# Multi-electron polarization effects and tunneling geometry in atoms and molecules

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# Outline

- Introduction
- Stark shifts and the effective potential
- Attoclock and tunneling geometry
- Conclusions and outlook

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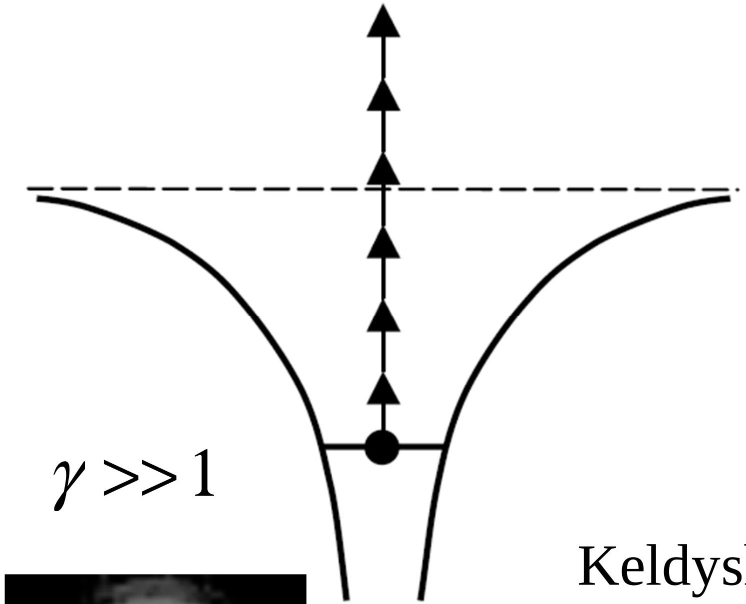
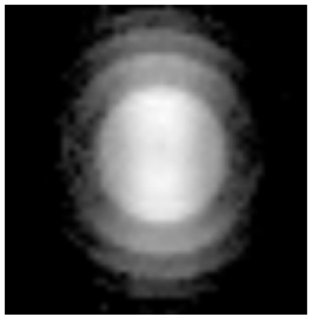
# Basic regimes in strong-field ionization

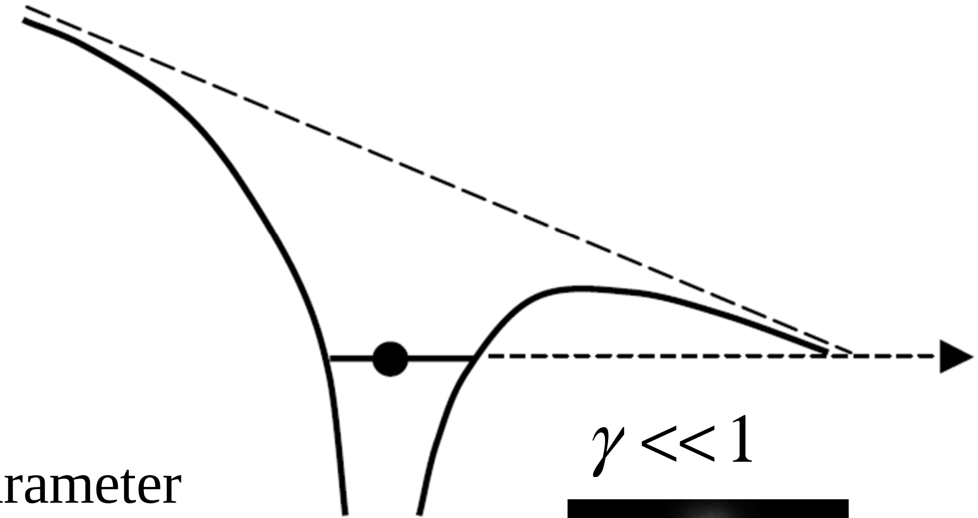
multiphoton regime



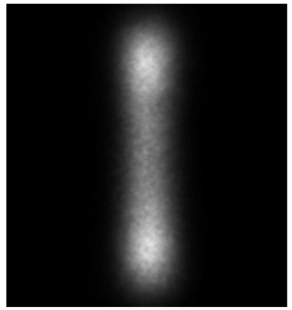
$\gamma >> 1$



tunneling regime



$\gamma << 1$

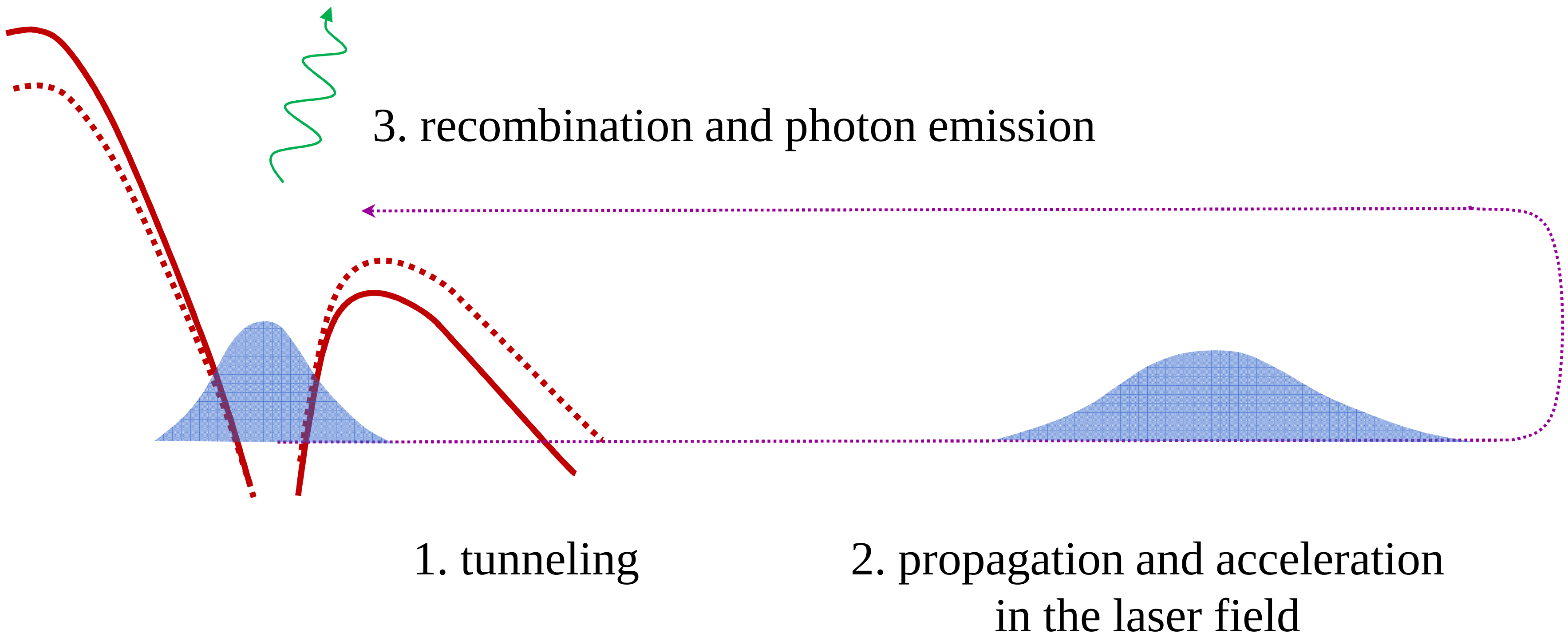


Keldysh parameter

$$\gamma = \frac{\omega\sqrt{2I_p}}{F}$$

[from L. Arissian *et al.*, PRL **105** 133002 (2010)]

# Three-step model, HHG and atosecond pulses




short femtosecond pulse → HHG → production of attosecond pulse

P. B. Corkum, PRL **71** 1994 (1993)

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# Static Stark shifts

To second order in field strength **F**, the total energy of the molecule/ion $E^{M/I}$ is

$$E^{\mathrm{M/I}}(\mathbf{F}) = E^{\mathrm{M/I}}(0) - \boldsymbol{\mu}^{\mathrm{M/I}} \cdot \mathbf{F} - \frac{1}{2}\mathbf{F}^{\top}\boldsymbol{\alpha}^{\mathrm{M/I}}\mathbf{F}$$

$\boldsymbol{\mu}^{M/I}$ –dipole moment $\boldsymbol{\alpha}^{M/I}$ – polarizability tensor
$E^{M/I}$ (0) – field free total energy

Ionization potential

$$I_p(\mathbf{F}) = E^{\mathrm{I}} - E^{\mathrm{M}} = I_p(0) + \boldsymbol{\Delta\mu} \cdot \mathbf{F} + \frac{1}{2}\mathbf{F}^{\top}\boldsymbol{\Delta\alpha}\mathbf{F}$$

$$\boldsymbol{\Delta\mu} = \boldsymbol{\mu}^{\mathrm{M}} - \boldsymbol{\mu}^{\mathrm{I}}, \quad \boldsymbol{\Delta\alpha} = \boldsymbol{\alpha}^{\mathrm{M}} - \boldsymbol{\alpha}^{\mathrm{I}}.$$

Ionization potential becomes function not only of |**F**|, but also on the relative orientation of **F** with respect to $\boldsymbol{\Delta\mu}$ and $\boldsymbol{\Delta\alpha}$.

# Adiabatic ansatz

In cases when the field is slowly-varying, we use the adiabatic approximation.

