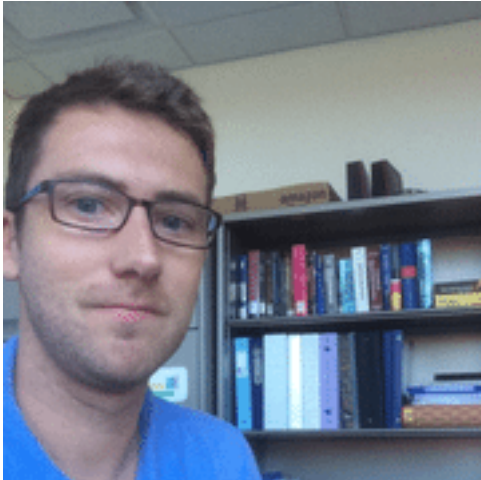
XSEDE
Extreme Science and Engineering
Discovery Environment

NSF support under PHY-1430245, PHY-1803844, and XSEDE-090031

Coherent Control and Attosecond Dynamics with Pulsed XUV and IR Radiation

more appropriate: **Nicolas Douguet** and Klaus Bartschat

Drake University, Des Moines, IA 50311, USA



XSEDE

Extreme Science and Engineering
Discovery Environment

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Overview of the Talk

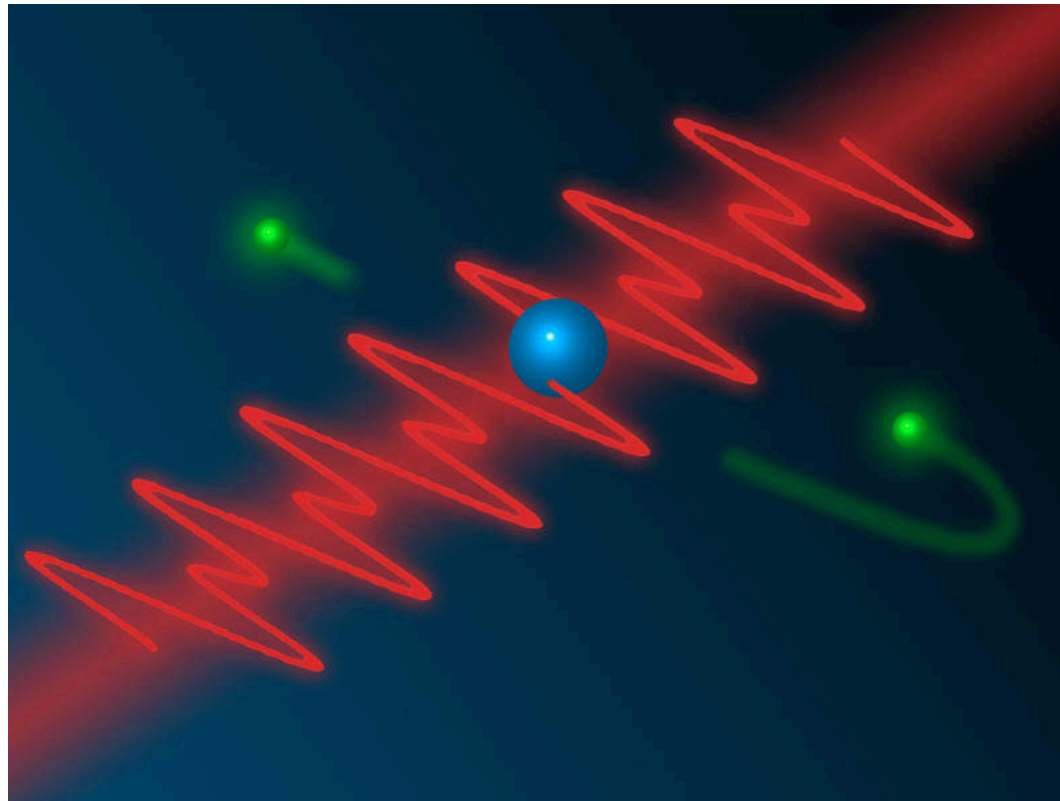
1. Light-induced Coherent Quantum Control

- (a) Interfering **one-photon** and **two-photon** ionization by XUV femtosecond pulses.
- (b) Overlapping XUV pulses with an **optical field** (XUV + IR).
- (c) Using **circularly polarized** XUV femtosecond pulses.

2. Multiphoton and Tunneling Ionization

- (a) **Circular dichroism** in two-color resonant multiphoton ionization of oriented He^+ .
- (b) Attoclock measurements of **tunneling time**.
- (c) Interpretation using **Bohmian Mechanics**

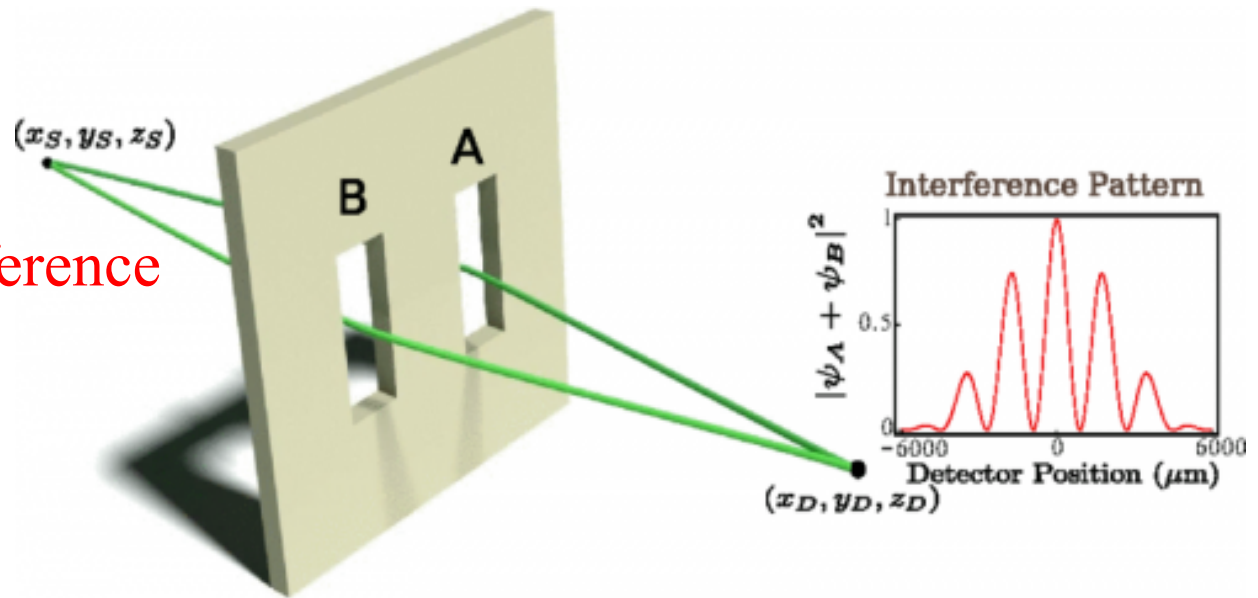
Light-induced Coherent Quantum Control



Motivation

- One of the goals of “quantum control” is to steer electrons into specific directions or locations (e.g., selected bond breaking in a molecule).

- Two-pathway interference is a way to achieve coherent control.

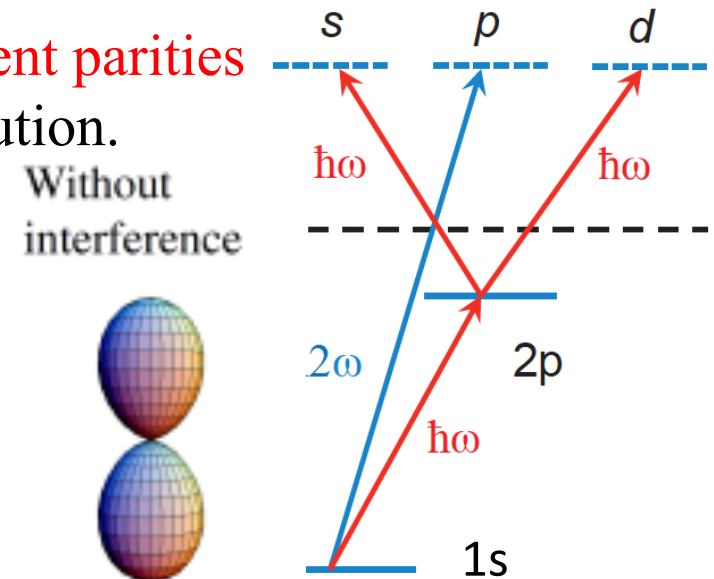
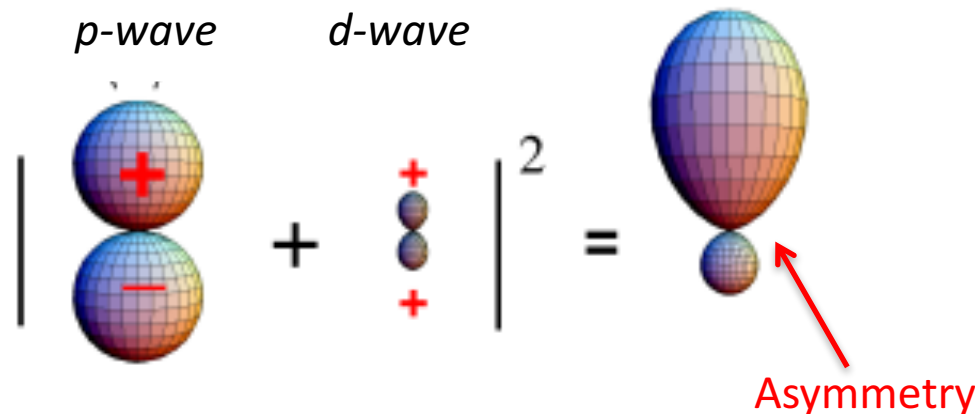
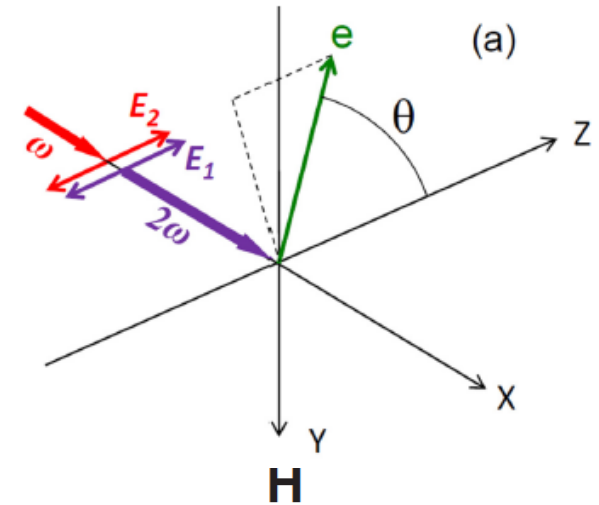


- Photoionization of an atomic system by the fundamental and the second harmonic ($\omega + 2\omega$) of a femtosecond VUV pulse is an example of coherent control of the photoelectron angular distribution.

Bichromatic Atomic Ionization with Linearly Polarized Light

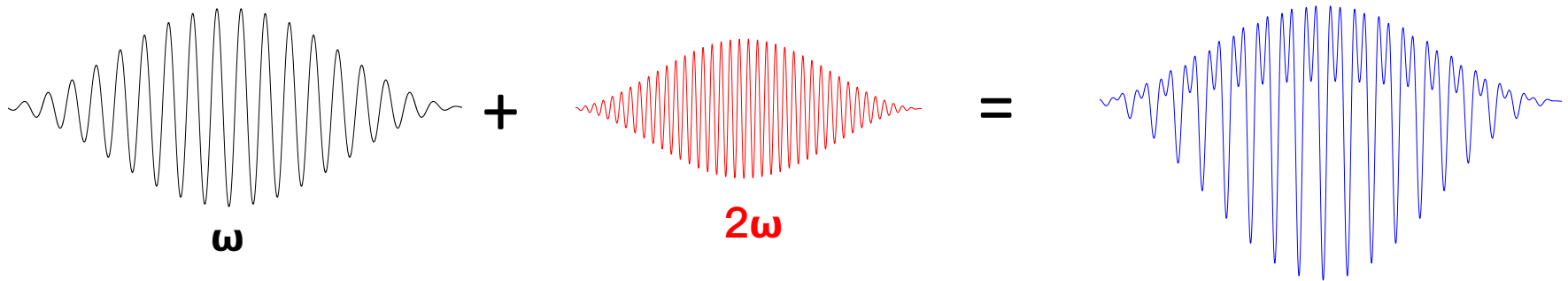
- In the case of linearly polarized light, the electric field is expressed as

$$E(t) = F(t) [\cos \omega t + \eta \cos(2\omega t + \phi)]$$
- Two-pathway interference** is enhanced by tuning the first harmonic near an intermediate state (e.g., 2p in H).
- Ionization leading to partial waves **with different parities** **can** cause an asymmetry in the angular distribution.



Control of the Photoelectron Angular Distribution (PAD)

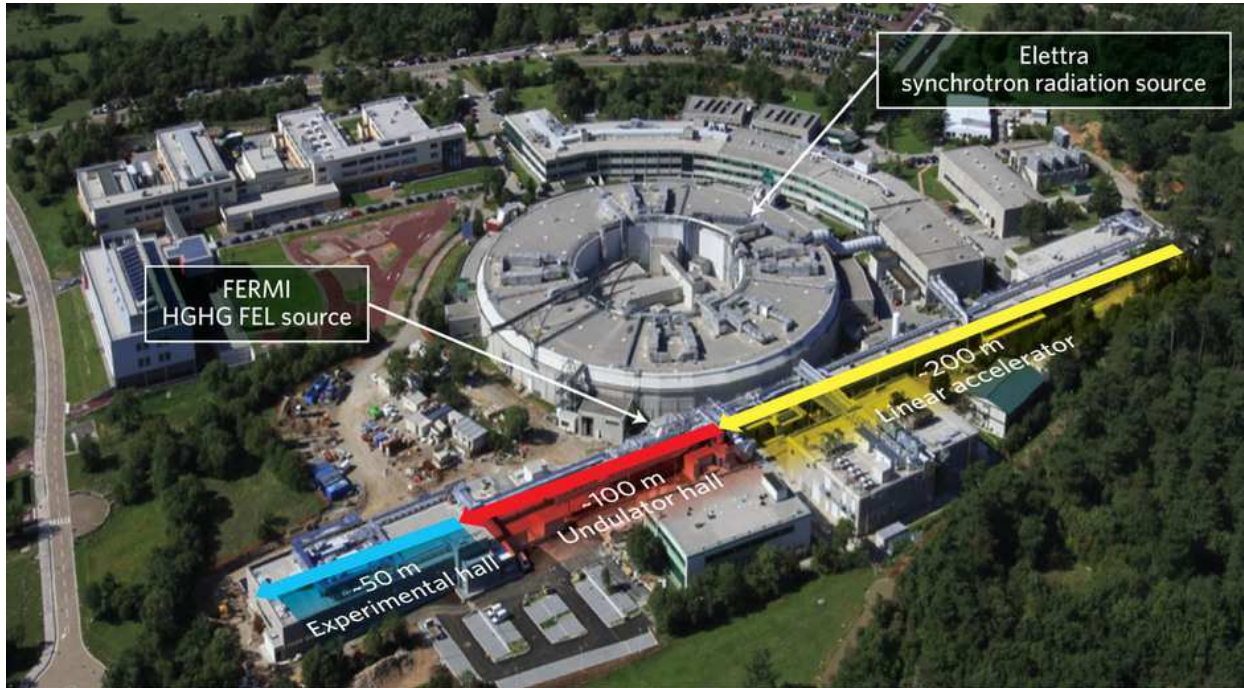
- The asymmetry in the PAD is the result of $\langle E^3 \rangle \neq 0$ of the electric field [N. B. Baranova and B. Ya. Zel'dovich, J. Opt. Soc. Am. B **8** 27 (1990)].



- The PAD takes the form:
$$W(\theta) = \frac{W_0}{4\pi} \left(1 + \sum_k \beta_k P_k(\cos \theta) \right)$$
- The **odd-rank anisotropy** parameters are responsible for the PAD asymmetry.
- The asymmetry is defined as:

$$A(0) = \frac{W(0) - W(\pi)}{W(0) + W(\pi)} = \frac{\sum_{k=1,3,\dots} \beta_k}{1 + \sum_{k=2,4,\dots} \beta_k}$$

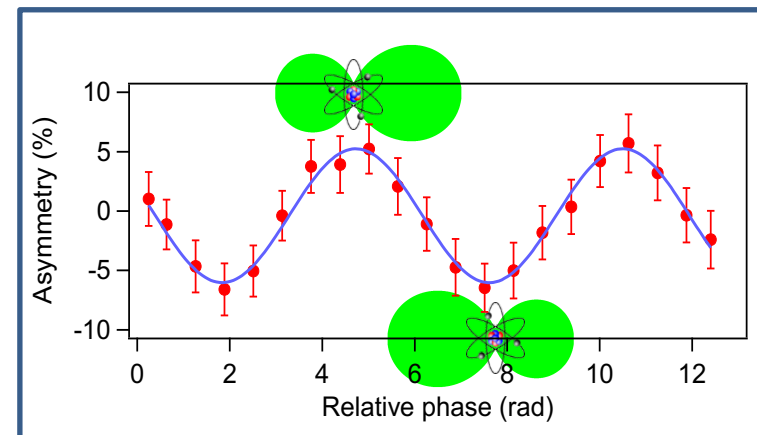
Experimental Setup at FERMI (Trieste, Italy)



Basic idea: Use $\text{Ne}(2p^6)$ as target and tune the fundamental to one of the $(2p^5 4s)_{J=1}$ states.

Results: The delay between the two pulses was controlled to a precision better than **3.1 attoseconds**. This is equivalent to controlling the phase ϕ to high precision [Prince *et al.*, Nat. Phot. **10** (2016) 176-179]

→ The asymmetry oscillates as a function of ϕ as predicted theoretically.



Numerical Approach

- We solve the **Time-Dependent Schrödinger Equation (TDSE)** in the Single-Active Electron (SAE) approach:

$$\hat{H}(\mathbf{r}, t)\Psi(\mathbf{r}, t) = \left(-\frac{\nabla^2}{2} + V(r) + \sqrt{\frac{4\pi}{3}} r \sum_{q=\pm 1} \mathcal{E}_q^*(t) Y_{1q}(\theta, \varphi) \right) \Psi(\mathbf{r}, t) = i \frac{\partial \Psi(\mathbf{r}, t)}{\partial t}$$

- The wavefunction is expanded in spherical harmonics. We solve the system of coupled equations using **finite differences, split-operator method, series expansion, Crank-Nicolson, matrix inversion, ...**, in both the length and velocity forms of the electric dipole operator, ...

