Recollision Scenario without Tunneling: Role of the Ionic Core Potential

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[3]Université de Sherbrooke – Canada

Financial supports:

Strong field double ionization physics: Insights from nonlinear dynamics

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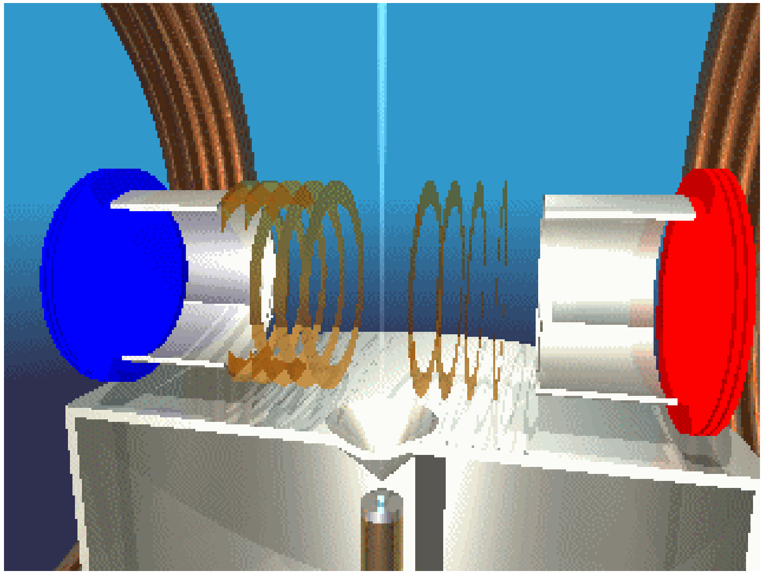
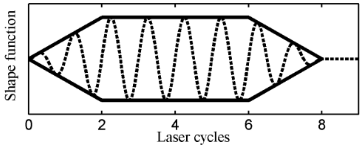
Introduction

- Physical system: Atom or molecule subjected to a strong laser pulse
  - too weak (< 10^{13} W·cm^{-2}): no ionization
  - too strong (> 10^{16} W·cm^{-2}): complete ionization
  - Intermediate intensities: very rich physics
- Experimental setup





Characteristics:

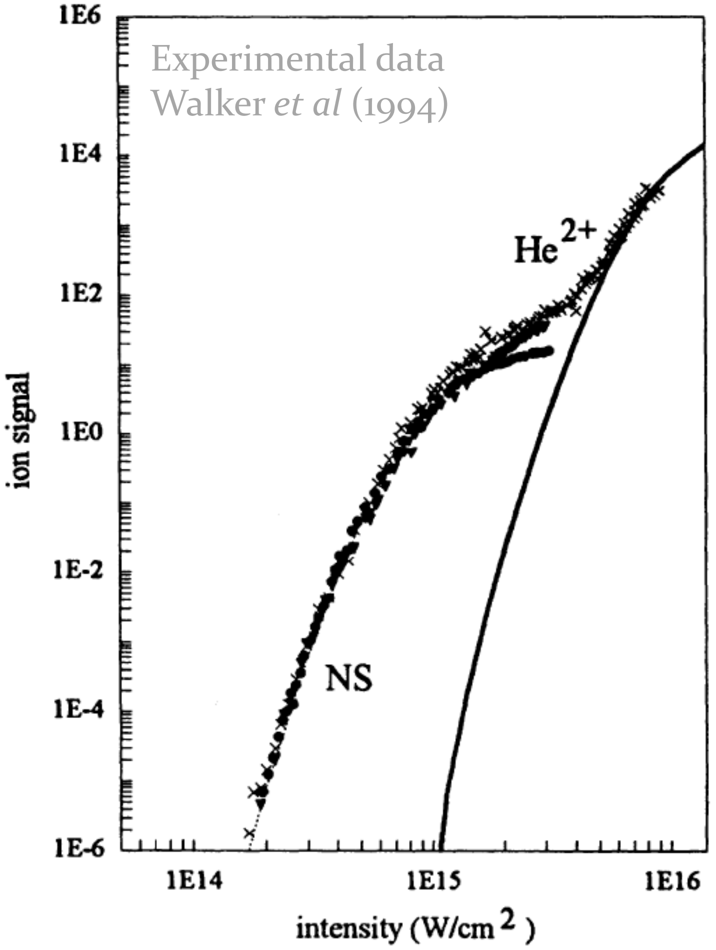
- Wavelength: ~800 nm (near infrared)
- Pulse duration on the order of 30 fs

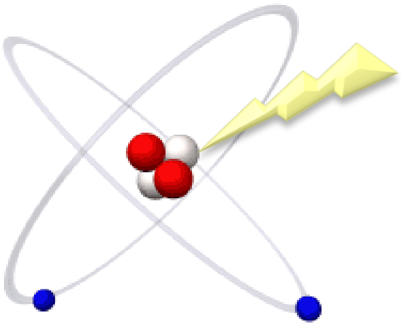
Measurements:

- Particle counts
- Momentum distributions
- Time of flight

- In this talk: Linear (LP) or Circular (CP) polarization

Double ionization: the puzzling knee-shape





Linear polarization
$\lambda = 780$ nm
T = 100 fs (10^{-13} s)

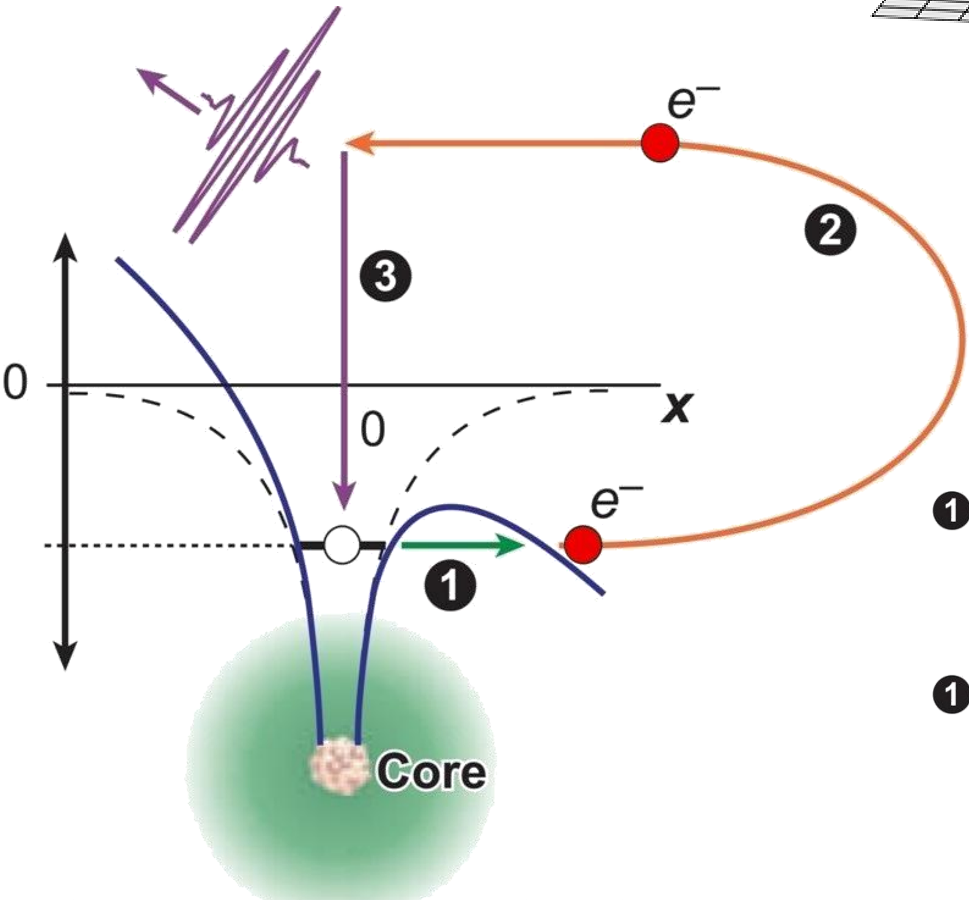
- Double Ionization
  - SDI

$$\mathrm{He} \rightarrow \mathrm{He}^{+} + e^{-} \rightarrow \mathrm{He}^{2+} + 2e^{-}$$

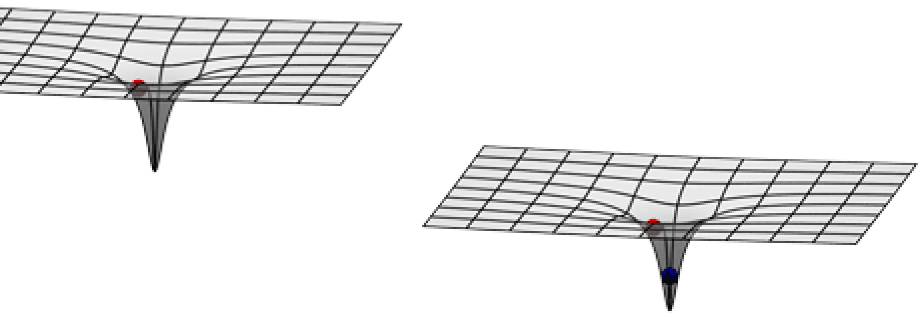
  - NSDI

$$\mathrm{He} \rightarrow \mathrm{He}^{2+} + 2e^{-}$$

Recollision with LP



Kling & Vrakking, Ann. Rev. Phys. Chem. **59**, 463 (2008)



Corkum - Schafer (1993)

Nonsequential double ionization

❶ $X \xrightarrow{h\upsilon} X^+ + e^-$, ❷ $[X^+ + e^-]^* \underset{\text{❸}}{\overset{h\upsilon}{\rightarrow}} X^{2+} + 2e^-$

High harmonic generation

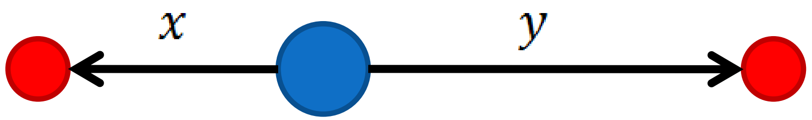
❶ $X \xrightarrow{h\upsilon} X^+ + e^-$, ❷ $[X^+ + e^-]^* \underset{\text{❸}}{\overset{h\upsilon}{\rightarrow}} X + nh\upsilon$

"Recollision or rescattering has turned out to be the keystone of strong-field physics"

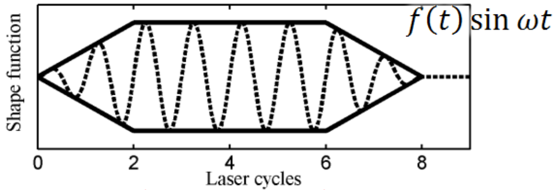
Becker & Rottke, Cont. Phys. **49**, 199 (2008)

Classical model

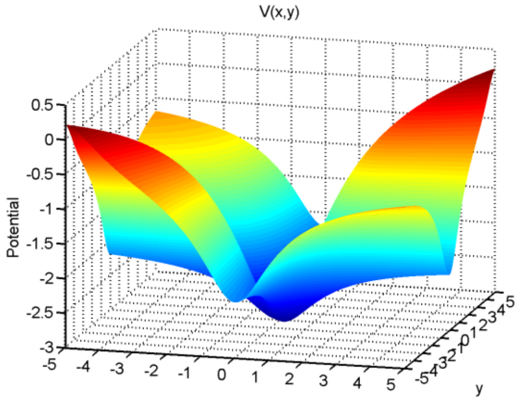
- Helium atom (*a*=*b*=1)



electron nucleus electron



- Laser interaction
  - dipole approximation
- Linear polarization (LP)
  - 1D model ↔ 2.5 d.o.f.
- Circular or elliptic polarization (CP/EP)
  - 2D model ↔ 4.5 d.o.f.



- Soft coulomb potential

Javanainen *et al.* Phys. Rev. A 38, 3430 (1988)

- Kinetic energy

$$H(\mathbf{x},\mathbf{y},\mathbf{p}_x,\mathbf{p}_y,t)=\frac{\|\mathbf{p}_x\|^2}{2}+\frac{\|\mathbf{p}_y\|^2}{2}+(\mathbf{x}+\mathbf{y})\cdot\mathbf{E}(t)\,f(t)$$

$$+\frac{1}{\sqrt{\|\mathbf{x}-\mathbf{y}\|^2+b^2}}-\frac{2}{\sqrt{\|\mathbf{x}\|^2+a^2}}-\frac{2}{\sqrt{\|\mathbf{y}\|^2+a^2}}$$

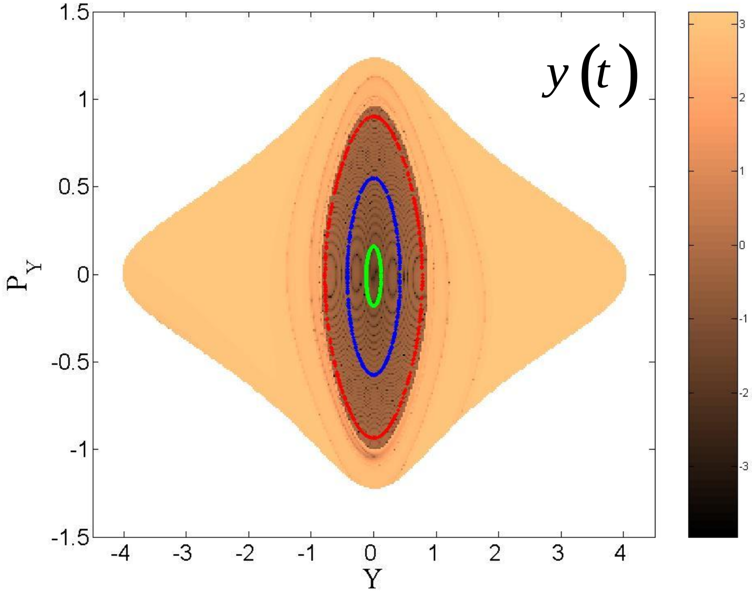
Nonsequential double ionization – recollision model

- Outer electron

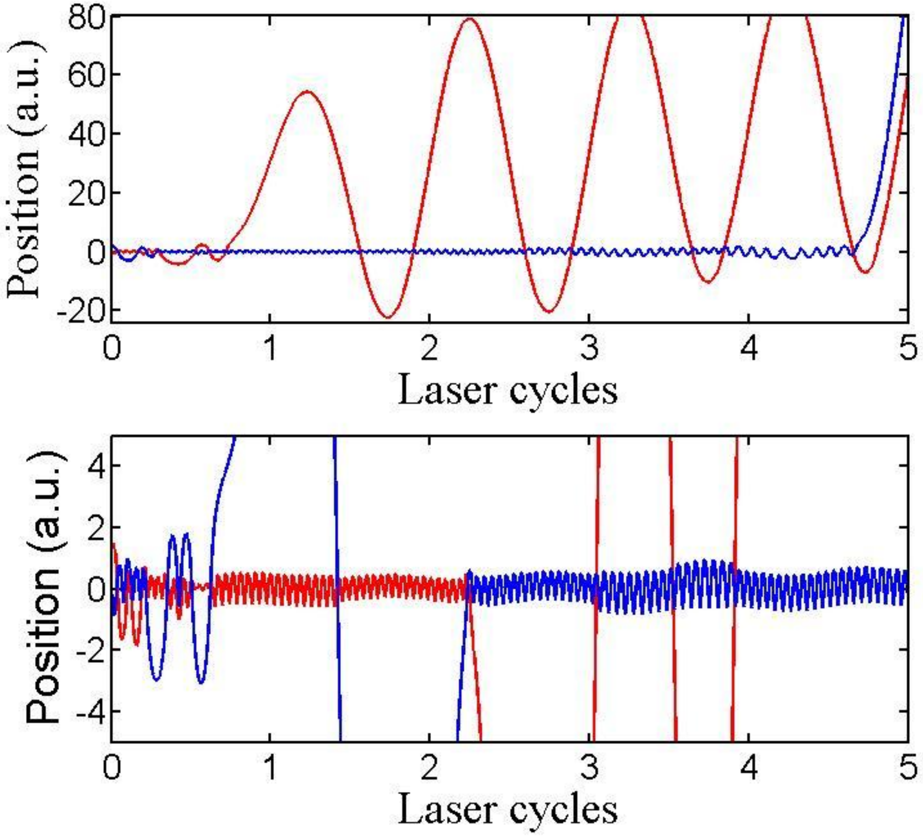
$$x\left(t\right) = x^0 + \left(p_x^0 - \frac{E_0}{\omega}\right) t + \frac{E_0}{\omega^2} \sin\omega t$$

$$\text{maximum } E_{\text{out}} = \kappa\frac{E_0}{4\omega^2} \text{ with } \kappa = 3.17\ \ldots$$