Then a (many-electron) bound state $|\Psi\rangle$ evolves according to

$$\exp\left(-i\int_{-\infty}^{t} dt' E(\mathbf{F}(t'))\right)|\Psi\rangle$$

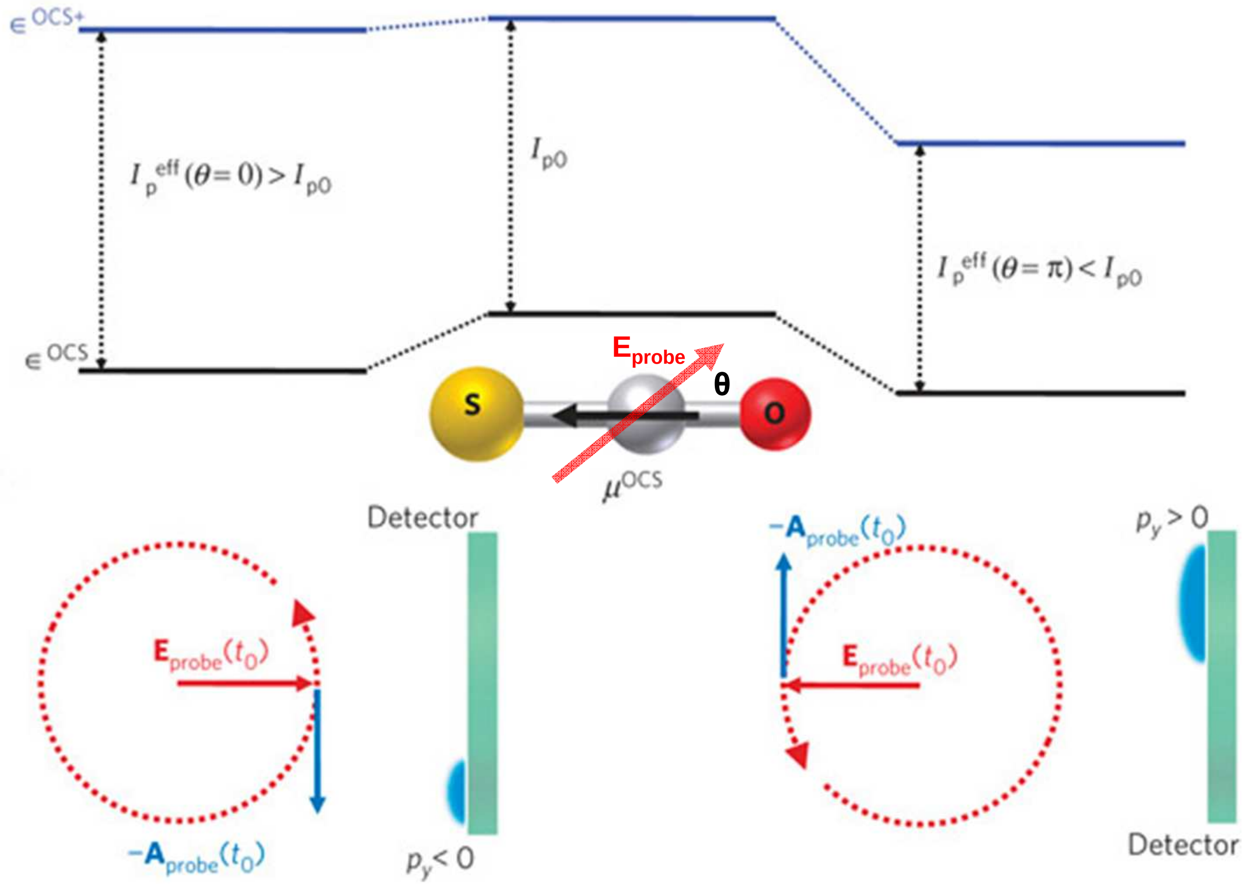
The Stark shifts enter in the exponent of the transition amplitude in the perturbation theory and in the SFA. [D. Dimitrovski *et al.*, PRA **82** 053404 (2010)]

The tunneling theory is corrected to account for the Stark shifts by modifying the binding energy in the tunneling exponent, so that

$$\exp\left(-\frac{2\kappa^3(\mathbf{F})}{3F}\right), \text{ where } \kappa(\mathbf{F}) = \sqrt{2I_p(\mathbf{F})}$$

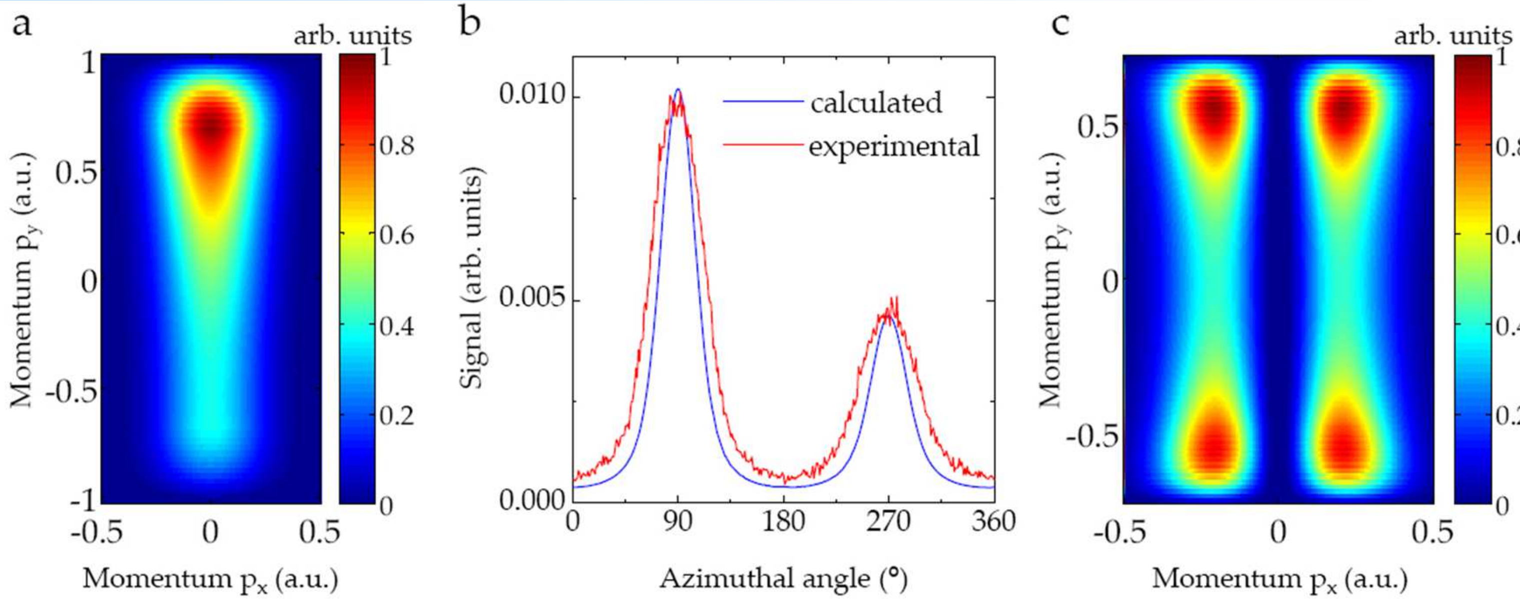
D. Dimitrovski *et al.*, PRA **83** 0523405 (2010)

# Asymmetry



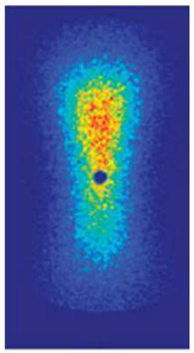

L. Holmegaard *et al*., Nat. Phys. **6**, 428 (2010)

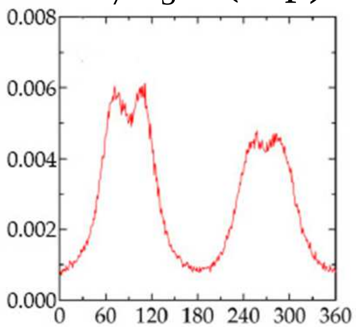
# Comparison between theory and experiment

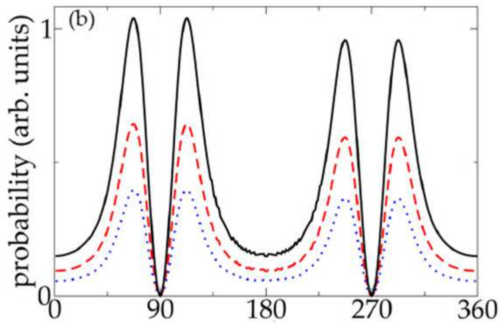


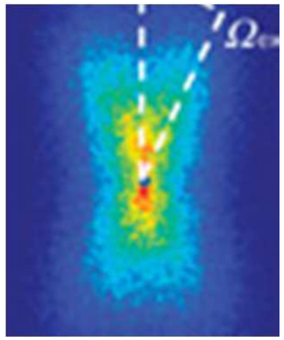

$C_7H_5N$ (exp)

$C_7H_5N$ (theo)







$\Omega_{exp}$=18°±1°

asymmetry for OCS: 0.651 (0.64)

asymmetry for $C_7H_5N$: 0.52 (0.55)

$$\Omega_{\text{theo}} = \arctan(2\omega/\sqrt{\pi F_0 \kappa}) \simeq 18.8^{\circ}$$

# Multielectron effects

Time-independent Schrödinger equation for a n-electron system

$$E^M(\mathbf{F})\Psi_n = \left(\sum_{j=1}^{n}(H_j + \mathbf{r}_j \cdot \mathbf{F}) + V_{ee}^n\right)\Psi_n$$ , where

$$H_j = -(1/2)\nabla_j^2 - \sum_{i=1}^{k} Z_i/|\mathbf{R}_i - \mathbf{r}_j|$$

$\mathbf{R}_i$ - nuclei coordinates, $Z_i$ - nuclei charges.