Numerical Approach

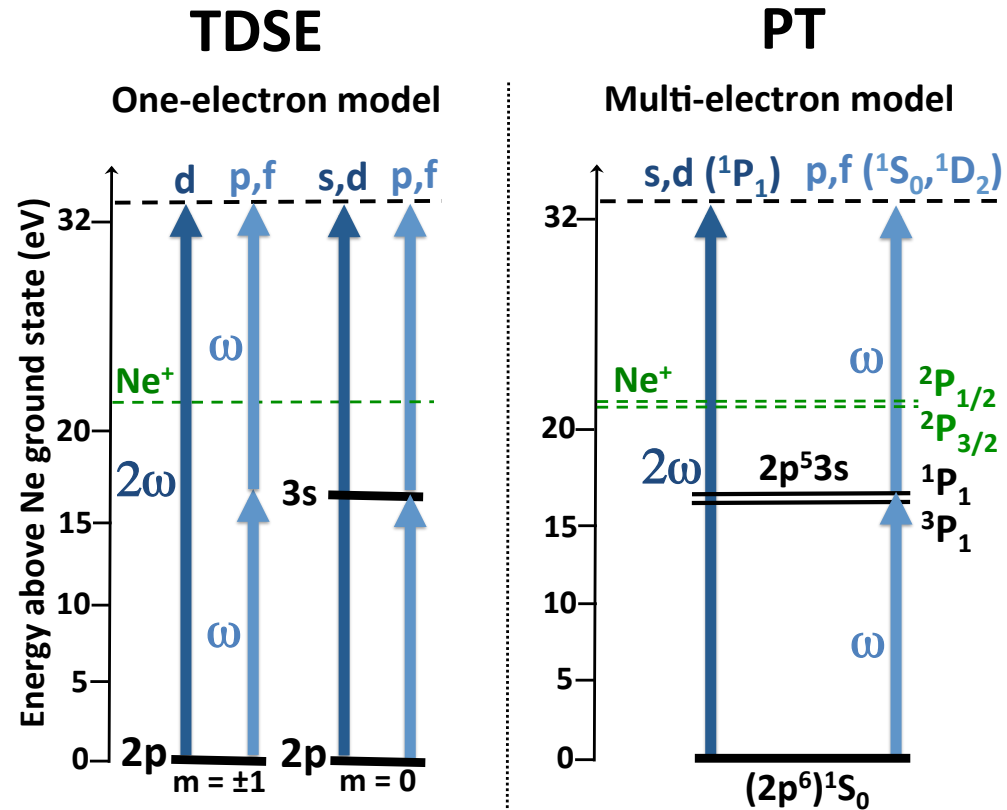
- We solve the **Time-Dependent Schrödinger Equation (TDSE)** in the Single-Active Electron (SAE) approach:

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- The wavefunction is expanded in spherical harmonics. We solve the system of coupled equations using **finite differences, split-operator method, series expansion, Crank-Nicolson, matrix inversion, ...**, in both the length and velocity forms of the electric dipole operator, ...
- The numerical issues are by no means trivial, and we spent a lot of time to ensure stability, accuracy, and efficiency.
- Our colleagues at Moscow State University (A.N. Grum-Grzhimailo, E.V. Gryzlova, E.I. Staroselskaya) use time-dependent **Perturbation Theory (PT)** to obtain the anisotropy parameters calculating the first-order (**one-photon absorption**) and second-order (**two-photon absorption**) ionization amplitudes.

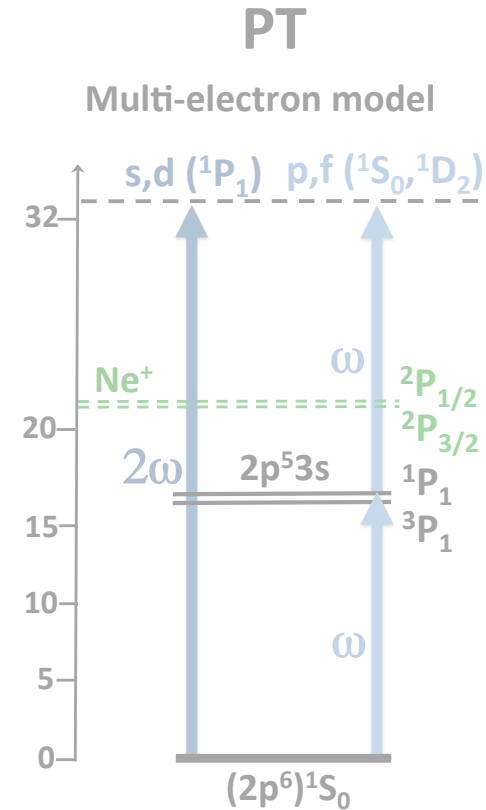
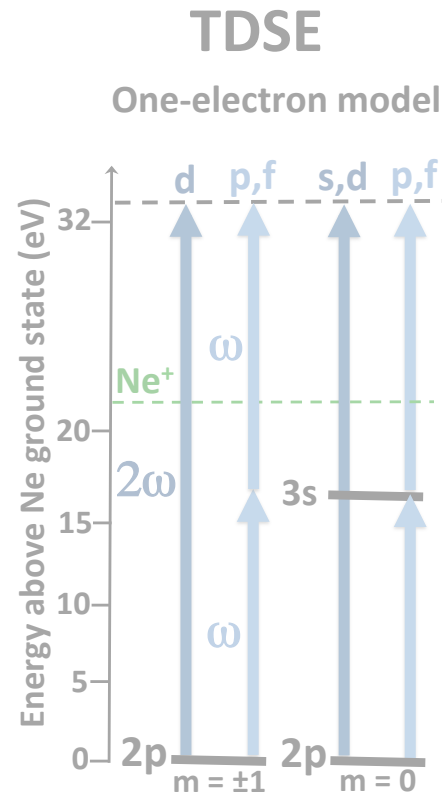
Using $2p^5 3s$ as Intermediate States (easier)

- We use the two $(2p^5 3s)$ $J=1$ states as stepping stones to enhance two-photon absorption.
- The TDSE calculations employ a **one-electron** model (no fine-structure), whereas PT uses a **multi-electron model**.
- LS -coupling \rightarrow Only one state can be significantly excited.
- Using PT we can obtain analytical expressions for the angular distribution and the anisotropy parameters β_1 , β_2 , β_3 , and β_4 . This allows us to scan the parameter space efficiently



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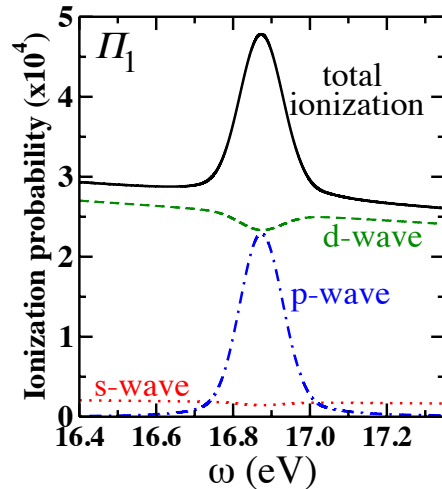


- Using PT we can obtain analytical expressions for the angular distribution and the anisotropy parameters β_1 , β_2 , β_3 , and β_4 . This allows us to scan the parameter space efficiently.
- **Consequently, it is very important to know whether PT is reliable.**

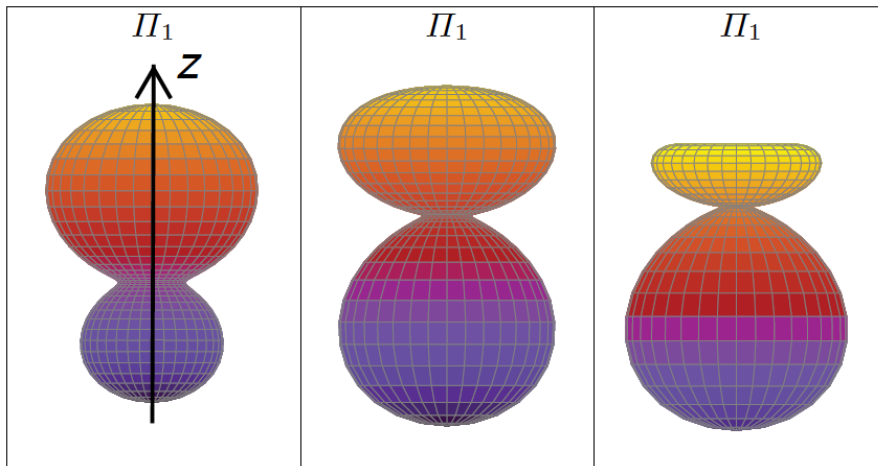
Theoretical Predictions

- We consider pulses of the form $E(t) = F(t) [\cos \omega t + \eta \cos(2\omega t + \phi)]$ with sine-squared pulse envelope $F(t)$ and fundamental peak intensity $I = 10^{12} \text{ W/cm}^2$.

Partial-wave
ionization
probability



3D PAD



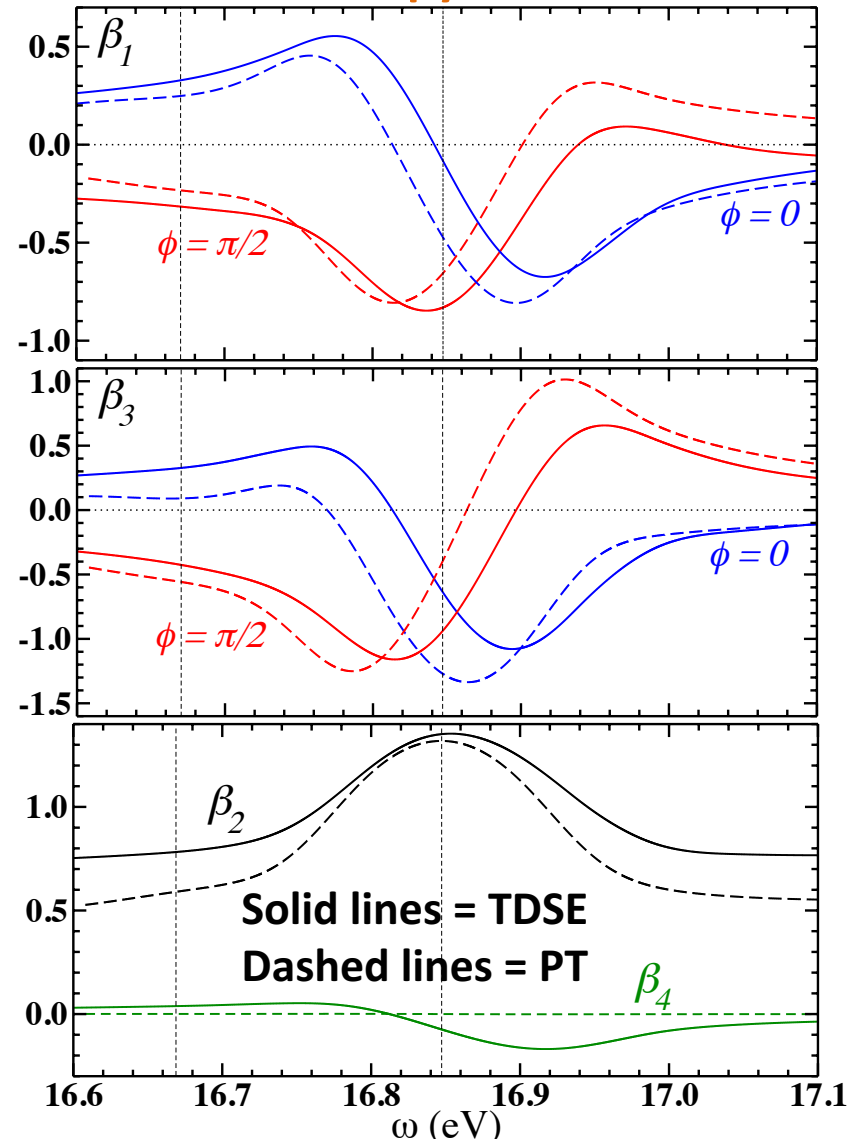
$\omega = 16.81 \text{ eV}$

$\omega = 16.85 \text{ eV}$

$\omega = 16.88 \text{ eV}$

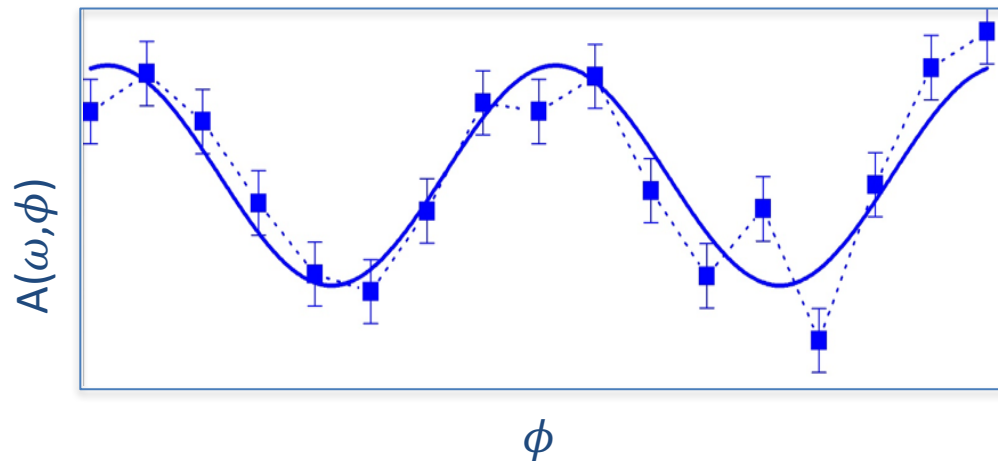
N. Douguet *et al.*, EPJD **72** (2017) 105

Anisotropy Parameters



Using $2p^54s$ as Intermediate States (tough)

- Experimentally, the two $(2p^54s)$ $J=1$ states were used as intermediate states. This complicates the situation due to:
 - 1) Strong mixture of triplet and singlet in the $4s$ and $4s'$ states.
 - 2) Presence of the $3d$ states in the vicinity and close-lying to the continuum.
- The maximum amplitude and associated phase of the asymmetry were determined by fitting the data to $A(\omega, \phi) = A_{\max}(\omega) \cos(\phi - \phi_{\max}(\omega))$

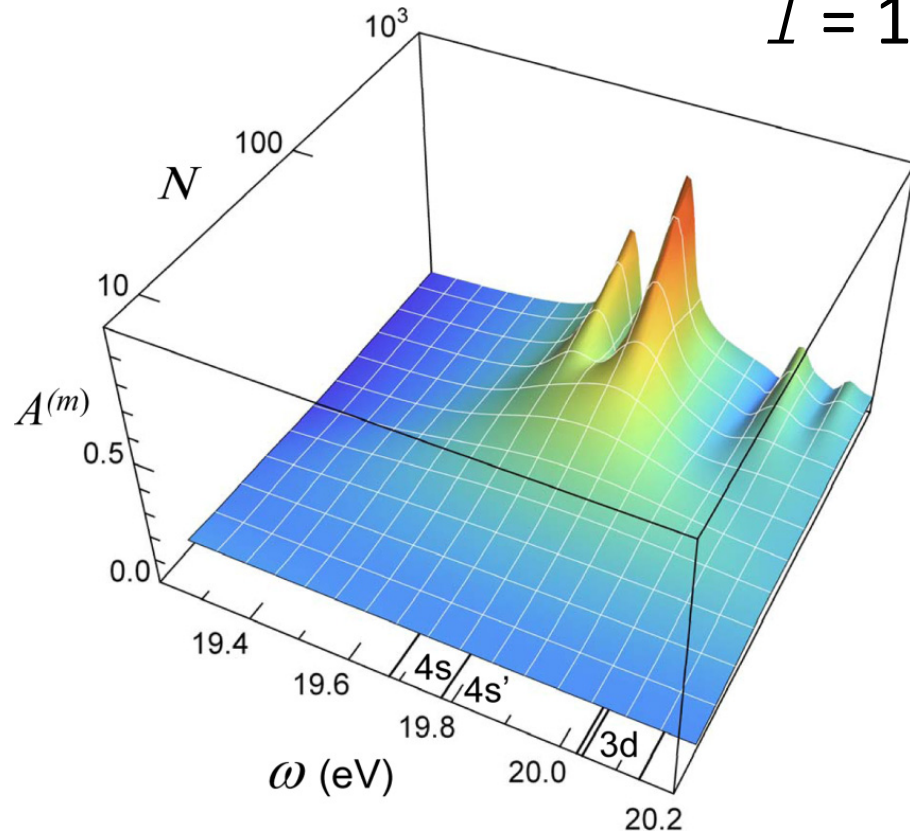


E. Gryzlova *et al.*, in preparation (2019)
G. Sansone *et al.*, “private communication”

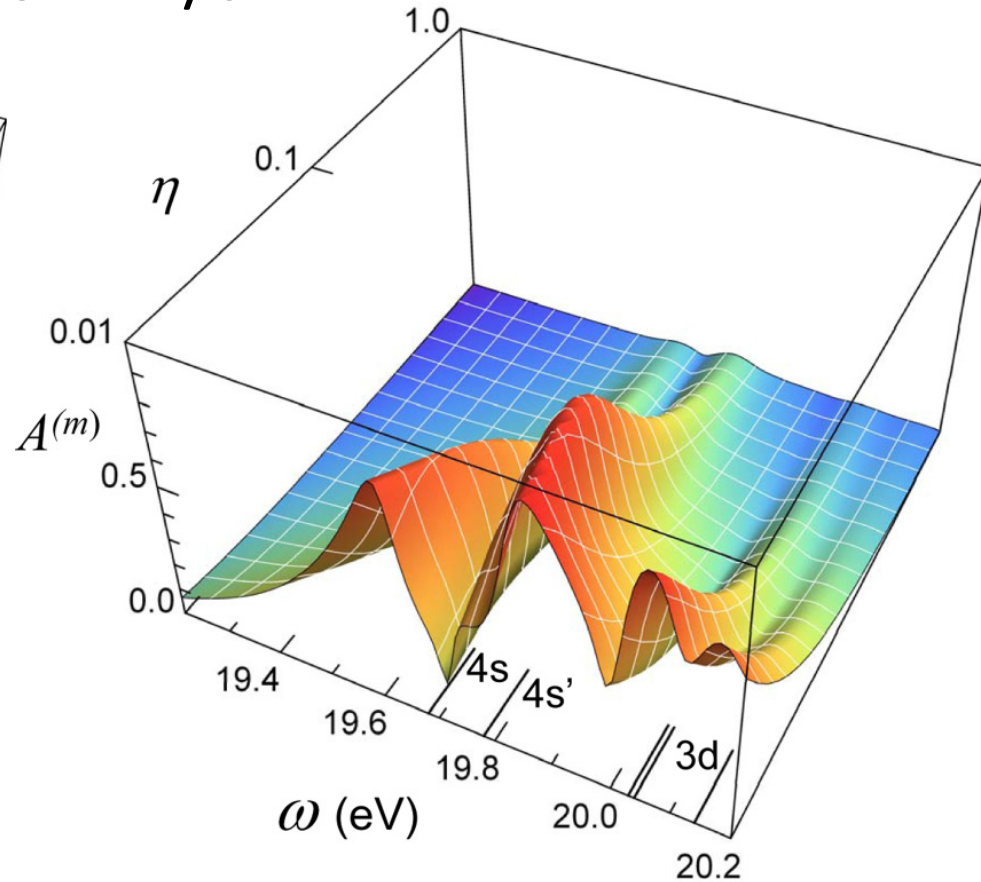
Dependence on Laser Parameters

[Gryzlova *et al.*, Phys. Rev. A **97**, 013420 (2018)]

