Atomic High Harmonic Generation and Ionization with Circular Polarization

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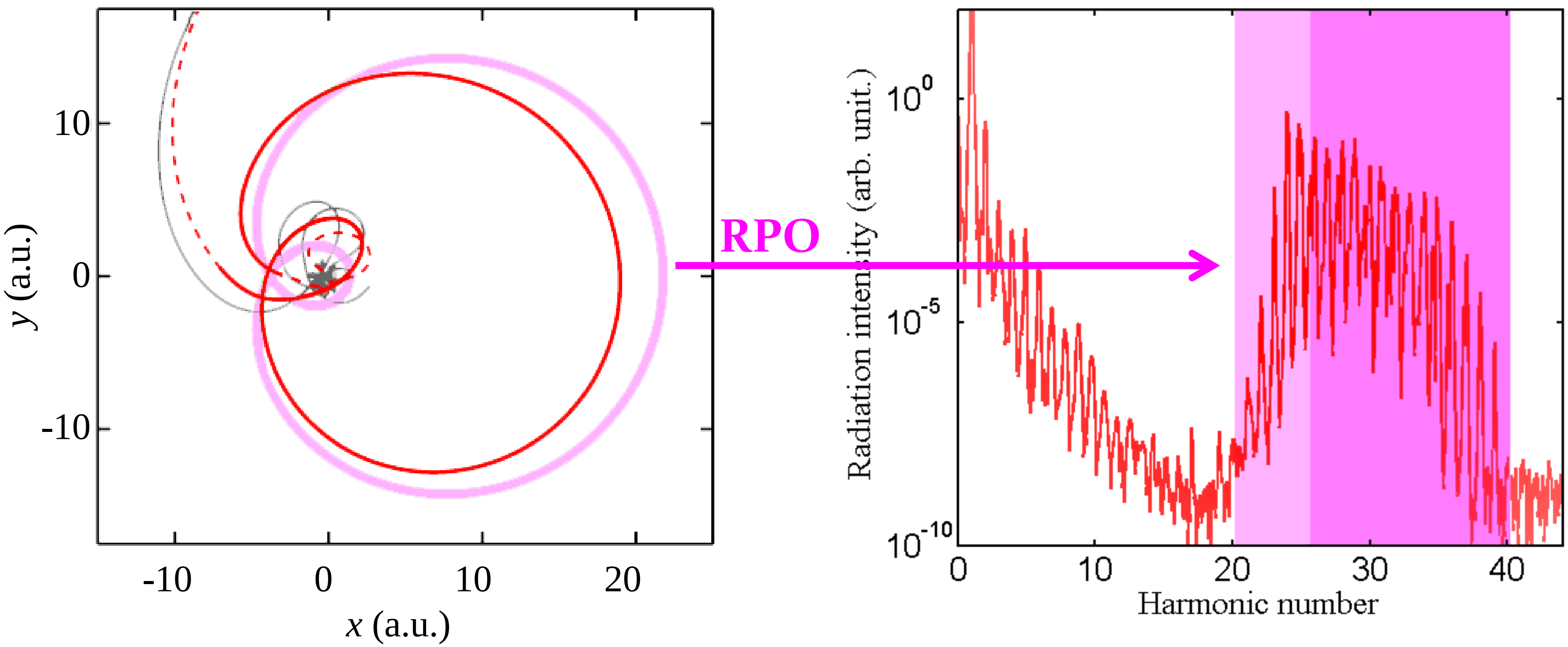
Financial supports:

Recollision with circular polarization … so what?

- Quantum/classical correspondence
  - Atomic recollision and HHG with circular polarization



- … but selection rules

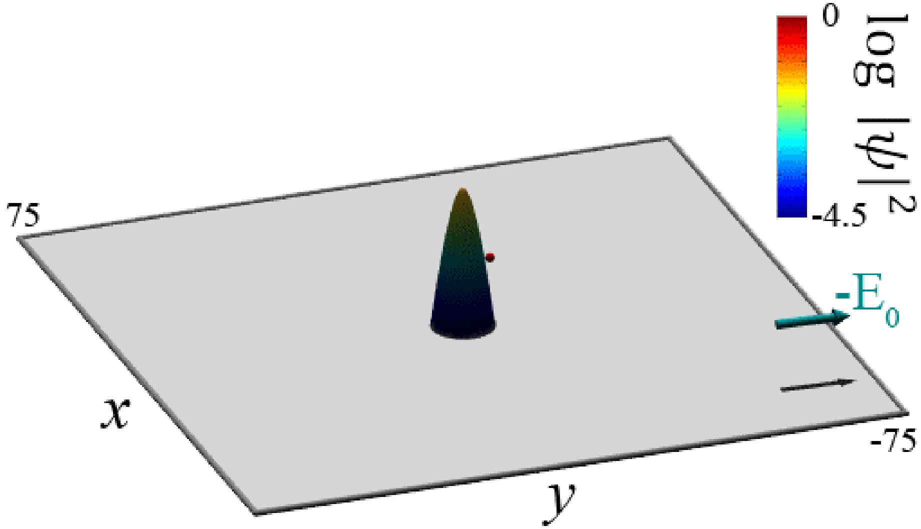
What selection rules have to say about it …