$V_{ee}^n = \sum_{l<j}^{n} 1/|\mathbf{r}_l - \mathbf{r}_j|$ - electron-electron interaction

Born-Oppenheimer-like ansatz is employed to decouple the motion of

(a) the residual, fast electrons – coordinates $\mathbf{r}_1, \ldots, \mathbf{r}_{n-1}$

(b) the slow electron that tunnels out – coordinate $\mathbf{r}_n$

T. Brabec *et al.*, PRL **95**, 073001 (2005)

D. Dimitrovski *et al.*, PRA **82**, 053404 (2010)

# Decoupling of slow and fast electronic coordinates

$$\Psi_n(\mathbf{r}_1, ..., \mathbf{r}_{n-1}, \mathbf{r}_n) = \Psi_{n-1}(\mathbf{r}_1, ..., \mathbf{r}_{n-1}; \mathbf{r}_n) \otimes \Psi_t(\mathbf{r}_n)$$

$\mathbf{r}_n$ is adiabatic parameter in $\Psi_{n-1}(\mathbf{r}_1, ..., \mathbf{r}_{n-1}; \mathbf{r}_n)$

The equation for the tunneling electron is

$$-I_p(\mathbf{F})\Psi_t = \left(-\frac{1}{2}\boldsymbol{\nabla}^2 + V_{ef}(\mathbf{r}_n; \mathbf{F}) + \mathbf{r}_n \cdot \mathbf{F}\right)\Psi_t$$

$V_{ef}$ is the effective potential

# The effective potential

$$V_{ef}(\mathbf{r}_n;\mathbf{F}) = -\frac{Z}{r_n} - \frac{(\boldsymbol{\mu}_T^I + \boldsymbol{\mu}_{ind}^I)\cdot\mathbf{r}_n}{r_n^3} + \frac{1}{2}\mathbf{d}^T\boldsymbol{\alpha}^I\mathbf{d}$$

$Z = m - n + 1$ - ion charge $m = \sum_{i=1}^{k} Z_i$ - nuclear charge

$\mathbf{d} = \mathbf{r}_n / r_n^3$ is the effective field felt by the residual *n-1* electrons

from the action of the outgoing tunneled electron.

$\boldsymbol{\mu}_T^I = \boldsymbol{\mu}^I + \frac{(\sum_{i=1}^{k} Z_i\mathbf{R}_i)\cdot\mathbf{r}_n}{r_n^3}$ permanent dipole of the ion

$\boldsymbol{\mu}_{\text{ind}}^I = \boldsymbol{\alpha}^I\mathbf{F}$, induced dipole of the ion

D. Dimitrovski *et al.*, PRA **82**, 053404 (2010)

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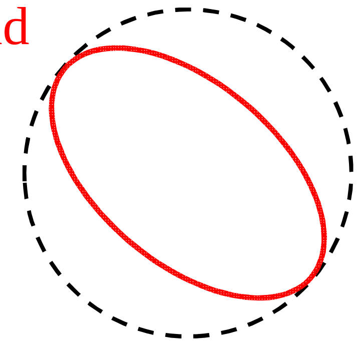
# Attoclock principle

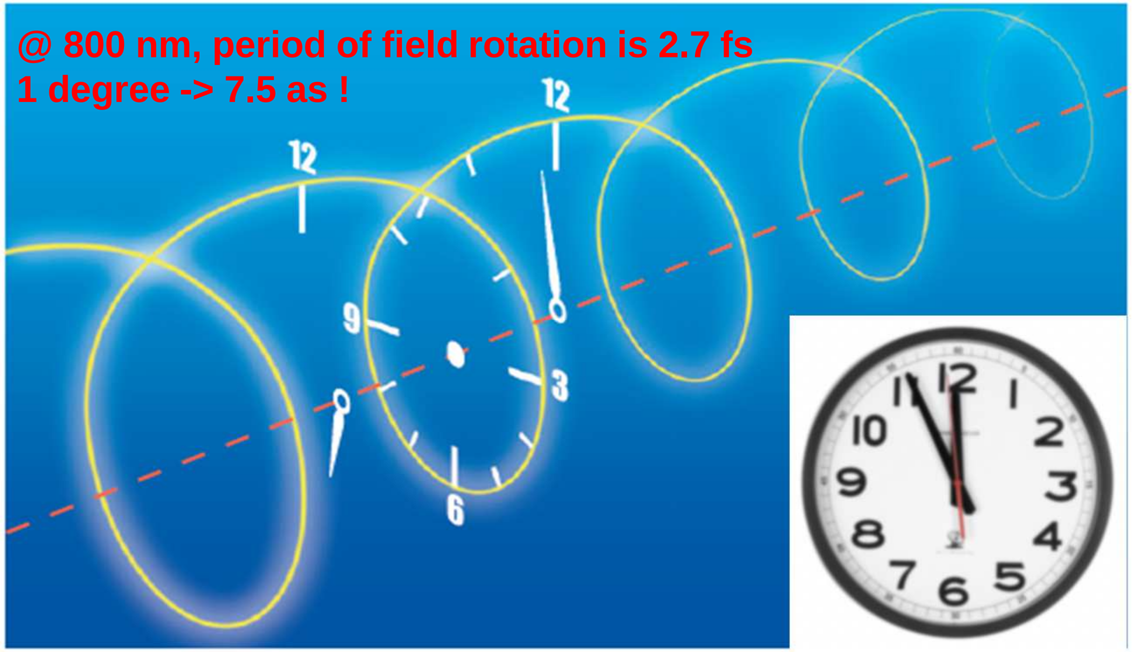
Attoclock: angular streaking technique



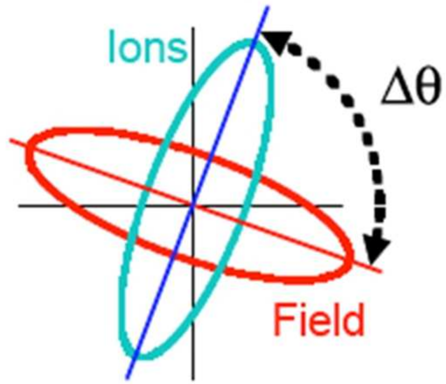

involves *single* near-circularly polarized femtosecond pulse

$$\mathbf{F}(t) = f(t)\left[\frac{1}{\sqrt{1+\varepsilon^2}}\cos(\omega t + \varphi_{CEO})\hat{\mathbf{e}}_x + \frac{\varepsilon}{\sqrt{1+\varepsilon^2}}\sin(\omega t + \varphi_{CEO})\hat{\mathbf{e}}_y\right]$$

time zero: time when the electric field points along the major axis



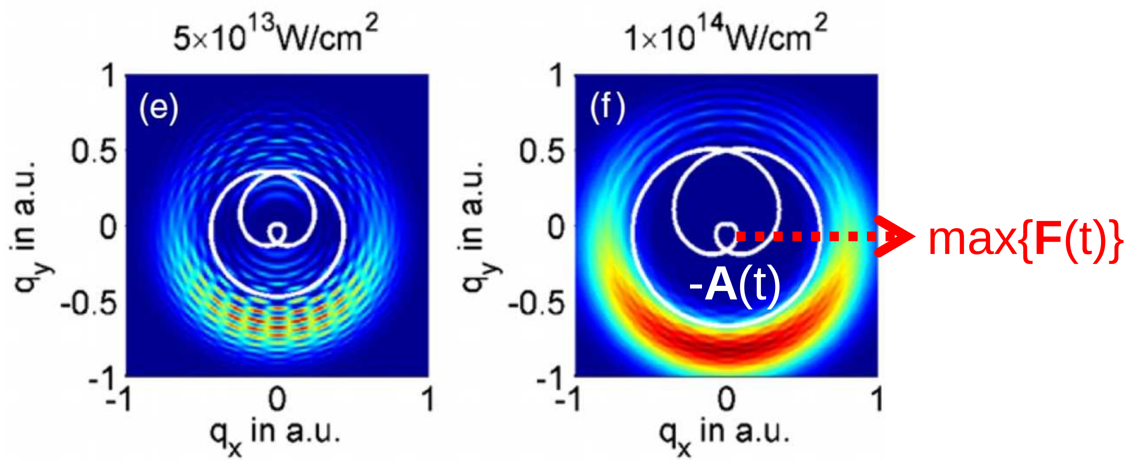

Measurement:
Momentum distribution of ions (electrons) in the polarization plane



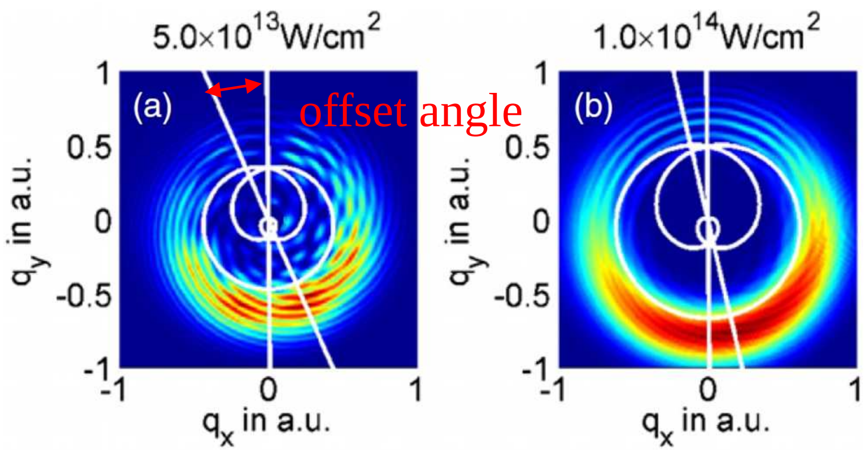

[from A. N. Pfeiffer *et al.*, Nat. Phys. 7, 428 (2011)]