- Inner electron

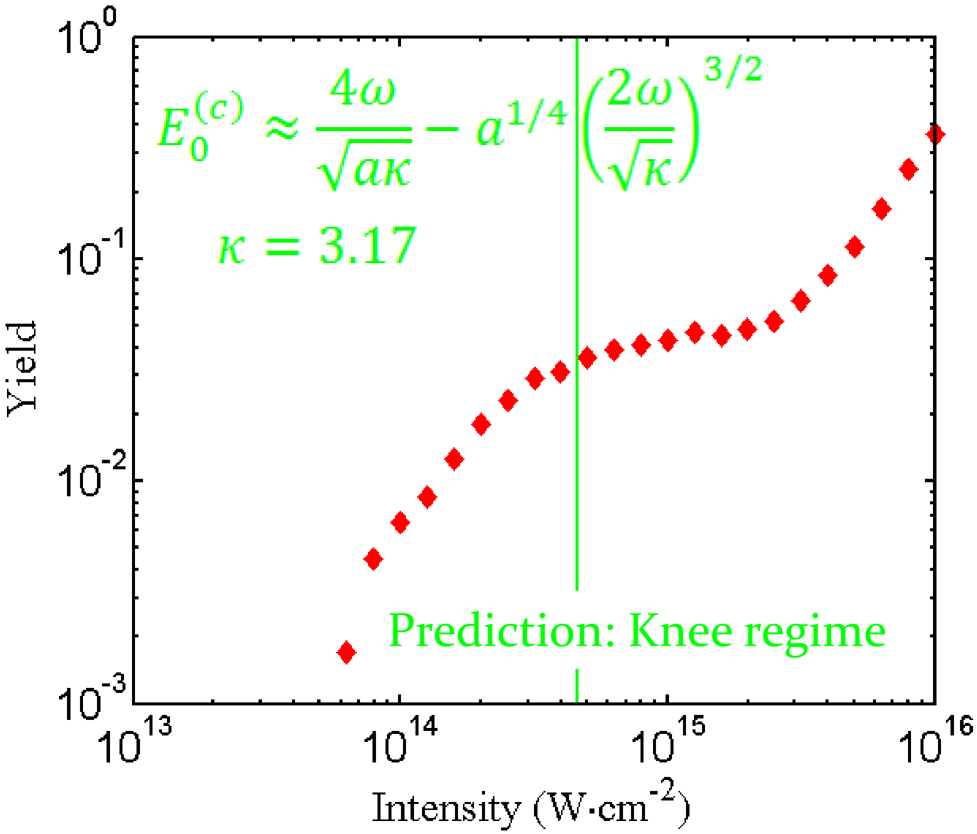


NSDI – Recollision model



- 1st phase
    - Ramp-up of the field
    - Selection of the outer electron
- 2nd phase
    - Return of the outer electron near the nucleus
    - Rescattering through the e-e interaction term
    - Possible NSDI

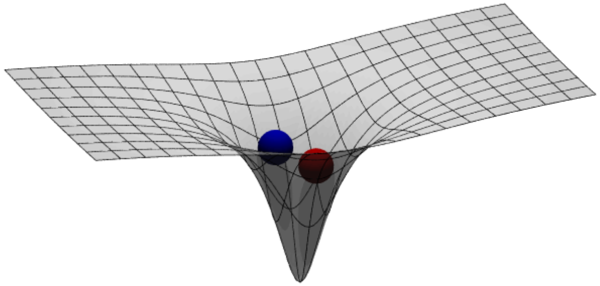
Recollision in phase space - linear polarization

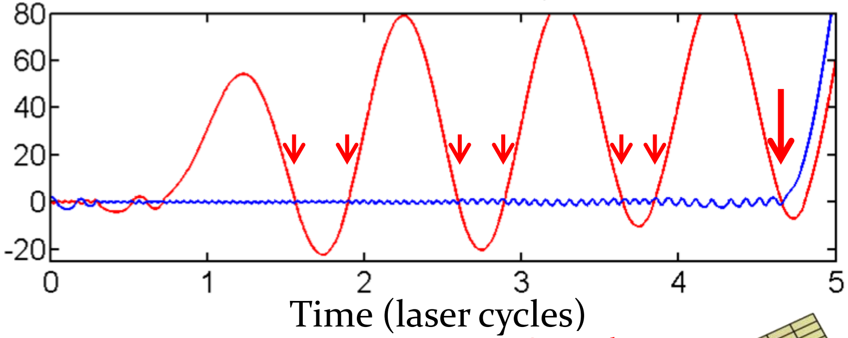


$$E_0^{(c)} \approx \frac{4\omega}{\sqrt{a\kappa}} - a^{1/4}\left(\frac{2\omega}{\sqrt{\kappa}}\right)^{3/2}$$

$\kappa = 3.17$

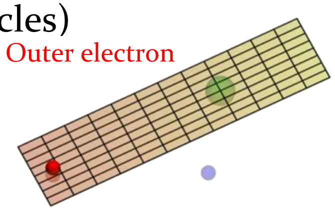
Prediction: Knee regime





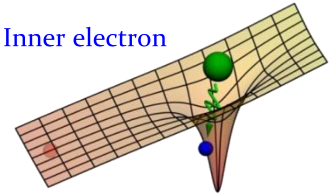
Corkum 3 step model

- Outer electron



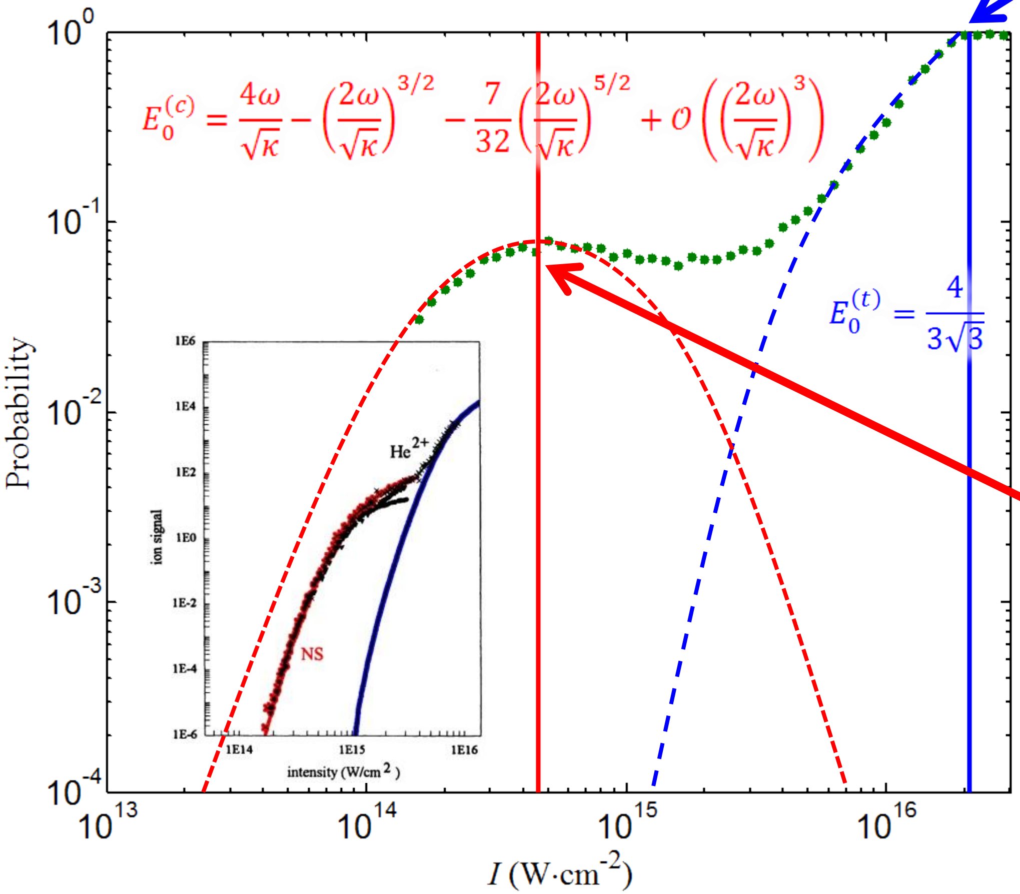
Complement the picture

- Inner electron



Recollision in phase space - linear polarization

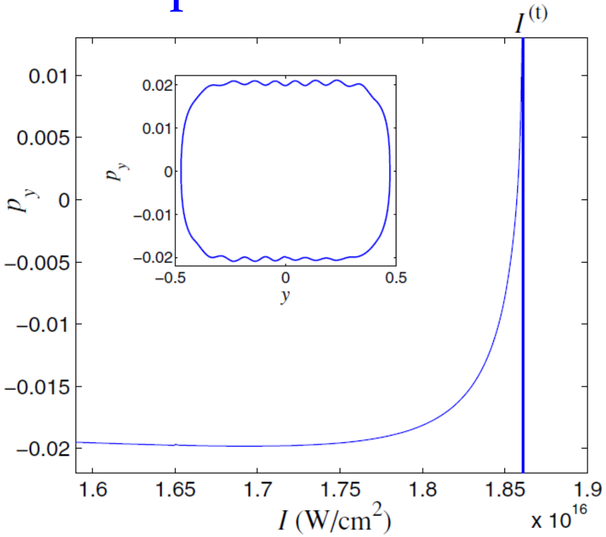
- Double ionization: separate NSDI and SDI components.



$$E_0^{(c)} = \frac{4\omega}{\sqrt{\kappa}} - \left(\frac{2\omega}{\sqrt{\kappa}}\right)^{3/2} - \frac{7}{32}\left(\frac{2\omega}{\sqrt{\kappa}}\right)^{5/2} + \mathcal{O}\left(\left(\frac{2\omega}{\sqrt{\kappa}}\right)^{3}\right)$$

$$E_0^{(t)} = \frac{4}{3\sqrt{3}}$$

- Complete SDI



- Maximum NSDI
  - Equal sharing relation

Mauger, Chandre & Uzer, PRL 102, 173002 (2009); JPB 42, 165602 (2009).

Recollisions and nonsequential double ionization with circular polarization

C. Chandre[1], A. Kamor[1,2], F. Mauger[3], T. Uzer[2]
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“If the polarization is circular, then as soon as any portion of the wavepacket emerges from the atom or molecule, it gets pulled by the field in constantly changing directions – first away from the ion, then laterally, and so on. The cusplike motion ensures that the wavepacket never returns to the ion of its birth”

Corkum, Physics Today 64, 36 (2011)

Conventional wisdom

“only a slight ellipticity of the laser polarization will ensure that the electron never returns to the environment of the ion”

Corkum, PRL 71, 1994 (1993)

“For a laser field with circular polarization and already a field with significant elliptical polarization (…) the recollision mechanism will lose its significance.”

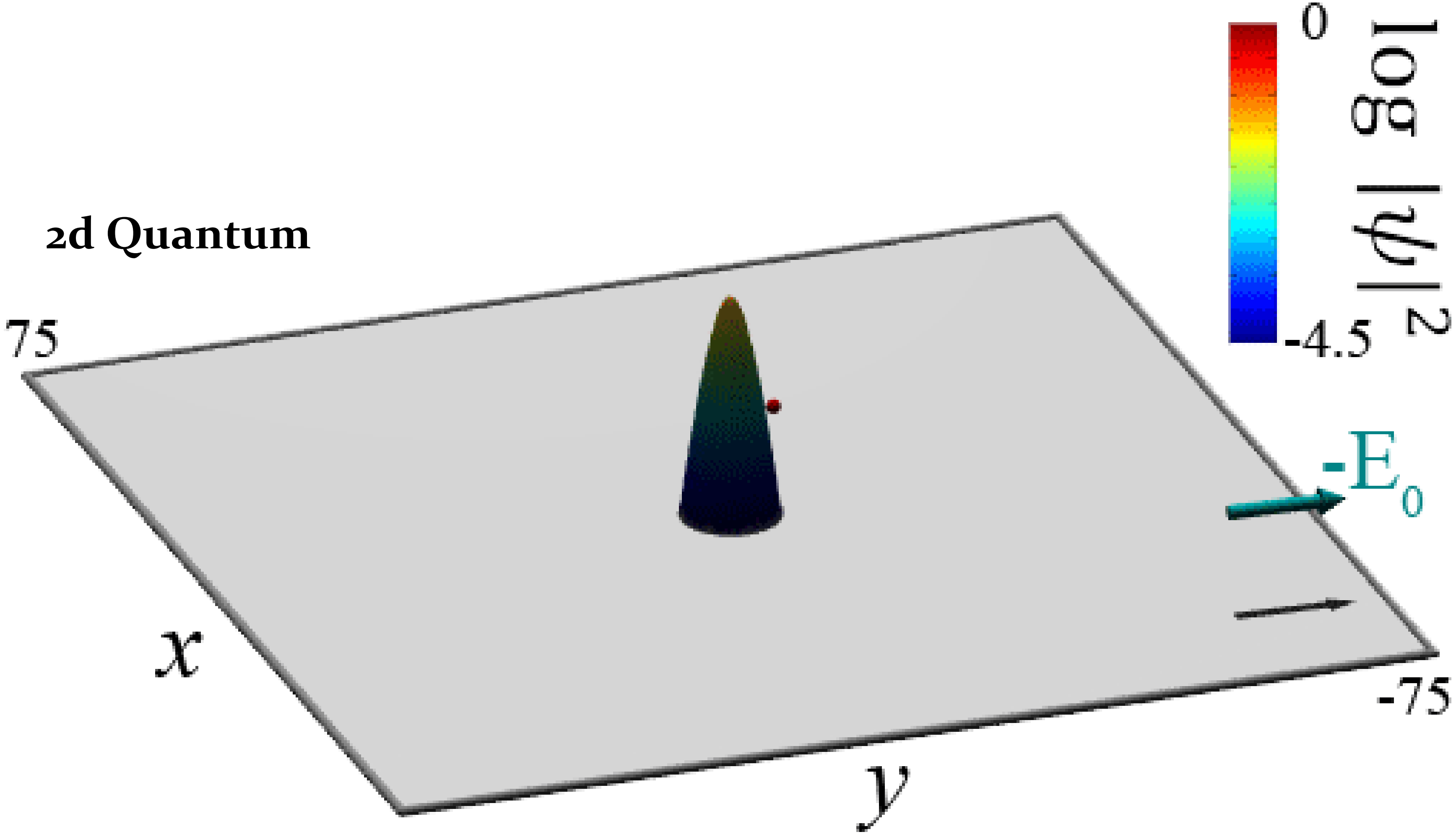
Becker & Rottke, Cont. Phys. 49, 199 (2008)