$$I = 10^{12} \text{ W/cm}^2$$



$$\eta = 0.1$$

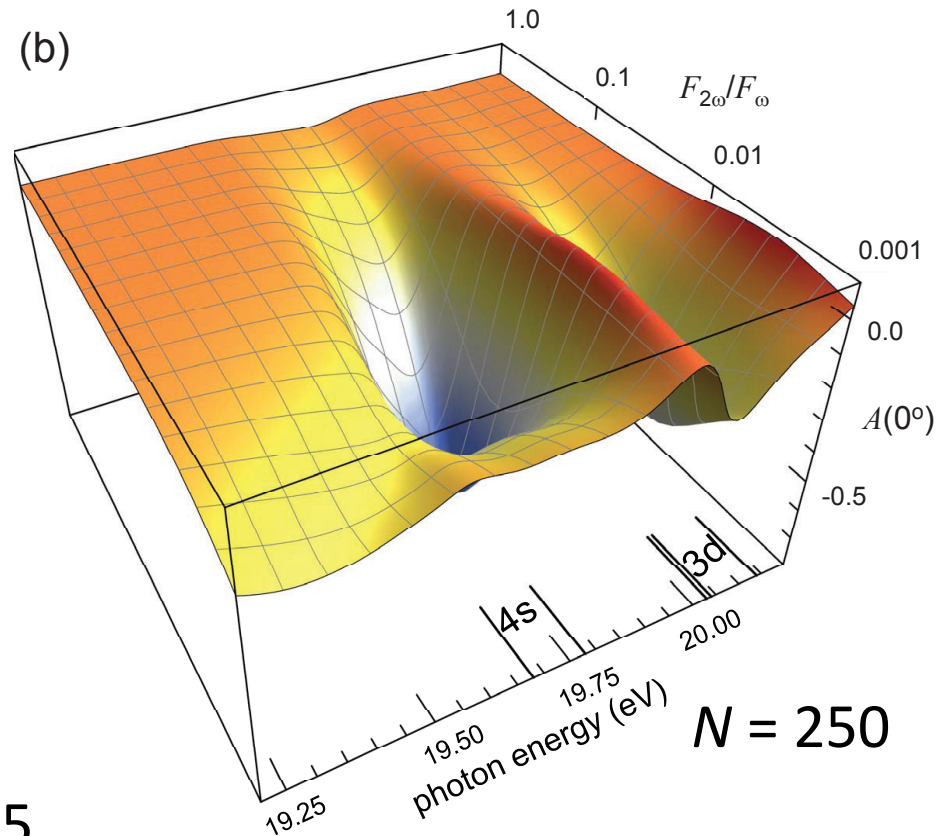
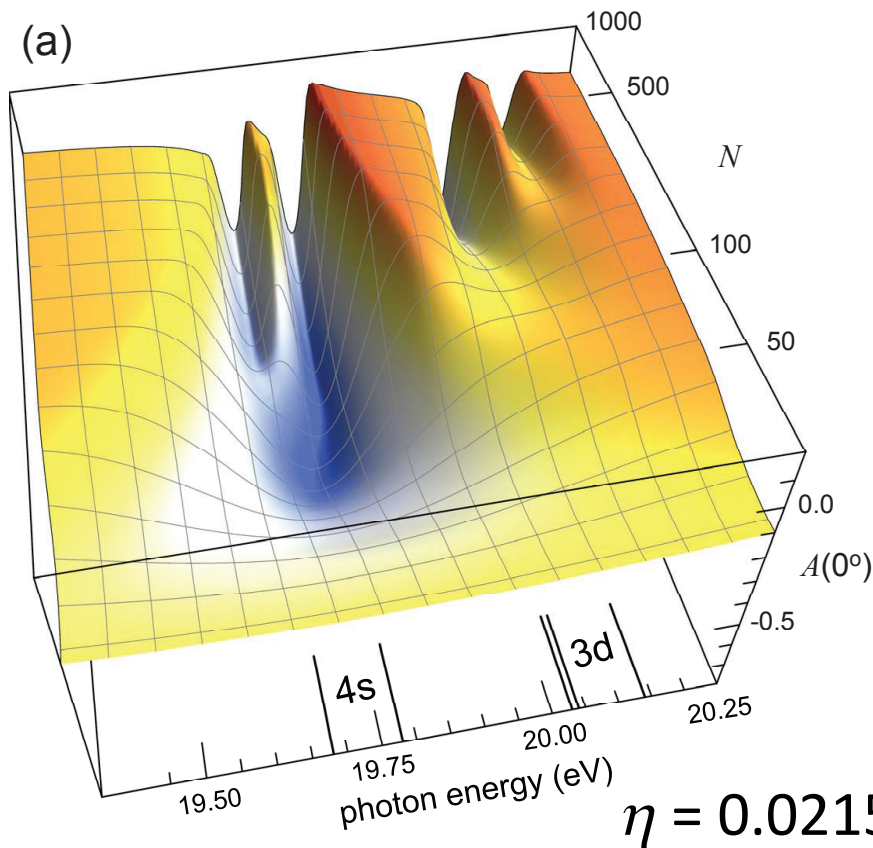


$$N = 500$$

Dependence on Laser Parameters

[Giannessi *et al.*, Sci. Rep. 8 (2018) 7774]

$$I = 10^{12} \text{ W/cm}^2$$



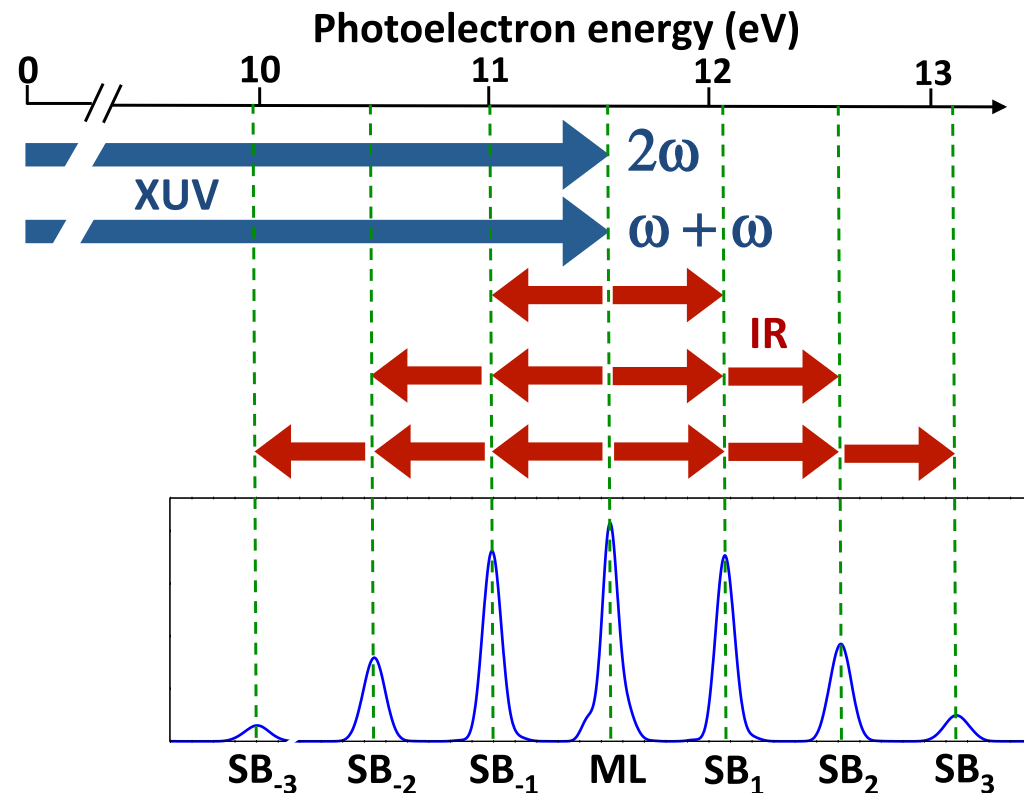
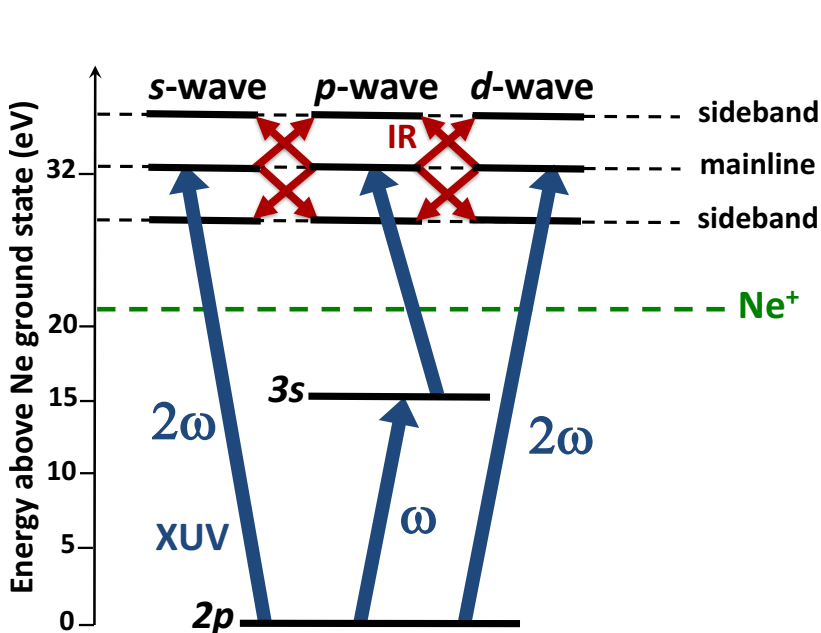
These parameters may be more realistic, but it is still **virtually impossible to directly compare experiment and theory**; what theory can handle is too difficult for experiment — and vice versa.

Overlapping XUV + IR fields

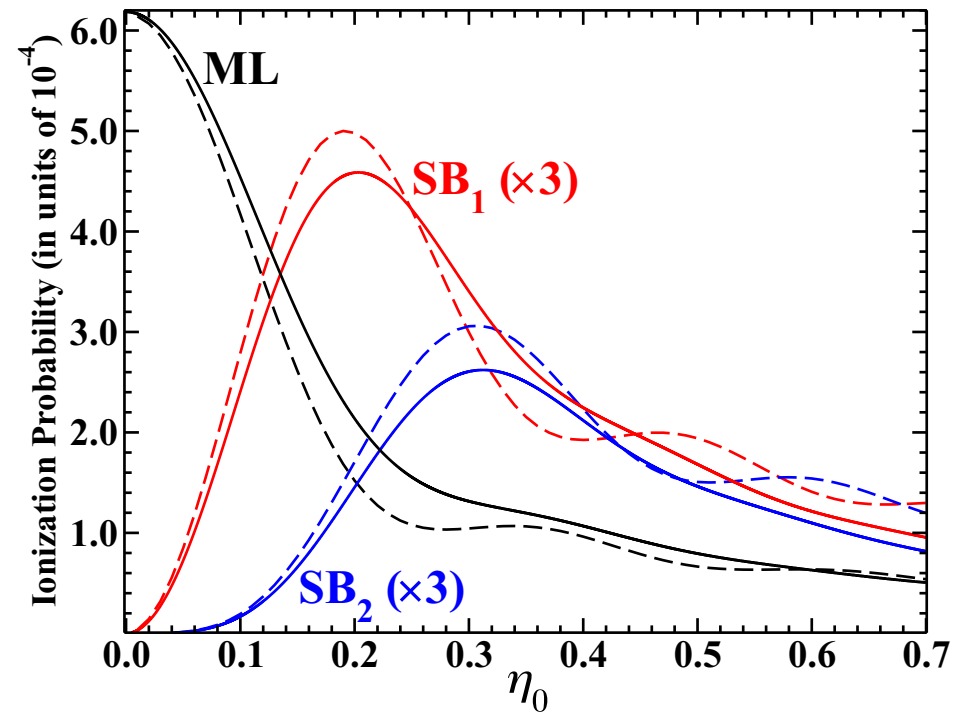
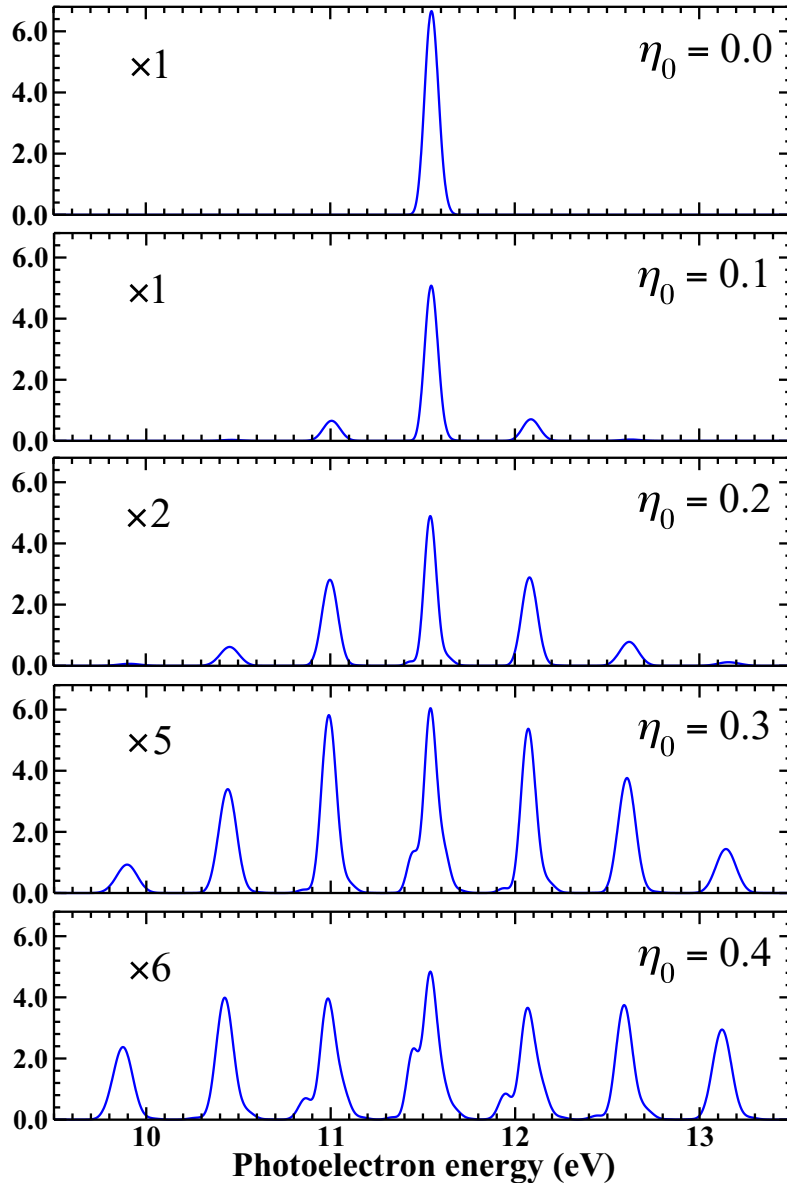
- Can we gain additional control in neon ionization by adding an **infrared field**?

$$\mathcal{E}(t) = \mathcal{E}_X(t) + \mathcal{E}_{IR}(t) \quad \mathcal{E}_X(t) = \bar{\mathcal{E}}_X f(t) [\cos(\omega t) + \eta_X \cos(2\omega t + \varphi_X)]$$

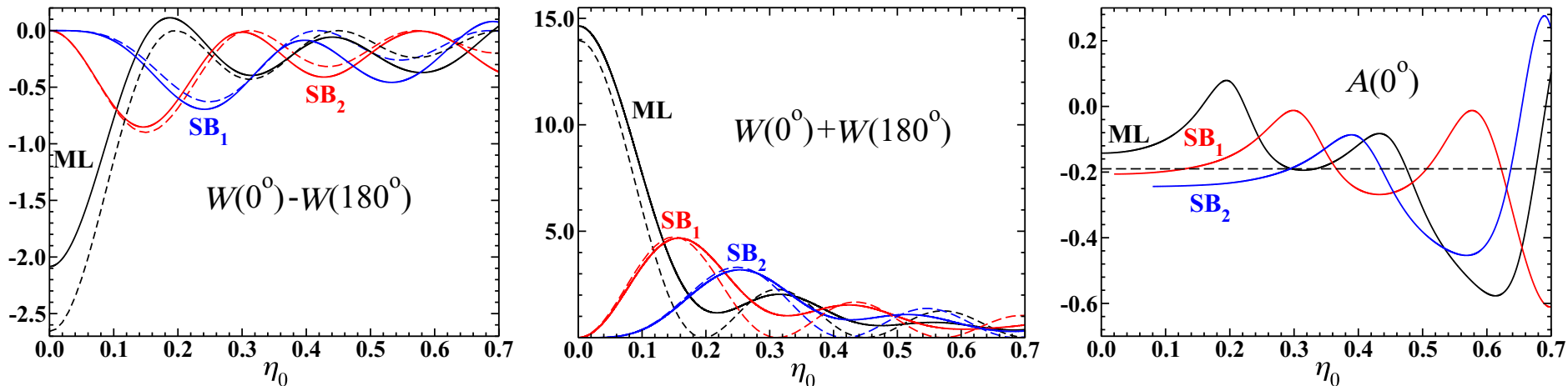
$$\mathcal{E}_{IR}(t) = \eta_0 \bar{\mathcal{E}}_X f(t) \cos(\Omega_0 t + \varphi_0)$$



Ionization at the Sidebands



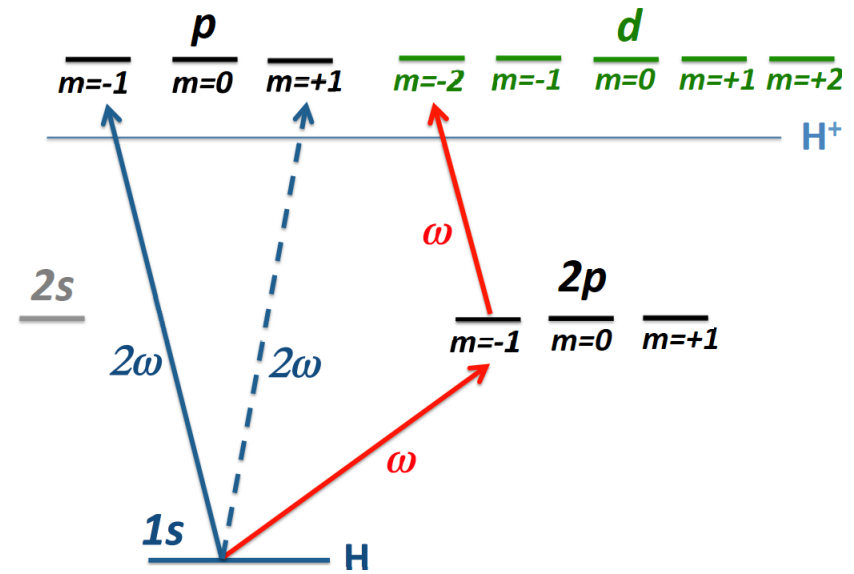
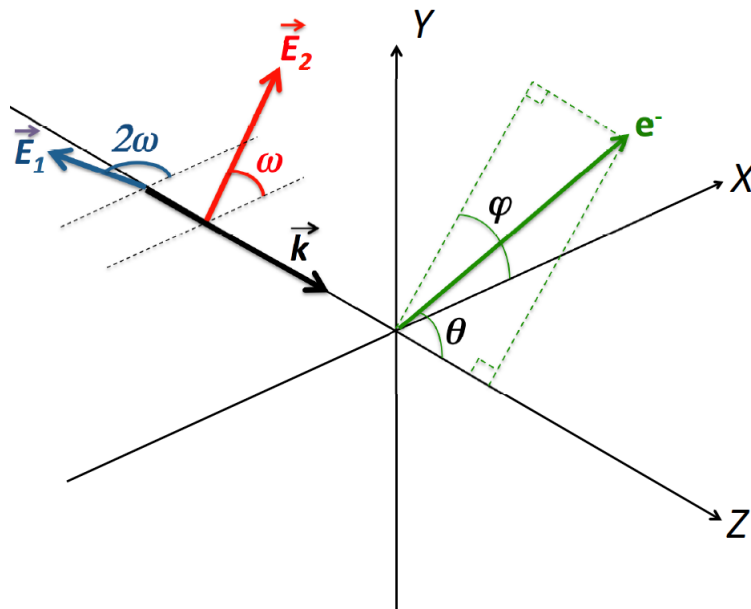
Asymmetry



- The SFA predicts an **asymmetry independent of the IR intensity** for a monochromatic pulse (about -0.2 in this case). Because the infrared field executes many symmetric oscillations, the asymmetry is simply **carried over from one sideband to another in the SFA model**.
- This is clearly not the case in the TDSE prediction ($N = 300$).
- We also showed that if the IR frequency **is tuned to a nearby transition** (e.g., $3s \rightarrow 3p$ in neon) then the asymmetry **can be manipulated through the IR frequency and intensity**.

Photoionization Scheme with Circularly Polarized Light in Atomic Hydrogen

- The electric field is in the XY plane and propagates along the Z axis.