# Offset angle

Without the Coulomb potential

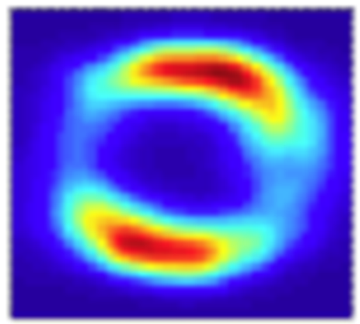



With the Coulomb potential

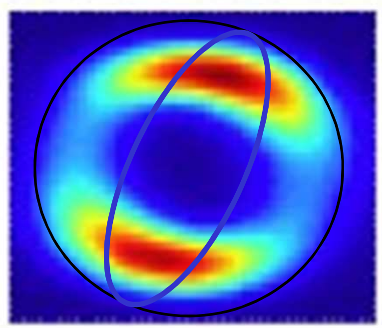



[from J. Phys. B 42 061001 (2009)]

Experimental momentum distribution



Maximum search



# Attoclock experiment

$\varepsilon \approx 0.8$-elipticity

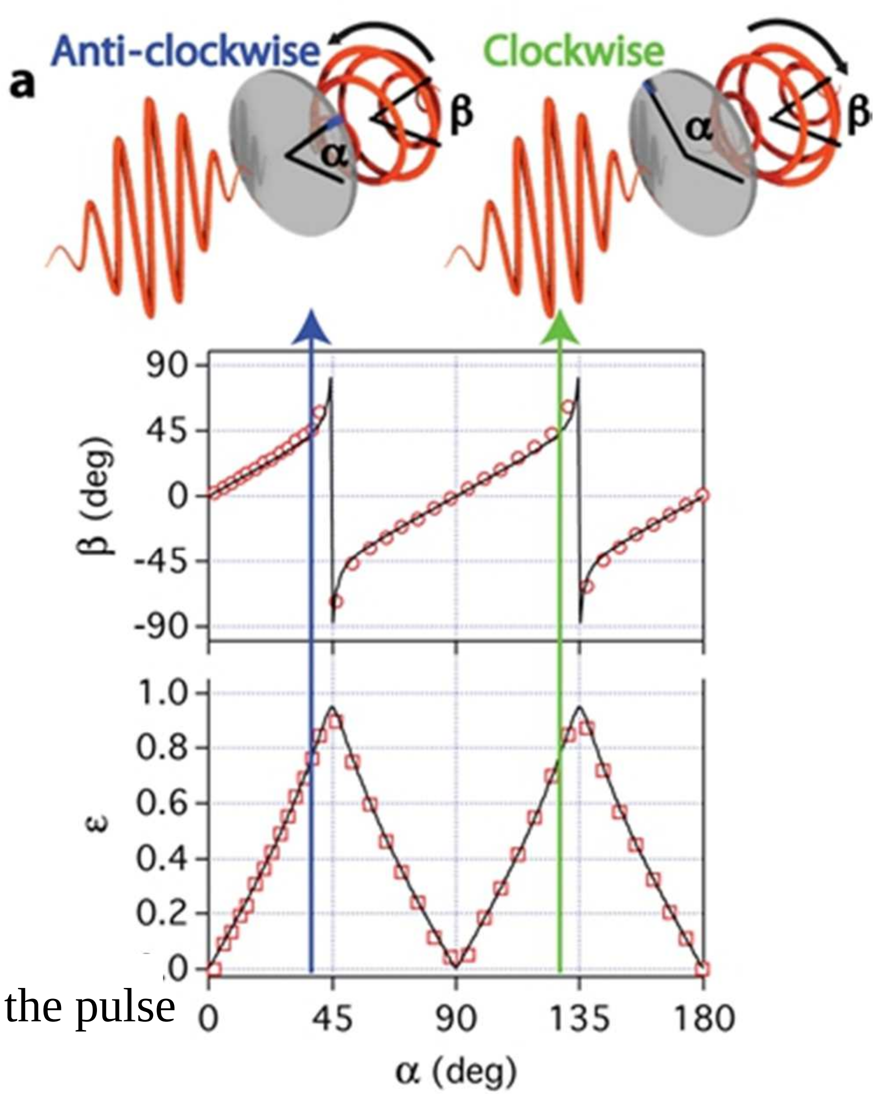
Pulse duration 7 fs, wavelength 740 nm.

Intensities $10^{14}$-$10^{15}$ W/cm$^2$

Atomic targets He and Ar

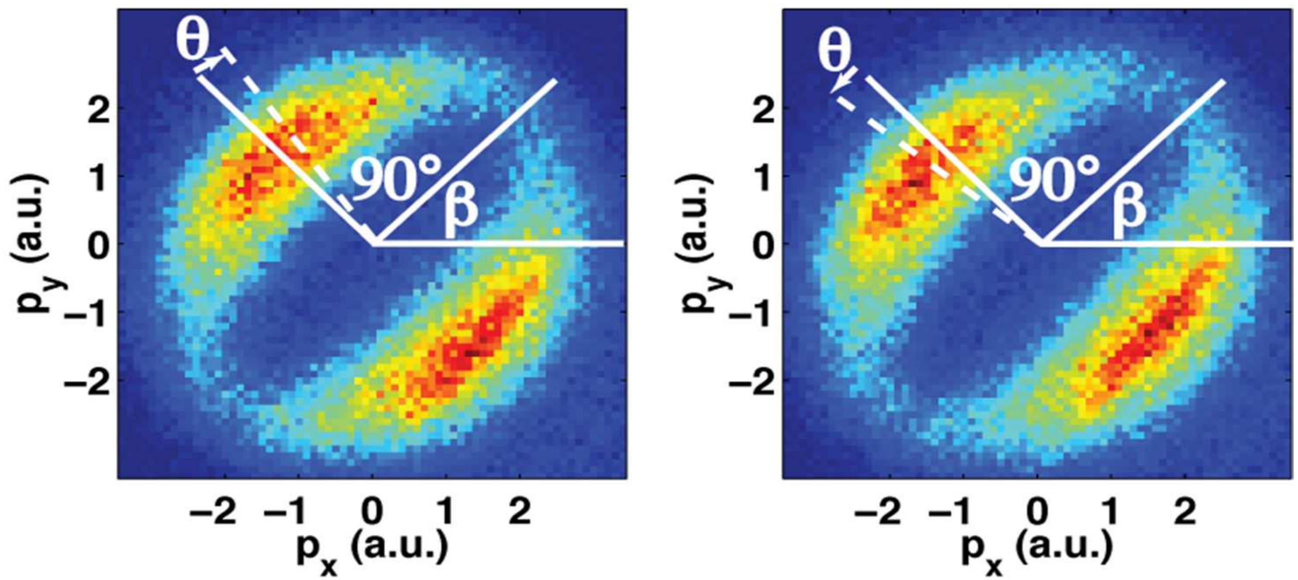
The position of the maximum of field ellipse is obtained independently