“If the polarization is circular, then as soon as any portion of the wavepacket emerges from the atom (…) the motion ensures that the wavepacket never returns to the ion of its birth”

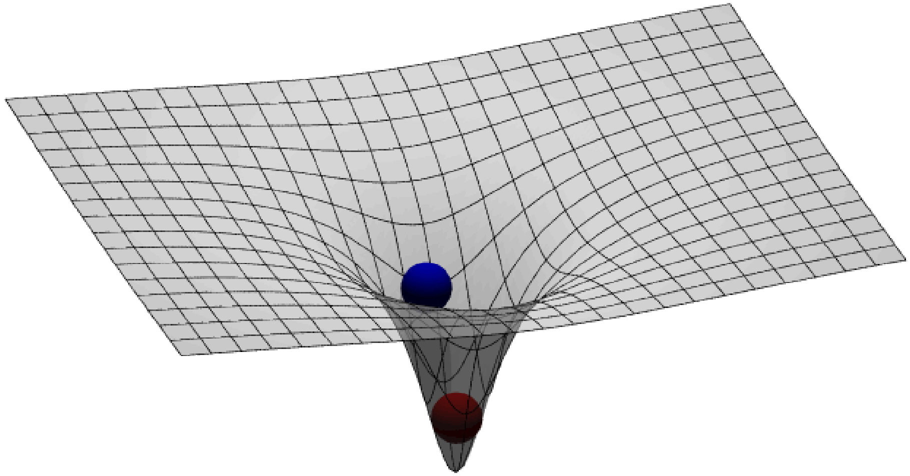
Corkum, Physics Today 64, 36 (2011)

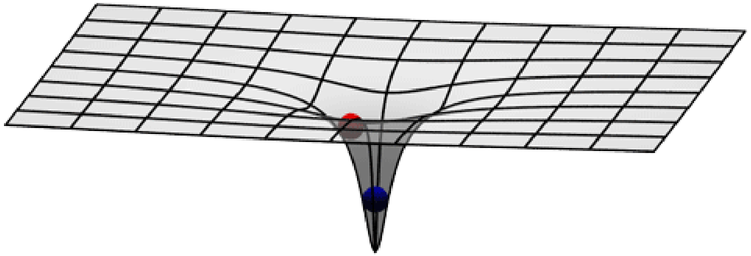
Conventional wisdom

2d Quantum

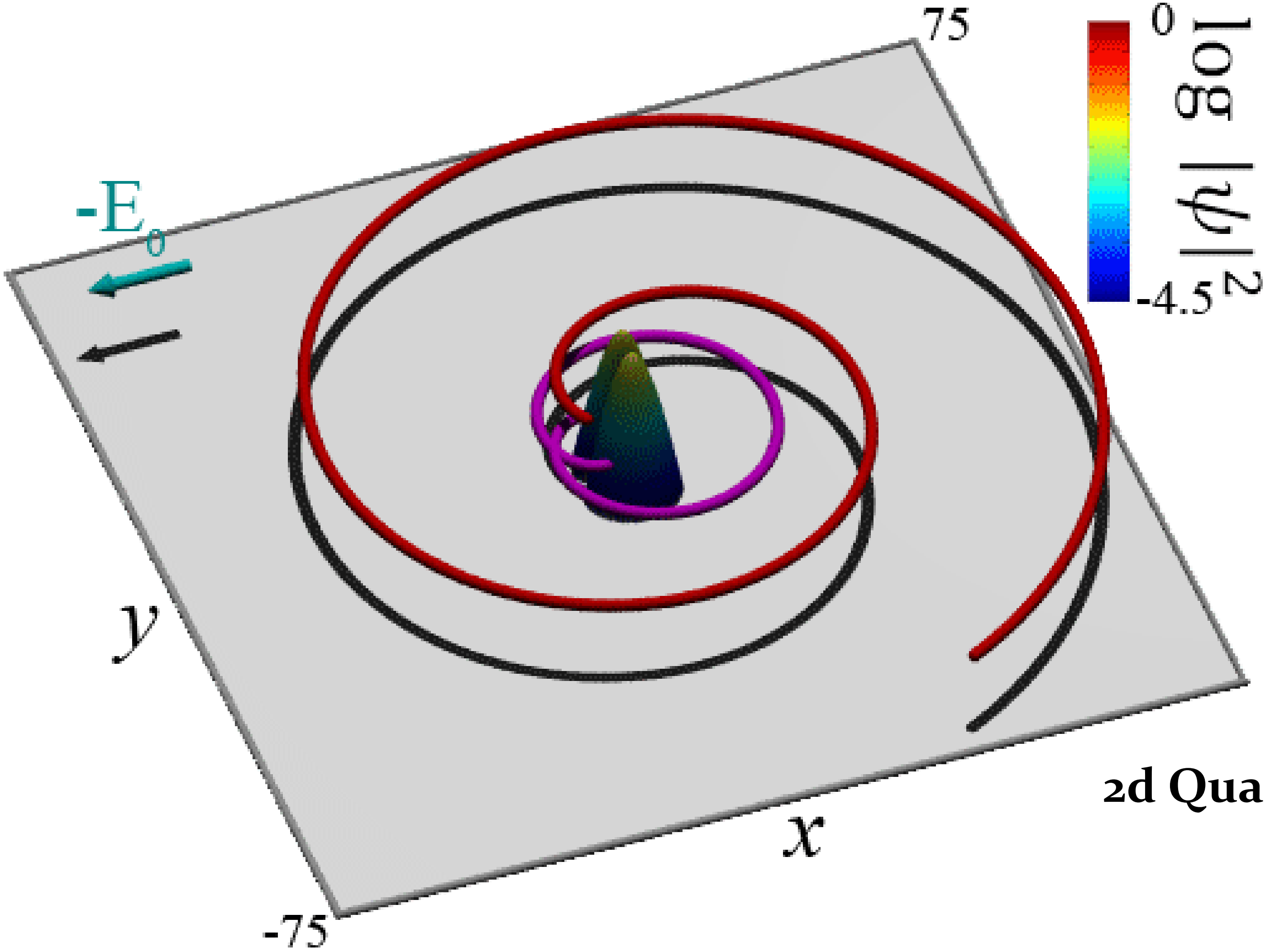


Conventional wisdom


CP

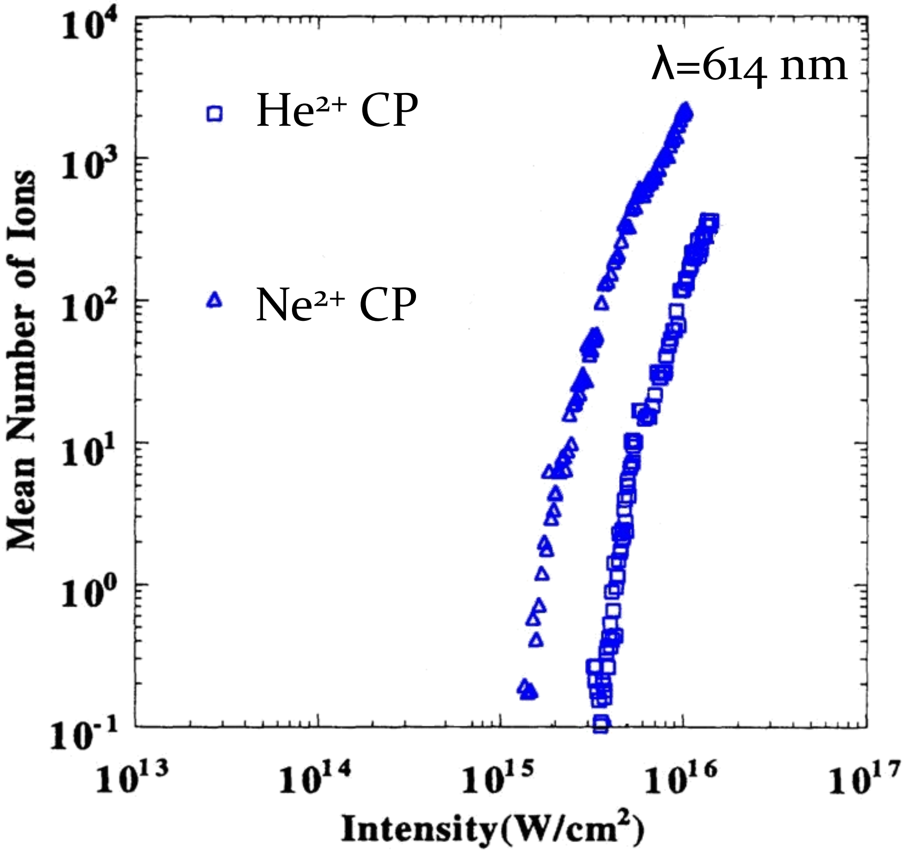

LP

What happens really

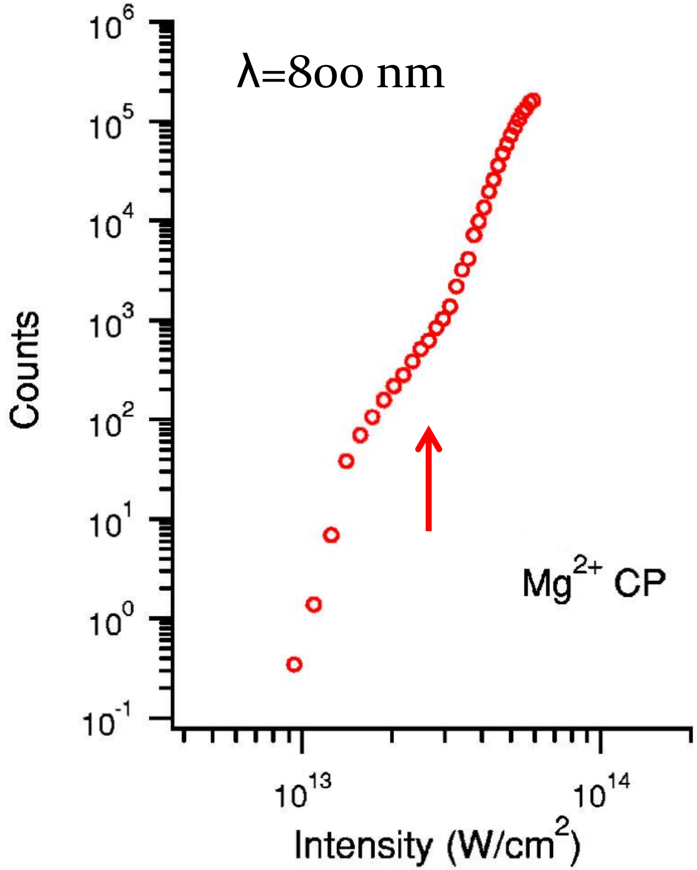


2d Quantum

Contradictory experiments with CP?



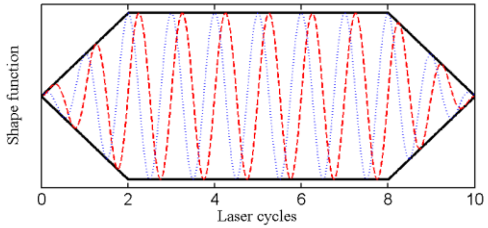
Fittinghoff *et al.*, PRA **49**, 2174 (1994)



Gillen *et al.*, PRA **64**, 043413 (2001)

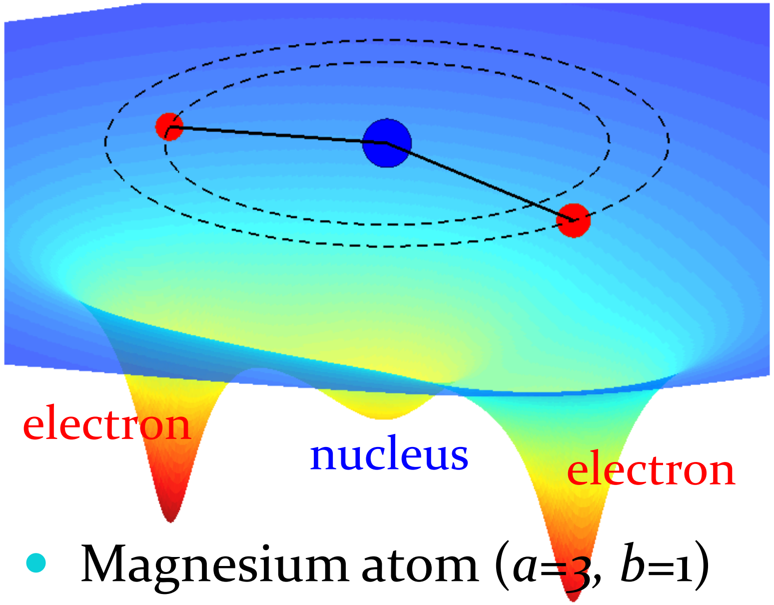
Classical model

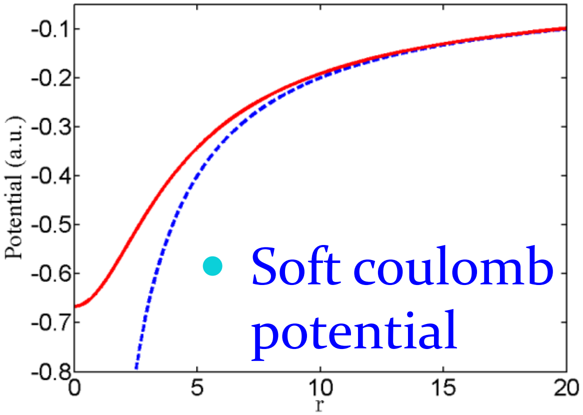
- Laser interaction
  - dipole approximation



- Circular or elliptic polarization (CP/EP)
  - 2D model ↔ 4.5 d.o.f.
- Kinetic energy

$$H(\mathbf{r}_1,\mathbf{r}_2,\mathbf{p}_1,\mathbf{p}_2,t) = \begin{pmatrix} x_1+x_2 \\ y_2+y_2 \end{pmatrix}\cdot\begin{pmatrix} \sin\omega t \\ \cos\omega t \end{pmatrix} E_0 f(t) + \frac{\|\mathbf{p}_1\|^2}{2} + \frac{\|\mathbf{p}_2\|}{2} + \frac{1}{\sqrt{\|\mathbf{r}_1-\mathbf{r}_2\|^2+b^2}} - \frac{2}{\sqrt{\|\mathbf{r}_1\|^2+a^2}} - \frac{2}{\sqrt{\|\mathbf{r}_2\|^2+a^2}}$$





