Hydrogen atoms under magnification*

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* https://doi.org/10.1103/PhysRevLett.110.213001

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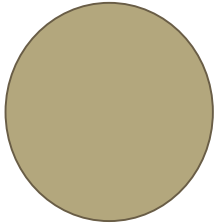
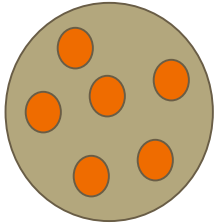
Introduction: Evolution of Atomic Models

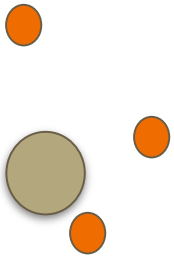
Dalton model (~1800):



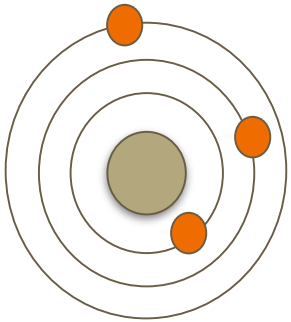
Thomson model (1897):



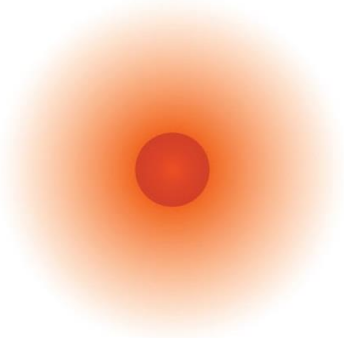
Rutherford model (1909):



Bohr model (1913):

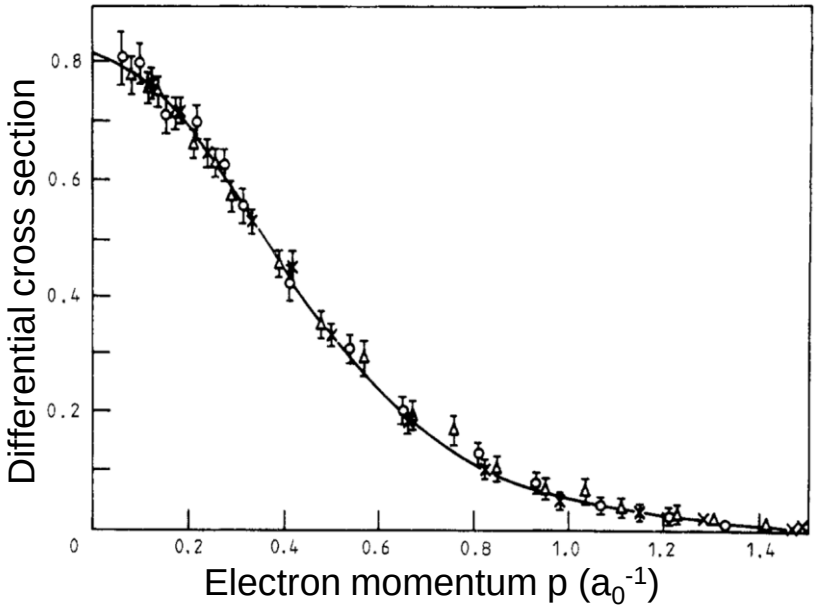


Electron cloud:



Background: Mapping of hydrogen using collision

- Non-coplanar (e,2e) spectroscopy
  - Hydrogen electron is removed by incident electron beam and momentum distribution measured
  - Experimental results match theoretical results for various electron beam energies



B. Lohmann, E. Weigold, Phys. Lett. A. 86, 3, 139-141 (1981).

Background: Rydberg wave packet emission

- Non-coplanar (e,2e) spectroscopy
  - Hydrogen electron is removed by incident electron beam and momentum distribution measured
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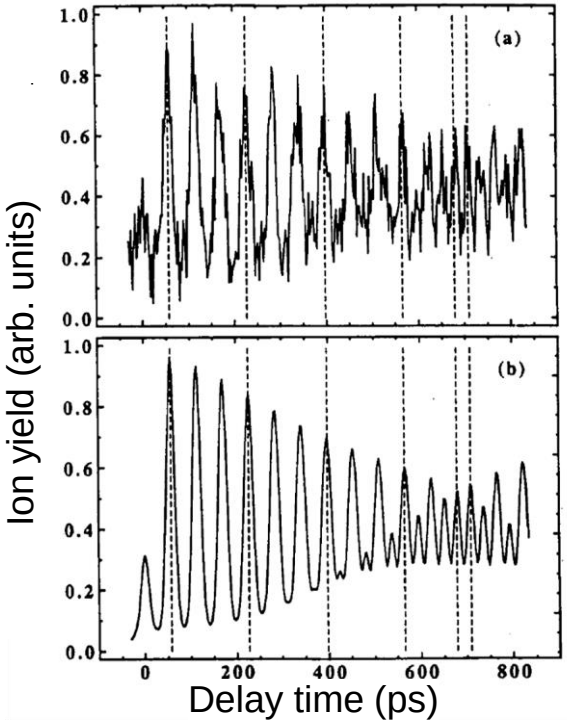
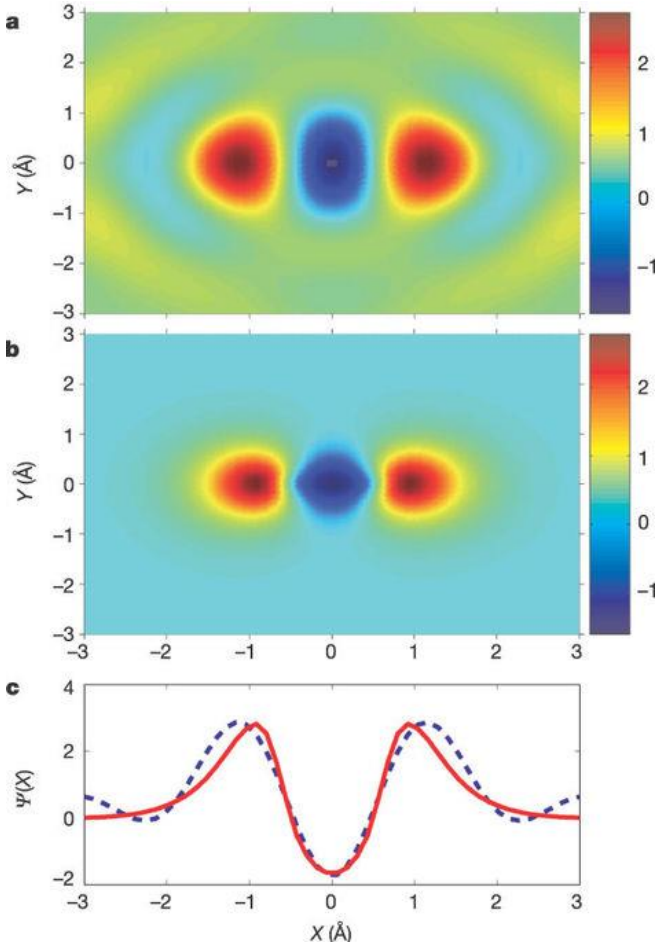
- Fluorescence measures precession of radial wave packet around nucleus
  - Ultrafast laser measured precession in K, which is compared to theoretical results



L. D. Noordam and R. R. Jones, J. Mod. Opt. 44, 2515(1997).

Background: Orbital tomography

- Non-coplanar (e,2e) spectroscopy
  - Hydrogen electron is removed by incident electron beam and momentum distribution measured
  - Experimental results match theoretical results for various electron beam energies
- Fluorescence measures precession of radial wave packet around nucleus
  - Ultrafast laser measured precession in K, which is compared to theoretical results
- High harmonic emission shows 3-dimensional shape of highest electronic orbital
  - Shown for nitrogen gas (N_2)



J. Itatani, J. Levesque, et al., Nature(London) 432, 867 (2004).

Photoionization Microscopy

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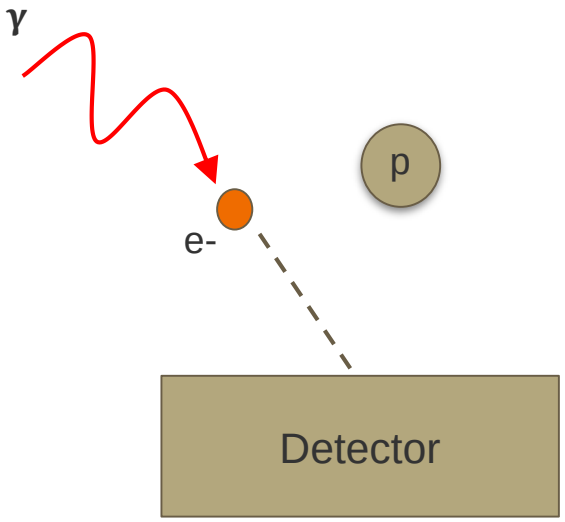
  - V. D. Kondratovich and V. N. Ostrovsky, J. Phys. B 23,3785 (1990).
- Enables image and measurements of radial wavefunctions



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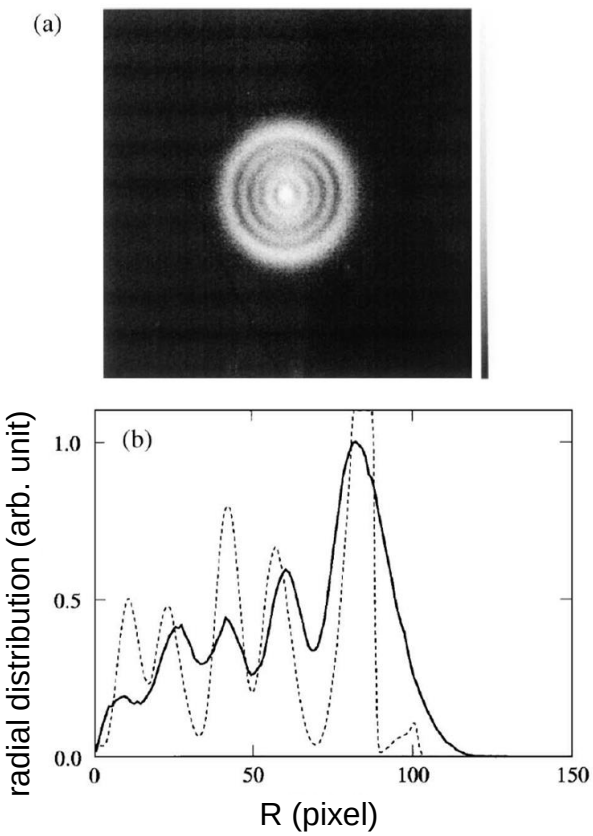