$\beta$ - defines the orientation of the major axis of the pulse



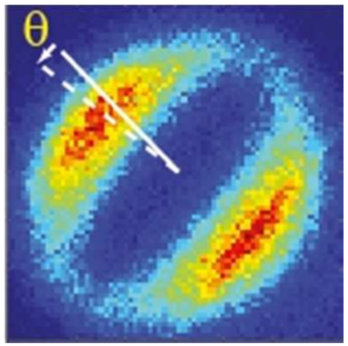

A. N. Pfeiffer *et al.*, Nat. Phys. (accepted 2011), DOI:1038/NPHYS2125

# Extracting the experimental offset angle

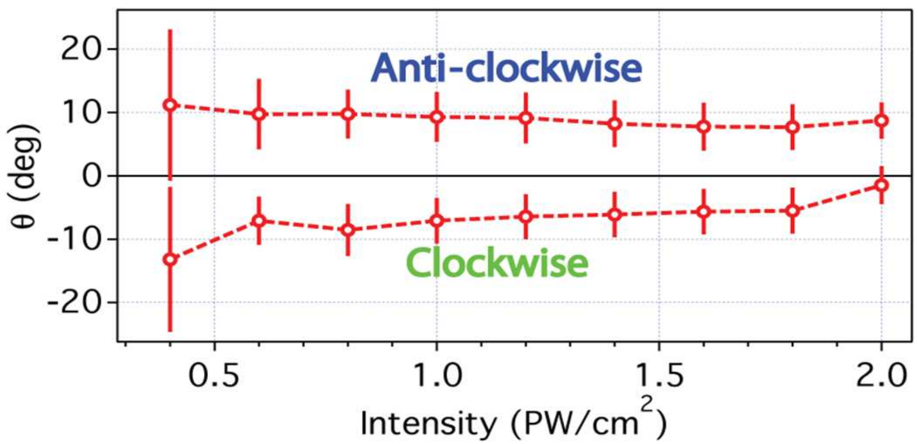



Clockwise

Anti-clockwise

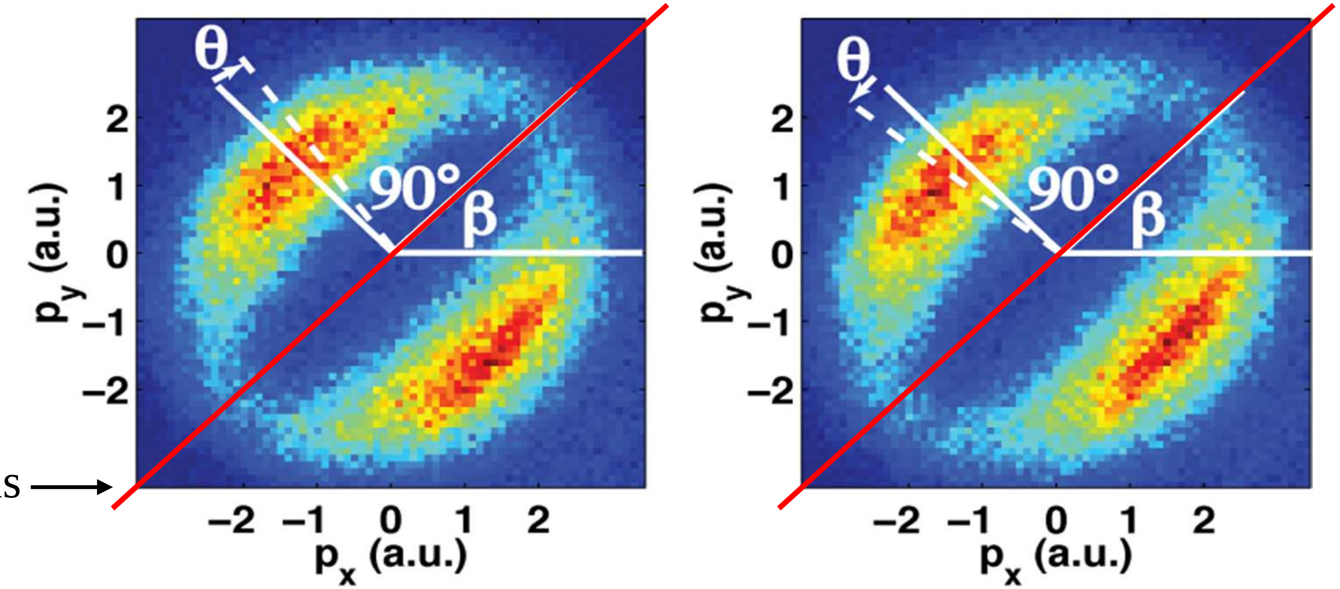



θ – offset angle



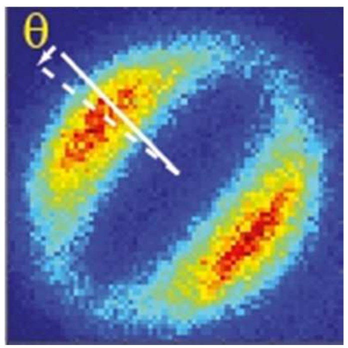
# Extracting the experimental offset angle

major axis →

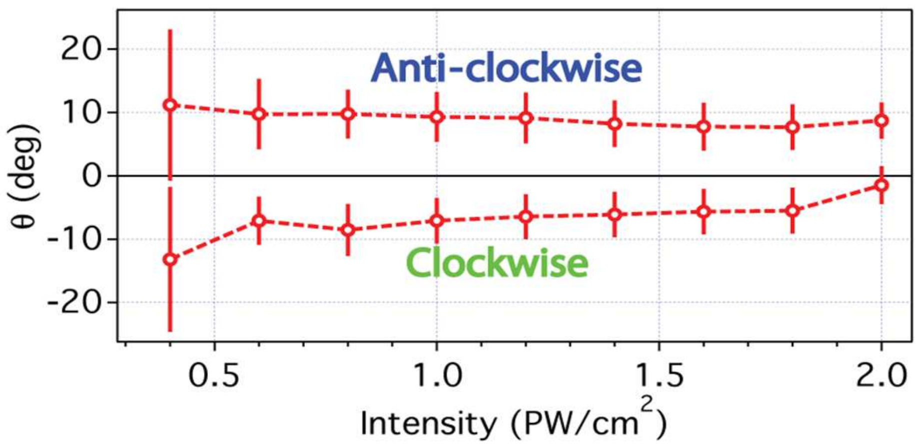



Clockwise

Anti-clockwise




θ – offset angle



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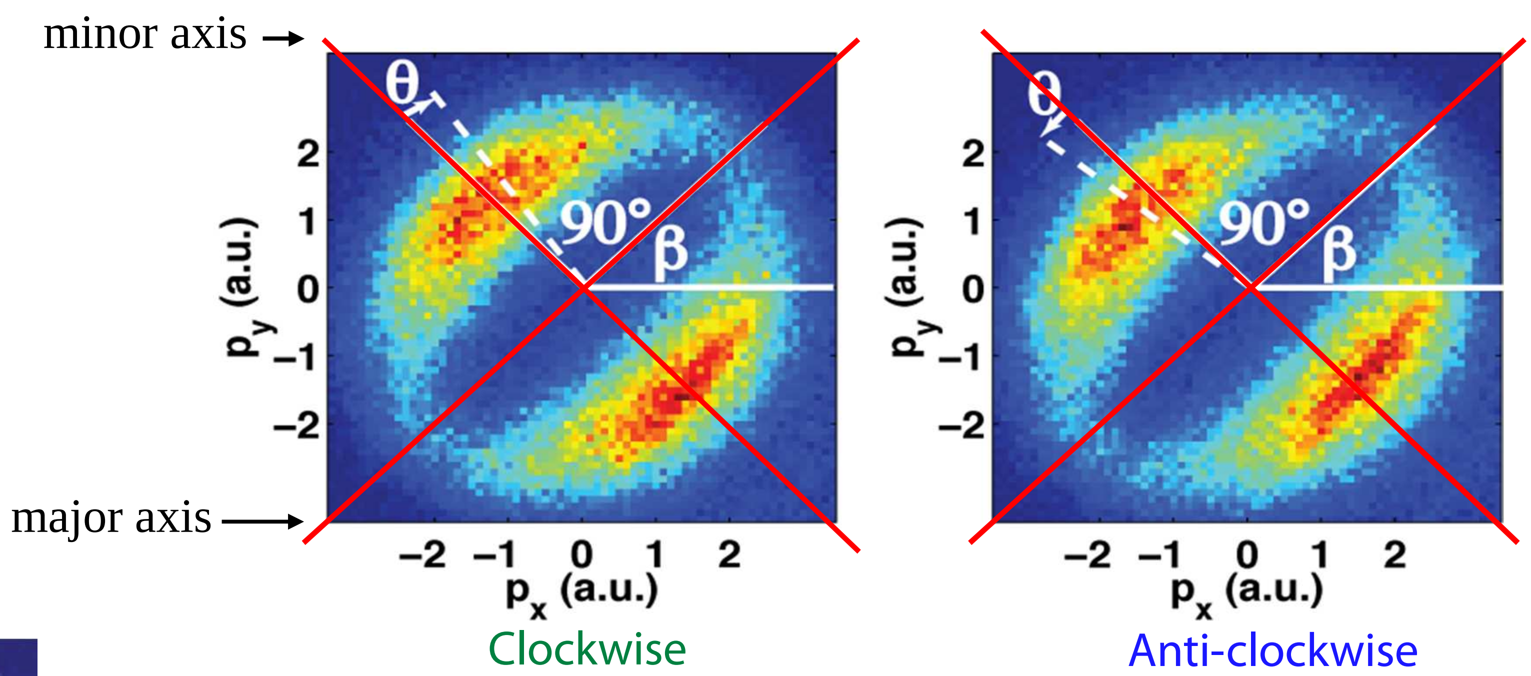
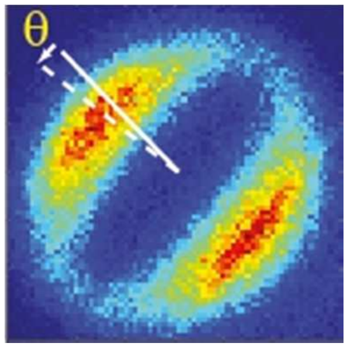
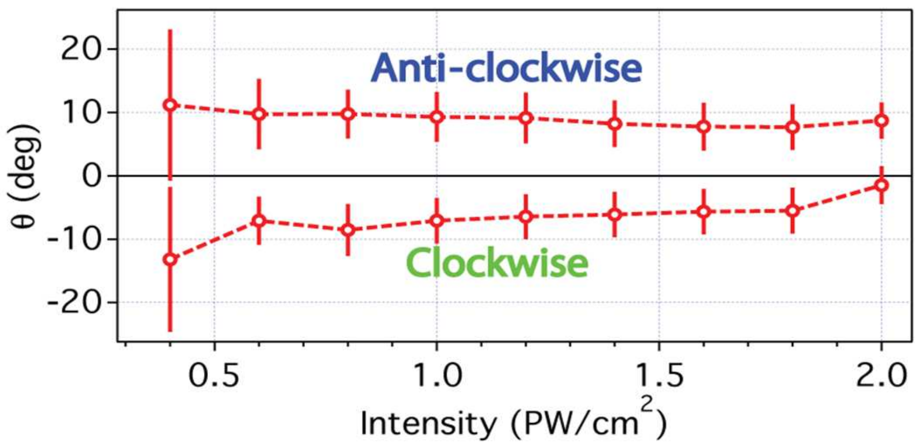