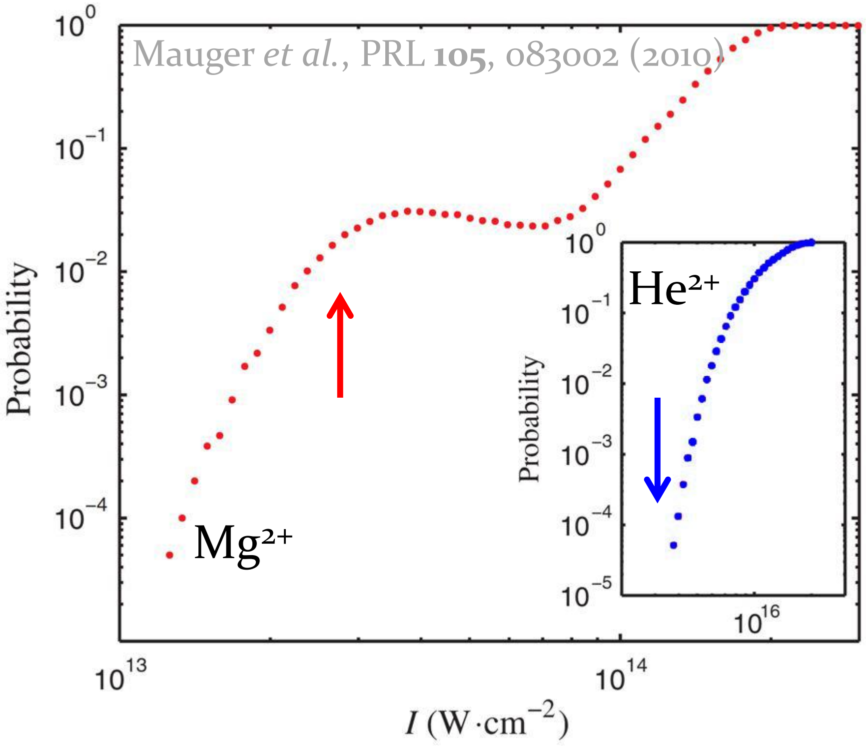
- Soft coulomb potential

Javanainen *et al.*, Phys. Rev. A 38, 3430 (1988)

- Magnesium atom (a=3, b=1)

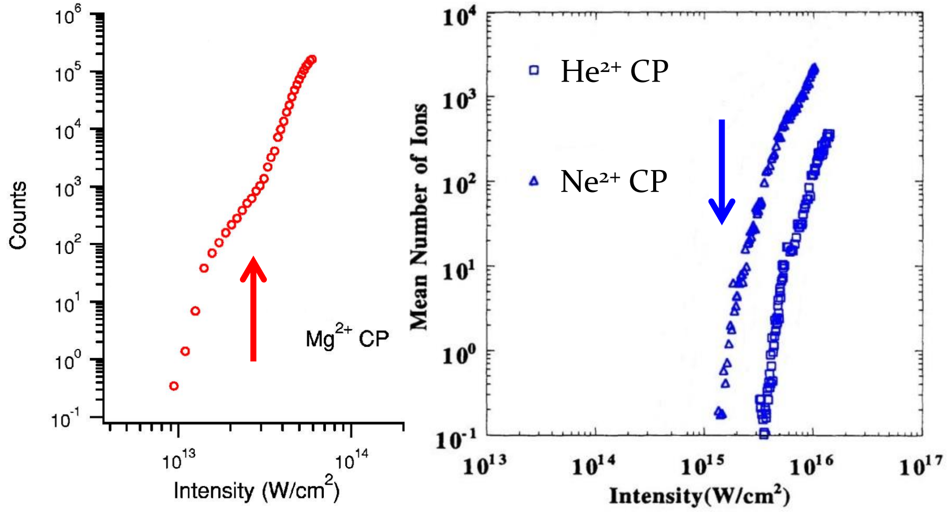
Numerical results – circular polarization

2+2d Classical



Mauger *et al.*, PRL **105**, 083002 (2010)

- Reproduce qualitatively experimental results
- No validation from quantum mechanics



- What is behind the knee?

> “enhanced double ionization for circular polarization contradicts simple rescattering theories” Gillen *et al.*, PRA 64, 043413 (2001)

Example: 2D atom

$$\mathcal{H} = \frac{1}{2}\left(p_x^2 + p_y^2\right) - \frac{1}{r} + \underbrace{F\left(x\cos\omega t + y\sin\omega t\right)}_{\text{CP}}$$

- Go to rotating frame

$$\begin{aligned} x' &= \quad x\cos\omega t + y\sin\omega t \\ y' &= -x\sin\omega t + y\cos\omega t \end{aligned}$$

- (Drop primes)

$$\mathcal{K} = \frac{1}{2}\left(p_x^2 + p_y^2\right) - \frac{1}{r} + F\,x - \underbrace{\omega\left(x\,p_y - y\,p_x\right)}_{\text{Coriolis}}$$

- (conserved) energy in rotating frame (Jacobi constant)

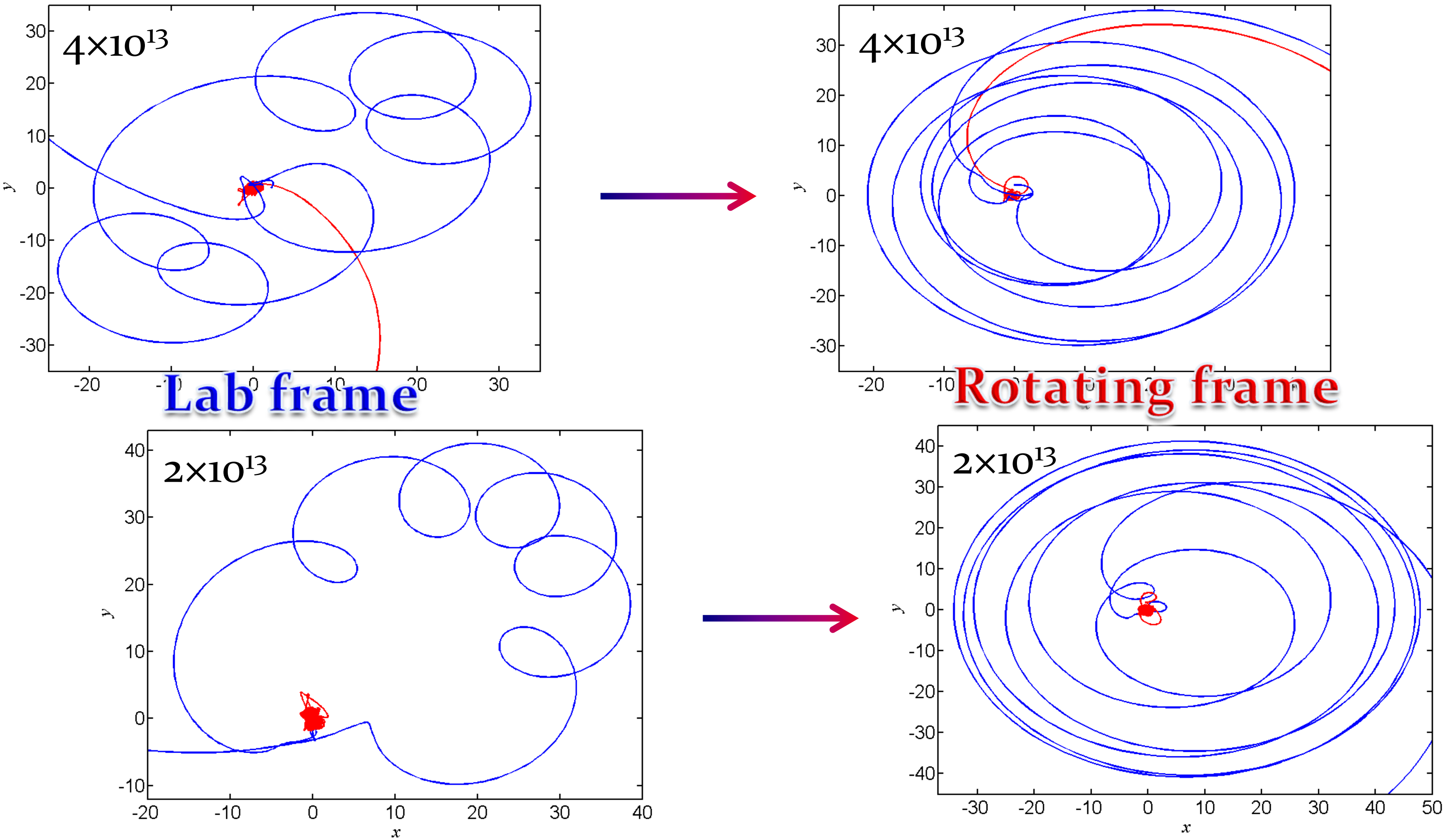
Recovering the "potential"

- Position-dependent momenta:

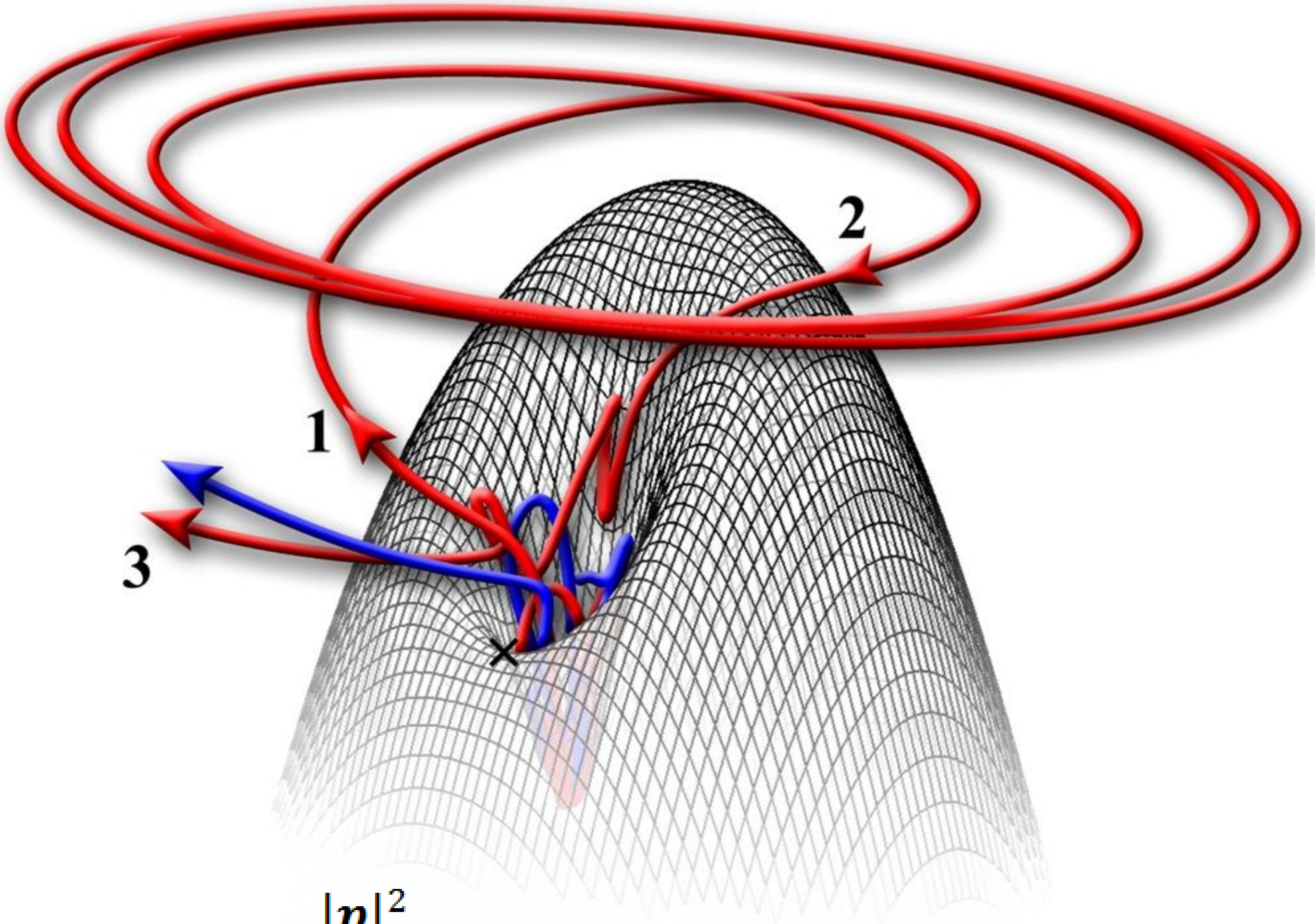
$$\begin{aligned}\dot{x} &= \frac{\partial \mathcal{H}}{\partial p_x} = p_x + \omega y\\ \dot{y} &= \frac{\partial \mathcal{H}}{\partial p_y} = p_y - \omega x\end{aligned}$$

$$\mathcal{H} = \underbrace{\frac{1}{2}(\dot{x}^2 + \dot{y}^2)}_{\textbf{Positive definite}} \underbrace{- \frac{1}{r} + F\,x - \frac{\omega^2}{2}(x^2 + y^2)}_{\textbf{New "potential": Zero-velocity surface (ZVS)}}$$