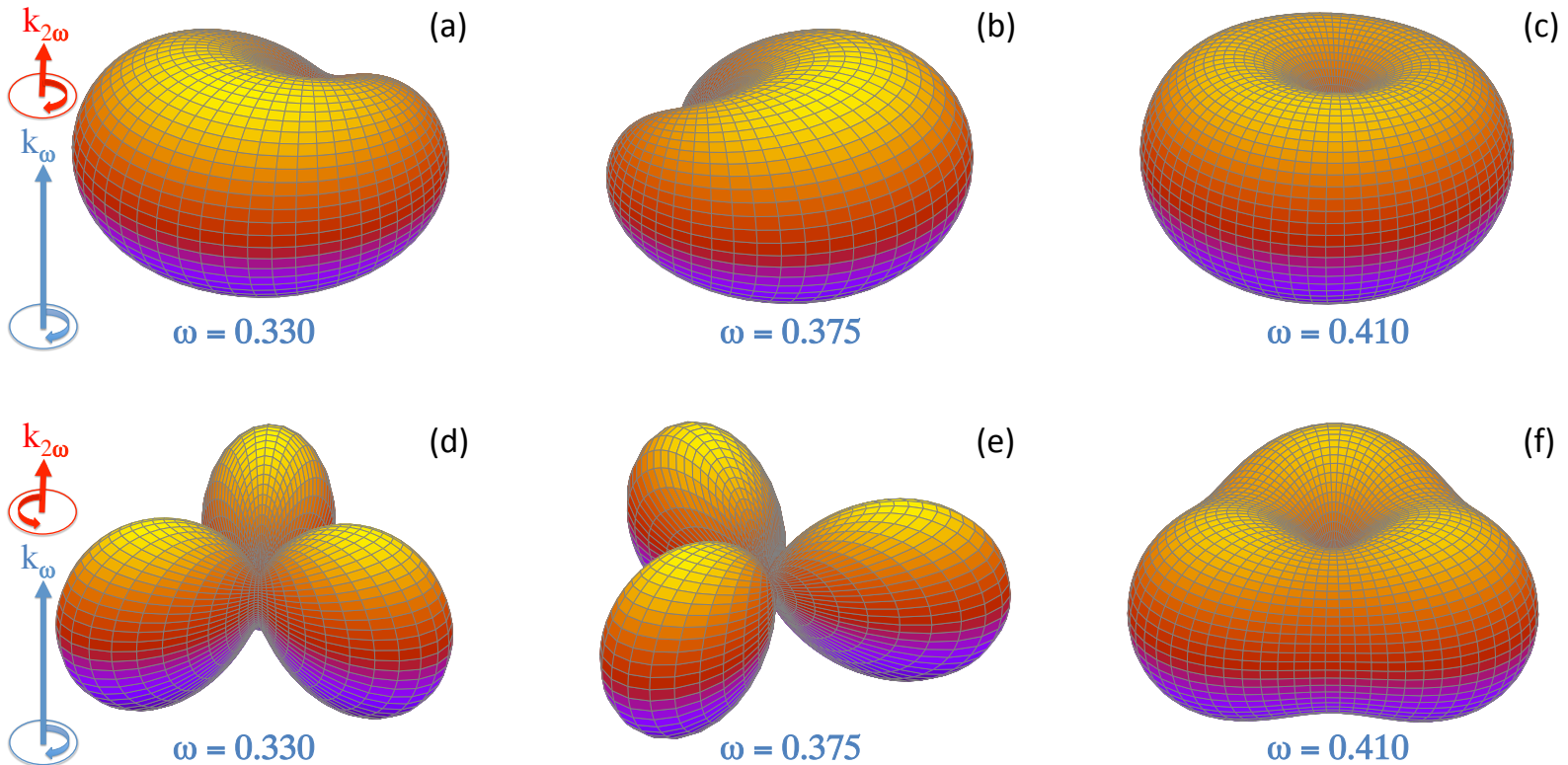
Pulse envelope Amplitude ratio CEP Helicity

$$\mathcal{E}(t) = F(t) \left[\underbrace{\cos(\omega t)\hat{x} - \sin(\omega t)\hat{y}}_{\text{First Harmonic}} + \eta \left\{ \underbrace{\cos(2\omega t + \phi)\hat{x} + \mathcal{H} \sin(2\omega t + \phi)\hat{y}}_{\text{Second Harmonic}} \right\} \right]$$

Electric field

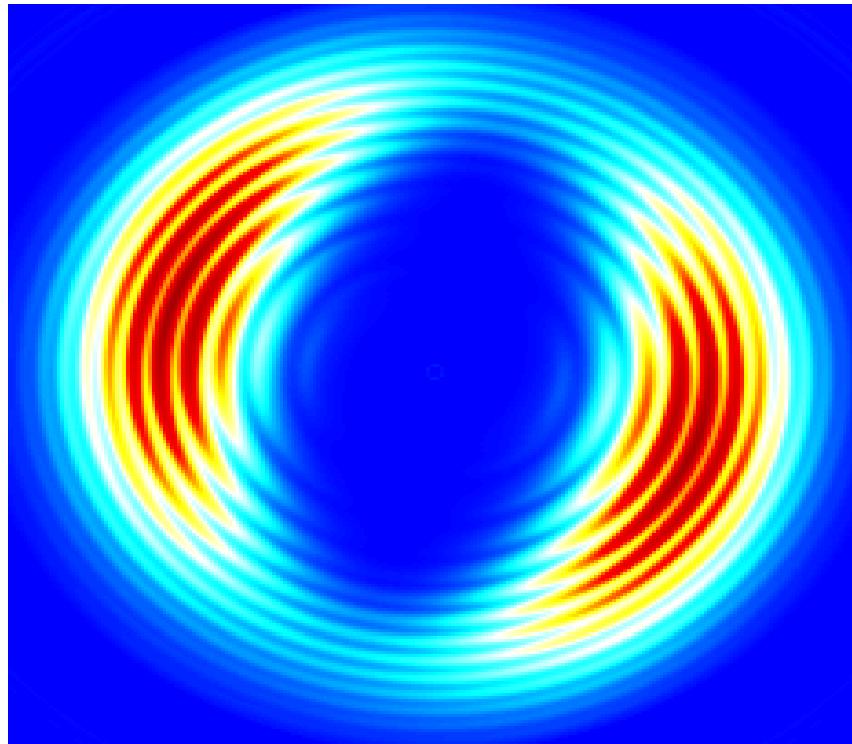
Visualizing the PAD in 3D

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



N. Douguet *et al.* Phys. Rev. A **93**, 033402 (2016)

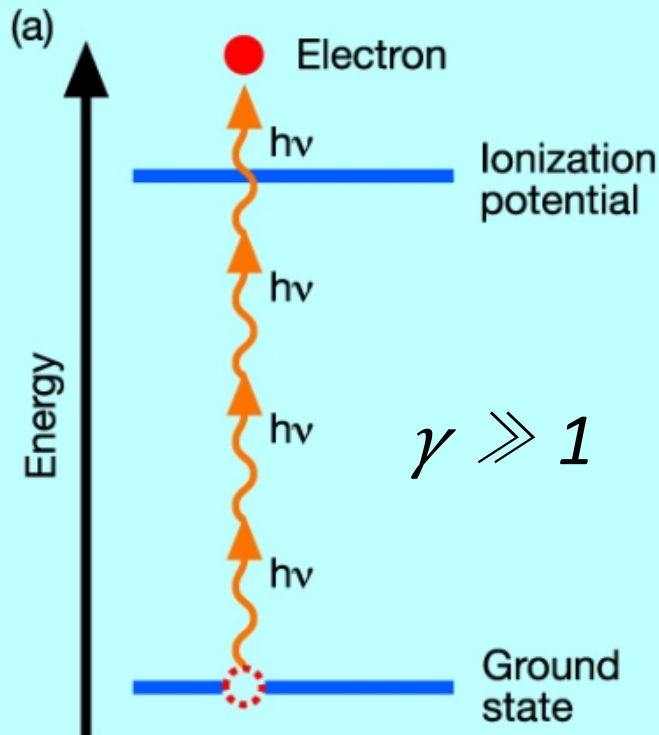
Multiphoton and Tunneling (?) Ionization



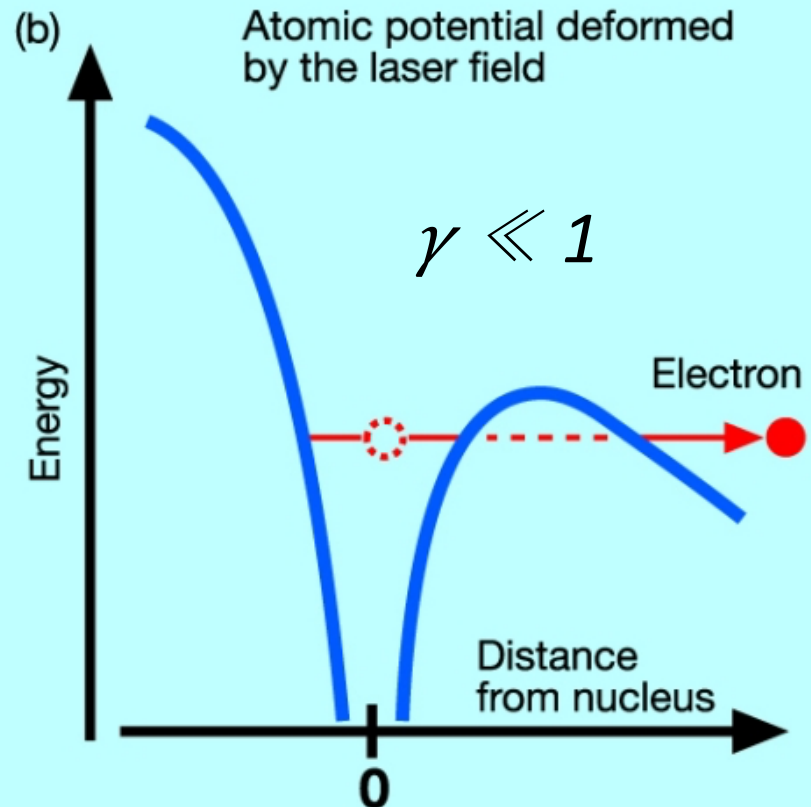
Multiphoton and Tunneling Ionization

- The **Keldysh parameter** $\gamma = (I_p/2U_p)^{1/2}$, with I_p the ionization potential and $U_p = I_{\max}/4\omega^2$ the ponderomotive energy, separates the cases.

Multiphoton Ionization

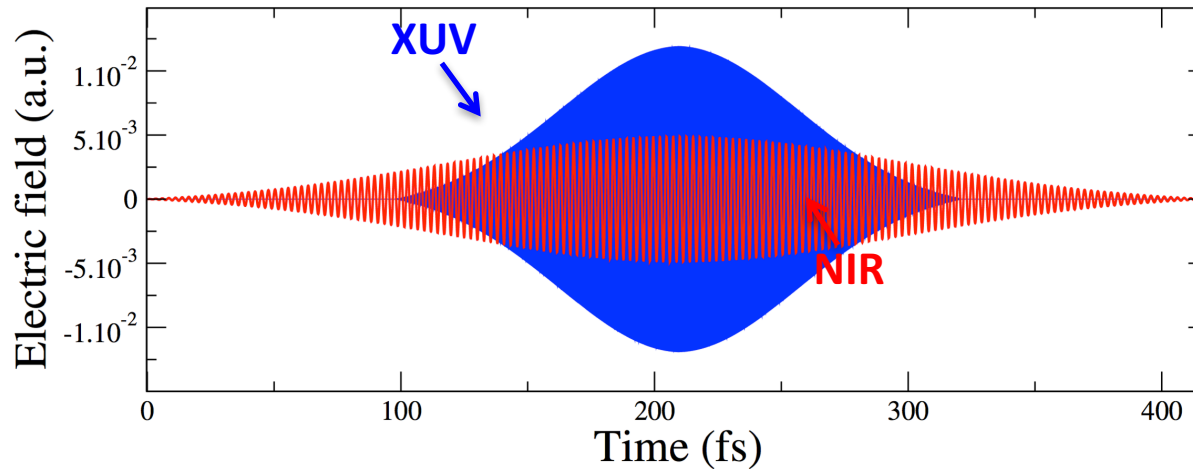


Tunneling Ionization



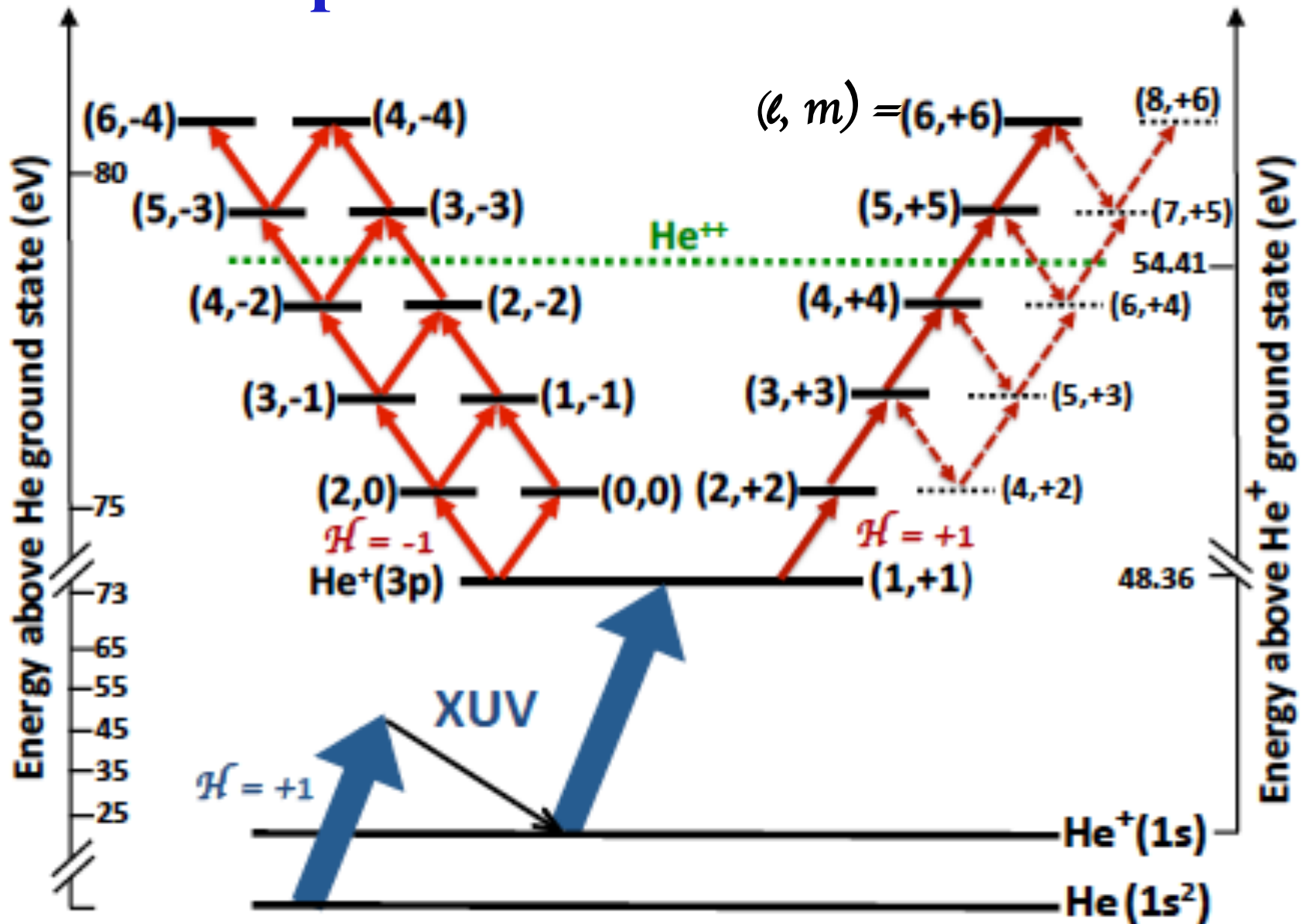
Circular Dichroism in Oriented He^+

- An overlapping circular XUV + NIR field is created at the FEL at FERMI

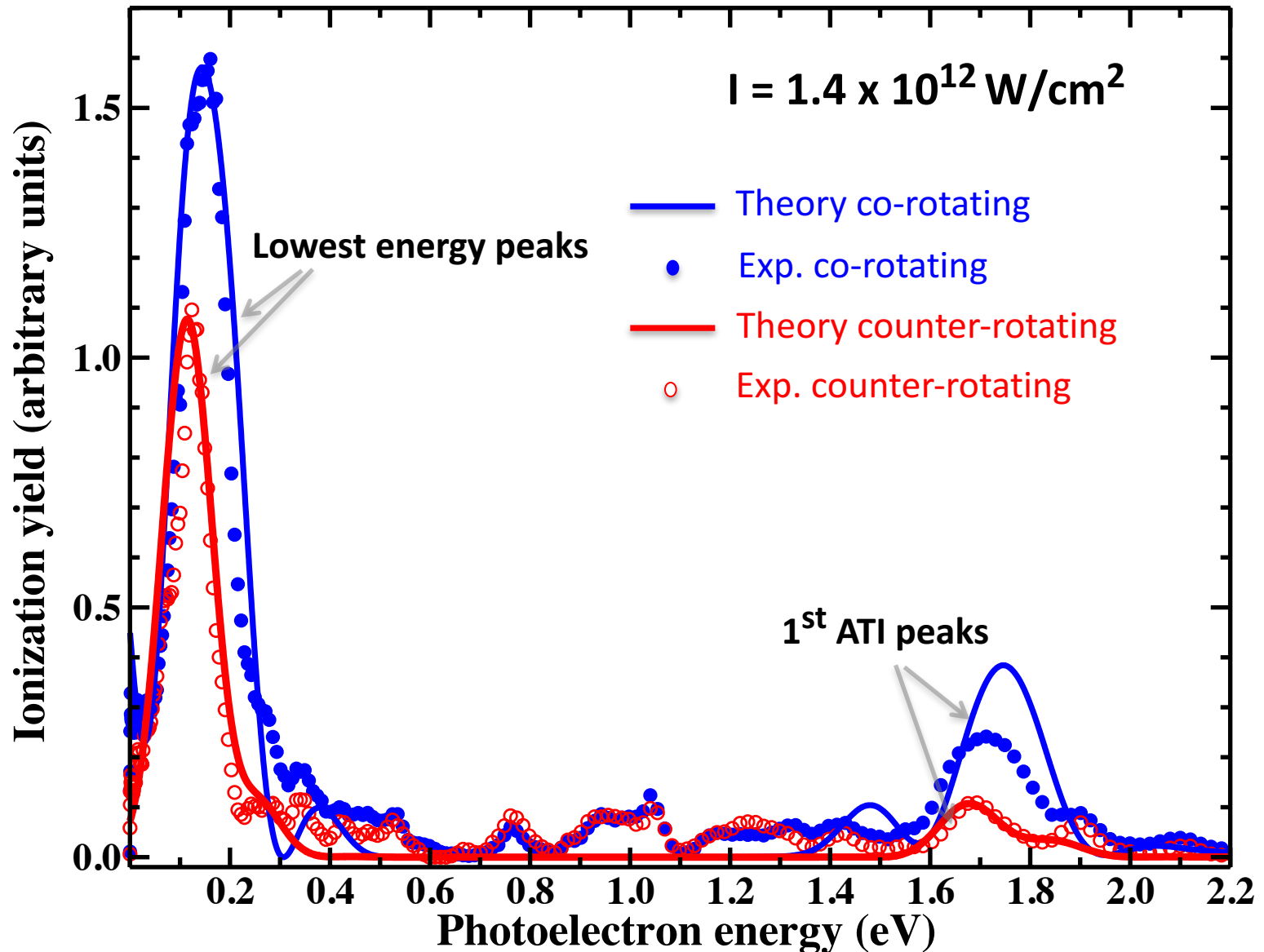


- The circularly polarized XUV pulse (FWHM = 100 fs and $I = 10^{13} \text{ W/cm}^2$ with positive helicity ($\mathcal{H} = +1$) creates oriented $\text{He}^+(3p; m = +1)$ via sequential absorption of two XUV photons:
 - Ionization**: $\text{He}(1s^2) + h\nu (48.37 \text{ eV}) \rightarrow \text{He}^+(1s) + e^-$
 - Pumping**: $\text{He}^+(1s) + h\nu (48.37 \text{ eV}) \rightarrow \text{He}^+(3p; m = +1)$
- The overlapping circularly polarized optical laser pulse (FWHM = 170 fs) with ($\mathcal{H} = +1$) or ($\mathcal{H} = -1$) ionizes the oriented $\text{He}^+(3p; m = +1)$ ion.
 - Multiphoton ionization**: $\text{He}^+(3p; m = +1) + 4 h\nu (1.58 \text{ eV}) \rightarrow \text{He}^{++} + e^-$