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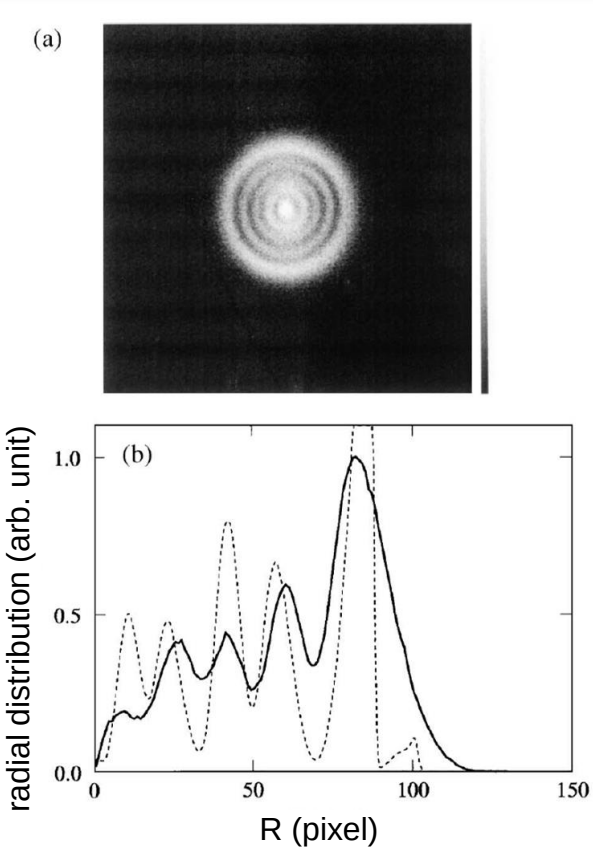
- Performed again with Li
  - S. Cohen, M. M. Harb, et al., Phys. Rev. Lett. 18, 183001, (2013).



Theory: Photoionization Microscopy and Stark effect

Basic set: hydrogen in electric field

$$\hat{H} = -\frac{\nabla^2}{2} - \frac{Z}{r} + Fz$$

where $Z = 1$. F =electric field.

Introduce parabolic coordinate:

$$\begin{aligned} \eta &= r - z \\ \xi &= r + z \\ \varphi &= tan^{-1}(\frac{y}{x}) \end{aligned}$$

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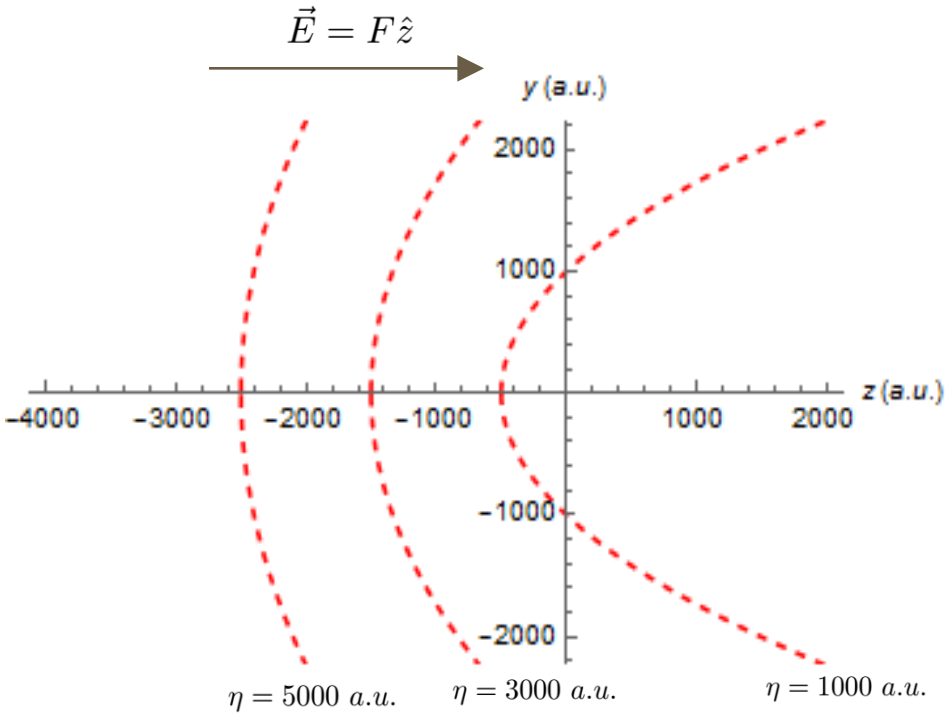

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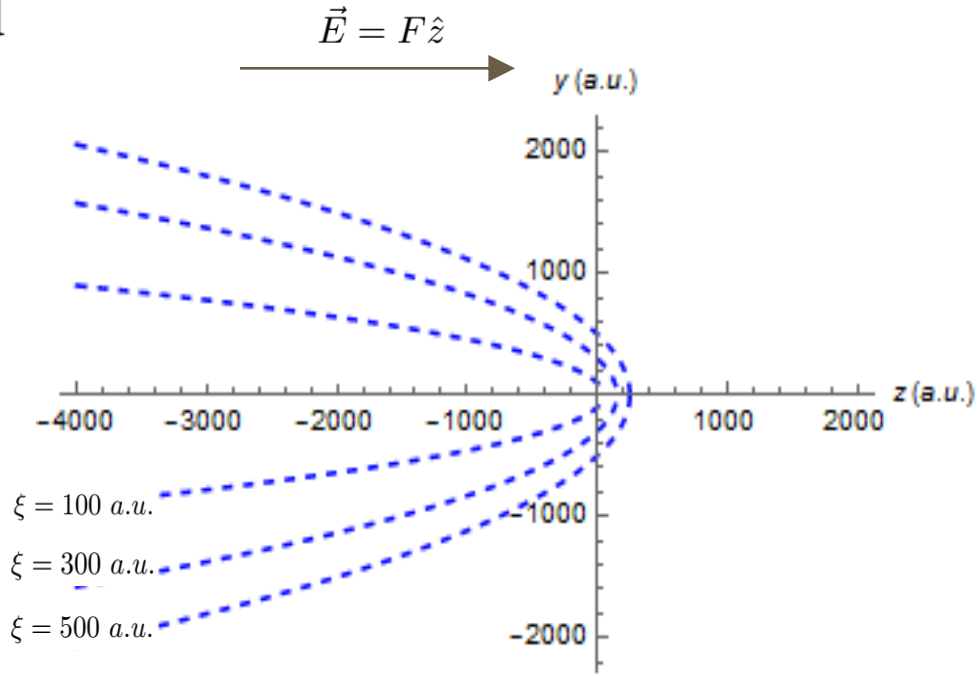

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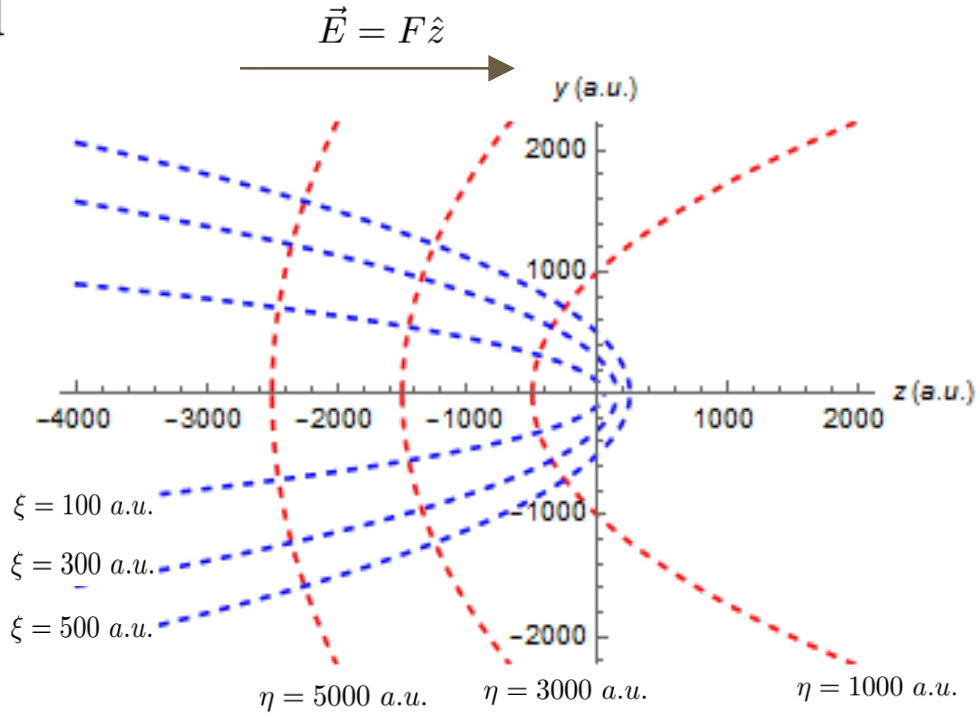

Quantum numbers $\{n_1,n_2,m\}$ of Stark state

Wave function becomes totally decoupled in parabolic coordinates:

$$\Phi(\xi,\eta,\varphi) = (2\pi\eta\xi)^{-1/2}\ \chi_1(\xi)\chi_2(\eta)e^{im\varphi}$$

Eigenstates are denoted by quantum numbers $\{n_1, n_2, m\}$ = nodes in $\{\xi, \eta, \varphi\}$.