Rotating frame

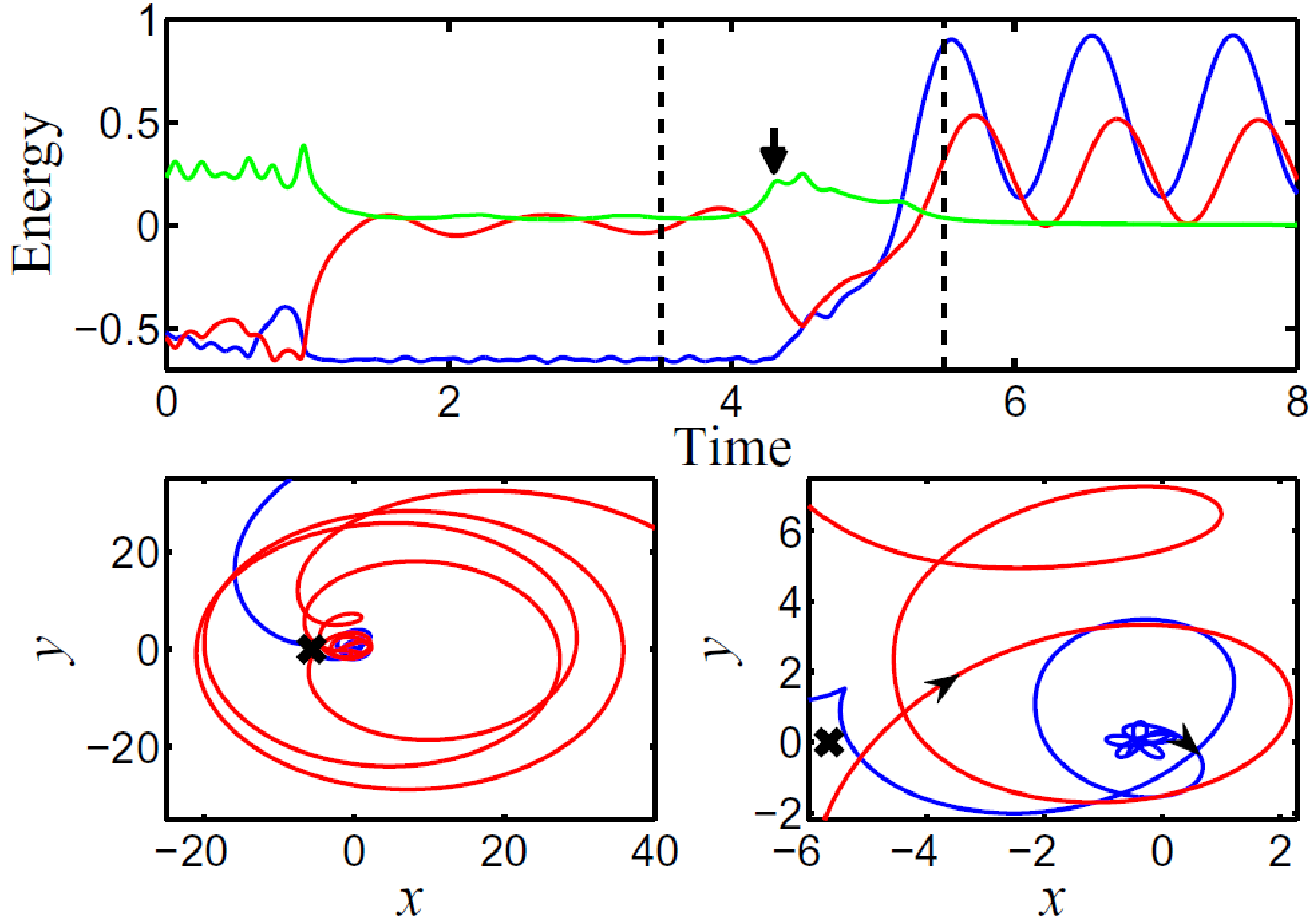


Ionization and recollision in circular polarization

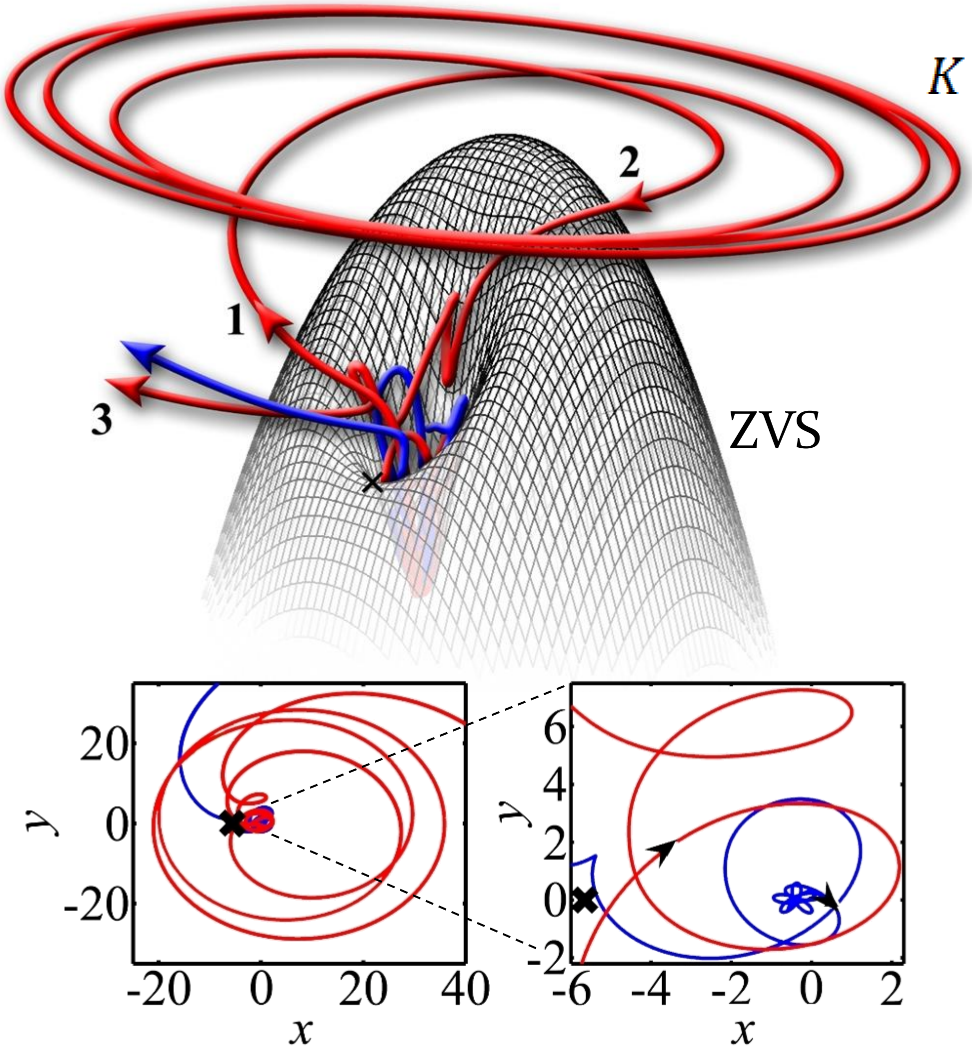


$$K = \frac{|\boldsymbol{p}|^2}{2} + V_{\mathrm{ne}}(|\boldsymbol{x}|) - \omega(\boldsymbol{x} \times \boldsymbol{p}) \cdot \boldsymbol{e}_z + E_0 x$$

Double ionization sample in circular polarization



Ionization and recollision in circular polarization



$$K = \frac{|\boldsymbol{p}|^2}{2} + V_{\mathrm{ne}}(|\boldsymbol{x}|) - \omega(\boldsymbol{x} \times \boldsymbol{p}) \cdot \boldsymbol{e}_z + E_0 x$$

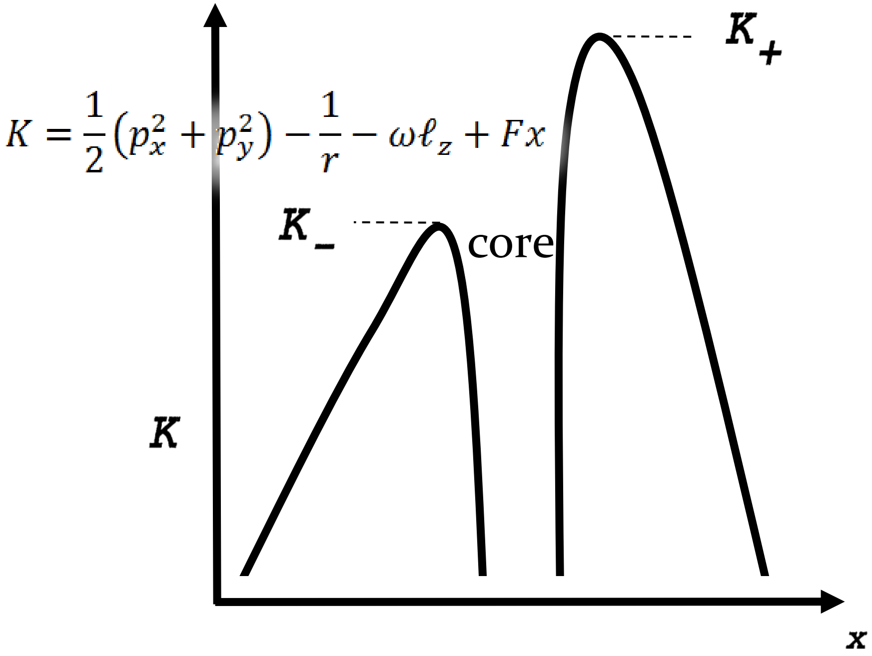
- Saddle point in phase space (×)
  - Entrance and exit door to the core region
- Heuristic criterion for recollision:
  - Two electron initial conditions
    Mg: both ionization and recollision accessible
    He: only ionization accessible
  - Double ionization probability
    Mg: knee
    He: no knee

Ionization Mechanism of Rydberg Atoms in a Circularly Polarized Microwave Field

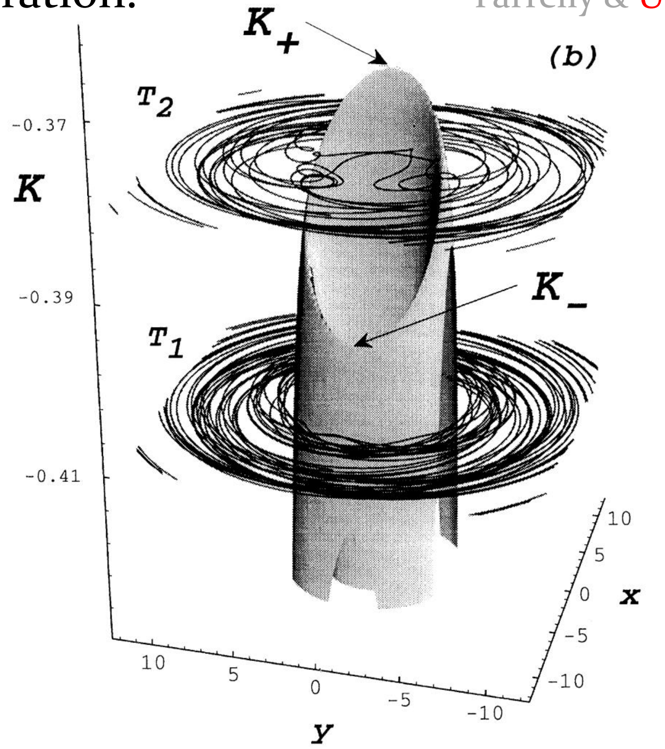
VOLUME 74, NUMBER 10 PHYSICAL REVIEW LETTERS 6 MARCH 1995

Placing a hydrogen atom in a circularly polarized microwave field exposes it to velocity-dependent forces that open new routes to chaotic ionization, access to which is controlled by the details of state preparation.

Farrelly & Uzer



$$K = \frac{1}{2}\left(p_x^2 + p_y^2\right) - \frac{1}{r} - \omega \ell_z + Fx$$



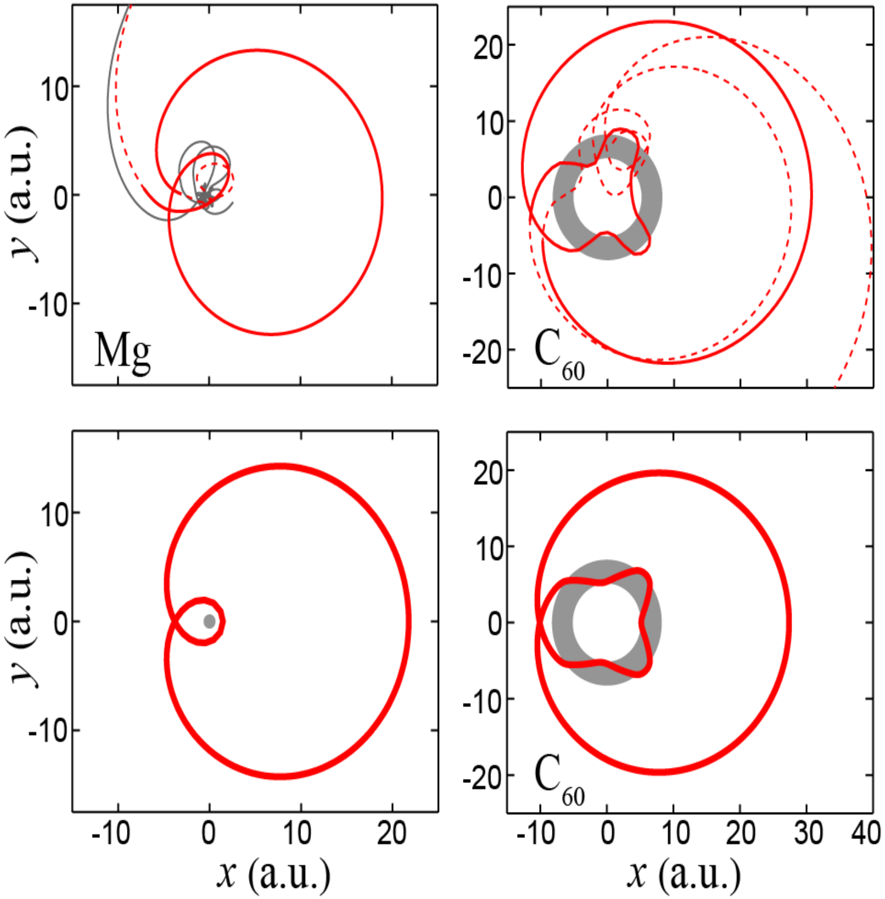
Let’s hear it from the master:

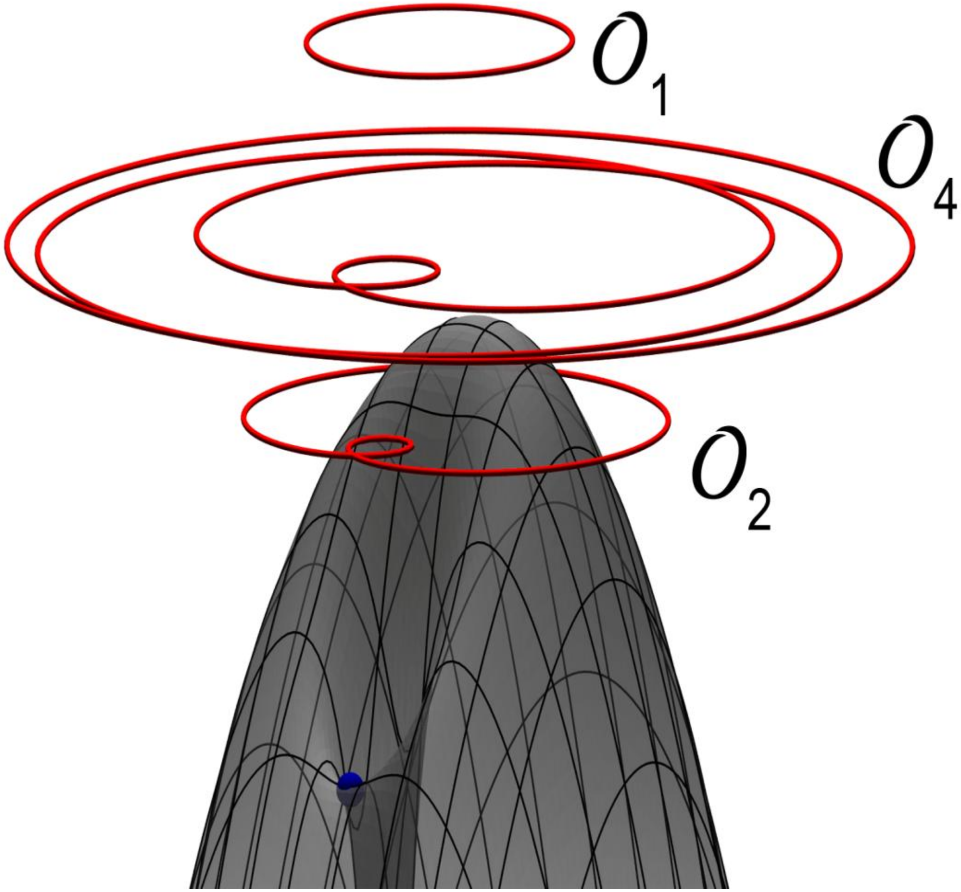
“Etant données des équations … et une solution particulière quelconque de ces équations, on peut toujours trouver une solution périodique (dont la période peut, il est vrai, être très longue), telle que la différence entre les deux solutions soit aussi petite qu'on le veut, pendant un temps aussi long qu'on le veut. D'ailleurs, *ce qui nous rend ces solutions périodiques si précieuses, c'est qu'elles sont, pour ainsi dire, la seule brèche par où nous puissions essayer de pénétrer dans une place jusqu'ici réputée inabordable.*”