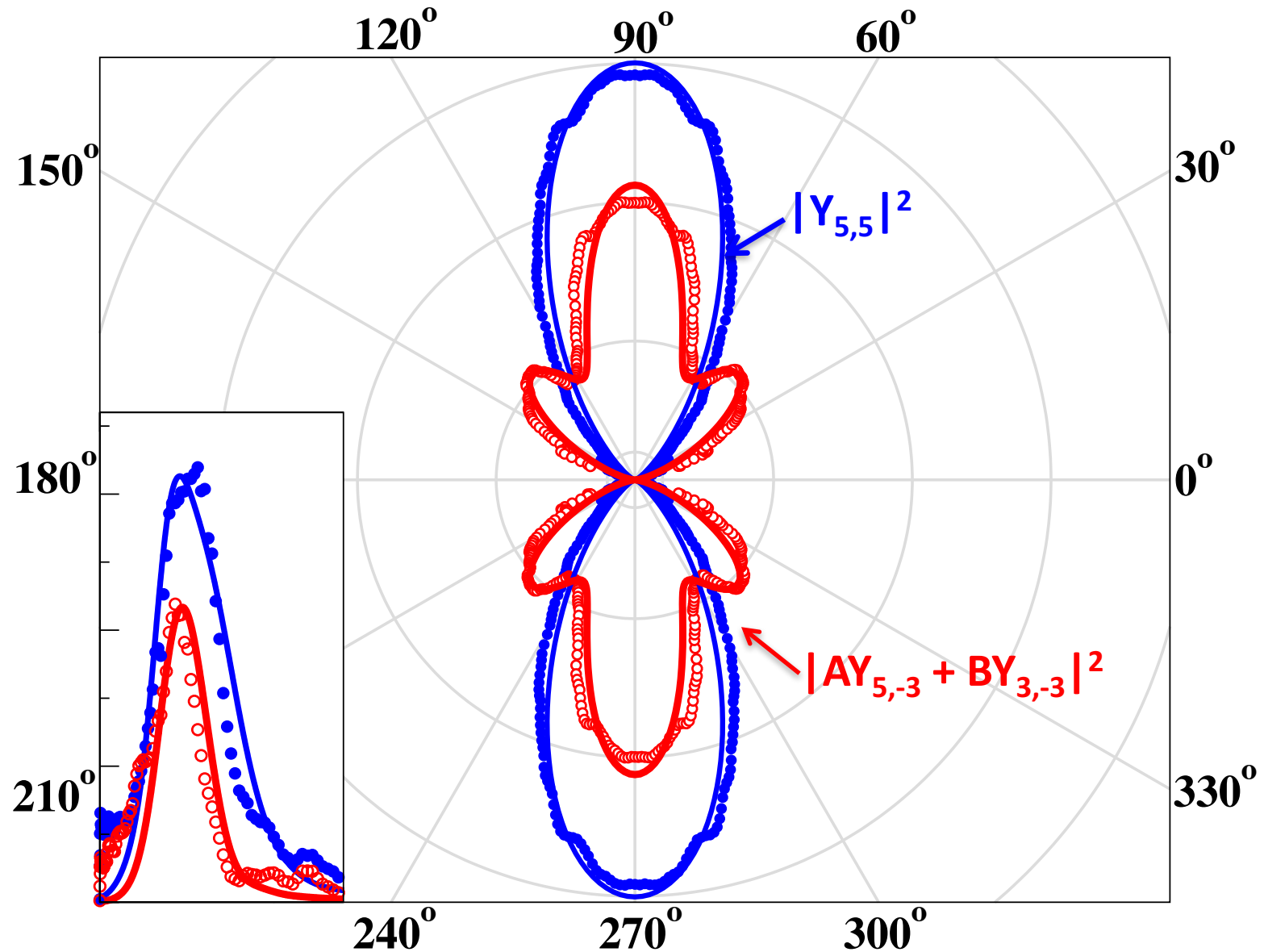
Multiphoton ionization scheme



Photoelectron spectrum



Photoelectron angular distribution



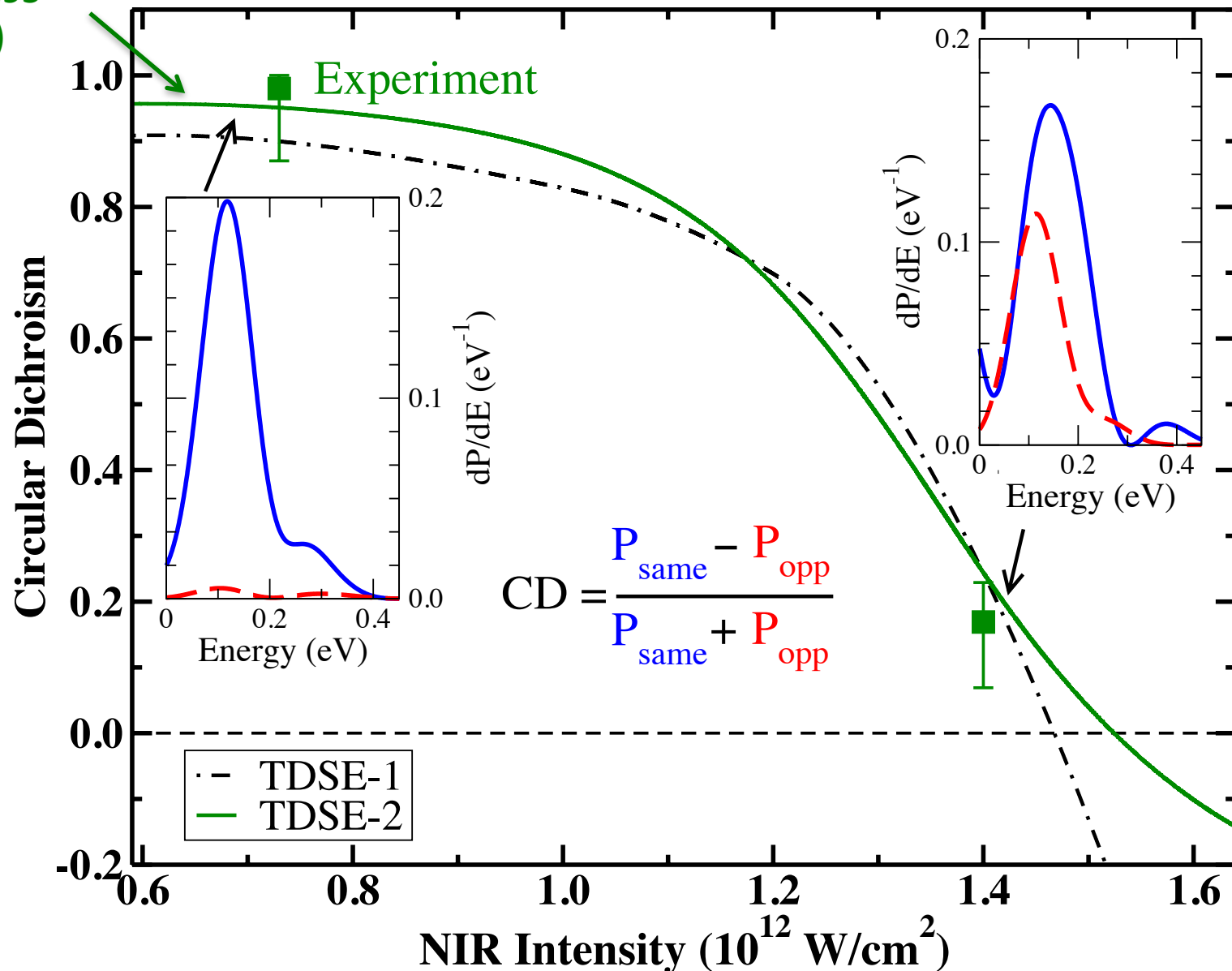
M. Ilchen, N. Douguet *et al.*, Phys. Rev. Lett. **118**, 013002 (2017)

Intensity dependence

- The photoionization spectrum was studied as a function of the **optical field intensity** from $I = 5 \times 10^{11} \text{ W/cm}^2$ to about $I = 2 \times 10^{12} \text{ W/cm}^2$.
- The ionization at the lowest peak was measured/calculated for both co-rotating and counter-rotating field helicities. **The circular dichroism** is defined as $\text{CD} = [\mathbf{P}_{\text{same}} - \mathbf{P}_{\text{opp}}] / [\mathbf{P}_{\text{same}} + \mathbf{P}_{\text{opp}}]$.

Circular Dichroism

CD ≈ 0.95
(LOPT)



M. Ilchen, N. Douguet *et al.*, Phys. Rev. Lett. **118**, 013002 (2017)

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- From LOPT, the ionization probability for co-rotating fields is expected **to be much larger** than for counter-rotating fields **at low intensity** since the angular factor is about **50 times larger** for the same field helicity.
- A negative CD was predicted by Barth and Sminorva [**PRA 84 0634153 (2011)**] in the tunneling ionization regime.

Intensity dependence

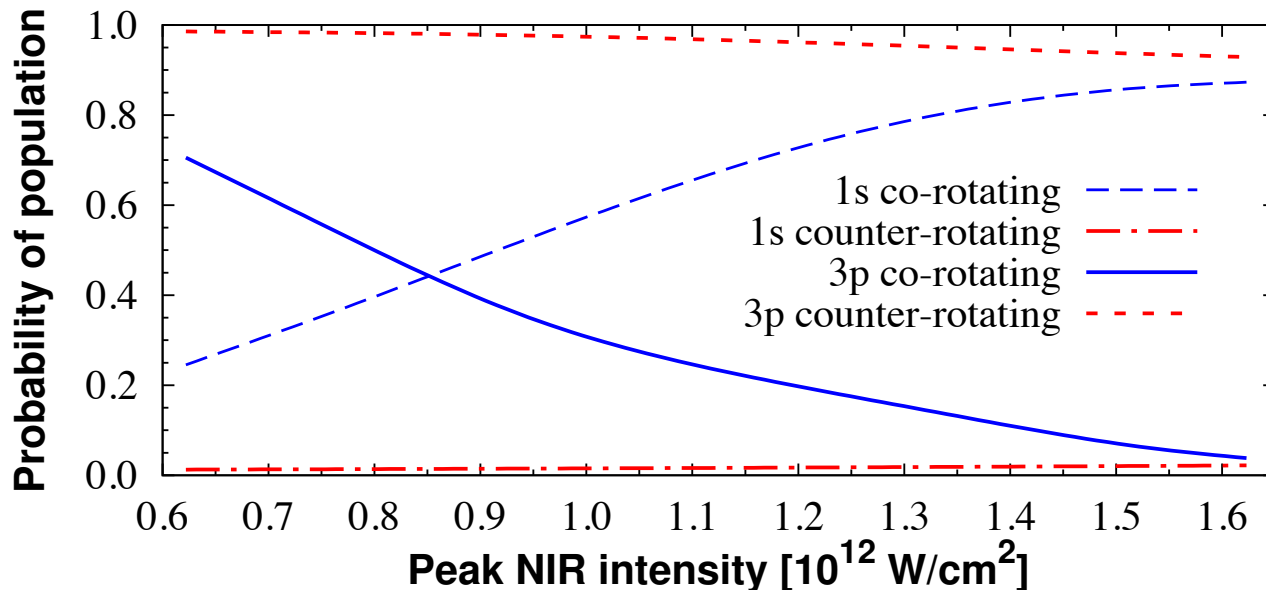
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- However, as the intensity is only slightly increased, the **CD decreases rapidly** and is predicted to become **negative** at only $I = 1.55 \times 10^{12} \text{ W/cm}^2$!

Intensity Dependence

- The photoionization spectrum was studied as a function of the **optical field intensity** from $I = 5 \times 10^{11} \text{ W/cm}^2$ to about $I = 2 \times 10^{12} \text{ W/cm}^2$.
- The ionization at the lowest peak was measured/calculated for both co-rotating and counter-rotating field helicities. **The circular dichroism** is defined as $\text{CD} = [P_{\text{same}} - P_{\text{opp}}] / [P_{\text{same}} + P_{\text{opp}}]$.
- From LOPT, the ionization probability for co-rotating fields is expected **to be way larger** than for counter-rotating fields **at low intensity**
→ The angular factor is about 50 times larger for the same field helicity!
- A negative CD was predicted by Barth and Sminorva [PRA 84 0634153 (2011)] in the tunneling ionization regime.
- However, as the intensity is only slightly increased, the **CD decreases rapidly** and is predicted to become **negative** at only $1.55 \times 10^{12} \text{ W/cm}^2$!
→ **Why do we observe a negative CD at low field intensity!?**

Discussion

- The behavior of the CD is most probably the result of several factors.
- Our analysis strongly suggests that two important factors play a role:
 - i. Changing the optical frequency strongly modifies the CD
→ Suggests **near-resonant** phenomena
 - ii. The **AC stark shift** of the 3p state is larger in the co-rotating case than in the counter-rotating case (confirmed by Fourier-analysis).
→ 3p state is not efficiently populated for co-rotating fields.



Counter-rotating

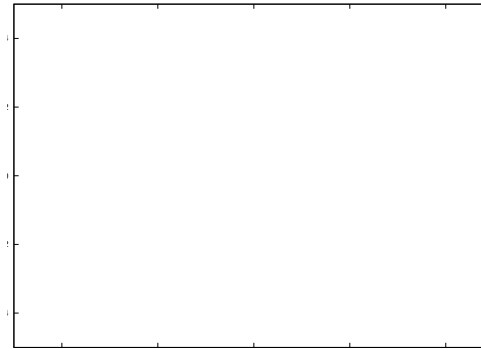
3p, m= +1

AC stark shift

Co-rotating

Illustration: Dichroism at $I = 10^{12} \text{ W/cm}^2$

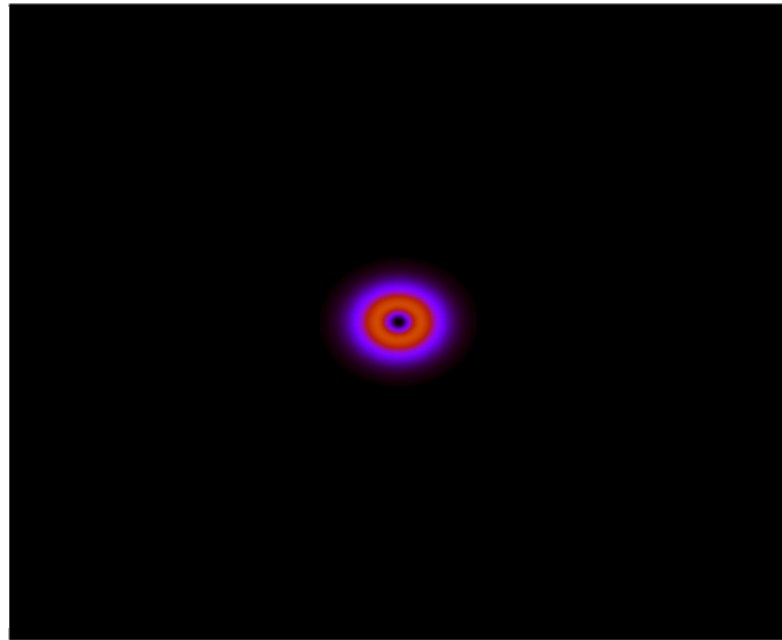
Pulse : 4 cycles and $\lambda = 780 \text{ nm}$
Target: Hydrogen $2p(+1)$ state.



$$\gamma = 11.1$$

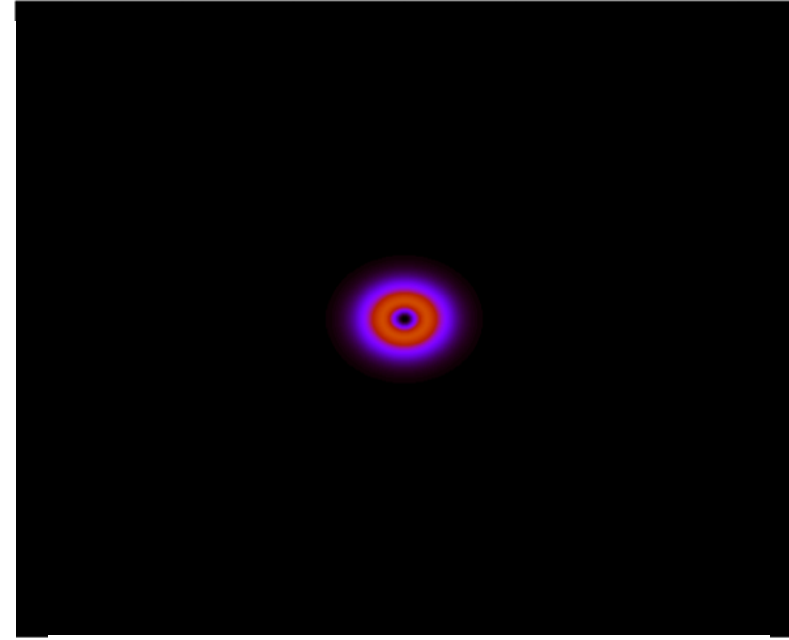


Co-rotating
($m = +1$)



Ionization Probability = 6.532×10^{-2}

Counter-rotating
($m = -1$)

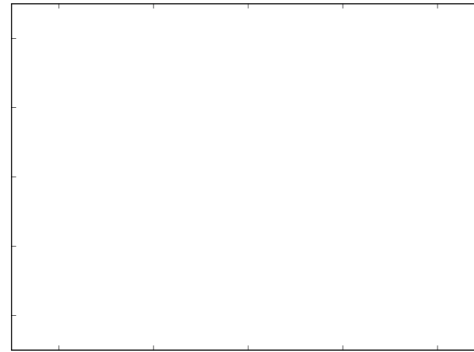


Ionization Probability = 1.572×10^{-2}

Illustration: Dichroism at $I = 10^{12} \text{ W/cm}^2$

Pulse : 4 cycles and $\lambda = 780 \text{ nm}$
Target: Hydrogen 2p state.