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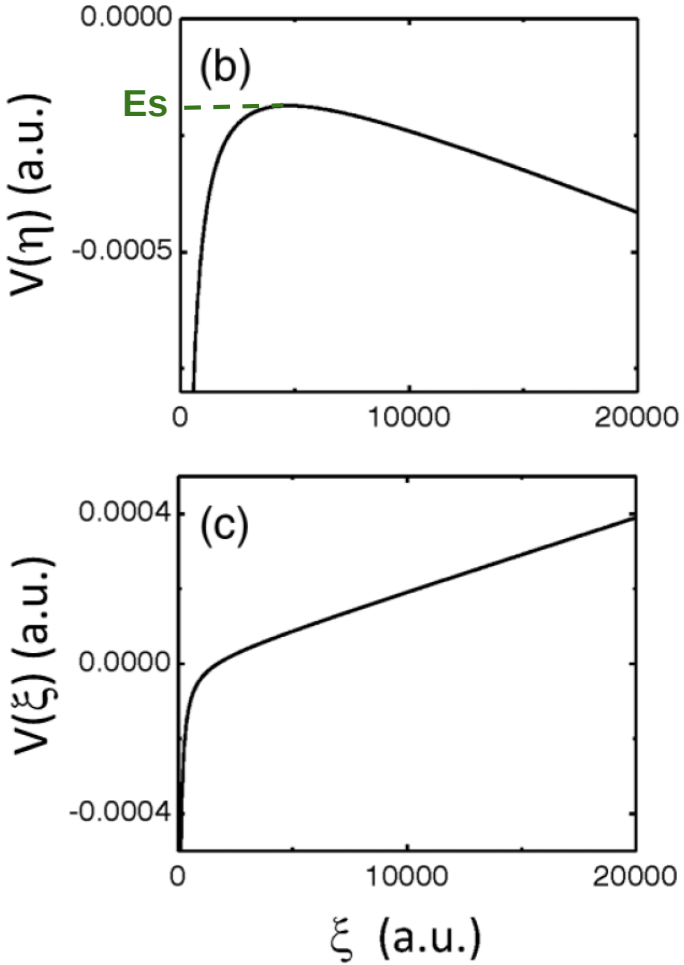
Each χ_1 and χ_2 satisfy 1D Schrodinger equation with effective potentials:

$$\begin{aligned} V(\eta) &\equiv -\frac{Z_2}{2\eta} + \frac{m^2-1}{8\eta^2} - \frac{F\eta}{8} \\ V(\xi) &\equiv -\frac{Z_1}{2\xi} + \frac{m^2-1}{8\xi^2} + \frac{F\xi}{8} \end{aligned}$$



Effective potential for m=0

$$V(\eta) \equiv -\frac{Z_2}{2\eta} + \frac{m^2-1}{8\eta^2} - \frac{F\eta}{8}$$

$$V(\xi) \equiv -\frac{Z_1}{2\xi} + \frac{m^2-1}{8\xi^2} + \frac{F\xi}{8}.$$

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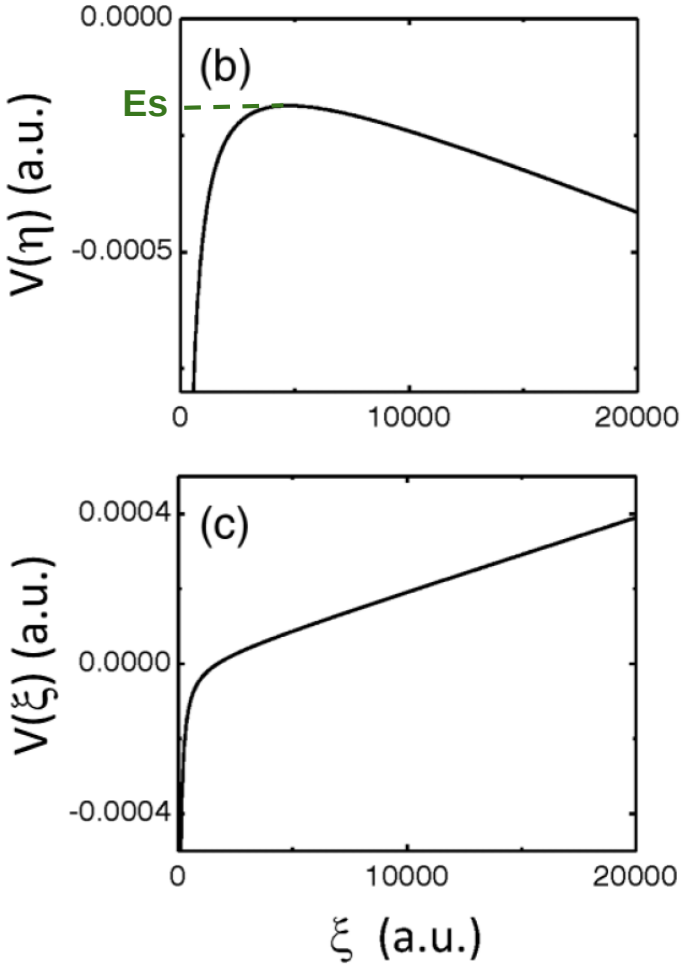

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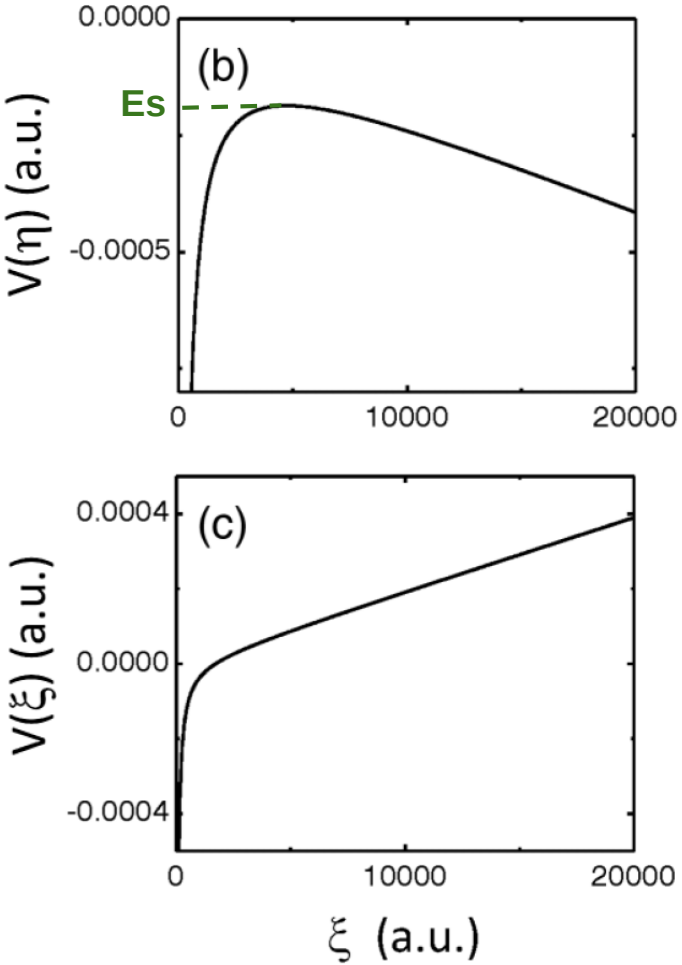
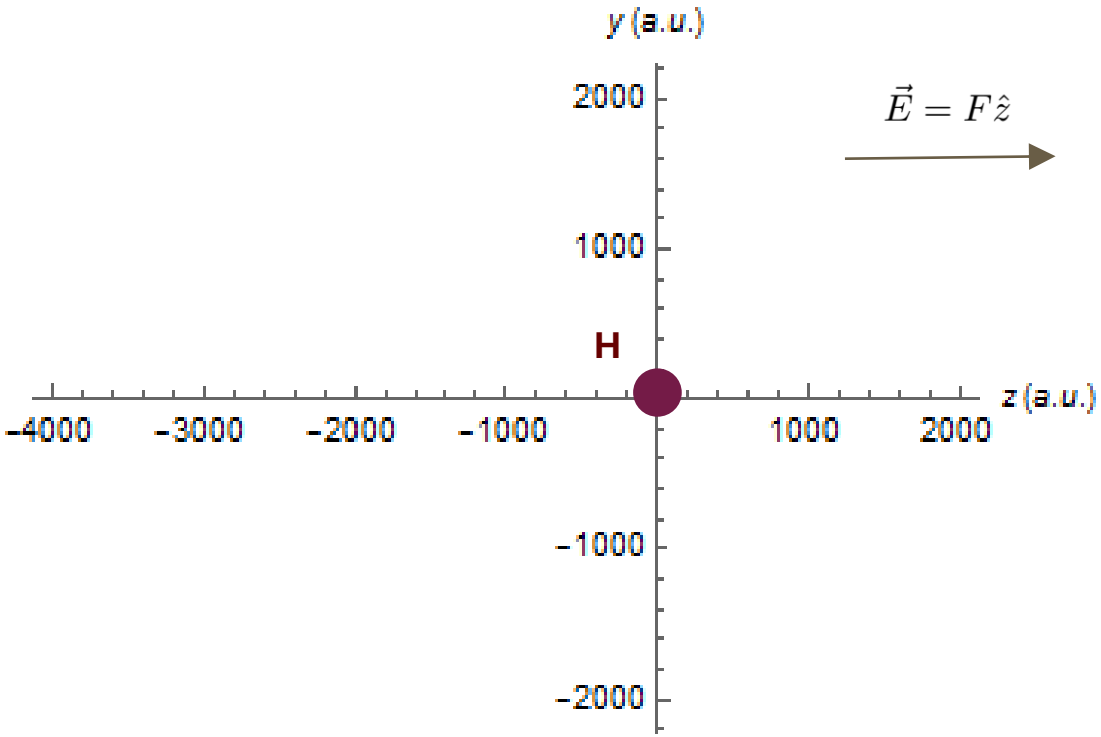
$$V(\xi) \equiv -\frac{Z_1}{2\xi} + \frac{m^2-1}{8\xi^2} + \frac{F\xi}{8}.$$

ξ direction is always bounded.
But e^- may release from η direction.