θ – offset angle



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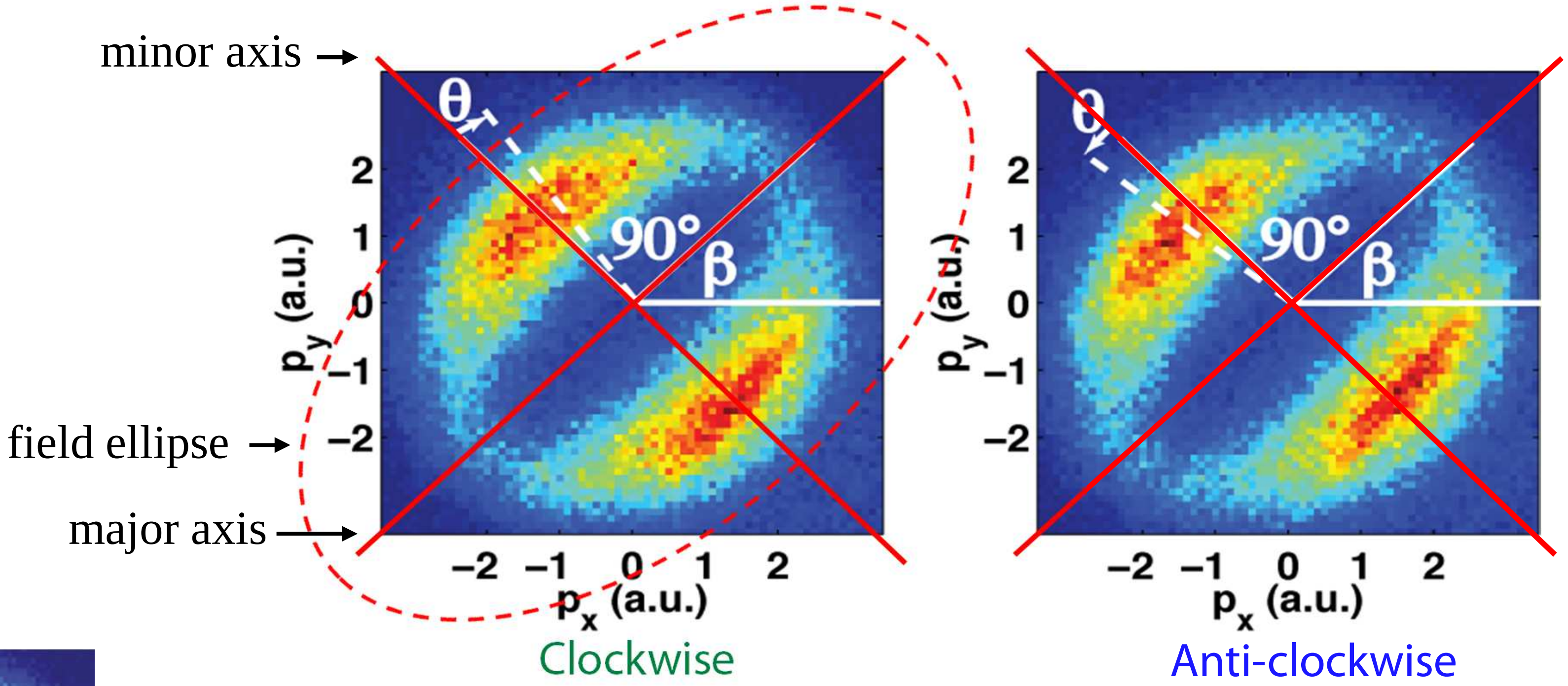
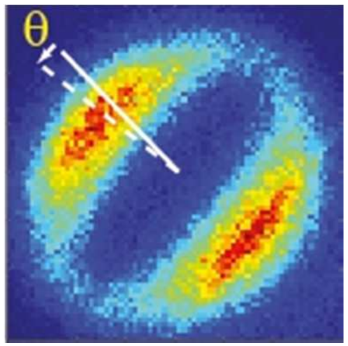
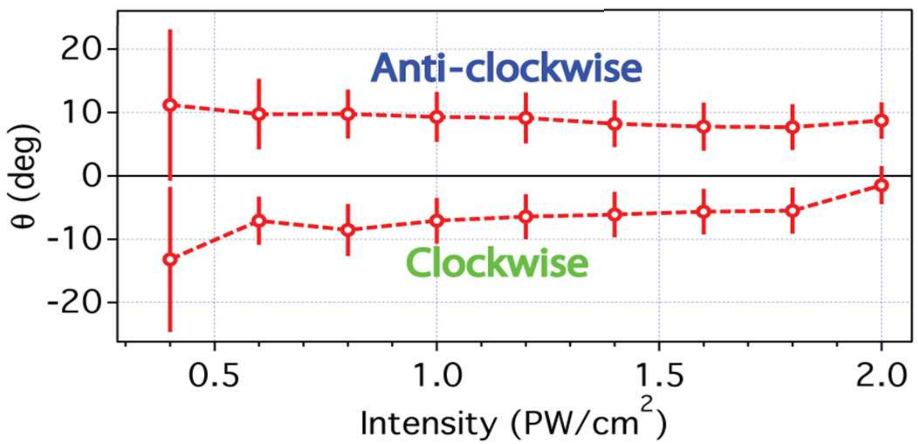



θ – offset angle

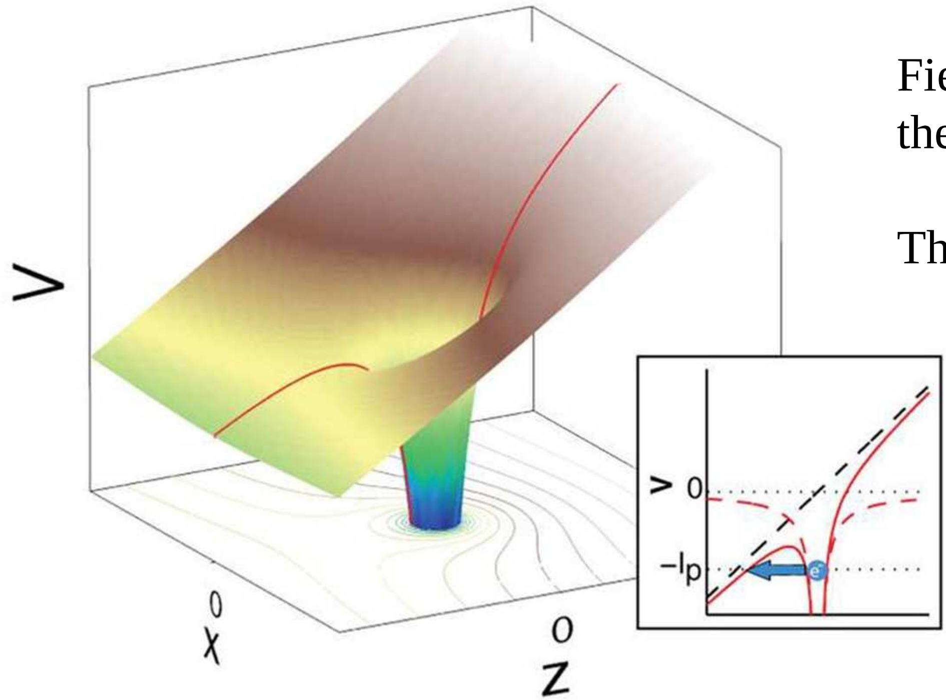


# Mapping emission time to offset angle

Mapping can be accomplished in the tunneling regime, using a semiclassical model.

First step tunneling, then propagation of classical trajectories $$\frac{d^2\mathbf{r}}{dt^2} = -\mathbf{F}(t) - \nabla(V_{ef}(\mathbf{r}))$$

Advantage: no rescattering with the parent ion for circularly and near-circularly polarized pulses




Field-direction model: only the potential along the field direction is considered.