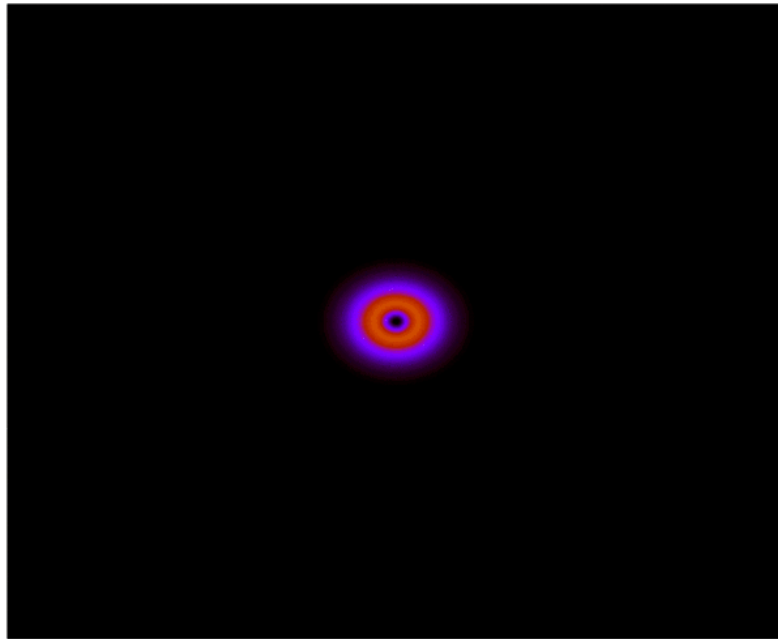
$$\gamma = 11.1$$



$$F = -eE(t)$$

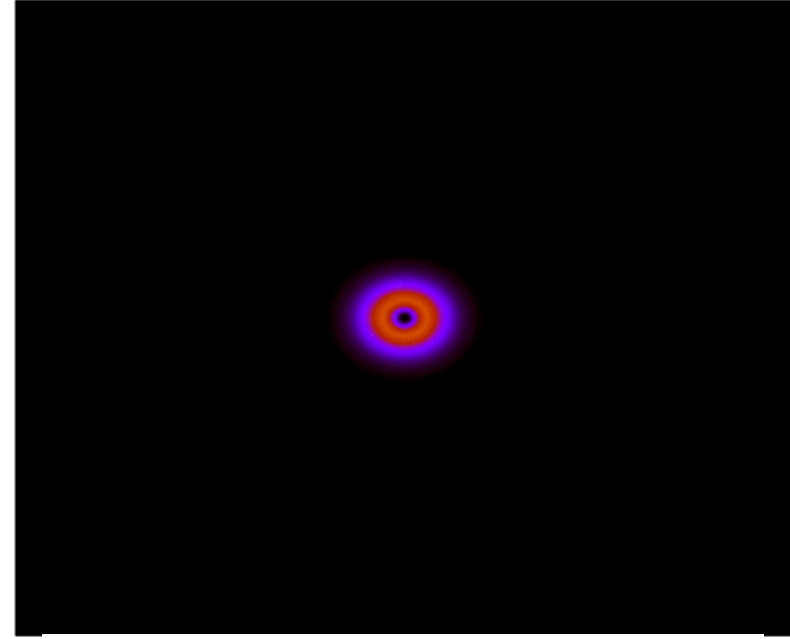


Co-rotating
($m = +1$)



Ionization Probability = 6.532×10^{-2}

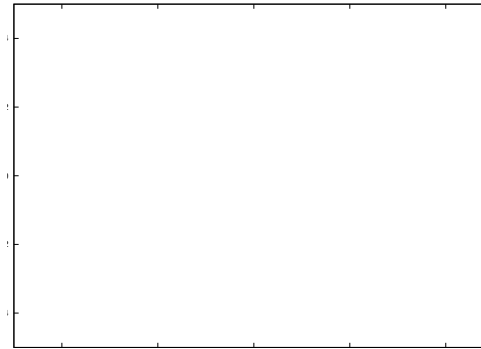
Counter-rotating
($m = -1$)



Ionization Probability = 1.572×10^{-2}

Illustration: Dichroism at $I = 4 \times 10^{13} \text{ W/cm}^2$

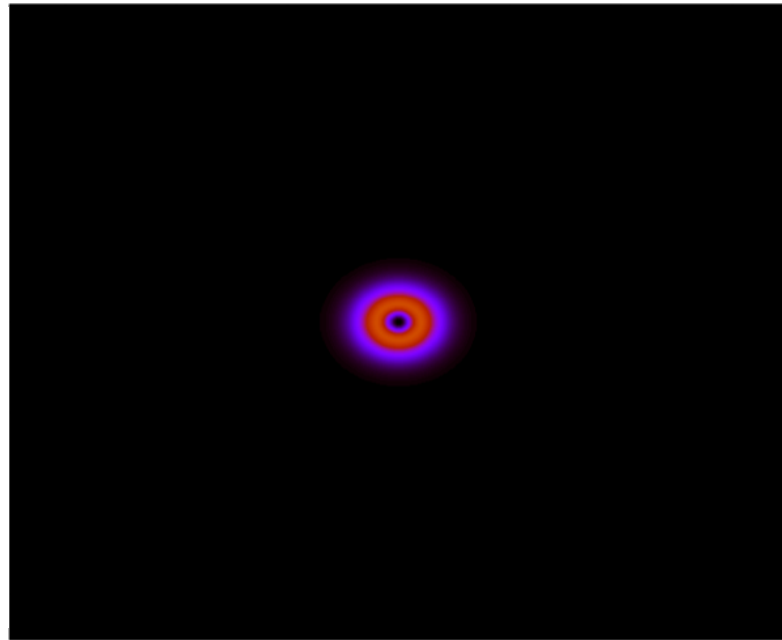
Pulse : 4 cycles and $\lambda = 780 \text{ nm}$
Target: Hydrogen 2p state.



$$\gamma = 1.75$$

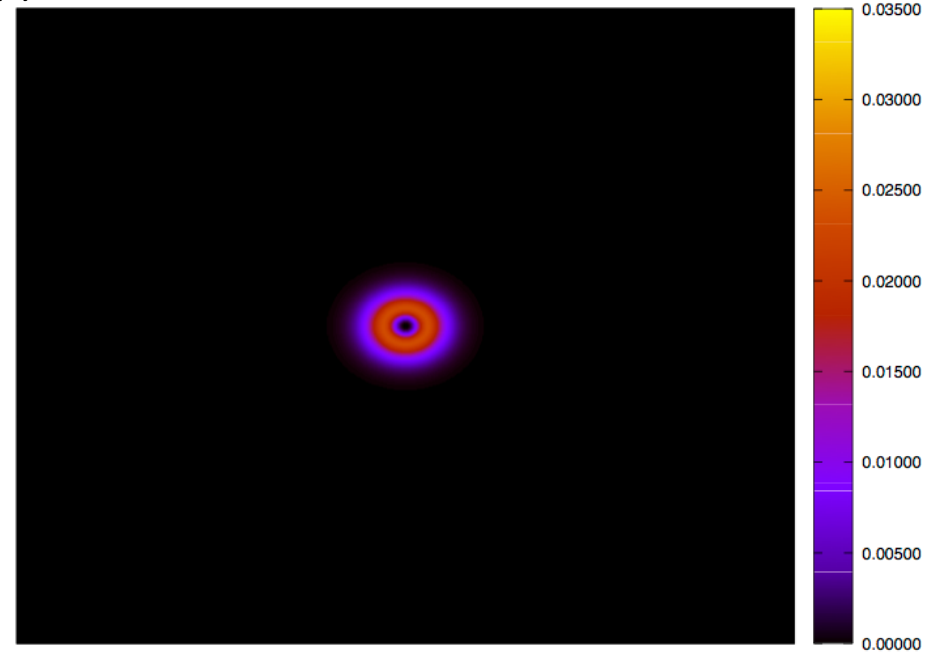


Co-rotating
($m = +1$)



Ionization Probability = $7.01\text{e-}01$

Counter-rotating
($m = -1$)

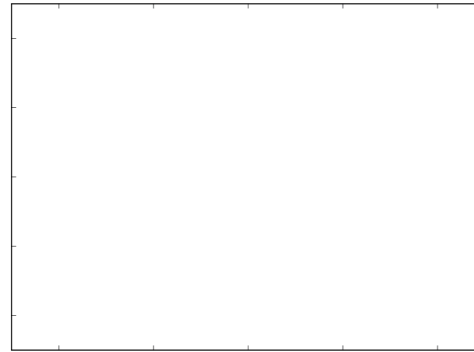


Ionization Probability = $7.72\text{e-}01$

Illustration: Dichroism at $I = 4 \times 10^{13} \text{ W/cm}^2$

Pulse : 4 cycles and $\lambda = 780 \text{ nm}$
Target: Hydrogen 2p state

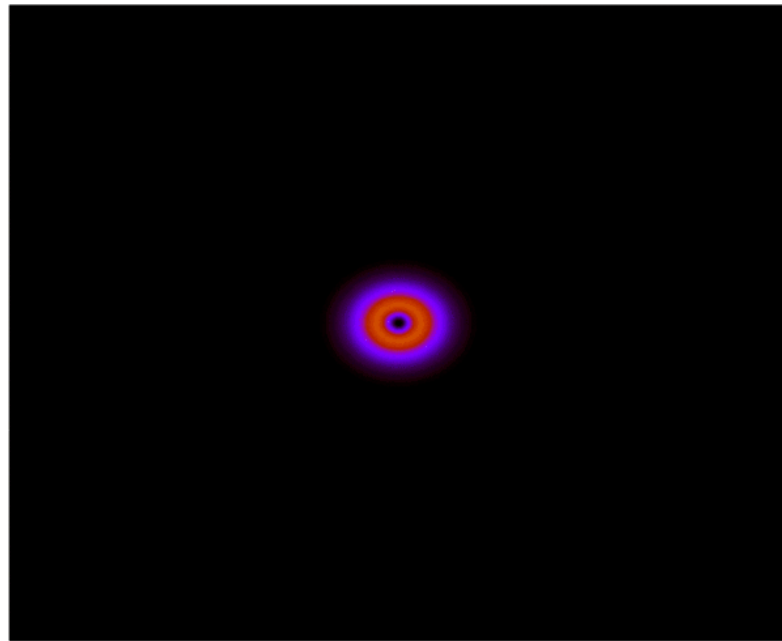
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$$F = -eE(t)$$

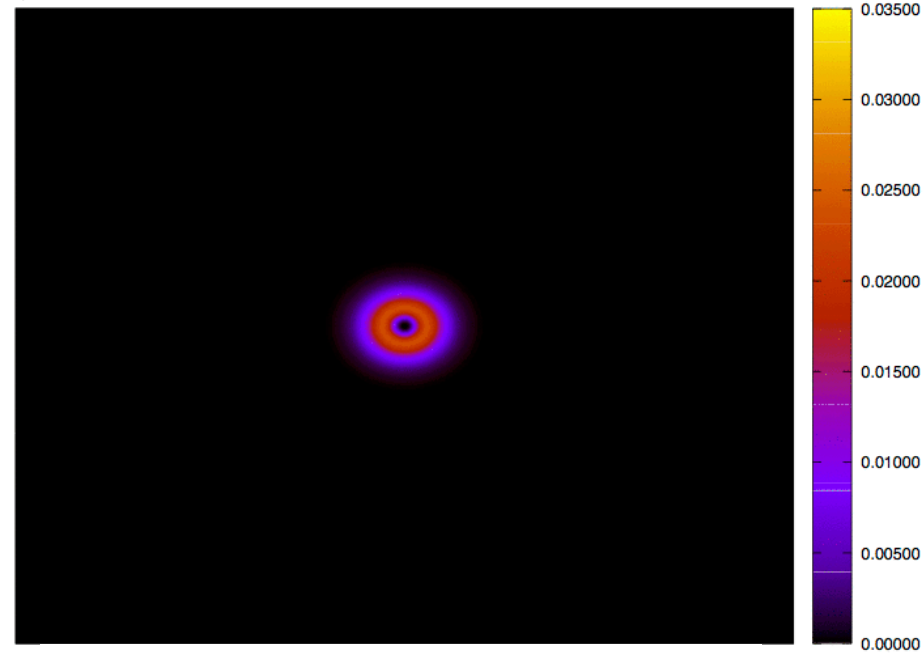


Co-rotating
($m = +1$)



Ionization Probability = $7.01\text{e-}01$

Counter-rotating
($m = -1$)



Ionization Probability = $7.72\text{e-}01$

Tunneling Time:

A somewhat (???) controversial topic

nature
physics

ARTICLES

PUBLISHED ONLINE: 25 MAY 2015 | DOI: 10.1038/NPHYS3340

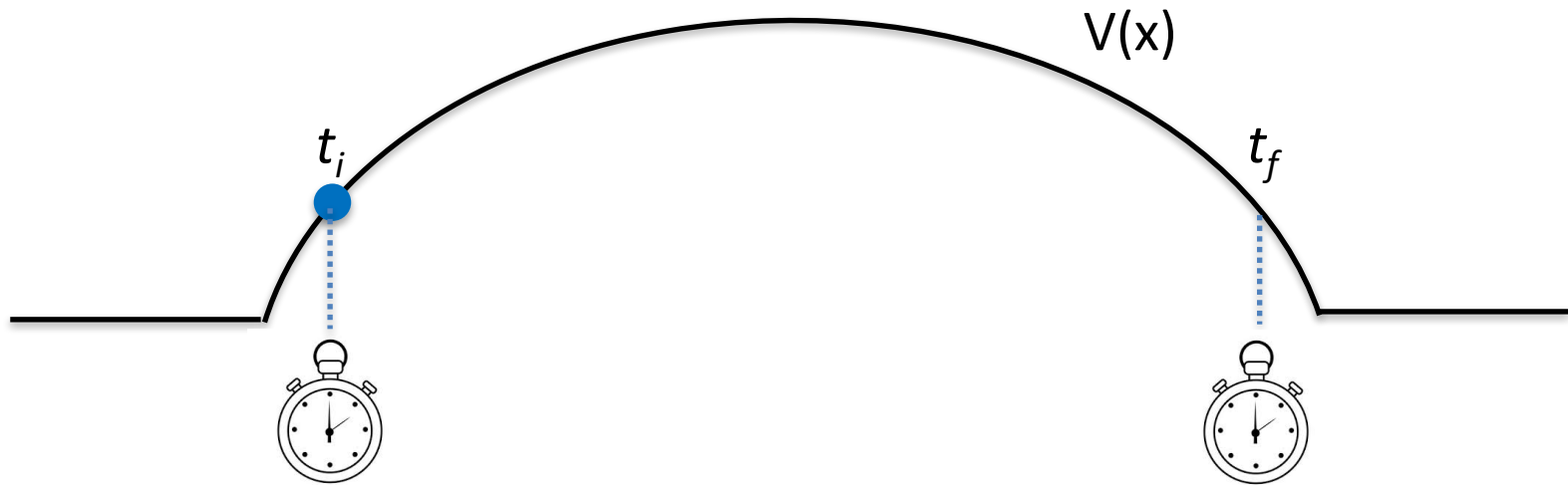
Interpreting attoclock measurements of tunnelling times

Lisa Torlina^{1†}, Felipe Morales^{1†}, Jivesh Kaushal¹, Igor Ivanov², Anatoli Kheifets², Alejandro Zielinski³, Armin Scrinzi³, Harm Geert Muller¹, Suren Sukiasyan⁴, Misha Ivanov^{1,4,5} and Olga Smirnova^{1★}

Resolving in time the dynamics of light absorption by atoms and molecules, and the electronic rearrangement this induces, is among the most challenging goals of attosecond spectroscopy. The attoclock is an elegant approach to this problem, which encodes ionization times in the strong-field regime. However, the accurate reconstruction of these times from experimental data presents a formidable theoretical task. Here, we solve this problem by combining analytical theory with *ab initio* numerical simulations. We apply our theory to numerical attoclock experiments on the hydrogen atom to extract ionization time delays and analyse their nature. Strong-field ionization is often viewed as optical tunnelling through the barrier created by the field and the core potential. **We show that, in the hydrogen atom, optical tunnelling is instantaneous.** We also show how calibrating the attoclock using the hydrogen atom opens the way to identifying possible delays associated with multielectron dynamics during strong-field ionization.

Measuring tunneling time?

- One of the goal of ultrafast physics is to investigate the electron dynamics in its **natural time-scale**. An obvious question:
→ Can we measure tunneling time??



Problems:

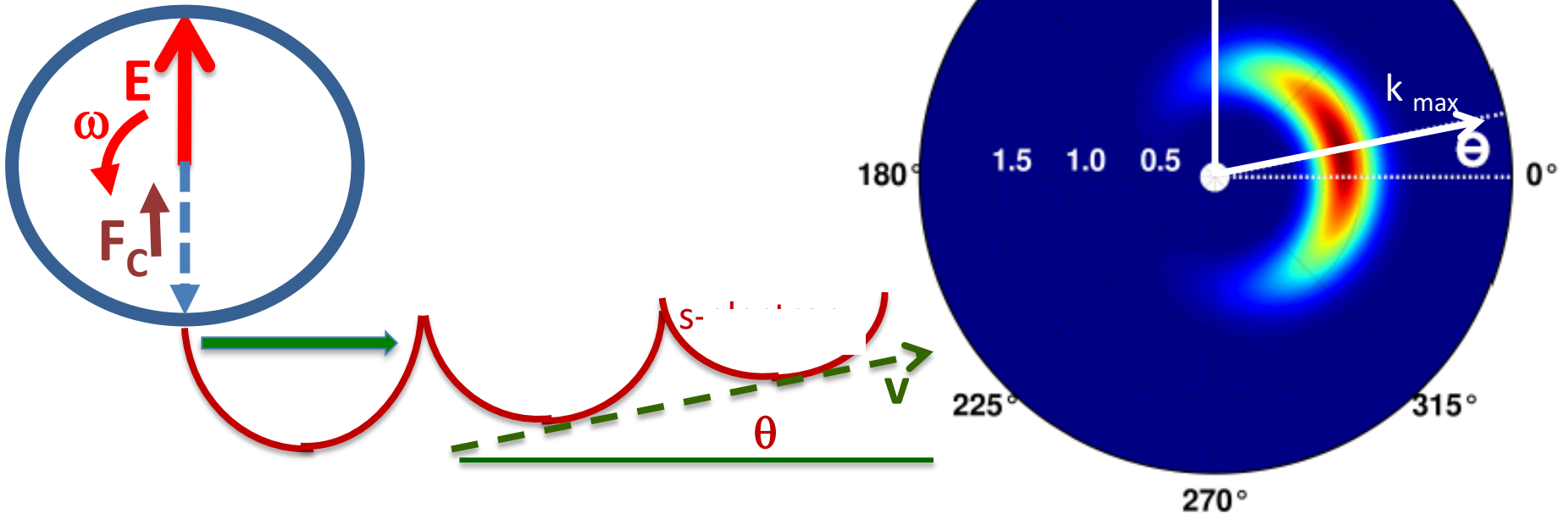
- “Time” is not a physical observable in Quantum Mechanics
- How do we define the starting and final moment t_i and t_f ?

Answer (?): The attoclock!

Tunneling Time (atto-clock?)