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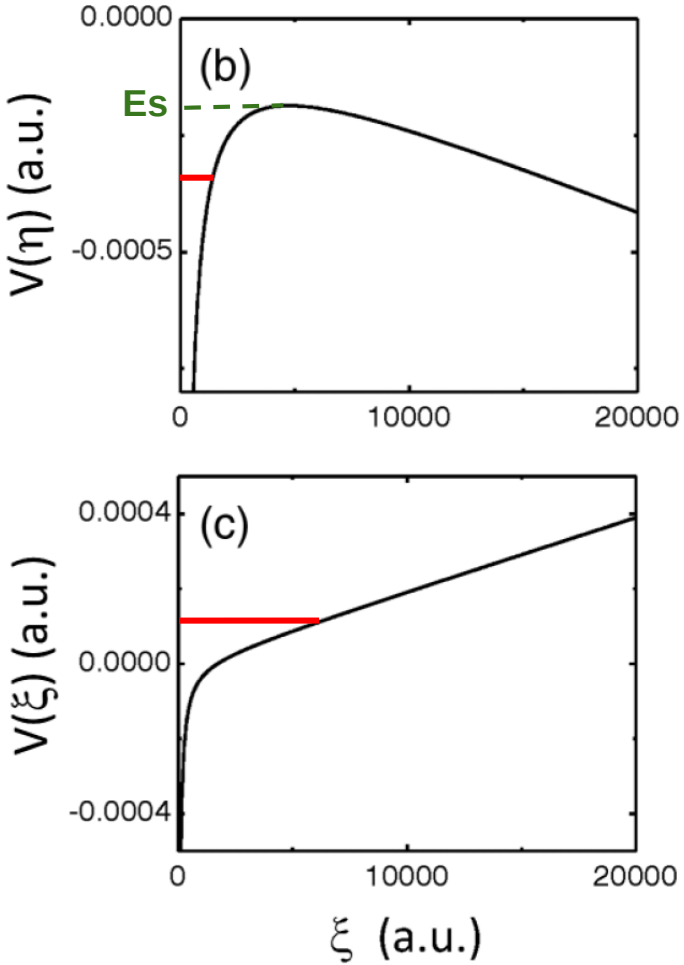
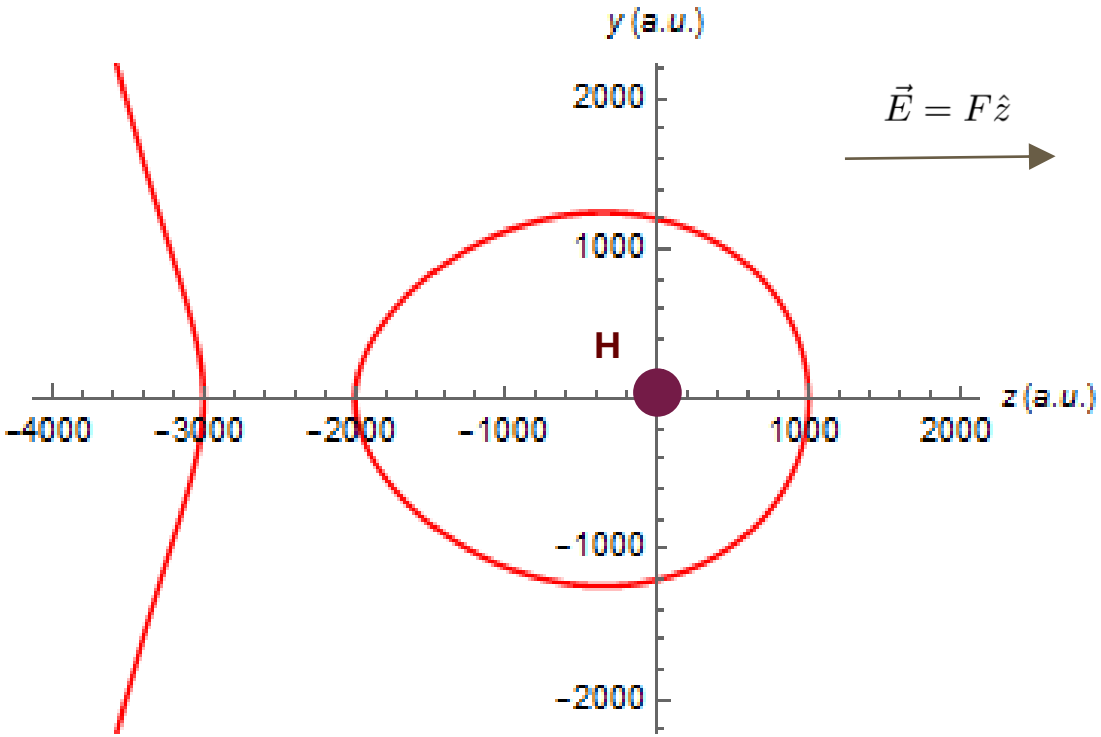


$E < E_s$: wavefunction is bounded.



Equal energy line of E

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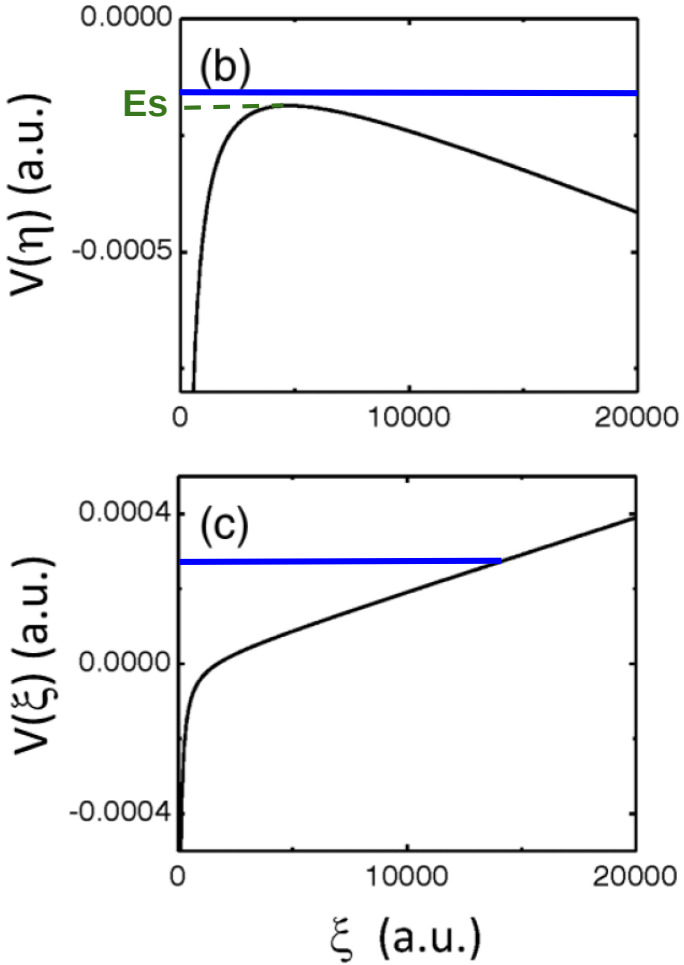
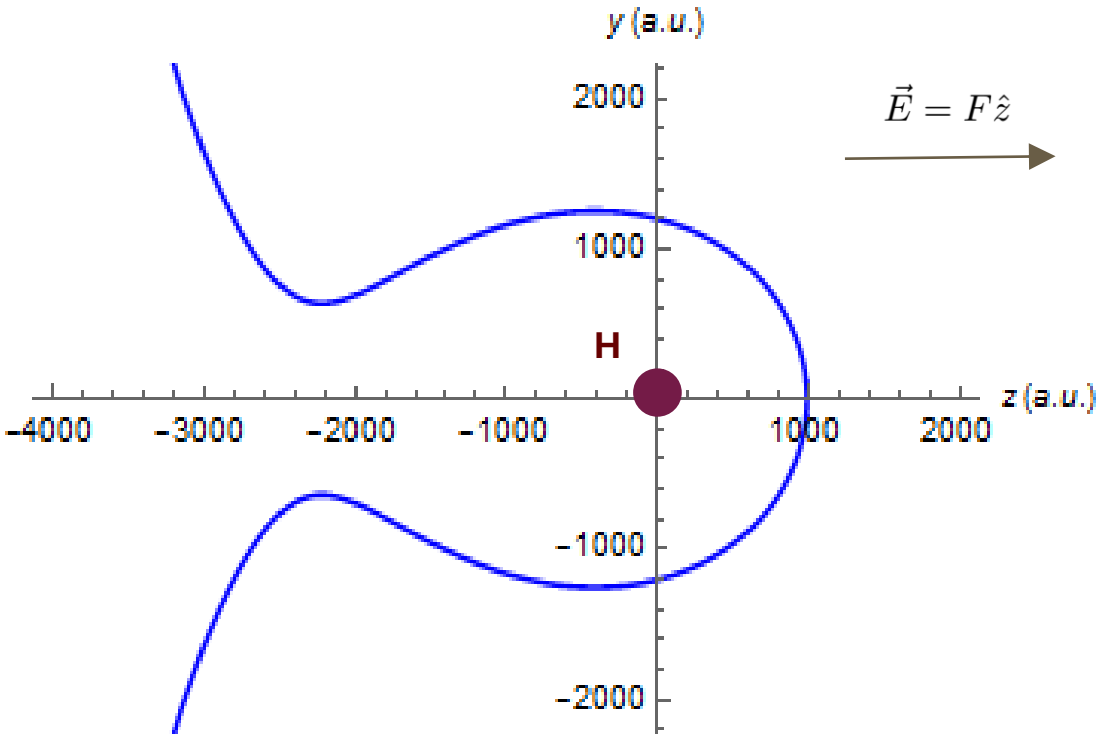


$E > E_s$: wavefunction can release to $\eta = \infty$.