The tunnel exit point is found from

$$F(t)z + V_{ef}(z) = -I_p$$

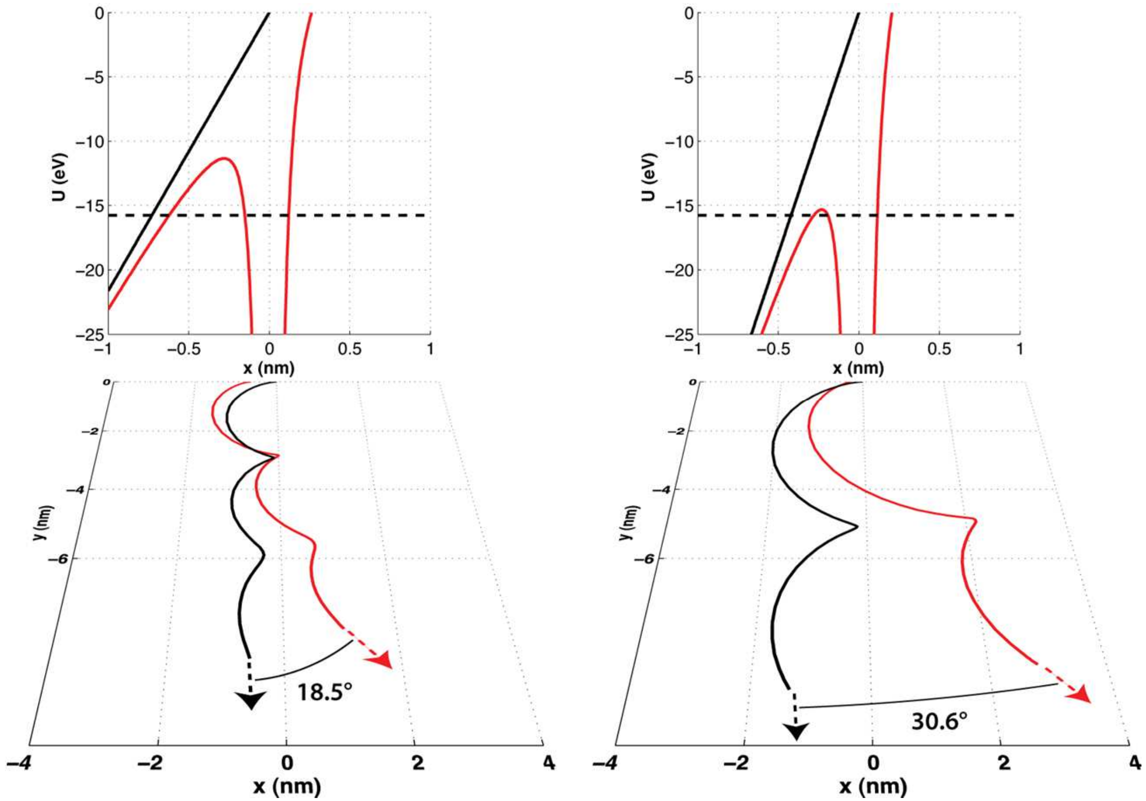
The underlying assumption: the potential in the transverse direction is constant.

# Field-direction model: trajectories

Weak fields: increasing intensity, the offset angle $\theta$ decreases.

Strong fields: increasing intensity, the offset angle $\theta$ increases.

The opposite trend is observed in the experiment.




well below the barrier

close to over the barrier (OBI)

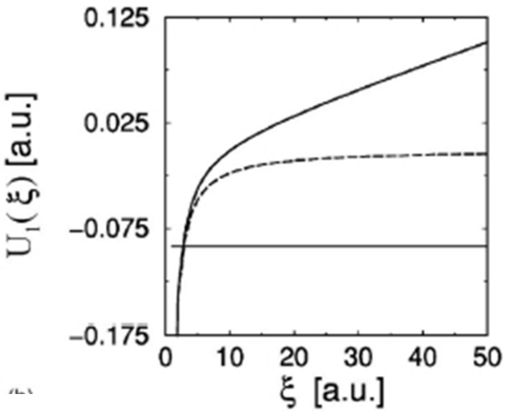
# Parabolic coordinates, separated problem

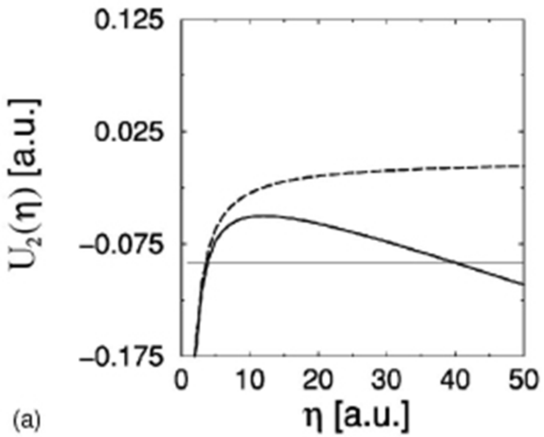
$$\left(-\frac{1}{2}\Delta-\frac{Z}{r}+\mathbf{F}\cdot\mathbf{r}\right)\psi=-I_p\psi$$

separates in parabolic coordinates

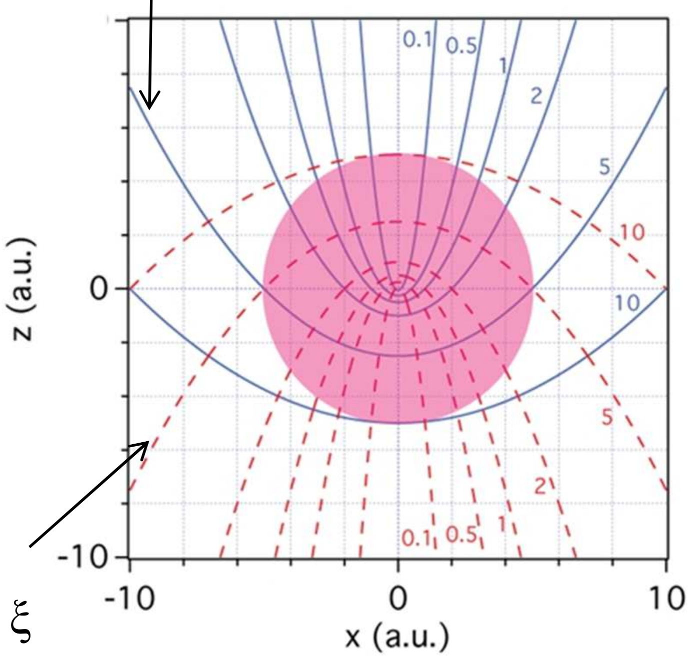
$$\xi = r + z$$
$$\eta = r - z$$
$$\phi = \arctan(y/x)$$

Analytic treatment of tunneling in 1D barrier along η




Lines of constant η




Lines of constant ξ

# Parabolic coordinates, induced dipole term

$$\left(-\frac{1}{2}\Delta + V_{ef}(\mathbf{r},\mathbf{F}) + \mathbf{F}\cdot\mathbf{r}\right)\psi = -I_p(\mathbf{F})\psi$$

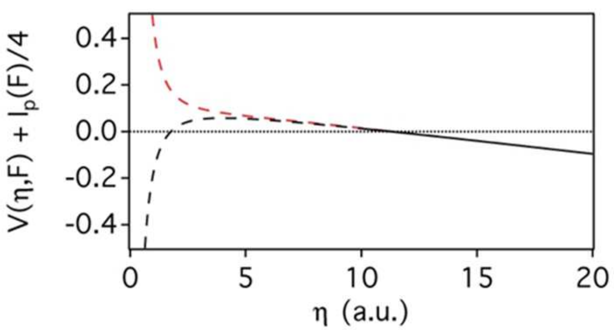
does not separate in parabolic coordinates

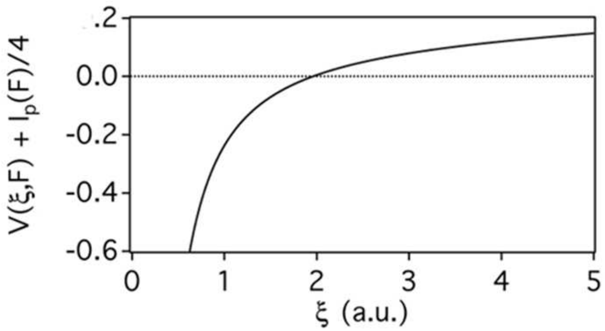
For atoms, $$V_{ef}(\mathbf{r},\mathbf{F}) = -\frac{Z}{r} - \frac{\alpha^I \mathbf{F}\cdot\mathbf{r}}{r^3}$$

Expanding $V_{ef}$(F,r) in the limit ξ/η<<1, the separation is possible.

The η part of the wave function satisfies $$\frac{d^2 f(\eta)}{d\eta^2} + 2\left(-\frac{I_p(F)}{4} - V(\eta;F)\right) f(\eta) = 0$$

$$V(\eta,F) = -\frac{(1-\sqrt{2I_p(F)}/2)}{2\eta} - \frac{1}{8}\eta F + \frac{m^2-1}{8\eta^2} + \frac{\alpha^I F}{\eta^2}$$