TDSE spectra

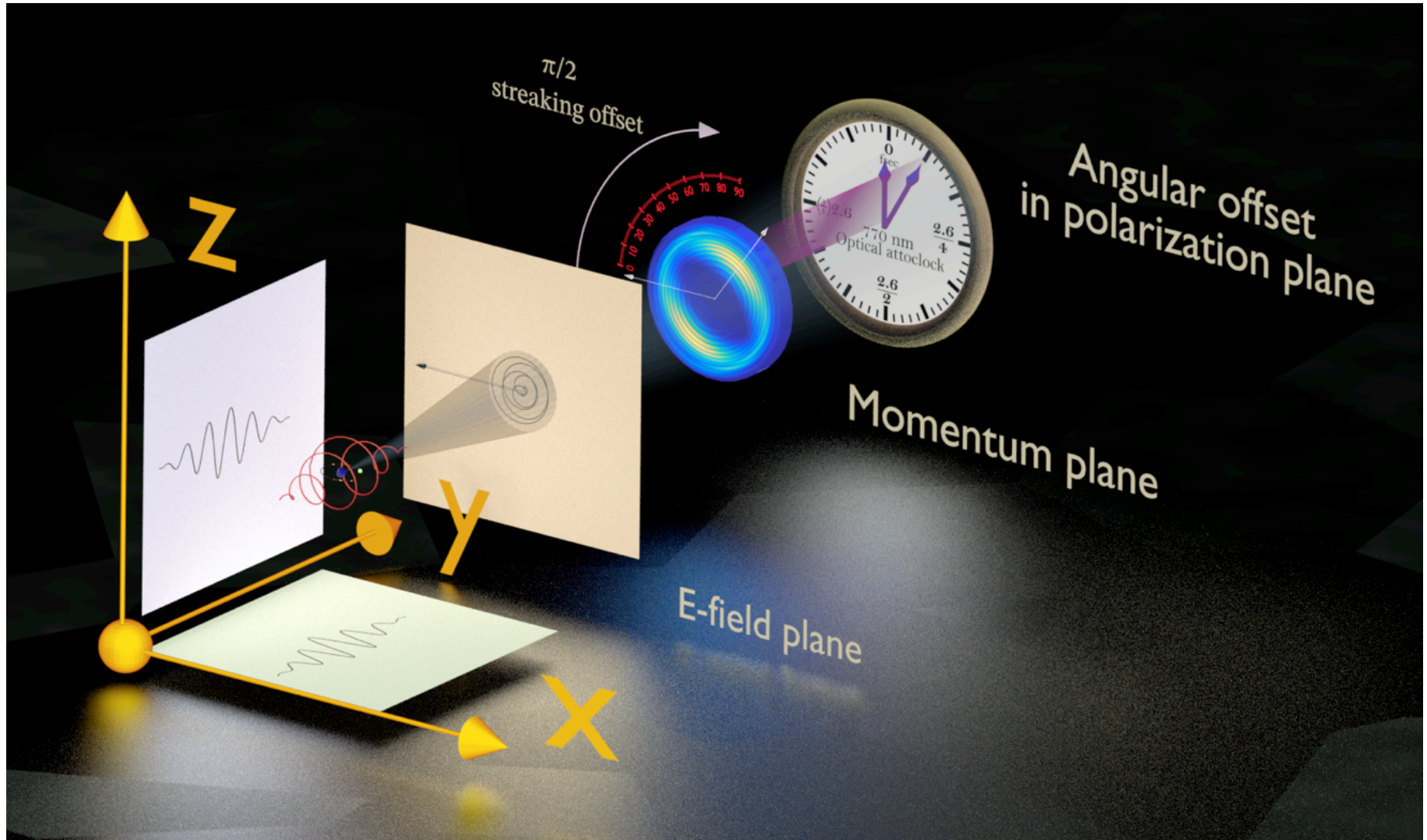
[adapted from Torlina *et al.*,
Nat. Phys. **11** (2016) 593]



- Assumption: Since the probability for tunneling ionization varies exponentially with the field strength, ionization occurs at the maximum of the field. From the offset angle (non-zero due to the long-range Coulomb potential), one hopes to read off the time (atto-clock).

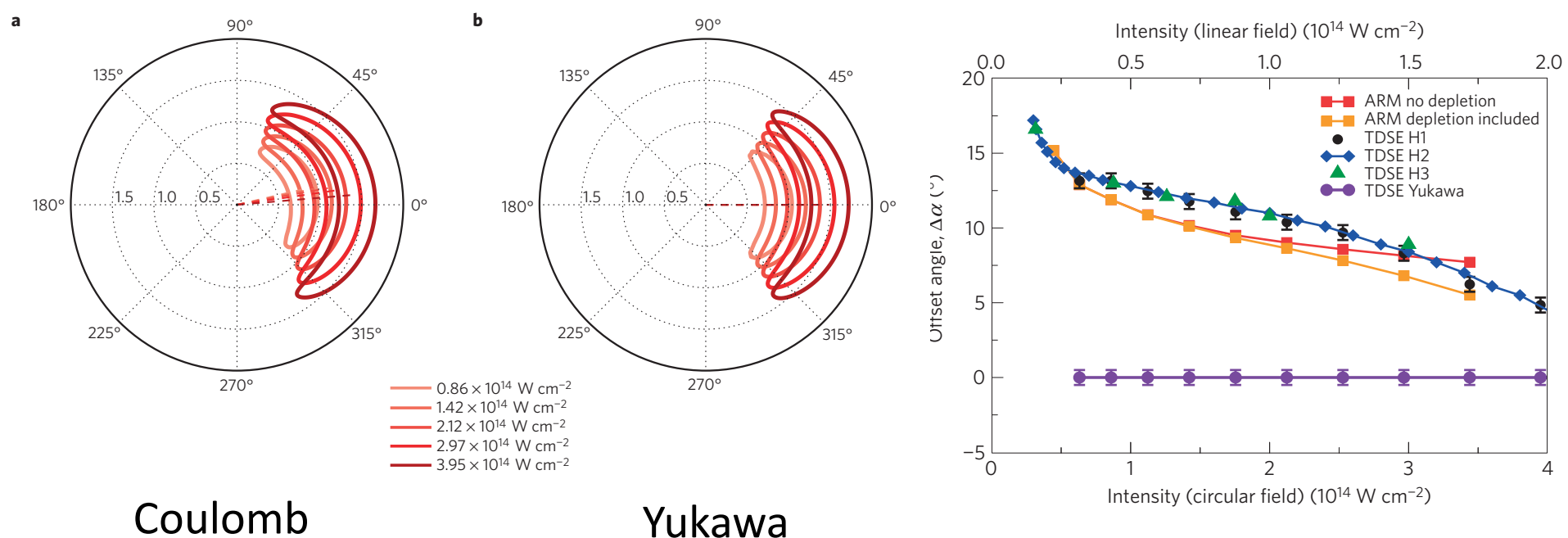
The Attoclock: Basic Idea

arXiv 1707.05445 (Griffith group)



Comparison with Short-Range Potential

- The offset angle can have **two origins**: (i) the effect of the long-range Coulomb potential, and/or (ii) the time it takes for the electron to tunnel through the barrier.
 - In order to answer this question, Torlina *et al.* performed calculations using a short-range **Yukawa potential** with the same energy of the 1s state.
- They found zero offset using the Yukawa potential and concluded that tunneling is **instantaneous** in atomic hydrogen. *Is this a valid conclusion?*



Some Comments about the Torlina *et al.* Calculations

- Even though “measurements” (?) is in the title of the paper, this was a **purely theoretical study**, with the principal goal of validating the analytical R-matrix theory (with approximations that allow for some further interpretation) against explicit numerical calculations.
- The **pulse shape** (“nearly” one cycle FWHM in intensity) was **unrealistic** – it was two cycles between the beginning and the end with very fast ramp-on and ramp off.

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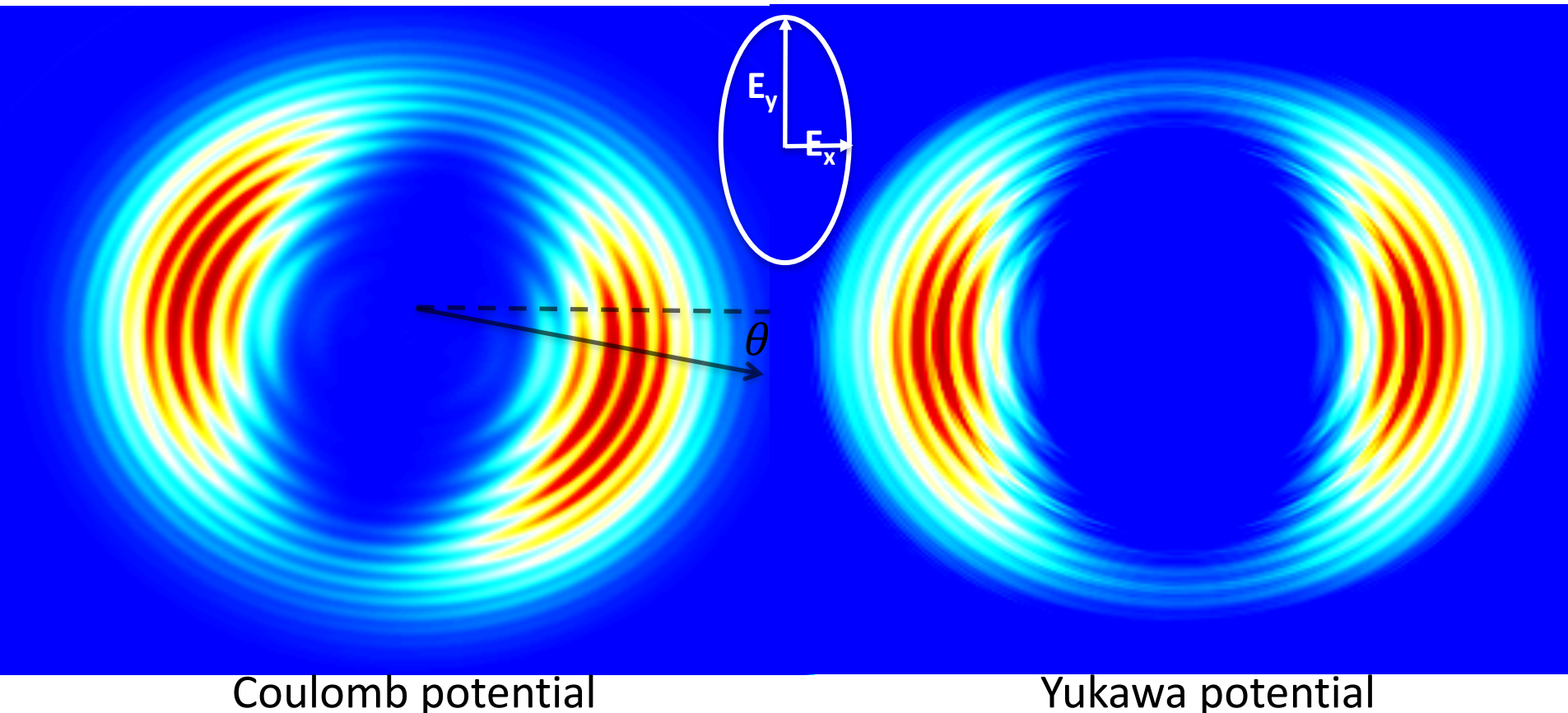
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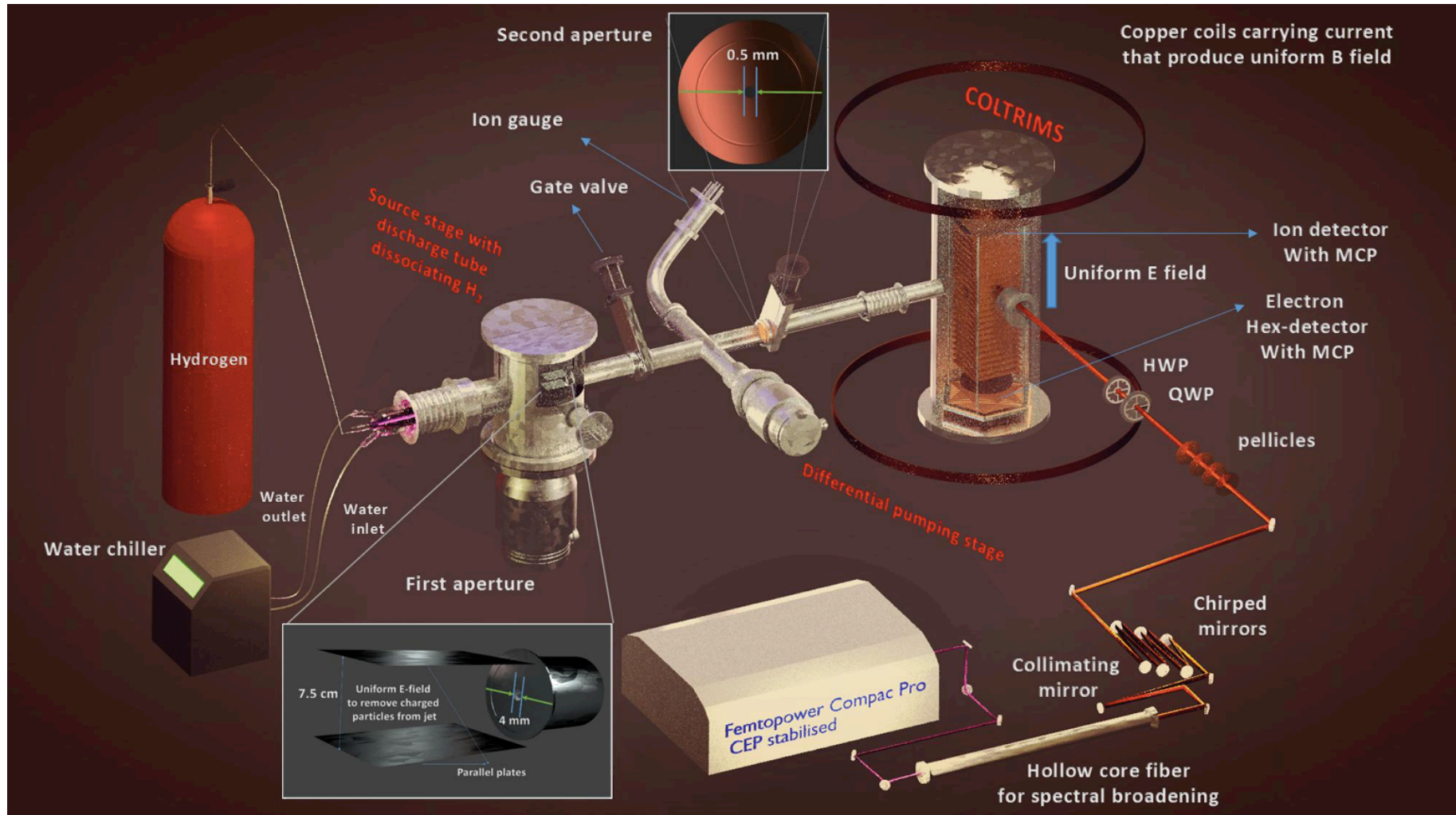
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- They presented a **cut** through the momentum distribution ($p_z = 0$).
- The **offset angle** was defined by the maximum of the momentum distribution (= a **single point in the (p_x, p_y) -plane**).
- **The conclusion of “zero tunneling time” was based on the comparison between the Coulomb and Yukawa calculations.**

Theoretical and Experimental Methods

- We collaborated with other theorists to describe a more realistic experiment performed at **Griffith University**. It uses a **6-cycle (FWHM) pulse** with wavelength $\lambda = 770 \text{ nm}$ and **ellipticity $\varepsilon = 0.84$** . The **CEP is not controlled** and must be averaged over. In the examples below, the peak intensity is $1.4 \times 10^{14} \text{ W/cm}^2$.

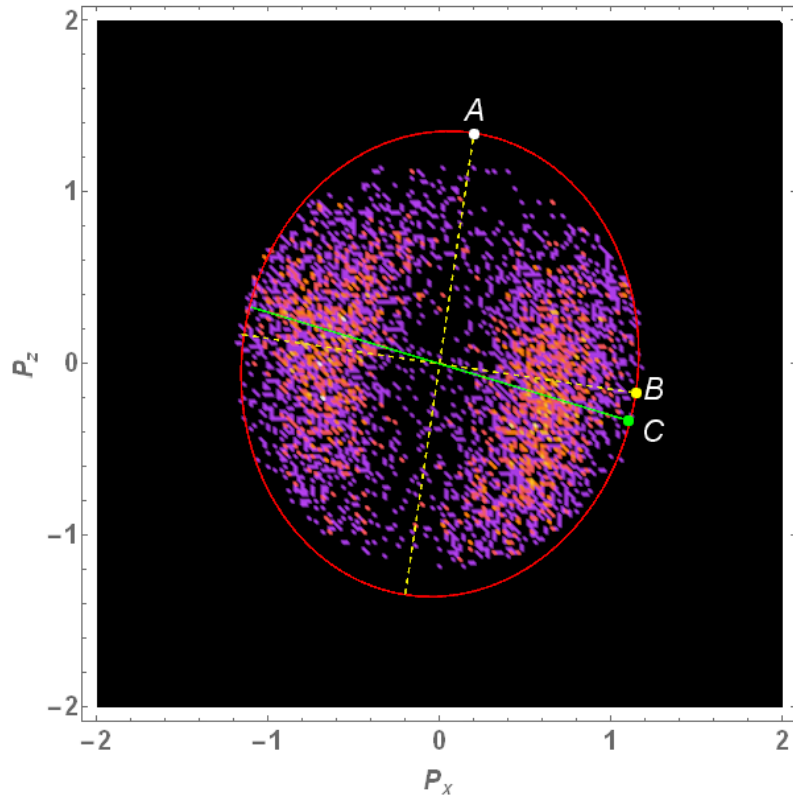


The Real Thing: Experimental Setup

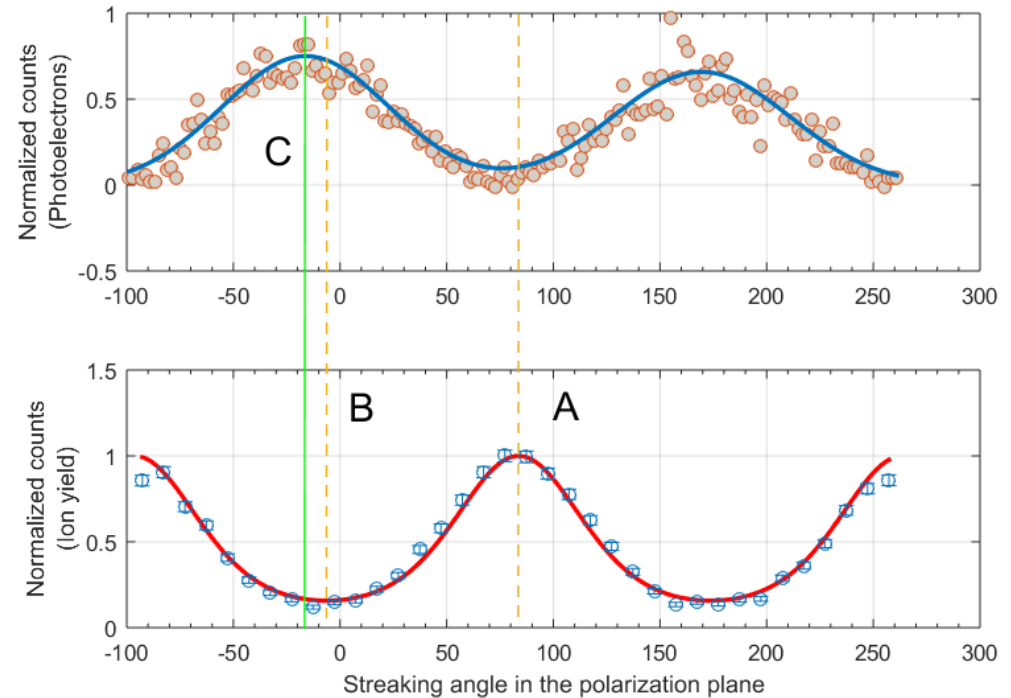


Raw Experimental Data and Processing

(a)



(b)