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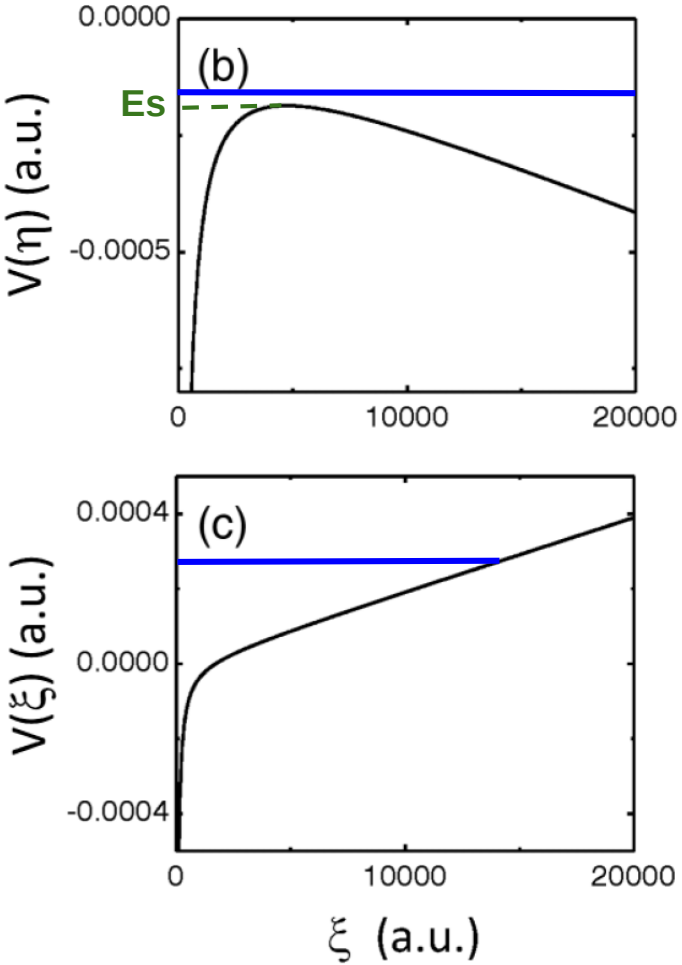

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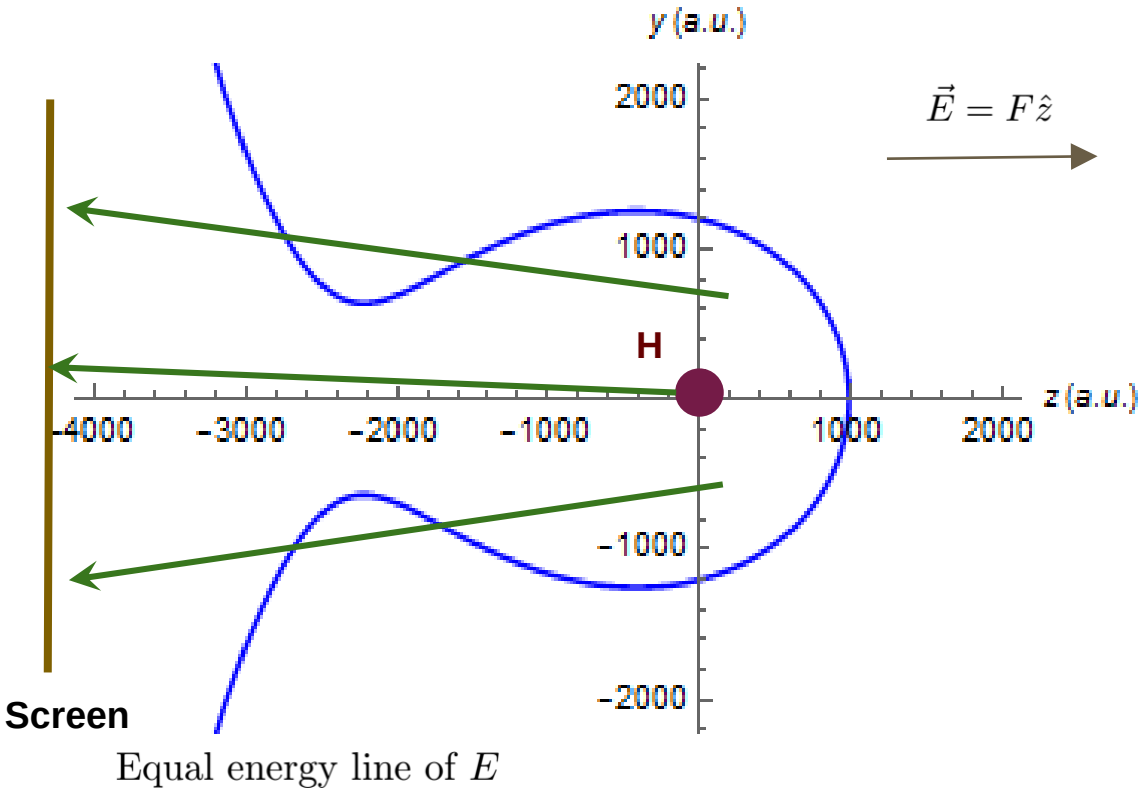

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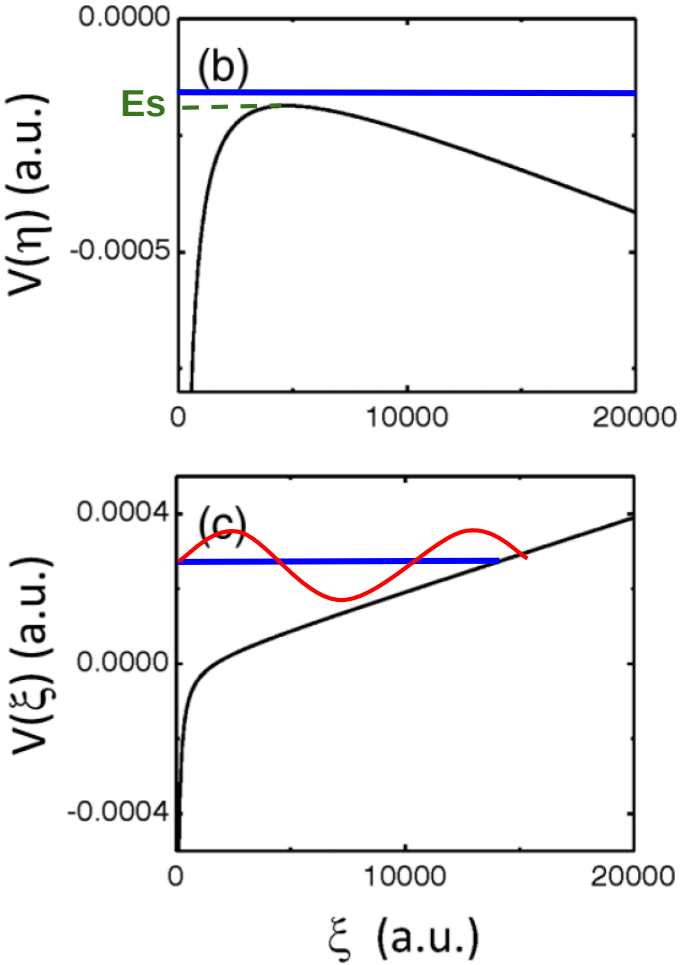
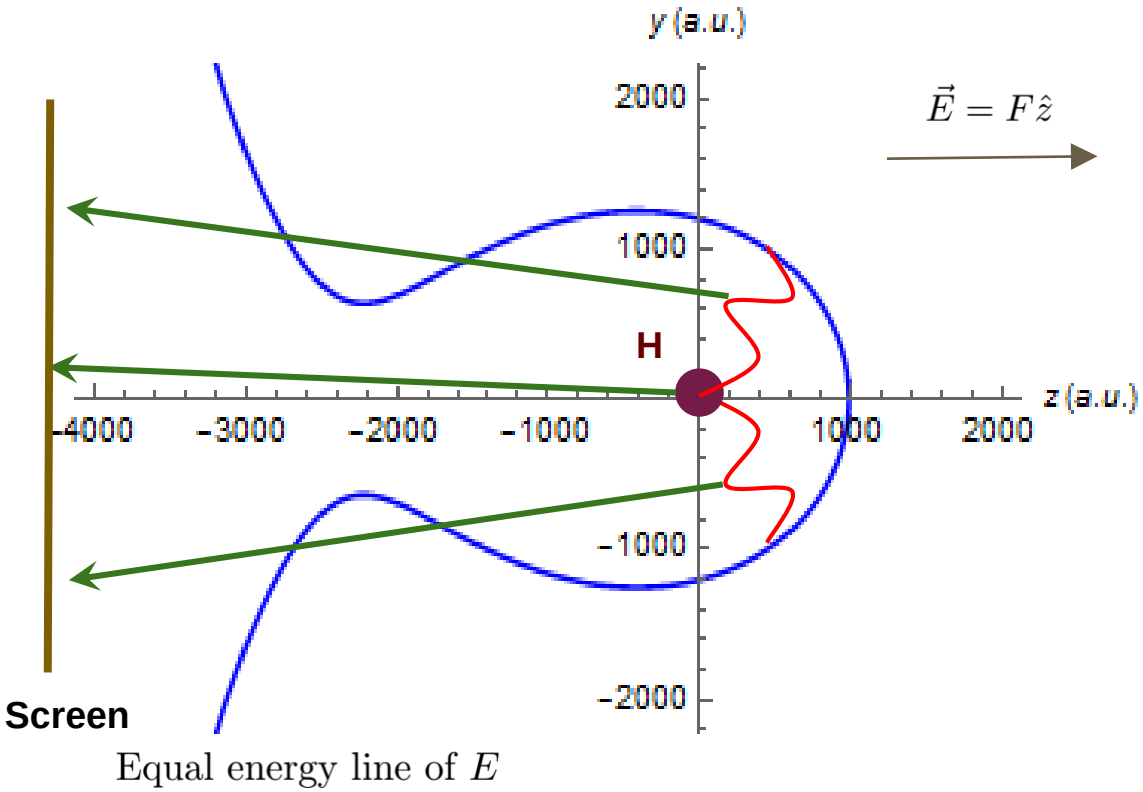
n_1 = number of dark rings on screen.