Poincaré

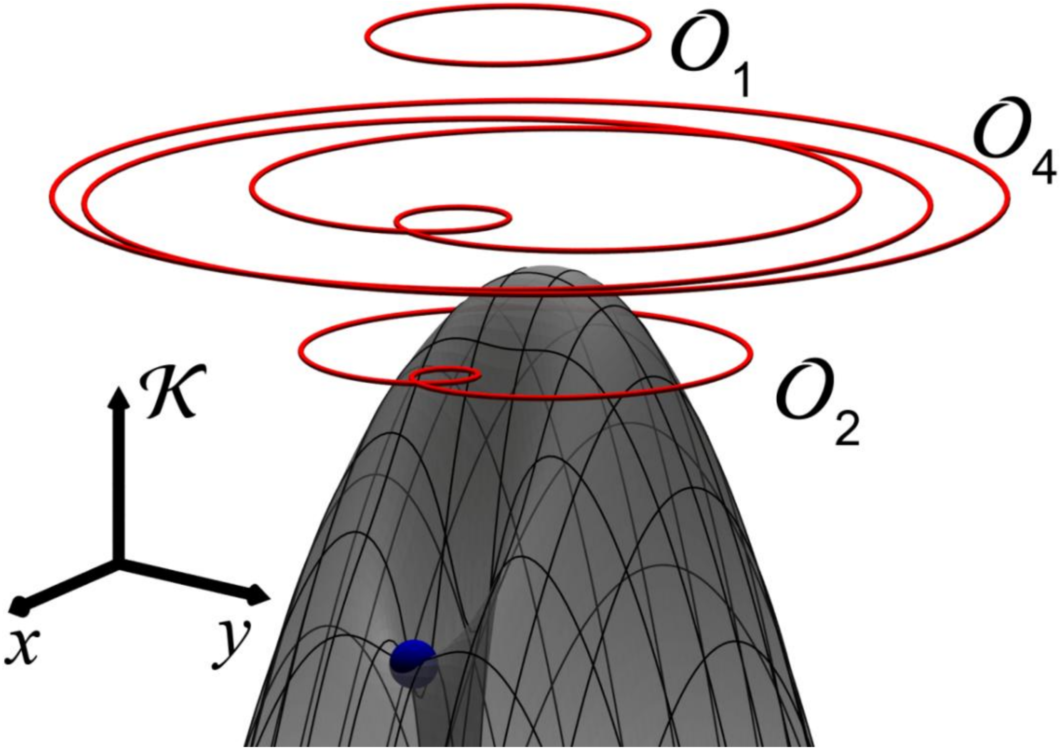
“*What makes these periodic solutions so valuable, is that the offer, in a manner of speaking, the only opening through which we might try to penetrate into the fortress which has the reputation of being impregnable.*”

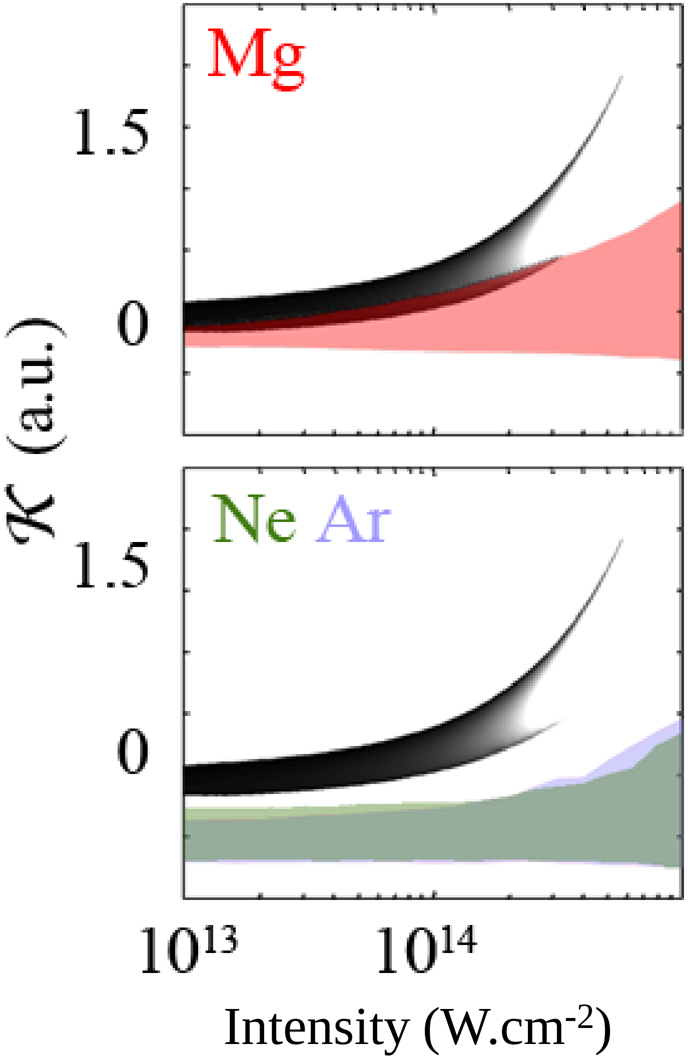
Skeleton of recollisions in circular polarization





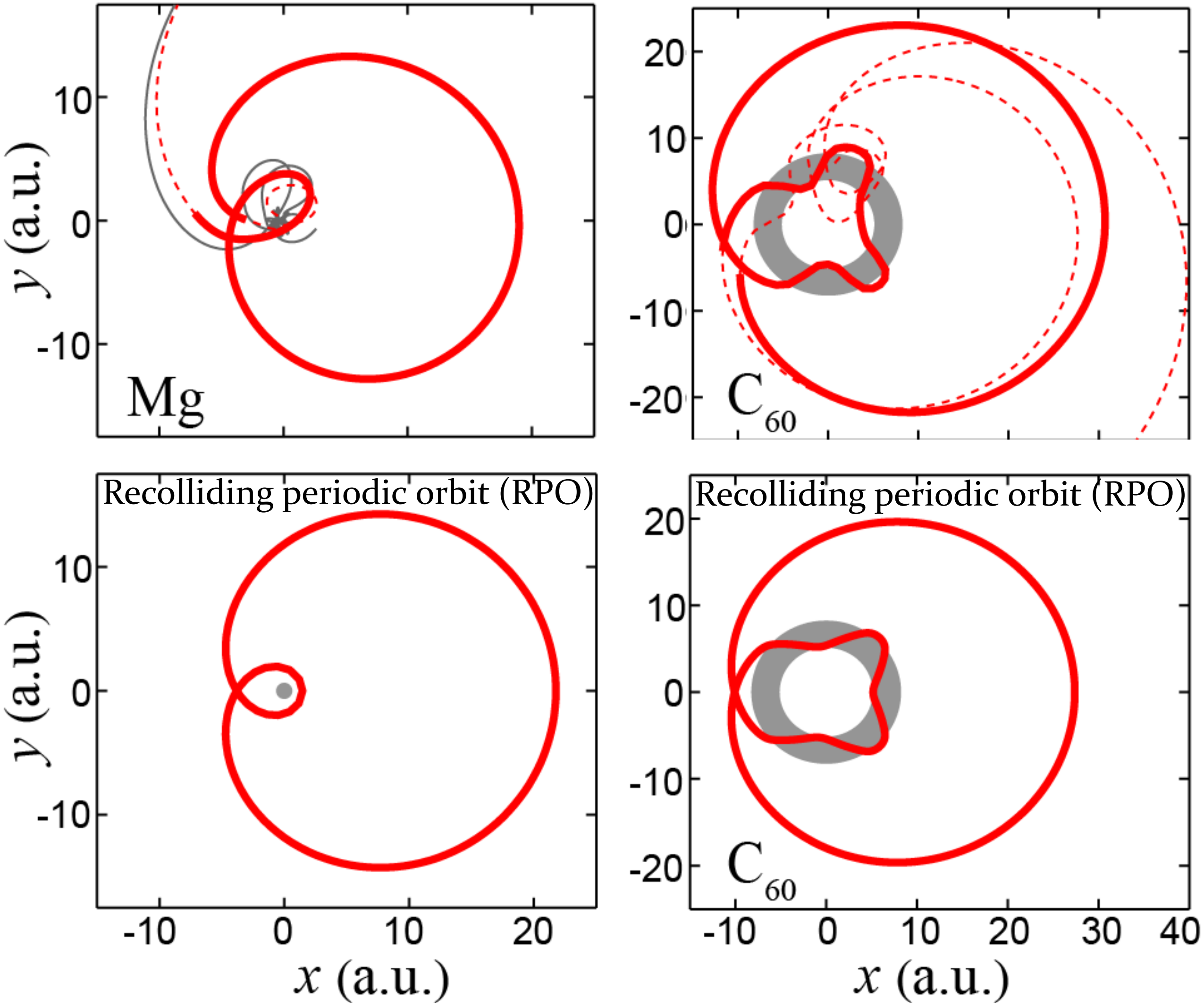
One electron – circular polarization



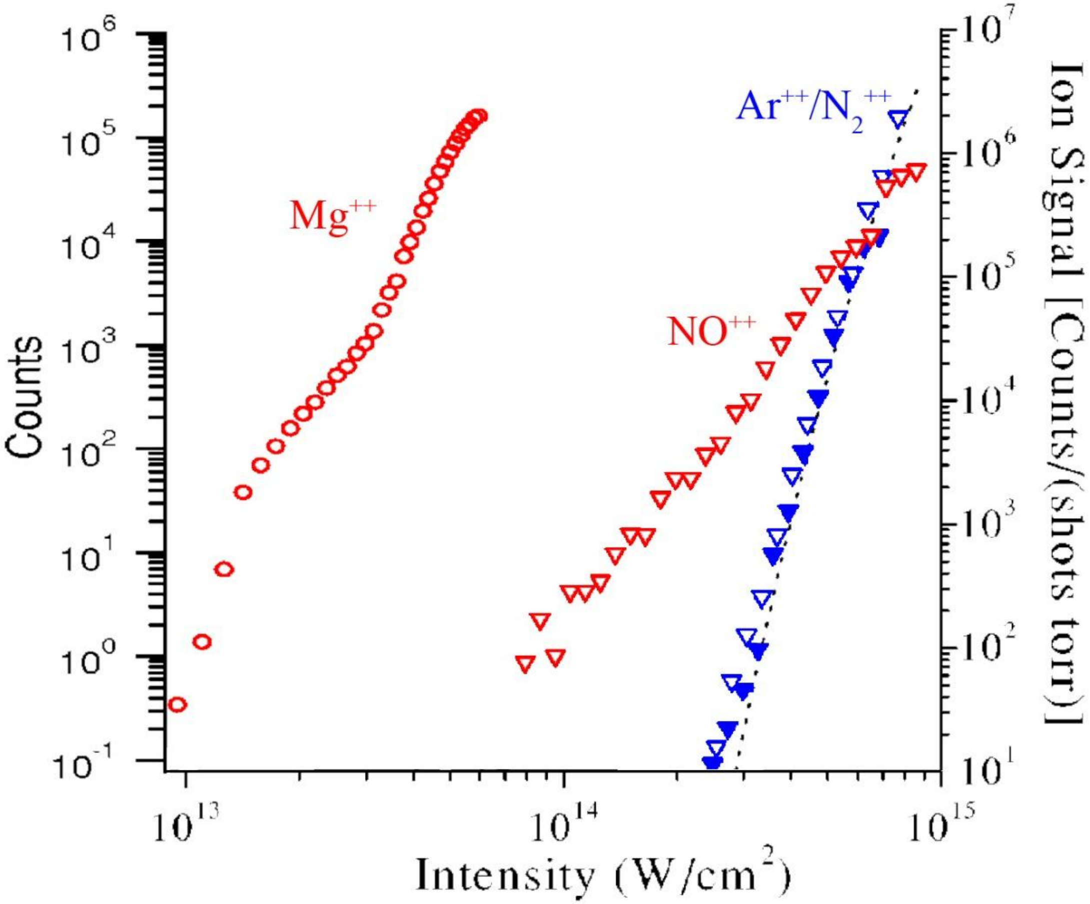


(Families of) RPOs organize recollisions

Two electrons – circular polarization



Molecular double ionization in circular polarization



Mg: Gillen *et al.*, PRA 64, 043413 (2001)
Ar/N2/NO: Guo & Gibson, PRA **63**, 040701(R) (2001)

- **NO** resembles **Mg**
- **N2** resembles **Ar**

1st Ip

- A knee in **NO**
- No knee in **N2**

BUT

- Knee for **NO** at larger intensity than Mg

2nd Ip

Recollision Scenario without Tunneling: Role of the Ionic Core Potential

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[3]Université de Sherbrooke – Canada

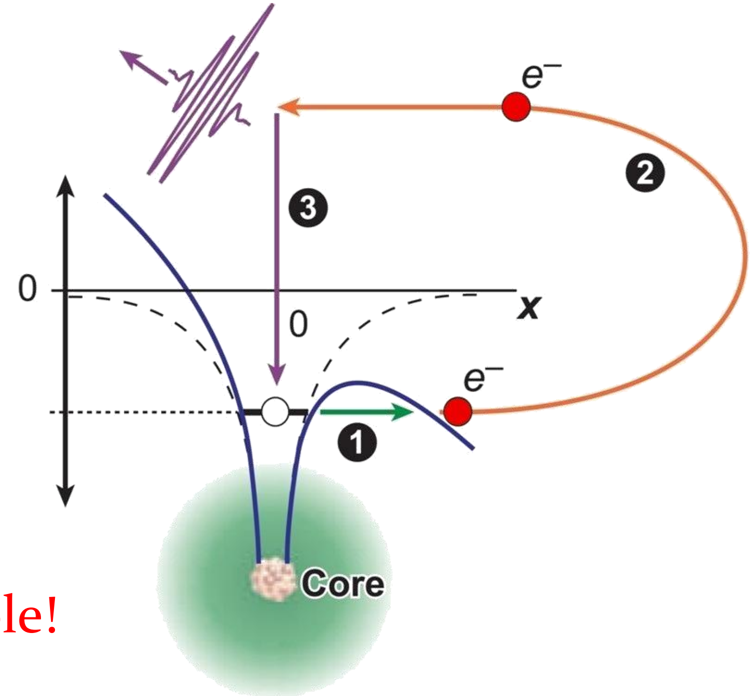
Financial supports:

Recollision – linear polarization

- Essentially a one electron process
- Three step: ❶ Tunneling (adiabatic assumption) ❷ Propagation of a (classical) field-driven electron which comes back to the core ❸ Recombination (HHG) or excitation/ionization of the remaining electron(s) (NSDI)
- In step ❷, the Coulomb field is neglected
- Consequence: maximum return energy

$$\frac{p^2_{\mathrm{return}}}{2} = \kappa \frac{I}{4\omega^2} \quad \text{where } \kappa \approx 3.17$$

- Problem: Coulomb interaction not negligible!