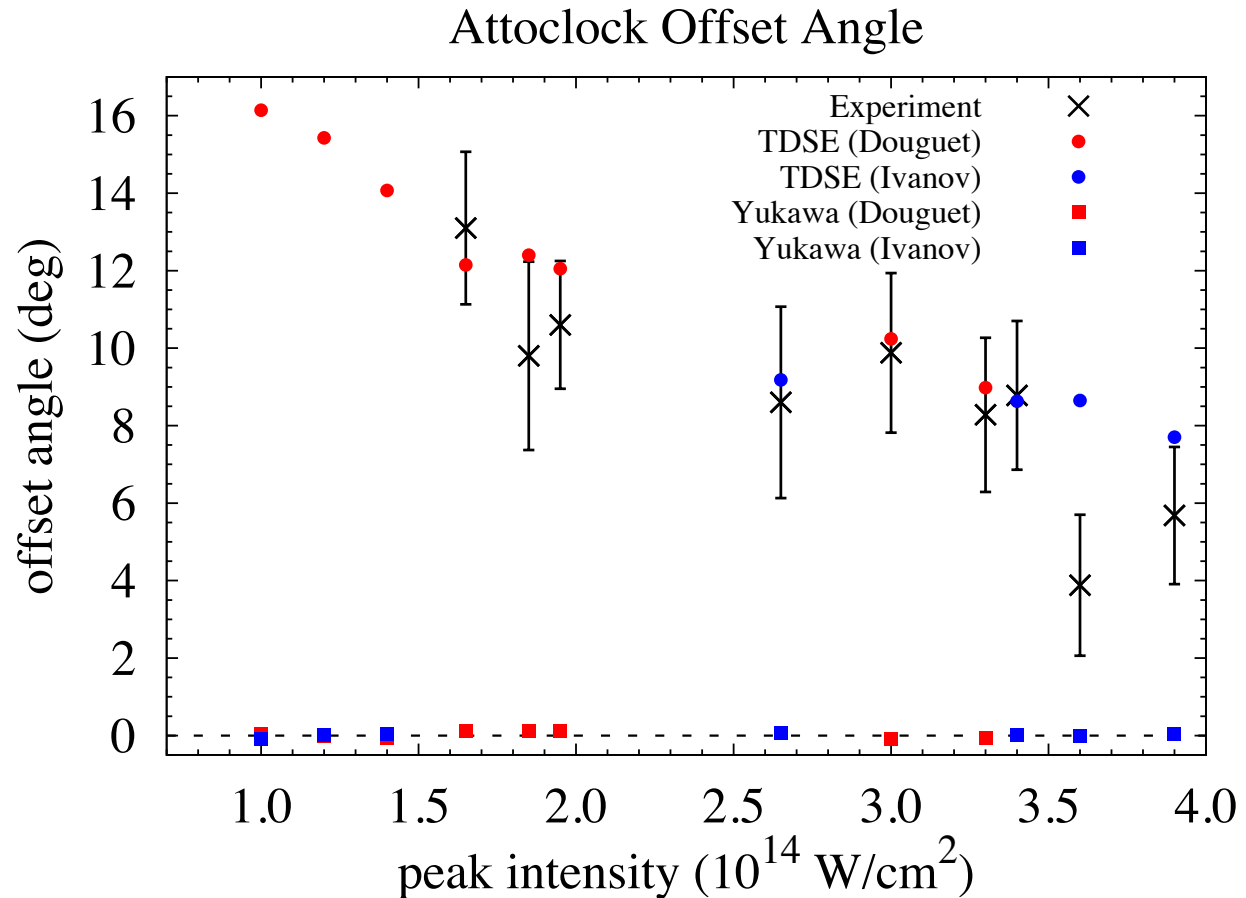
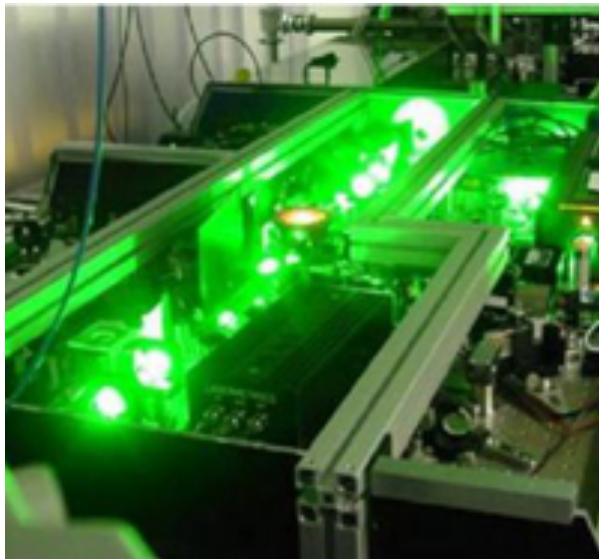


- First the **axes need to be calibrated** to find the **maximum of the E-field**.
- **Without tunneling delay or the long-range Coulomb interaction**, the maximum of the electron distribution would be **expected perpendicular to that direction**.
- **The difference between the actual and the expected angle is the offset.**

Comparison with Experimental Data

(S. Satya, I. Litvinyuk, R. Sang, ...); arXiv 1707.05445

The Australian AttoSecond
Facility at Griffith University



- **Good agreement** was observed between experiment and theory, which provides confidence in both.
- The hope is that the results can be used to **calibrate the attoclock** for future studies on more complex systems.

(finally) published a few days ago (March 18, 2019)

Received: 26 July 2017; Accepted: 7 January 2019;
Published online: 18 March 2019

LETTER

<https://doi.org/10.1038/s41586-019-1028-3>

Attosecond angular streaking and tunnelling time in atomic hydrogen

U. Satya Sainadh¹, Han Xu^{1*}, Xiaoshan Wang², A. Atia-Tul-Noor¹, William C. Wallace¹, Nicolas Douguet^{3,6}, Alexander Bray⁴, Igor Ivanov⁵, Klaus Bartschat³, Anatoli Kheifets⁴, R. T. Sang^{1*} & I. V. Litvinyuk^{1*}

The tunnelling of a particle through a potential barrier is a key feature of quantum mechanics that goes to the core of wave-particle duality. The phenomenon has no counterpart in classical physics, and there are no well constructed dynamical observables that could be used to determine ‘tunnelling times’. The resulting debate^{1–5} about whether a tunnelling quantum particle spends a finite and measurable time under a potential barrier was reignited in recent years by the advent of ultrafast lasers and attosecond metrology⁶. Particularly important is the attosecond angular

field at the moment of ionization. The instant of maximum field thus serves as well-defined ‘time zero’ of the attoclock, while the instant of ionization—which might be regarded as ‘tunnel exit’—is encoded onto the free electron’s momentum. We find that the time interval between those two instances, often interpreted as tunnelling delay, is zero for atomic hydrogen.

Although angular streaking works best with circularly polarized few-cycle pulses, the angle at which the electric field (and hence the tunnelling ionization probability) reaches its maximum depends on

Some Comments about the Evaluation of the Offset Angle

- For the real case of a Coulomb potential, **the result depends (strongly!!!) on the way the offset angle is determined**. Using theoretical predictions, we found a dependence on:
 - the cut-off momentum (last fringe that can be resolved)
 - maximum of the distribution vs. peak in the momentum-integrated spectrum
 - cut ($p_z = 0$) vs. integral over all p_z
 - Pulse parameters: peak intensity, length, ramp-up & ramp-down, CEP

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- **As a result, it seems impossible to predict the offset angle by a simple model that does not account for the above dependencies.**

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 - Pulse parameters: peak intensity, length, ramp-up & ramp-down, CEP
- **As a result, it seems impossible to predict the offset angle by a simple model that does not account for the above dependencies.**
- **In the previous graph, the same procedure was used to process the experimental and theoretical raw data.**

Current Conclusions

- For a sufficiently short-range Yukawa potential (e.g., a range parameter of $1a_0$ for the 1s orbital, the angle is zero, independent of those parameters.
- Hence it appears as if a non-zero offset angle (however it is determined) is entirely due to the long-range Coulomb interaction.

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- For a sufficiently short-range Yukawa potential (e.g., a range parameter of $1a_0$ for the 1s orbital, the angle is zero, independent of those parameters.
- Hence it appears as if a non-zero offset angle (however it is determined) is entirely due to the long-range Coulomb interaction.
- This was also the conclusion of Torlina et al., as well as of Ni et al. (Phys. Rev. A **97** (2018) 013426), who studied a model (reduced-dimension) helium atom with one active electron.

(B) While the velocity criterion $k_{\parallel} = 0$ gives zero tunneling exit time, different position criteria $r = r_{\text{exit}}$ give nonzero tunneling exit time, mainly because the nonadiabatic behavior of tunneling dynamics is not taken into account. The nonzero tunneling exit time was (mis)interpreted in the past as a nonzero time delay in the tunneling process for atoms with a single active electron.

Explain “Tunneling” by Bohmian Mechanics?

- Bohmian Mechanics can be useful in interpreting results obtained in a fully quantum mechanical approach.
- The basic idea (illustrated here in 1D) is the following:

Suppose $\varphi(x, t) = R(x, t) \exp [iS(x, t)]$ is the solution of the TDSE. Then $\rho(x, t) = R(x, t)^2$ is the probability density, $v(x, t)$ is the velocity field, and $V_C(x, t)$ and $V_Q(x, t) = -0.5\Delta R(x, t)/R(x, t)$ are the classical and quantum potentials, respectively.

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- The velocity field can be obtained from the flux and charge densities.
- Bohmian trajectories, labeled by their starting point x_0 , are calculated as in Classical Mechanics with $v_0 = 0$.
- The quantum potential allows for motion in the classically forbidden region.

Dynamics of tunneling ionization using Bohmian mechanics

Nicolas Douguet^{1,2} and Klaus Bartschat¹

¹*Department of Physics and Astronomy, Drake University, Des Moines, Iowa 50311, USA*

²*Department of Physics, University of Central Florida, Orlando, Florida 32816, USA*

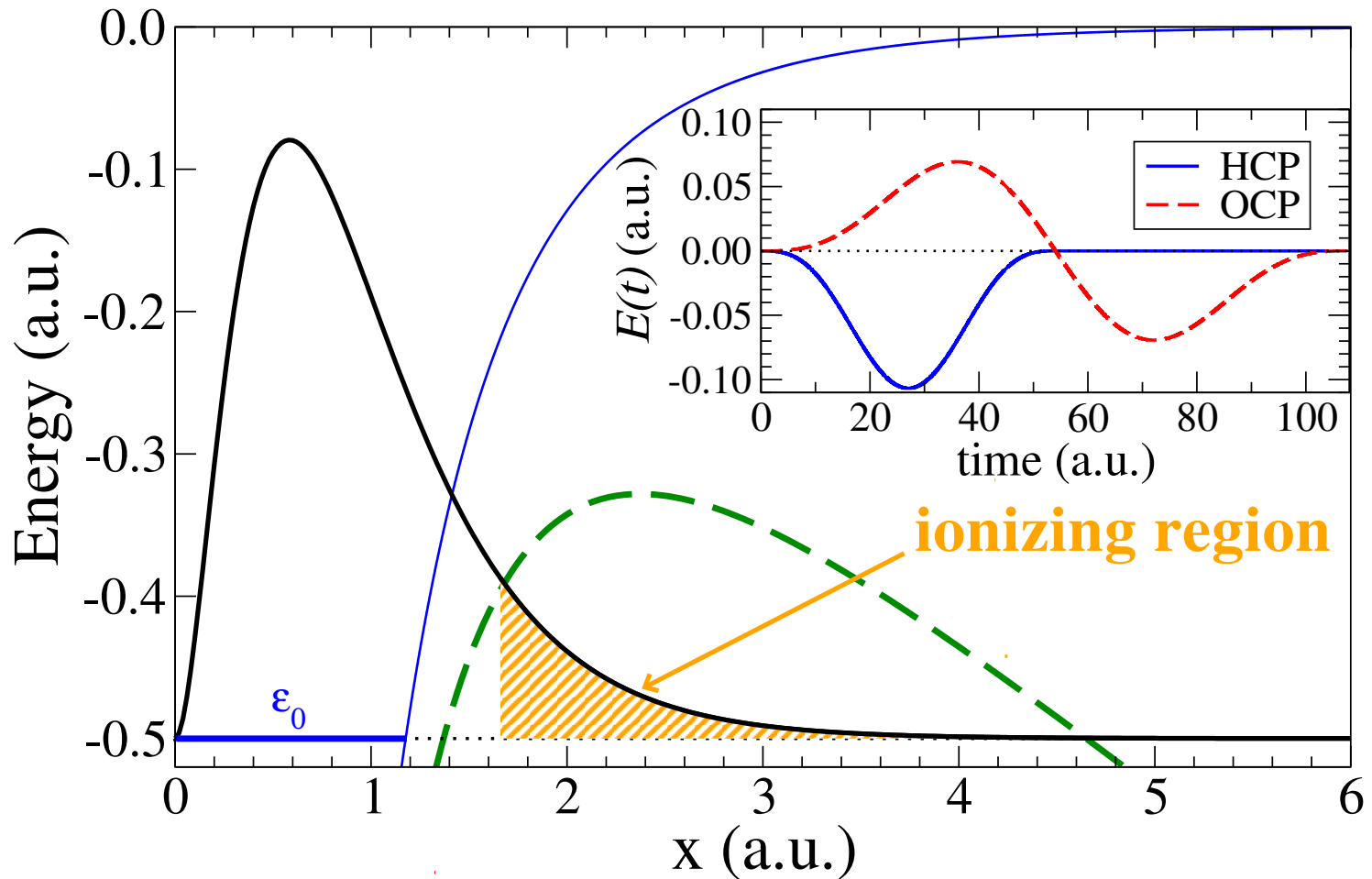


(Received 20 April 2017; published 8 January 2018)

Recent attoclock experiments and theoretical studies regarding the strong-field ionization of atoms by few-cycle infrared pulses revealed features that have attracted much attention. Here we investigate tunneling ionization and the dynamics of the electron probability using Bohmian mechanics. We consider a one-dimensional problem to illustrate the underlying mechanisms of the ionization process. It is revealed that in the major part of the below-the-barrier ionization regime, in an intense and short infrared pulse, the electron does not tunnel through the entire barrier, but rather starts already from the classically forbidden region. Moreover, we highlight the correspondence between the probability of locating the electron at a particular initial position and its asymptotic momentum. Bohmian mechanics also provides a natural definition of mean tunneling time and exit position, taking account of the time dependence of the barrier. Finally, we find that the electron can exit the barrier with significant kinetic energy, thereby corroborating the results of a recent study [N. Camus *et al.*, [Phys. Rev. Lett.](#) **119**, 023201 (2017)].

The next few slides show the main results for the 1D Yukawa potential and half- (HCP) or one-cycle (OCP) pulses.

Escaping from the classically forbidden region ...



thin solid blue line: field-free 1D Yukawa potential

green dashed line: potential at maximum field (4×10^{14} W/cm²)

thick black line: ground state probability distribution.

Conclusions from Bohmian Mechanics

- It is unlikely for electrons to tunnel through the entire barrier, unless the intensity gets close to the “over the barrier” value.
- Many of the free electrons seen after the pulse will likely have started already in the classically forbidden regime.

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- These ideas, and their consequences, need to be studied in more realistic cases than in 1D.

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- Many of the free electrons seen after the pulse will likely have started already in the classically forbidden regime.
- Bohmian Mechanics also provides a tool to investigate tunneling times and exit points. [See our manuscript for details.]
- These ideas, and their consequences, need to be studied in more realistic cases than in 1D.
- It is hoped that Bohmian Mechanics (a very popular approach recently) will be able to provide further insight regarding the understanding, and ultimately, **the control of ultrafast dynamics in atoms, molecules, and solids.**

New Proposal:

Negative ions instead of Yukawa

(For neutral targets, Yukawa is wrong for both small and large r)

PHYSICAL REVIEW A **99**, 023417 (2019)

Attoclock setup with negative ions: A possibility for experimental validation

Nicolas Douguet¹ and Klaus Bartschat²

¹*Department of Physics, University of Central Florida, Orlando, Florida 32789, USA*

²*Department of Physics and Astronomy, Drake University, Des Moines, Iowa 50311, USA*



(Received 14 September 2018; revised manuscript received 29 October 2018; published 14 February 2019)

The presumed connection in attoclock setups between the electron tunneling time and its asymptotic momentum has triggered vigorous debates. In neutral atomic systems investigated so far, the action of the long-range Coulomb potential on the electron momentum hinders extracting the effect of the tunneling process on the offset angle of the asymptotic electron momentum. We propose and investigate an attoclock experiment using F^- or Cl^- to circumvent this difficulty. Our calculations, performed with realistic laser parameters in the tunneling regime, could be checked directly against experiment and predict essentially a “zero” offset angle with no detectable effect of polarization.

Specifically: F^- and Cl^-

- Both of these negative ions have electron affinities of about 3.5 eV
- As a result, a wavelength of 1,500 nm is more appropriate
- We also use a realistic pulse length (6 cycles)
- Finally, we include polarization effects due to the field and the ejected electron.
- Such an experiment, and hence a direct test of the theoretical predictions is possible.

$$V(\mathbf{r}, t) = -\frac{Z}{r}e^{-r/a} - \left[\frac{\alpha_d \mathbf{E}(t) \cdot \mathbf{r}}{r^3} + \frac{\alpha_d}{2r^4} \right] \xi_c(r)$$

In contrast to Yukawa, this model contains all the main physics.

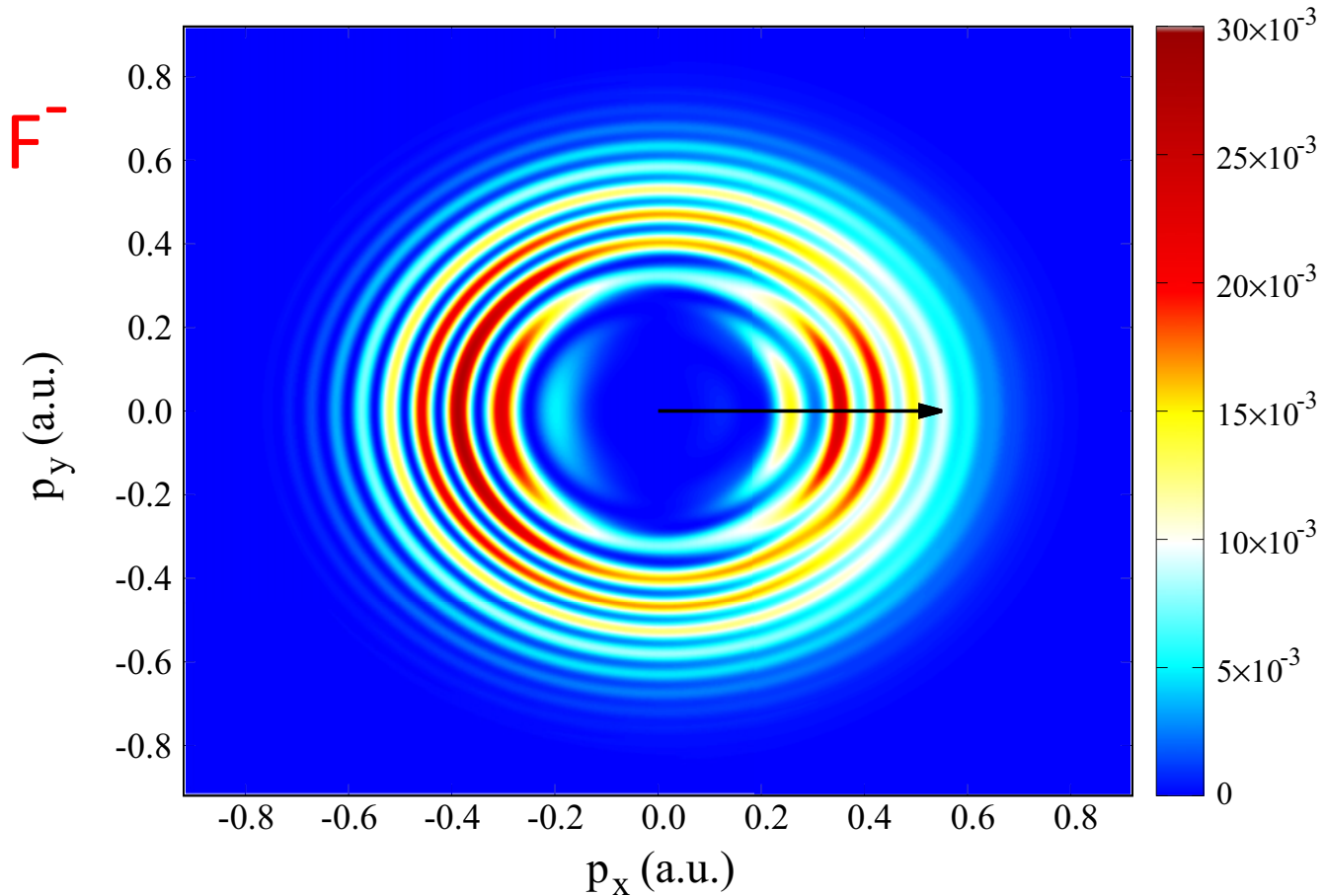


FIG. 2. PMD for a 1500-nm elliptically polarized ($\varepsilon = 0.84$) 6-cycle pulse with a \sin^2 envelope and a peak intensity $I_0 = 10^{13}$ W/cm². The arrow defines the positive x axis.

The situation is much more complex, but the angle remains essentially zero.

The End

THANK YOU FOR YOUR ATTENTION !