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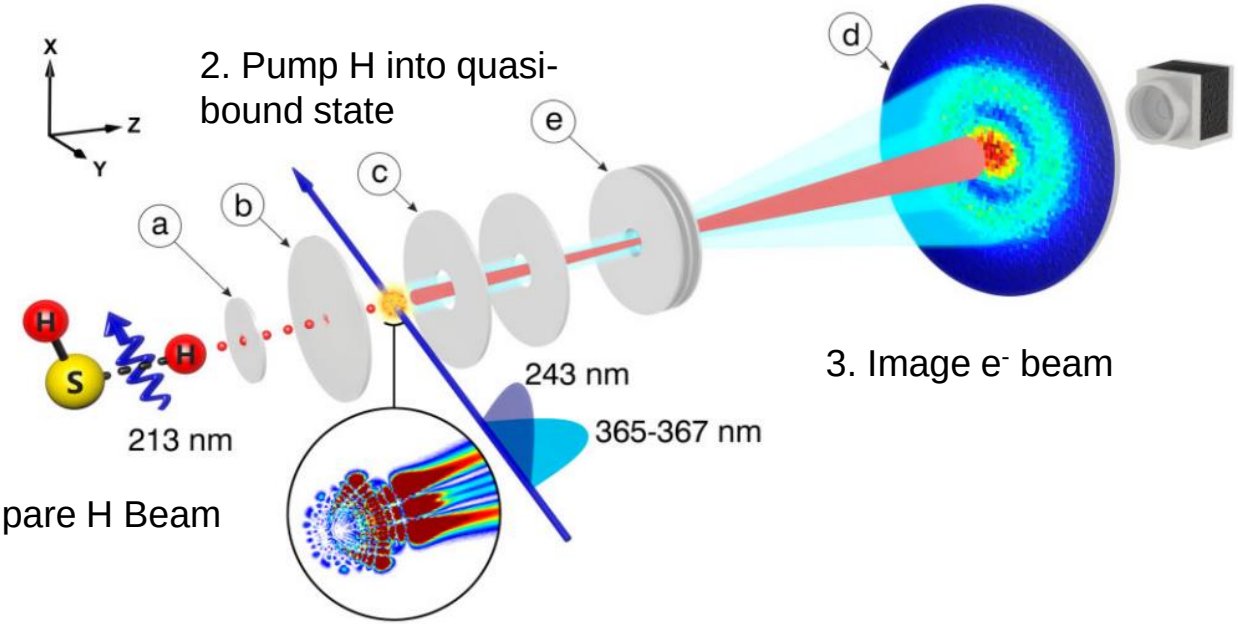
Setup for Photoionization of Hydrogen



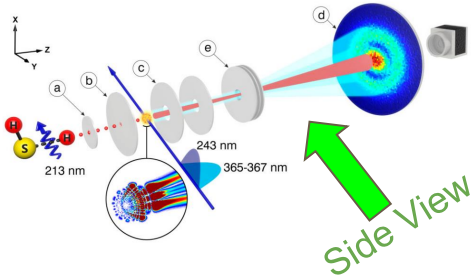
2. Pump H into quasi-bound state

3. Image e^- beam

1. Prepare H Beam

Stodolna, Rouze, Lépine, et al, Phys. Rev. Lett. 110, 213001 (2013).

Results: Imaging the nodal structure of electron density

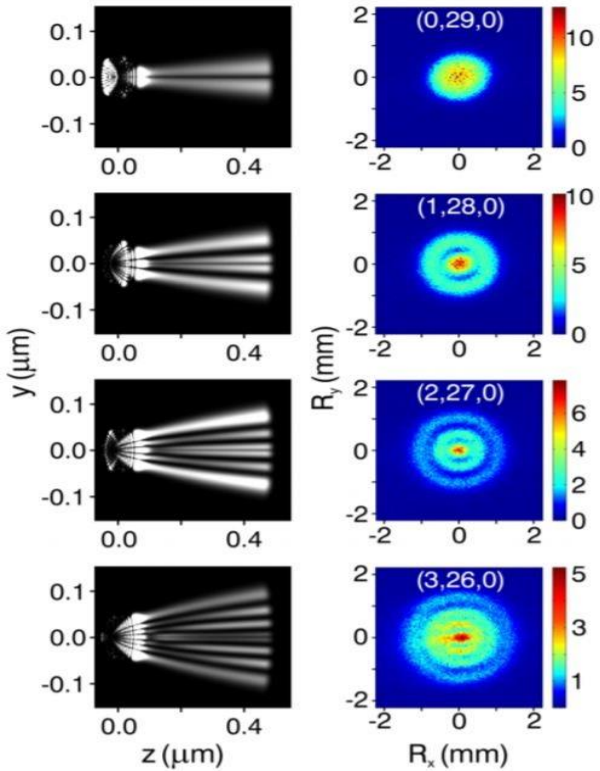


n1=0
n2=29

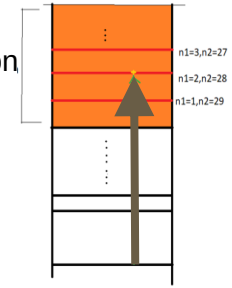
n1=1
n2=28

n1=2
n2=27

n1=3
n2=26



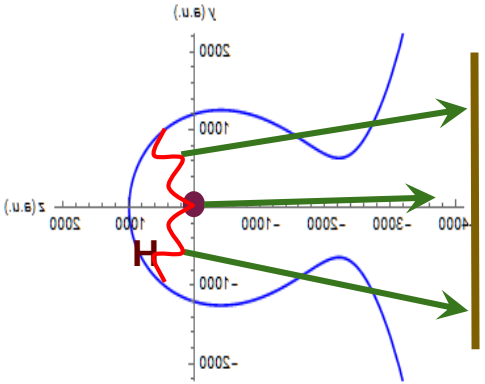
Ionization Region

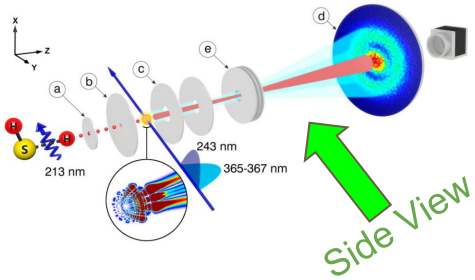


"Quasi-bound states"

Stodolna, Rouze, Lépine, et al, Phys. Rev. Lett. 110, 213001 (2013).

Results: Quasibound States Tunnels to screen preserving Nodal Structure





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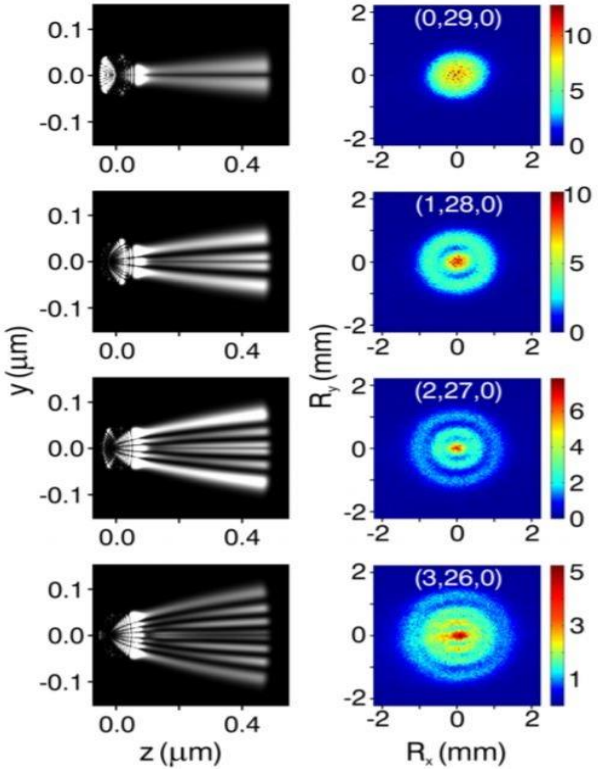
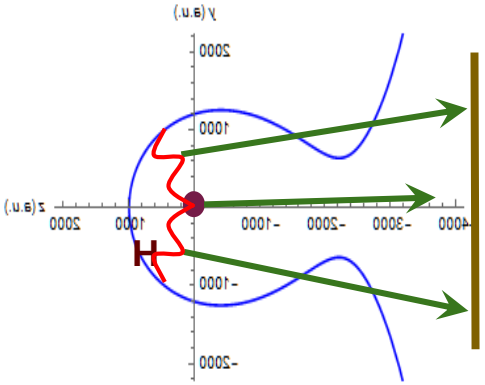
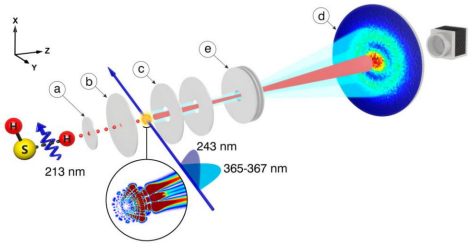


Stodolna, Rouze, Lépine, et al, Phys. Rev. Lett. 110, 213001 (2013).