# T.I.P.I.S

Two novel elements included

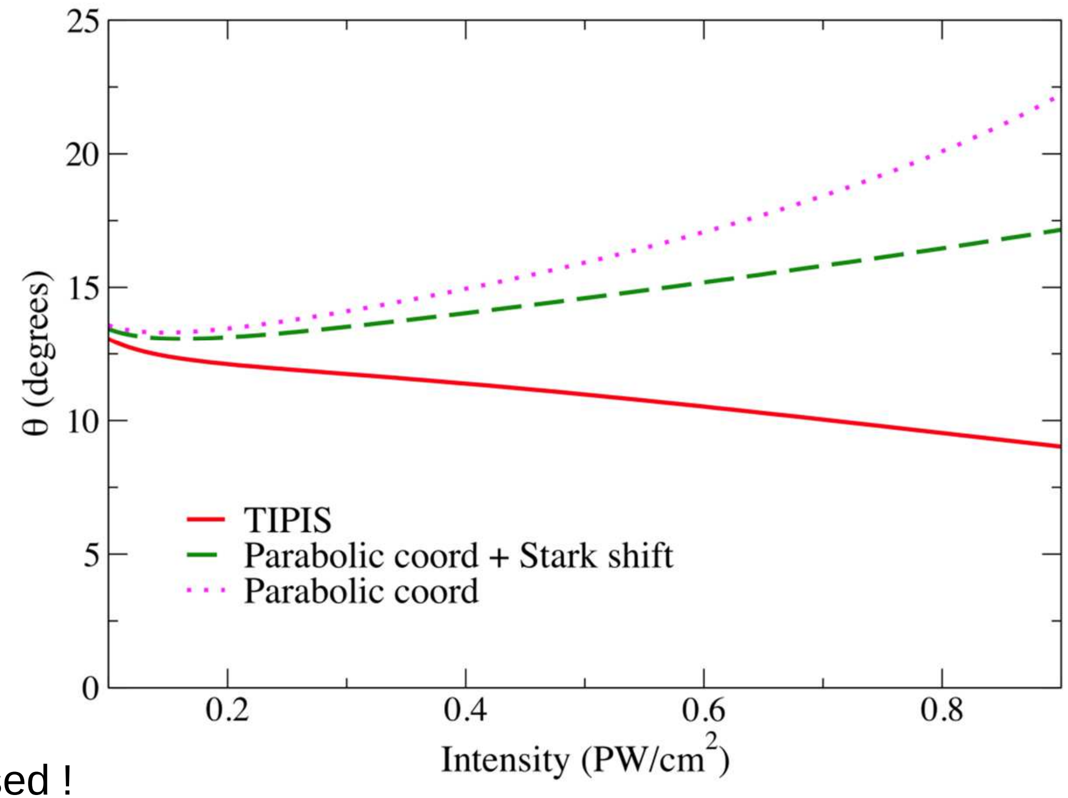
1. Stark shifts

$$I_p(F) = I_p(0) + \frac{1}{2}(\alpha^N - \alpha^I)F^2 > I_p(0)$$

2. increase of the η- barrier due to the induced dipole of the ion

find the exit point in η

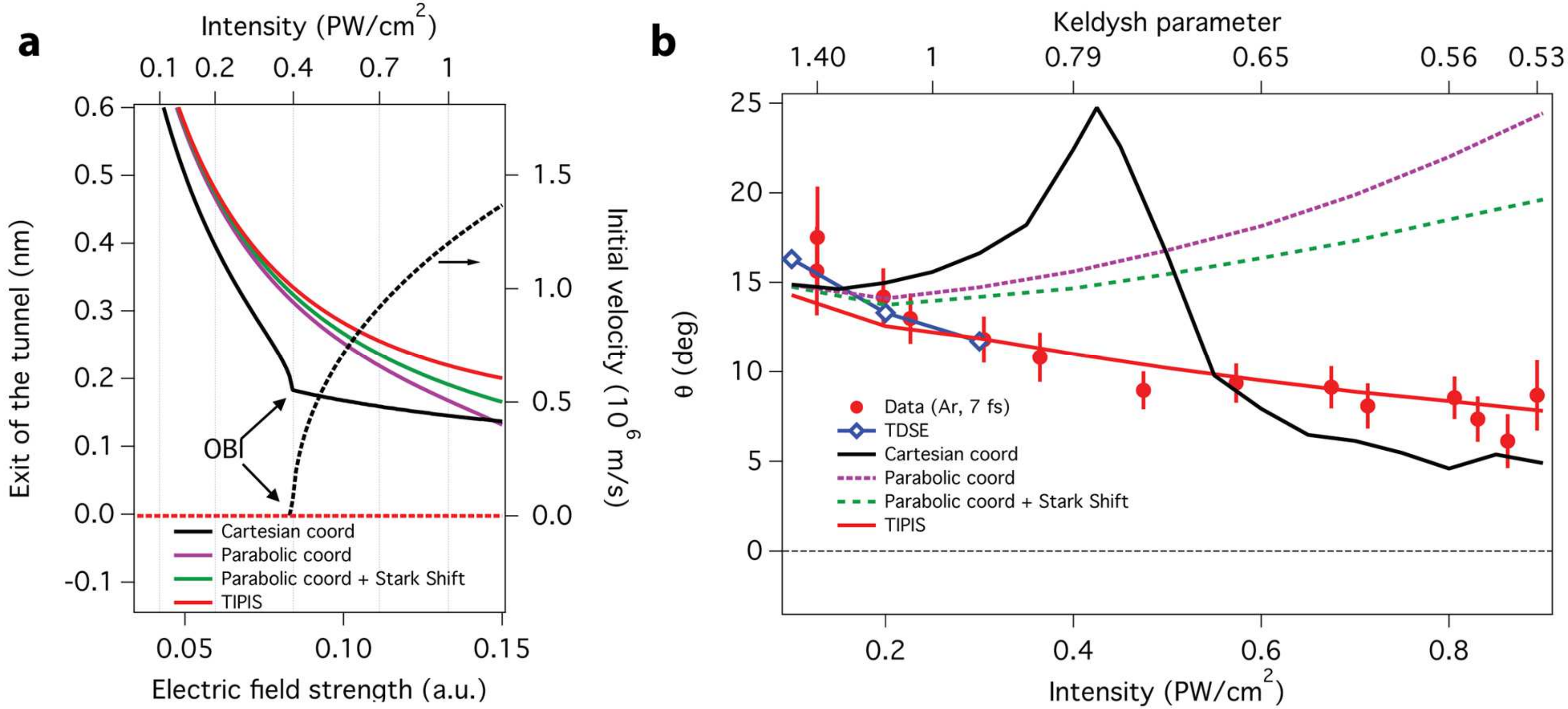
put z=-η/2



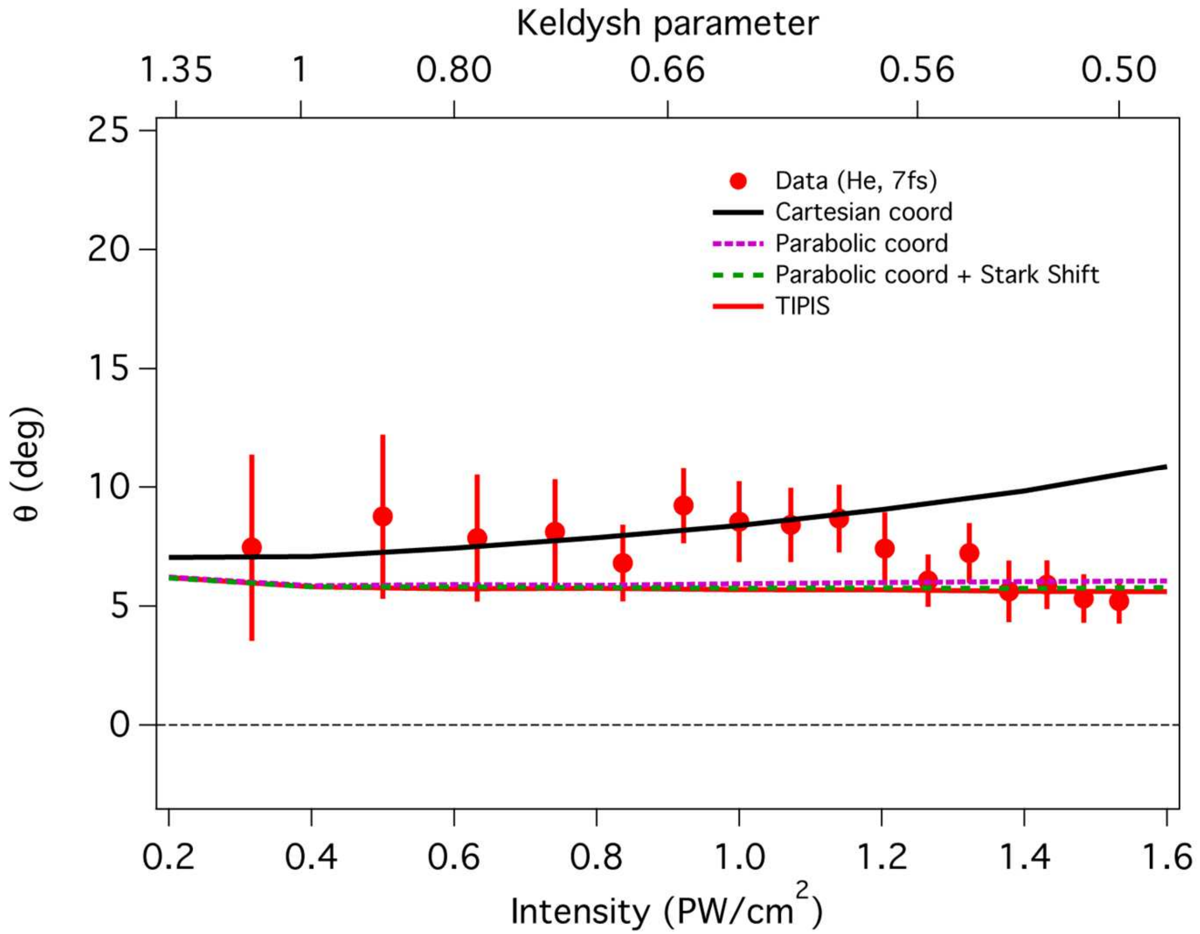

Over-the-barrier intensity is increased !

Tunnel Ionization in Parabolic coordinates with Induced dipole and Stark shift = TIPIS

# Comparison with experiment (Ar)



# Comparison with experiment (He)



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# Conclusions

•The multielectron effects are quantified by simple model

•The natural coordinates of the laser-induced tunneling current flow are the parabolic coordinates; experimental confirmation of the tunneling geometry.

•The present findings indicate that over-the-barrier intensity for atoms might be larger.

• The force terms identified here contribute to the more precise description of the tunneling step and post-ionization dynamics in strong fields.

# Outlook

•Angular shifts for molecules?

•Over-the-barrier ionization?

•Modification of orbitals in strong fields: can we do better to estimate the influence of the field beyond the effective potential?

• What if the field is not adiabatic?