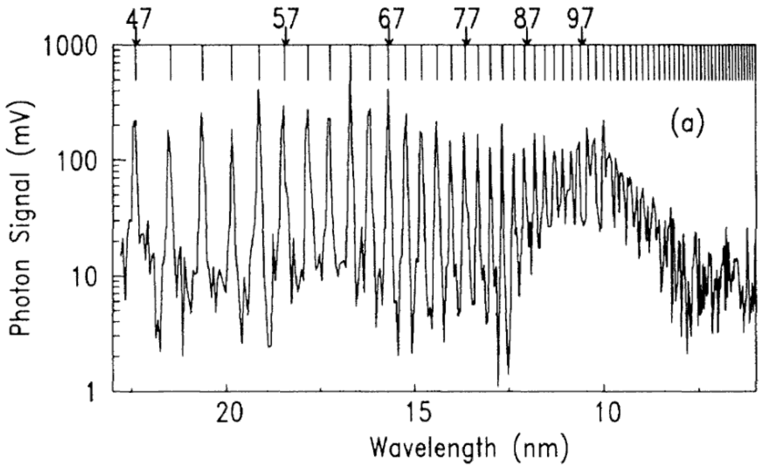


“Recollision or rescattering has turned out to be the keystone of strong-field physics”

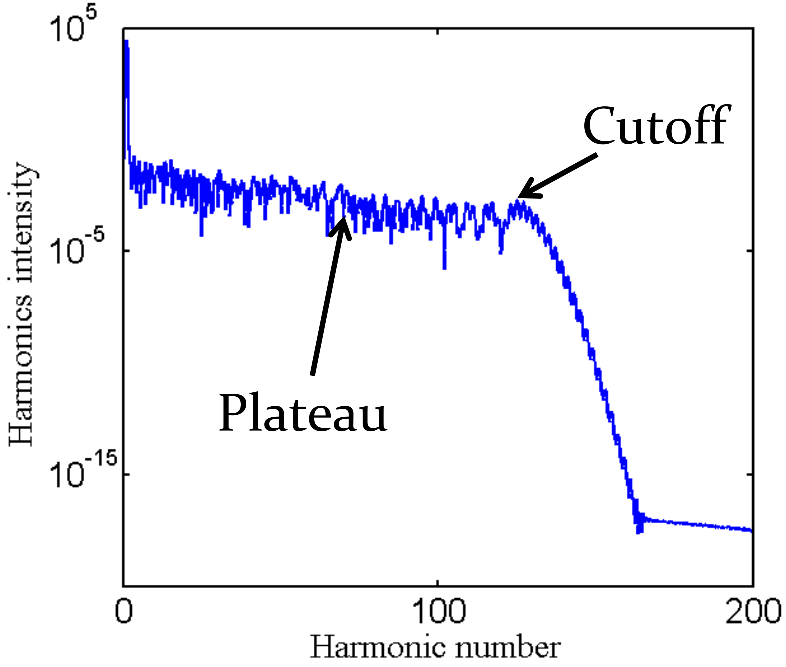
Becker & Rottke, Cont. Phys. 49, 199 (2008)

High harmonic generation with linear polarization

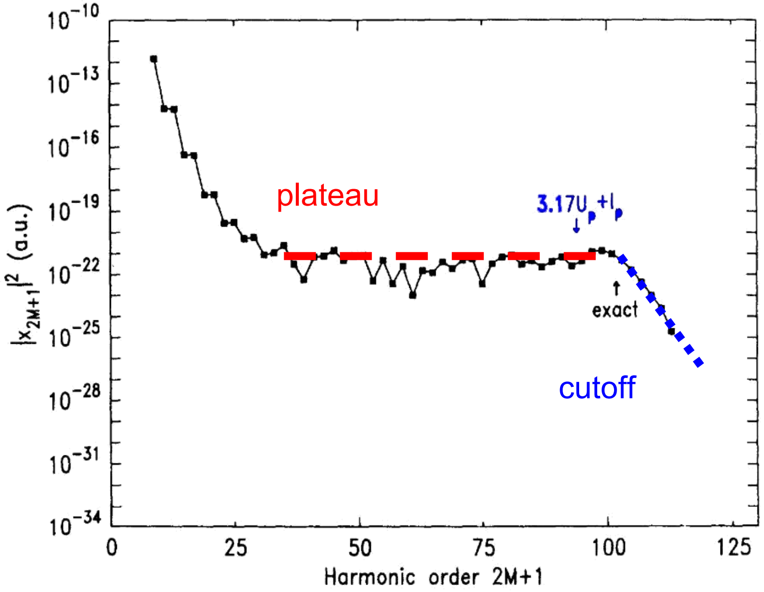
- Applications of HHG $X \xrightarrow{h\nu} X^+ + e^-, \quad [X^+ + e^-]^* \xrightarrow{h\nu} X + nh\nu$
  - Coherent sources with short wavelength
  - Generation of attosecond pulses
  - Probing of molecular dynamics



L'Huillier, *et al.* PRL (1993)



High harmonic generation with linear polarization



Lewenstein *et al.*, PRA 49, 2117 (1994)

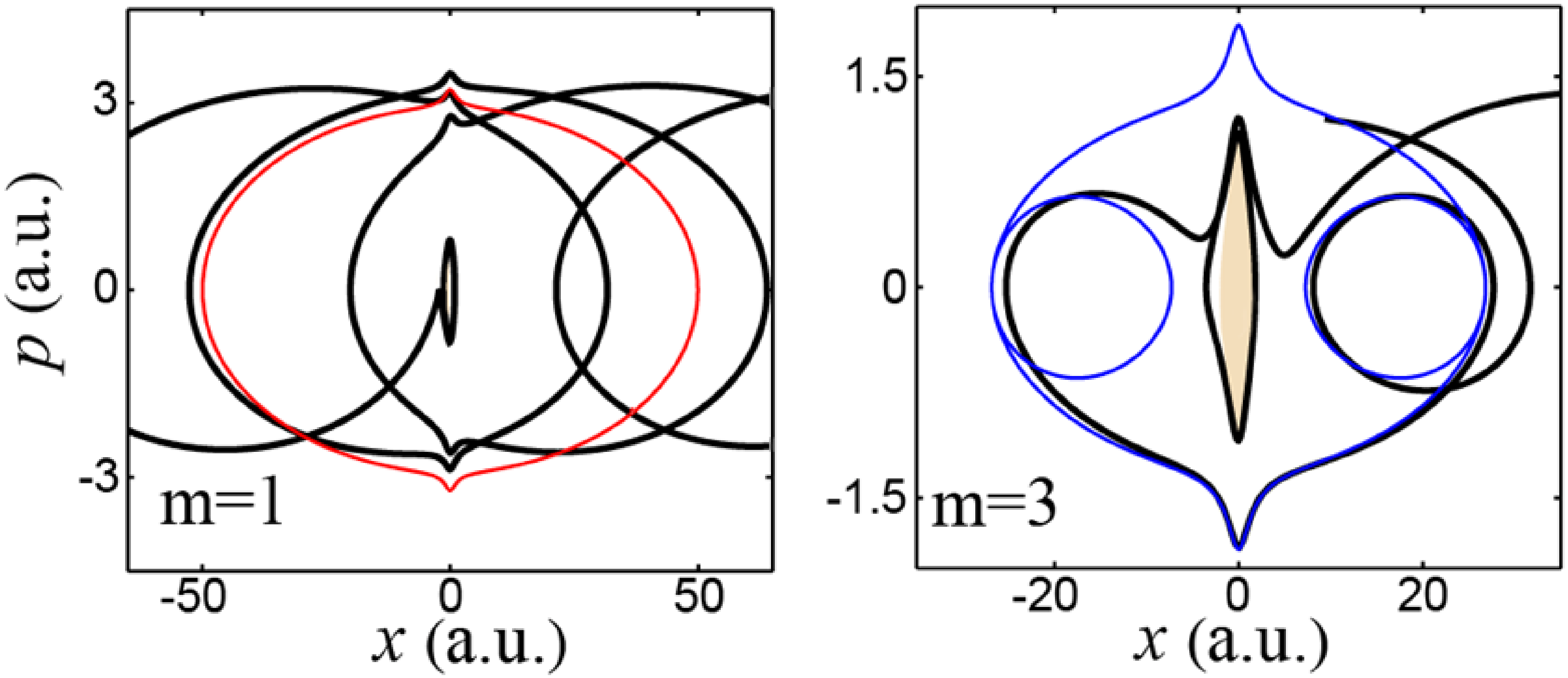
- HHG spectrum:
  - Only odd harmonics
  - Plateau in the spectrum
  - Cutoff at

- Applications:
  - Coherent sources with short wavelength (UV, XUV, …)
  - Generation of very short pulses (attosecond)
  - Molecular probe

WHY ALWAYS 3.17?

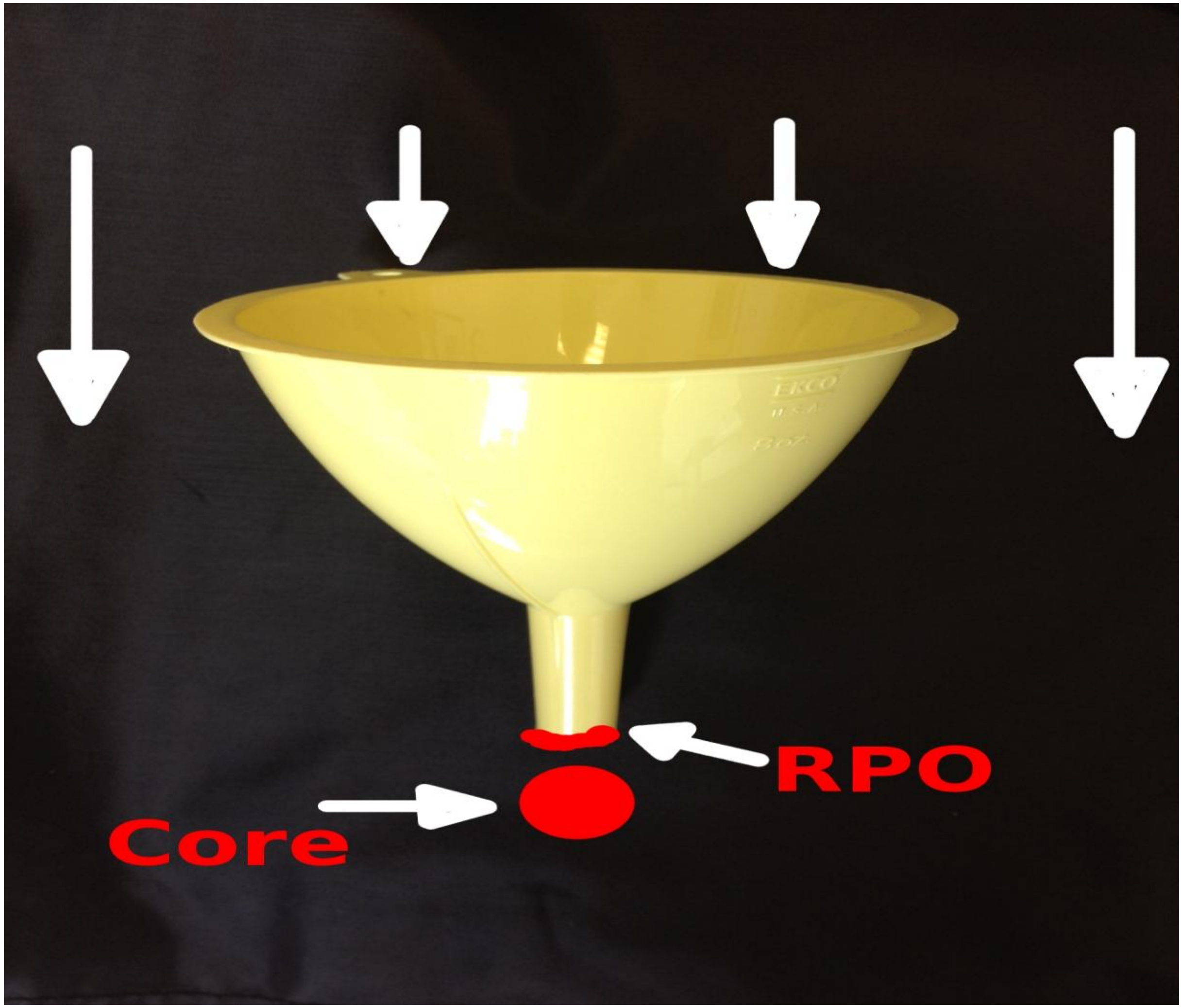
Kamor *et al.*, Phys. Rev. Lett. 112, 133003 (2014)

One electron model – linear polarization

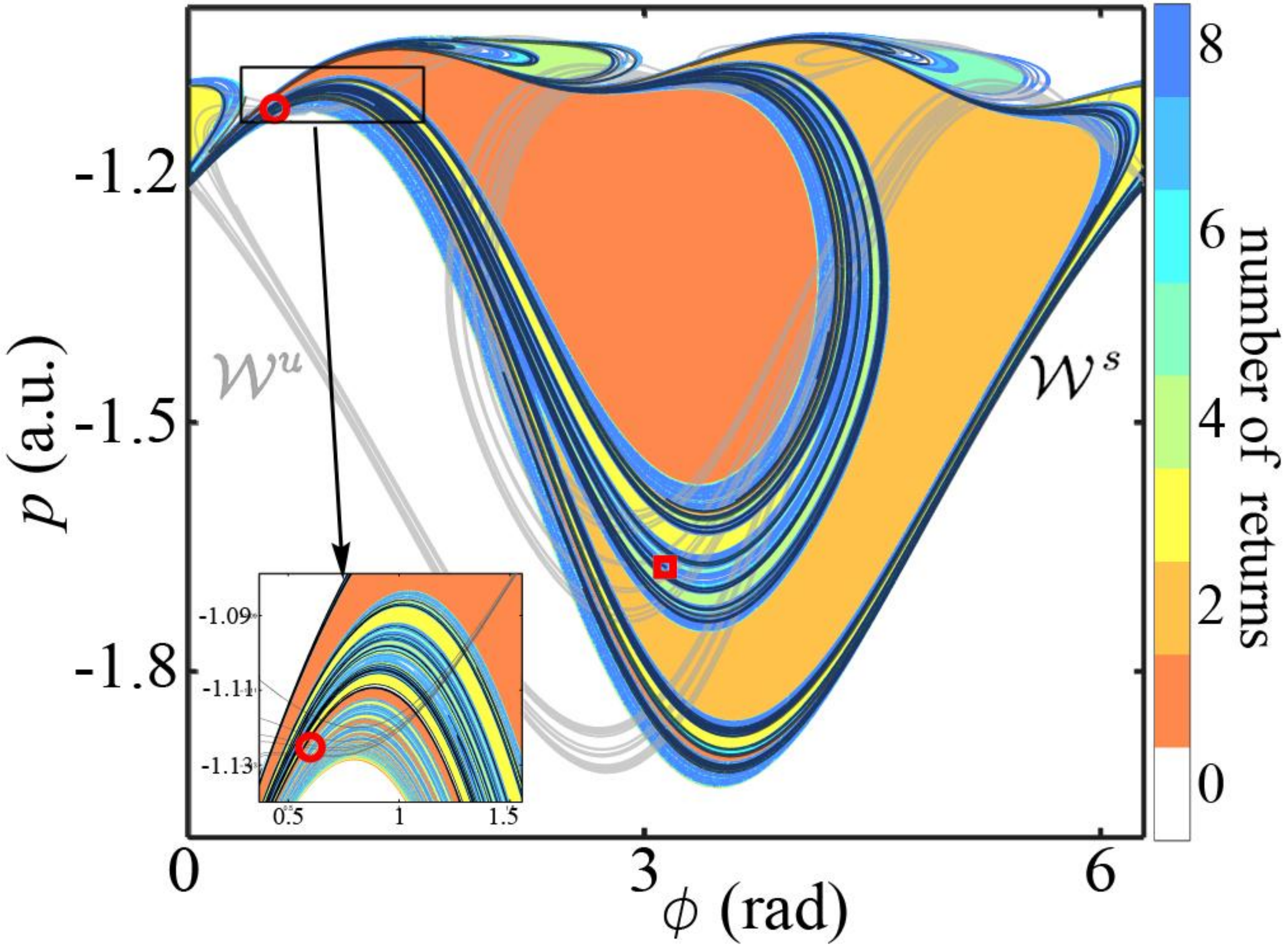


- Two lowest order resonance periodic orbits influencing the dynamics
- Recolliding Periodic Orbits - RPO
  - Must ionize and return to the nucleus
  - Short and weakly hyperbolic periodic orbits