Results: Comparing with Theory





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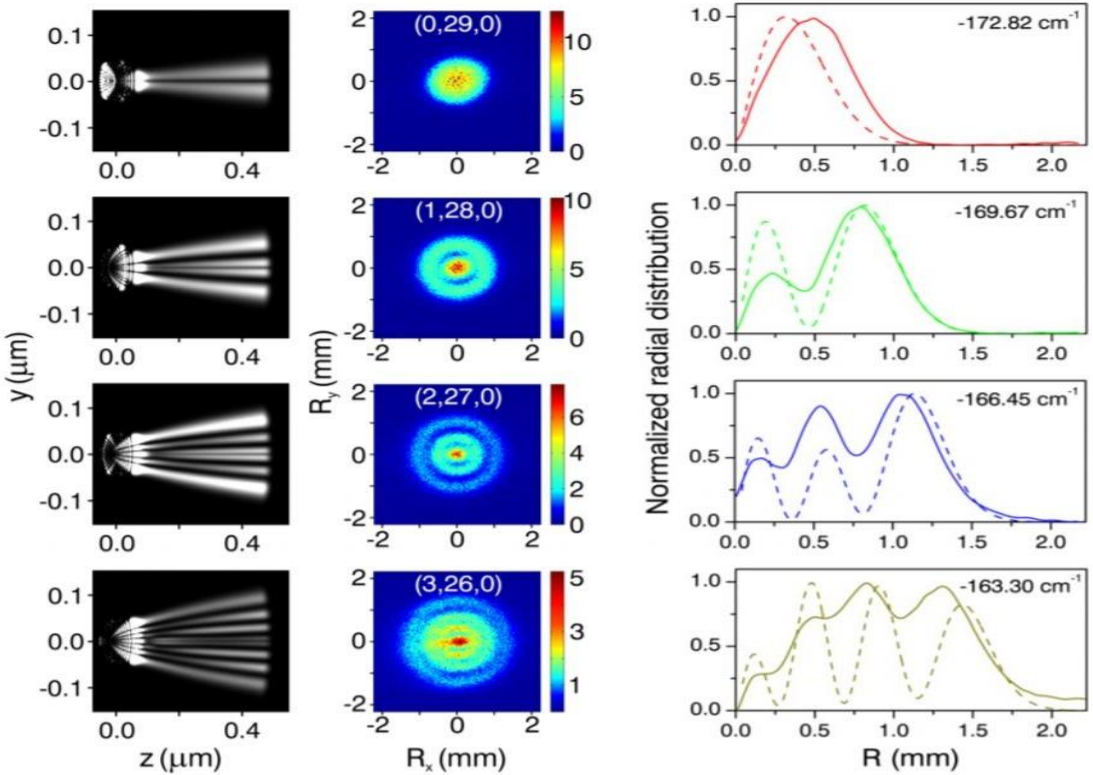
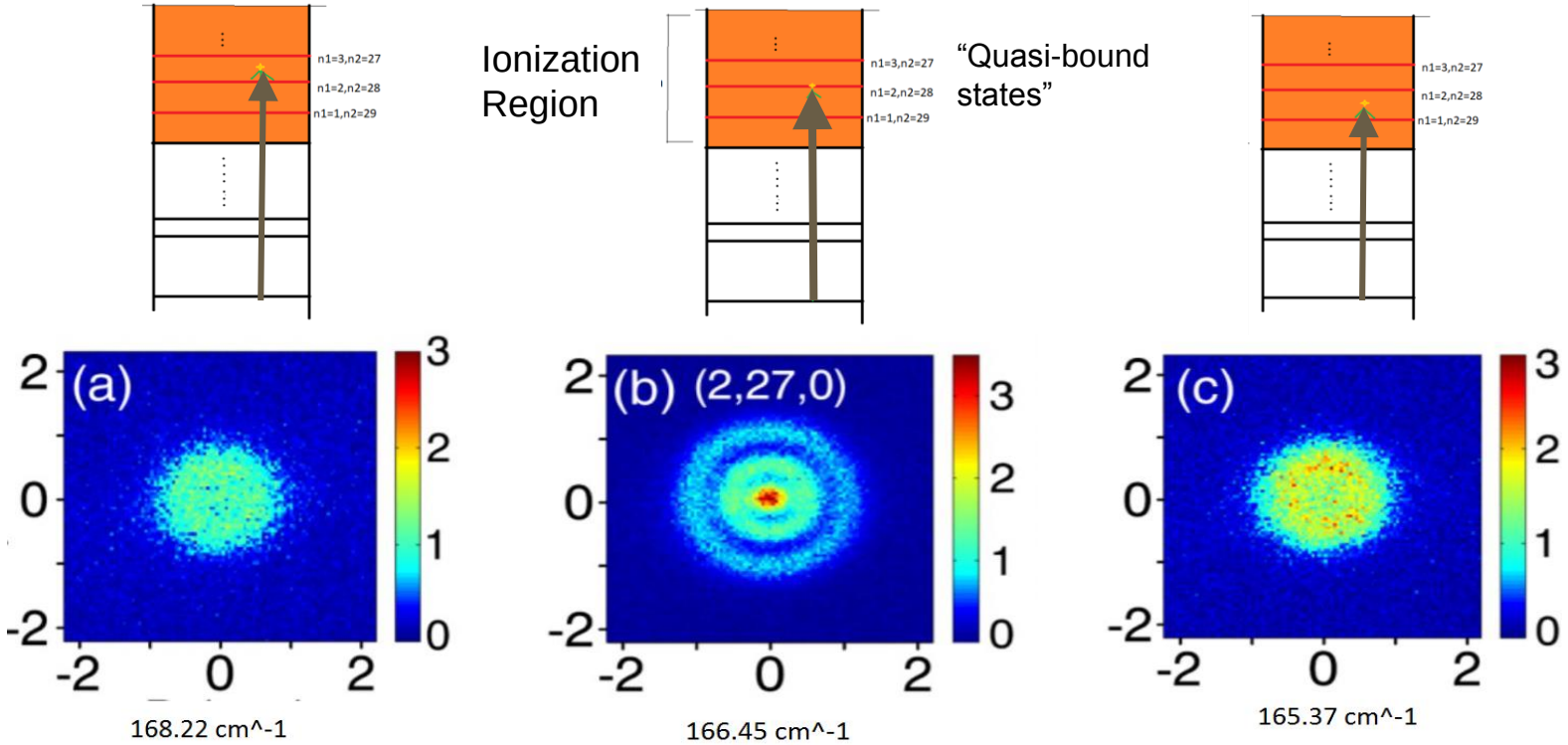


Stodolna, Rouze, Lépine, et al, Phys. Rev. Lett. 110, 213001 (2013).

Results: Loss of fringes at classical Ionization



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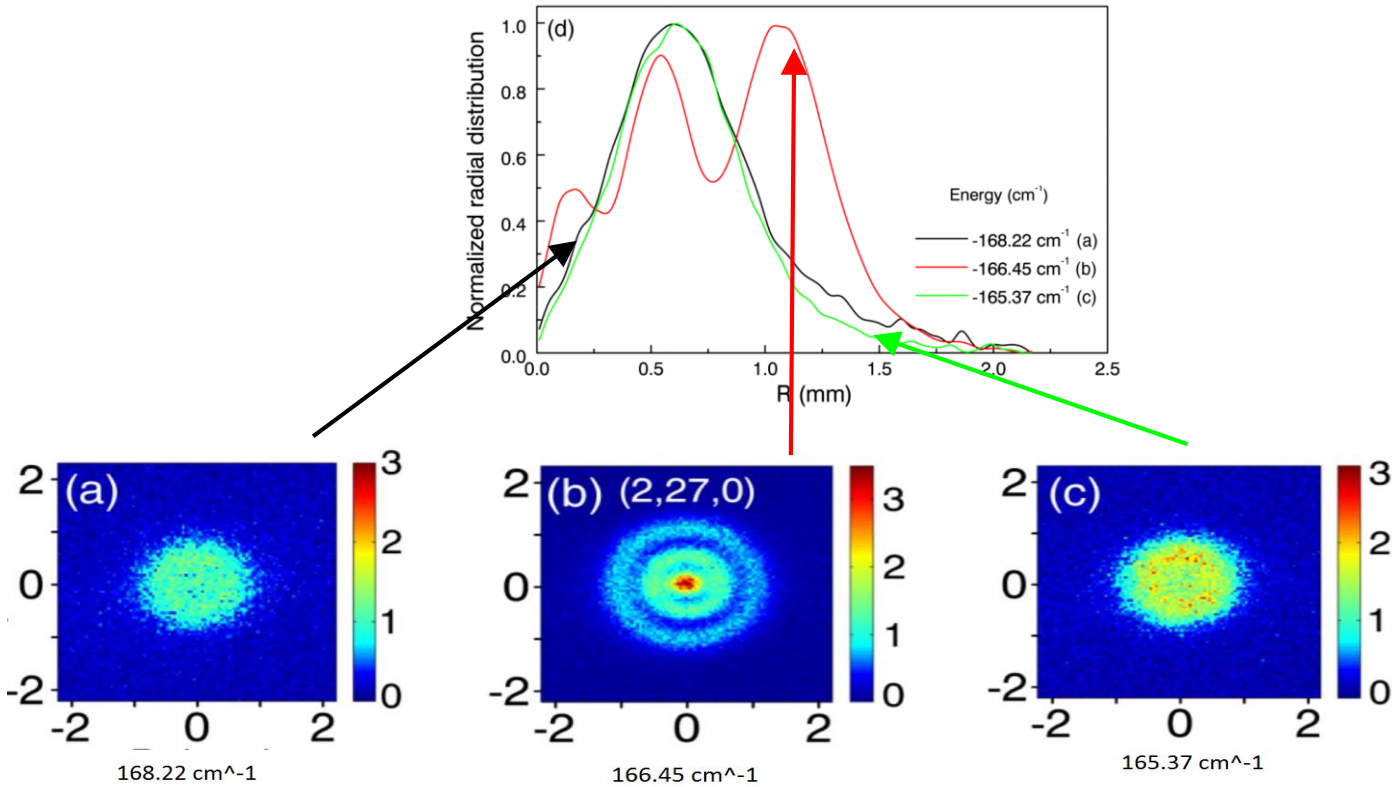
Results: Departure of Tunnel effect from Classical Ionization



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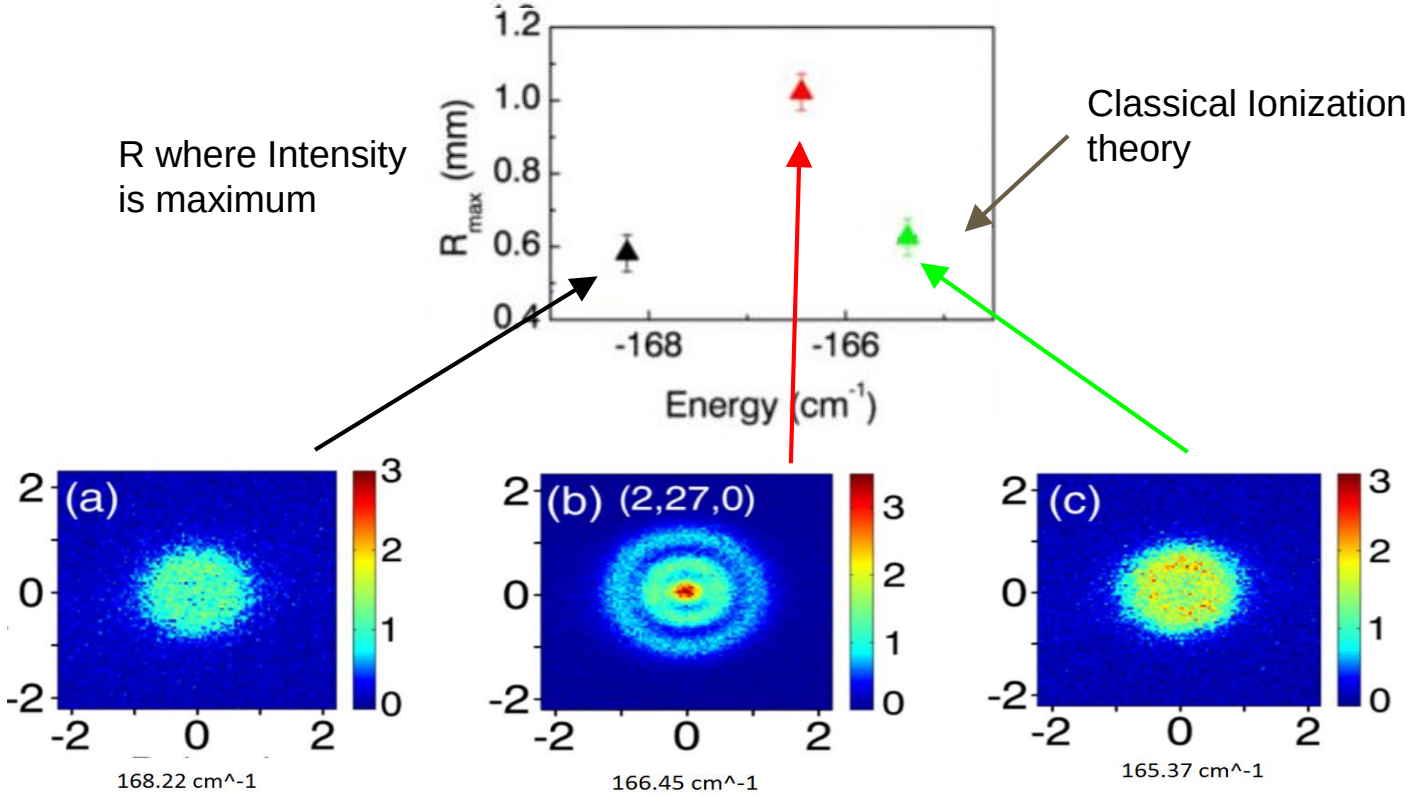
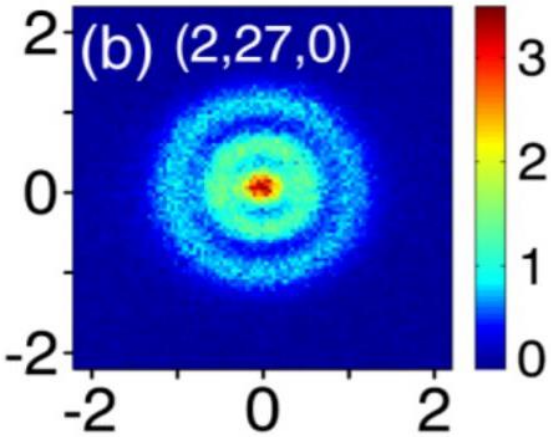


Stodolna, Rouze, Lépine, et al, Phys. Rev. Lett. 110, 213001 (2013).

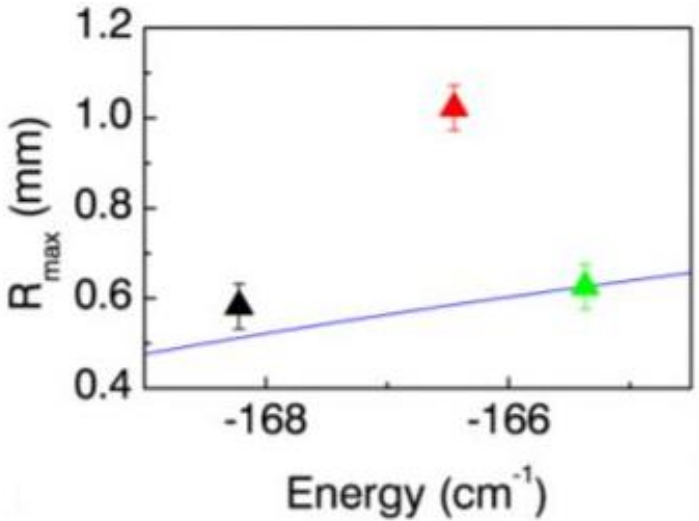
Results: Summary

1) Imaging the nodal structure



166.45 cm^-1

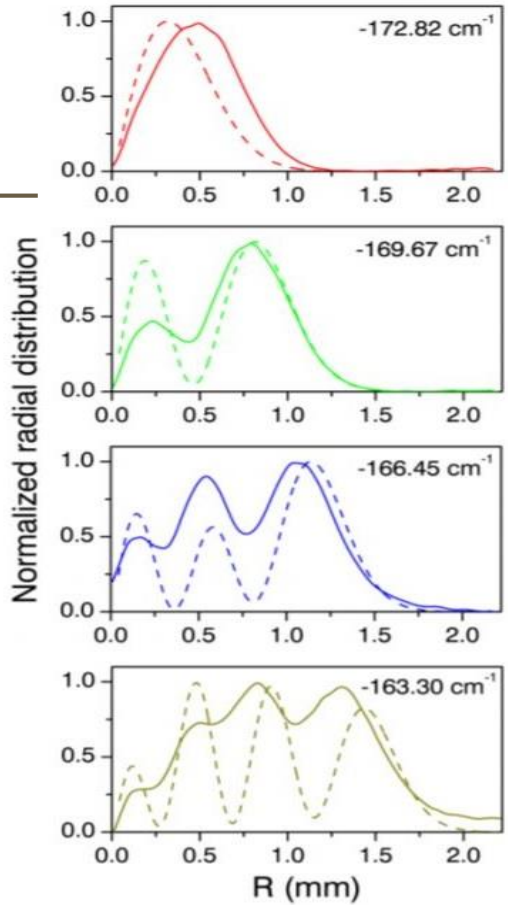
2) Verification of “remarkable tunnel effect”



Stodolna, Rouze, Lépine, et al, Phys. Rev. Lett. 110, 213001 (2013).

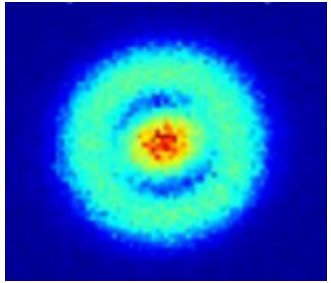
Critiques

- Differences between observed and calculated results are poorly explained.
- Not enough experimental details, even in the supplemental material.
- This was the first time the photoionization microscopy (proposed 30 years ago) was implemented for the hydrogen atom.
- It validated predictions made ~30 years ago (tunnel effect).



Stodolna, Rouze, Lépine, et al, Phys. Rev. Lett. 110, 213001 (2013).

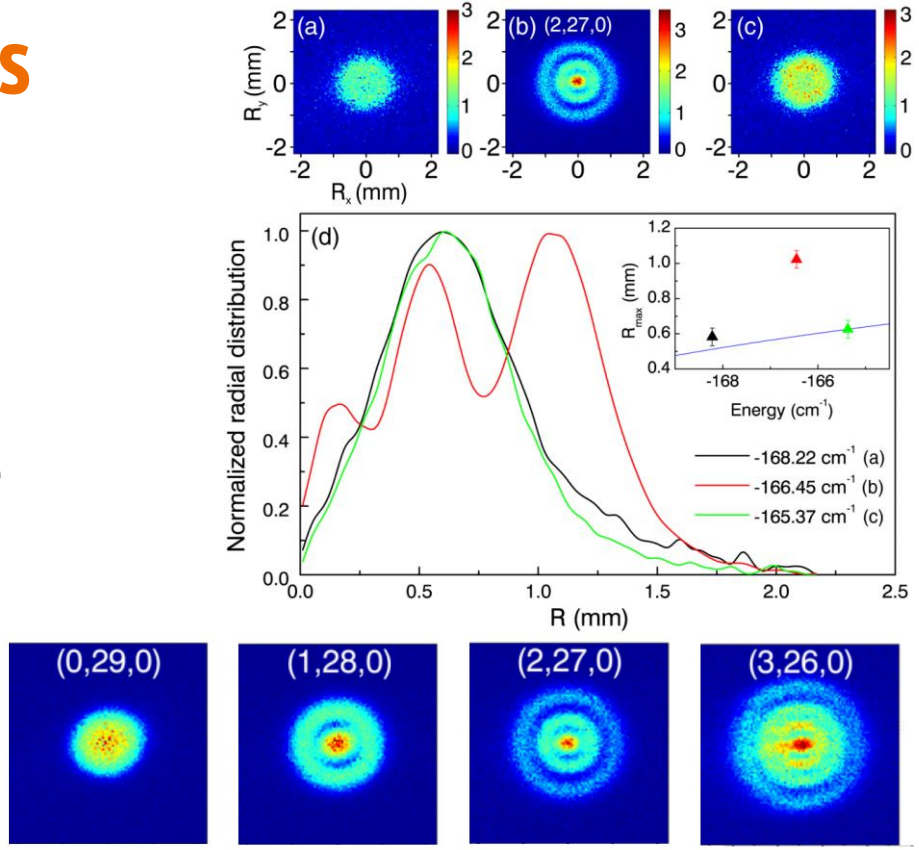
Impact and related work



- Cited by 36 papers to date (most cited in 2015).
- Shared even in social media:
  - Facebook - Shares, Likes & Comments: 237
  - Google+ - +1s: 282
  - Twitter - Tweets: 21
- Subsequent work:
  - Visualization of coupling between Stark states. (Stodolna, Lépine, Bergeman, 2014)
  - Imaging photoelectron momentum distribution of Hydrogen atom. (Murakami, Chu, et al, 2016)

Summary and questions

- Photoionization microscopy implemented for the first time for a hydrogen atom.
- Observation of nodal structure and tunnel effect (showing the quantum nature of the hydrogen atom).



Stodolna, Rouze, Lépine, et al, Phys. Rev. Lett. 110, 213001 (2013).