One electron model – linear polarization

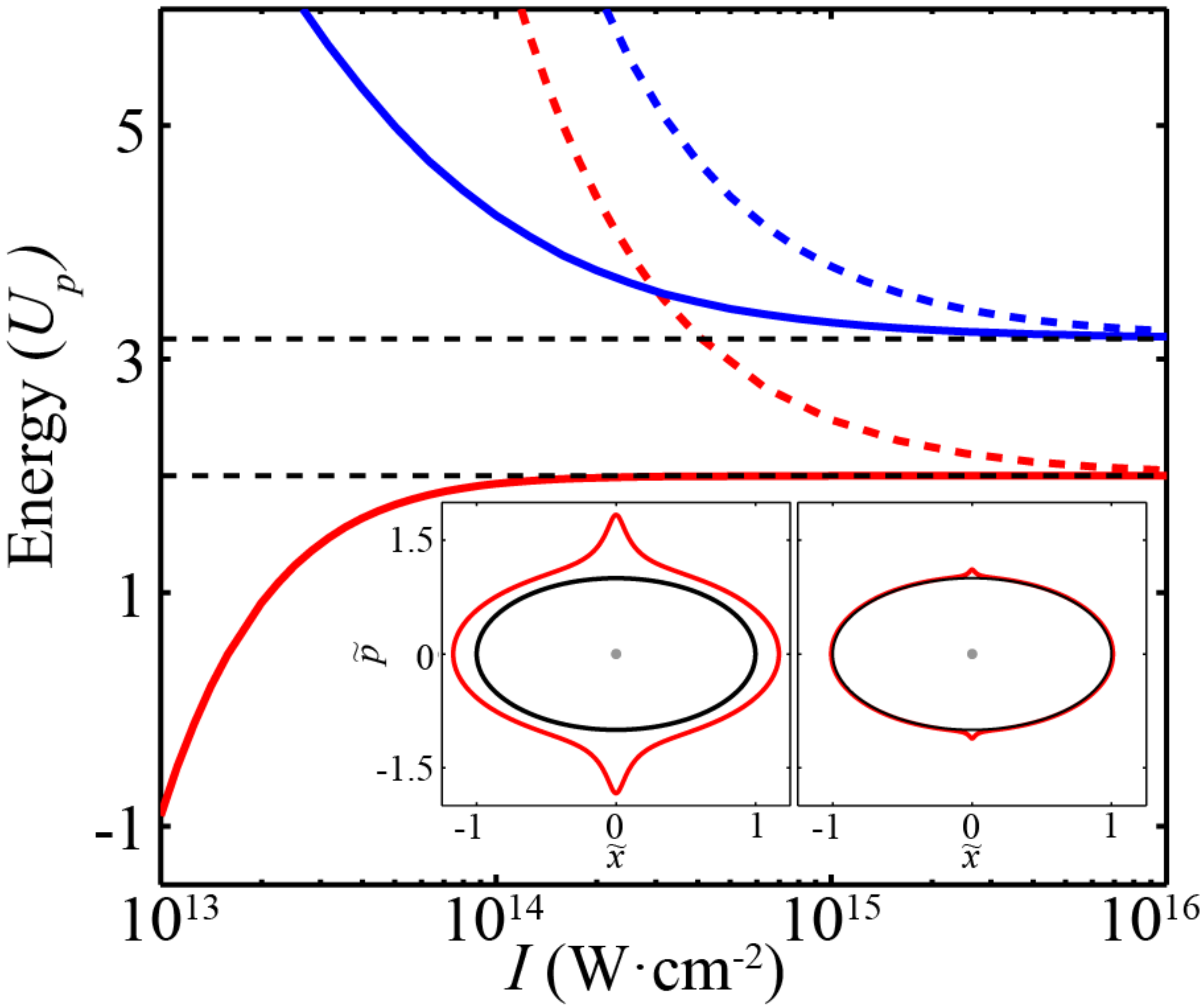


One electron model – linear polarization



The manifolds of the RPO drive the recollision process

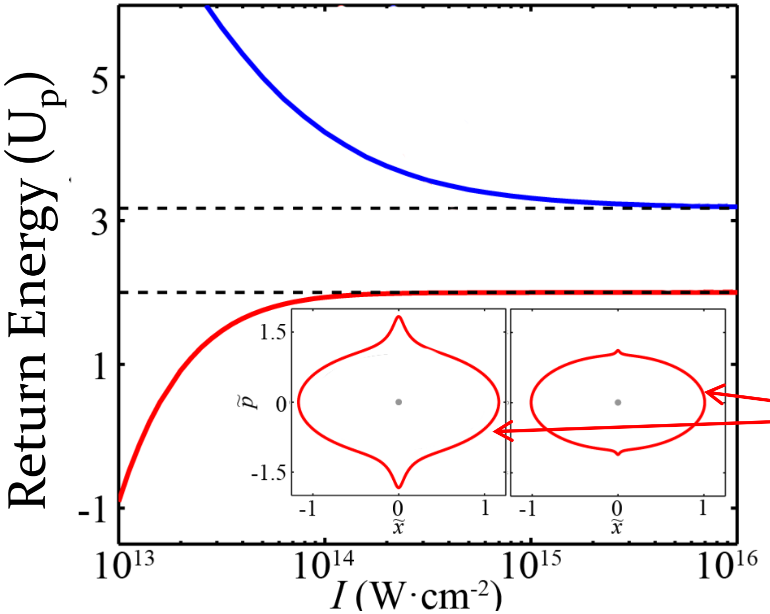
Why always 3.17?



Kamor *et al.*, Phys. Rev. Lett. 112, 133003 (2014)

Why always 3.17

- If the electron is initially close to the stable manifold, it returns to the core



Max return energy

Max return energy along RPO

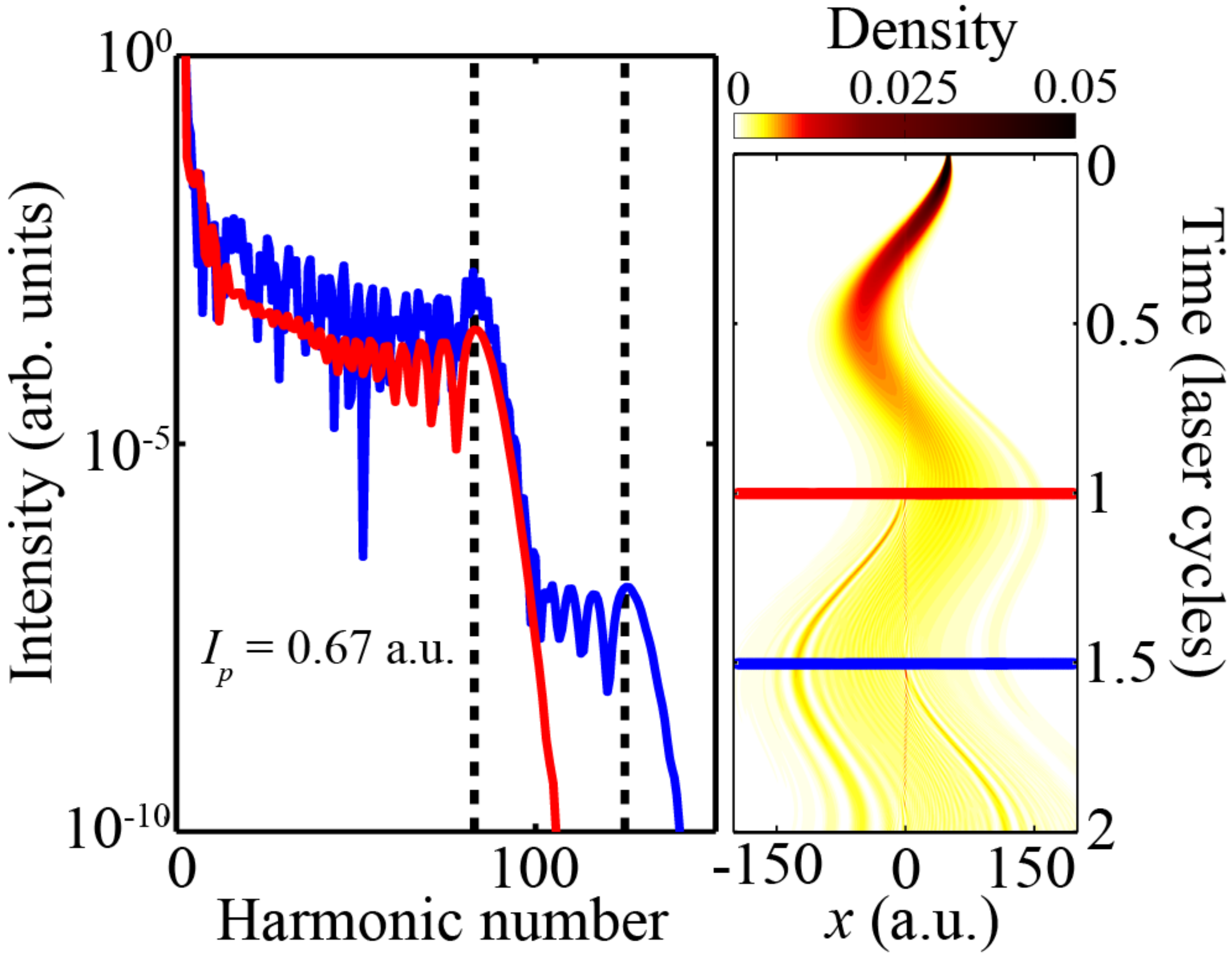
RPO A_1

- Maximum return energy is not constant
- Purely classical recollision scenario (no tunneling)
- Continuous process vs three step model
- Coulomb interaction fully taken into account all along

$$\kappa(I,\omega) \approx \kappa_0 + \kappa_1 \frac{\omega^2}{I}$$

3.17 !

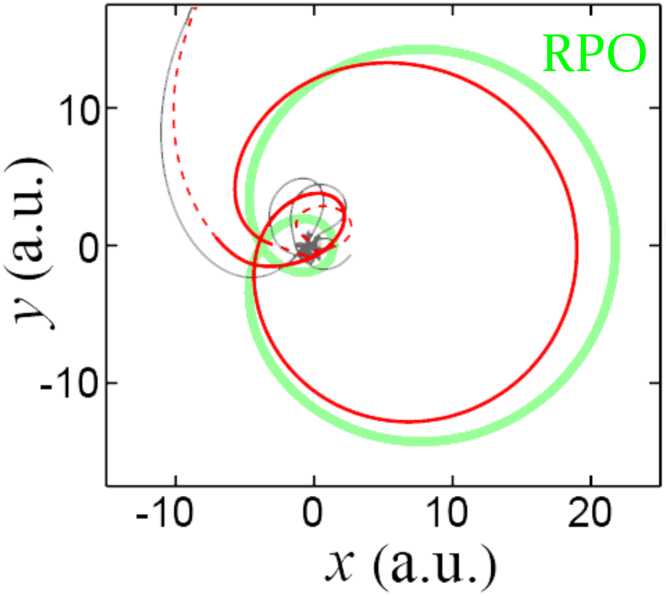
High harmonic generation – linear polarization



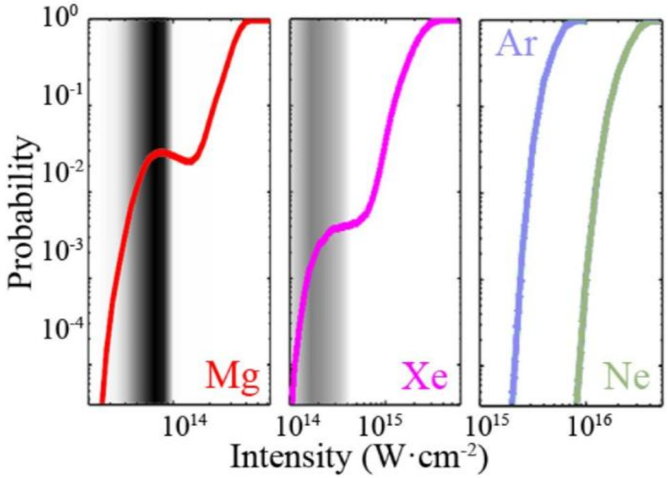
Kamor *et al.*, Phys. Rev. Lett. 112, 133003 (2014)

Mechanism of recollision with circular polarization

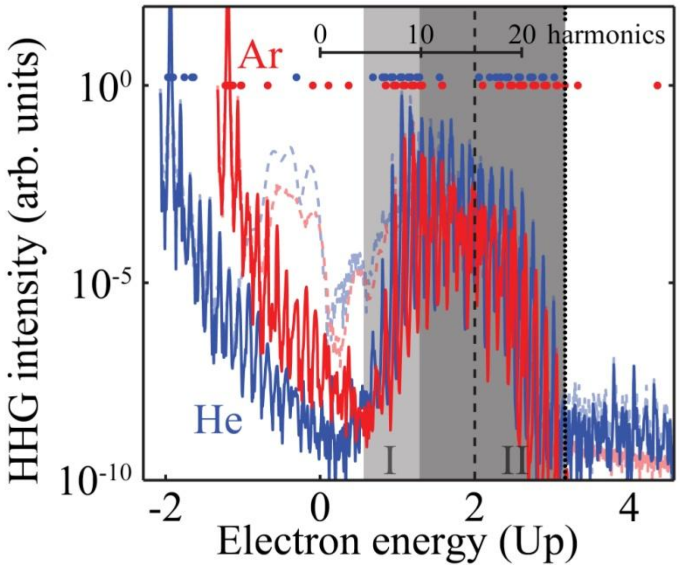
Recollision is organized by RPOs
(Recolliding Periodic Orbits)



NSDI



HHG



What does nonlinear dynamics bring?

- Searches for the collective behavior of trajectories
- Find structures (like Periodic Orbits) which drive the collective behavior.

- Here we considered electron physics rather than atomic physics
- Interactions that matter are:
  - Laser-Electron
  - Electron-Electron
- Energy transfers during recollisions are large compared to level spacing
- Electron spends most time in continuum
- Most importantly…it works!

Conclusion

- Recollision mechanism for LP: role of the Coulomb interaction
- There is recollision for CP!
- Dynamical organization by Recolliding Periodic Orbits
- Take-home message: nonlinear dynamics is a powerful tool to analyze those strong laser-driven systems

… and thanks to

Our publications

- Inner and outer electron models
  - PRL 102, 173002 (2009) & JPB 42, 165602 (2009)
- Mapping model for recollision dynamic
  - PRL 104, 043005 (2010) & PRA 81, 063425 (2010)
- Delayed double ionization
  - PRL 108, 063001 (2012) & PRE 85, 066205 (2012)
- Recollision with circular polarization
  - PRL 105, 083002 (2010)
- Recolliding periodic orbits
  - PRL 110, 253002 (2013) & JPB 47, 041001 (2014) & PRL 112, 133003 (2014)