"For a given qth harmonic process ($q \geq 3$) involving q photons, selection rules in the weak field approximation (Delone and Krainov 1985) prohibit the generation of the harmonic by only the circularly polarized term. That is because for the q-photon up transition from the ground state using circularly polarized field, we have $\Delta M = \pm q$ where M is the magnetic quantum number. For one-photon qth harmonic generation, the down transition from the upper state with $M = \pm q$ back to the ground state is forbidden because $\Delta M \neq \pm 1$ (note that for simplicity we assume $J=0$ and $M=0$ in the ground state, where J is the total angular momentum quantum number). On the other hand, harmonic generation by the purely linearly polarized term is always permitted because one can always create the upper state with $J = \pm 1$ and $M=0$ from the general q-photon transition selection rules $\Delta J = q, q-1, \ldots, -q$; $\Delta M = 0$ so that the selection rules for one harmonic down transition, $\Delta J = \pm 1$ and $\Delta M = 0$, can always be satisfied."

Liang *et al.*, JPB **27**, 1276 (1994)

No recollision with circular polarization?

- Quantum/classical (2D)



- Experimental confirmation

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

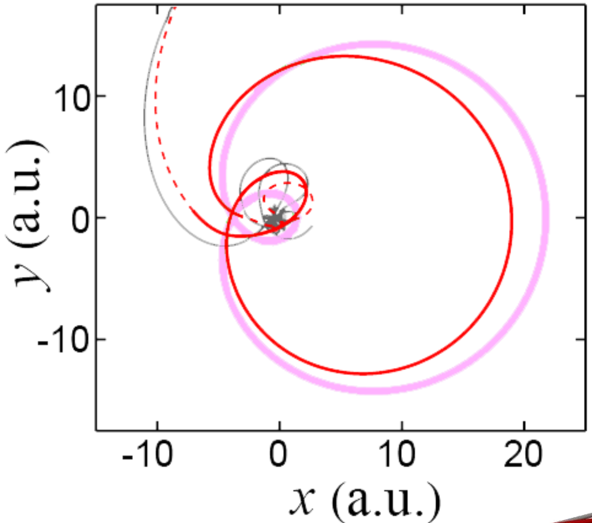
- Strong field approximation:

"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 <u>wavepacket never returns to the ion of its birth</u>"

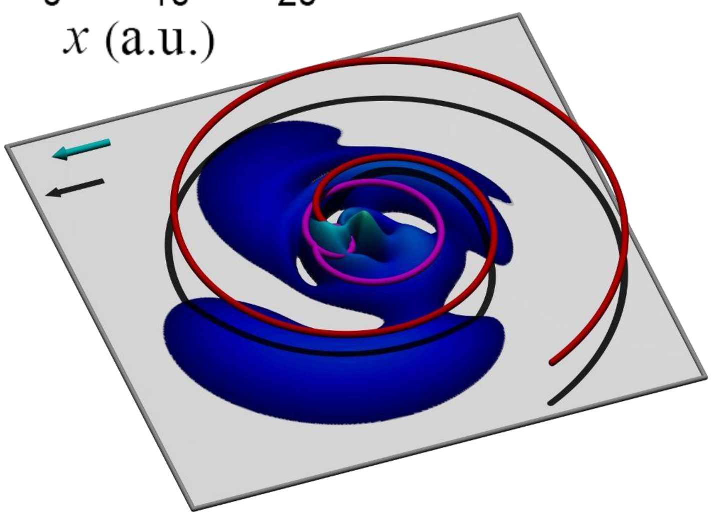
Corkum, Physics Today **64**, 36 (2011)

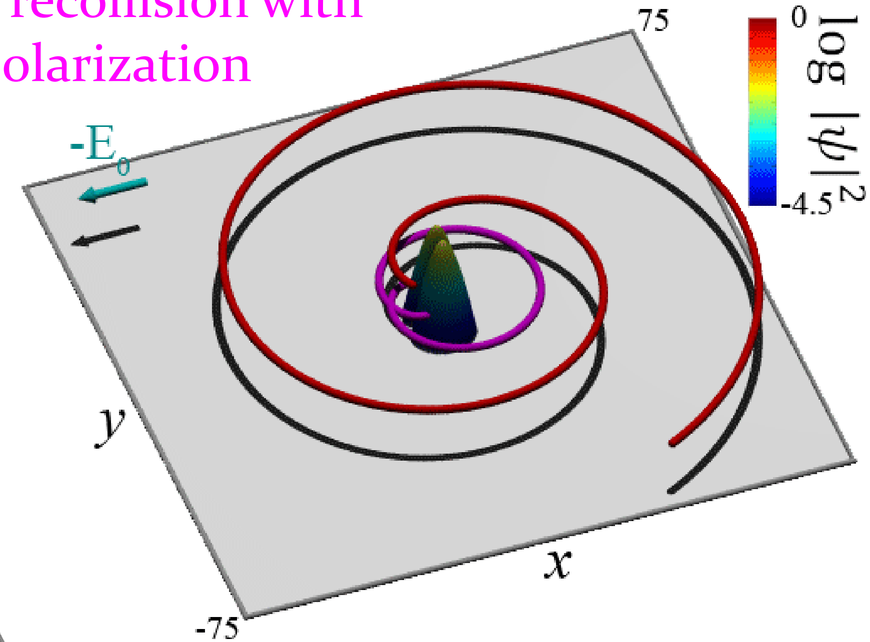
Recollision with circular polarization

- What is missing in the SFA: **the Coulomb potential**



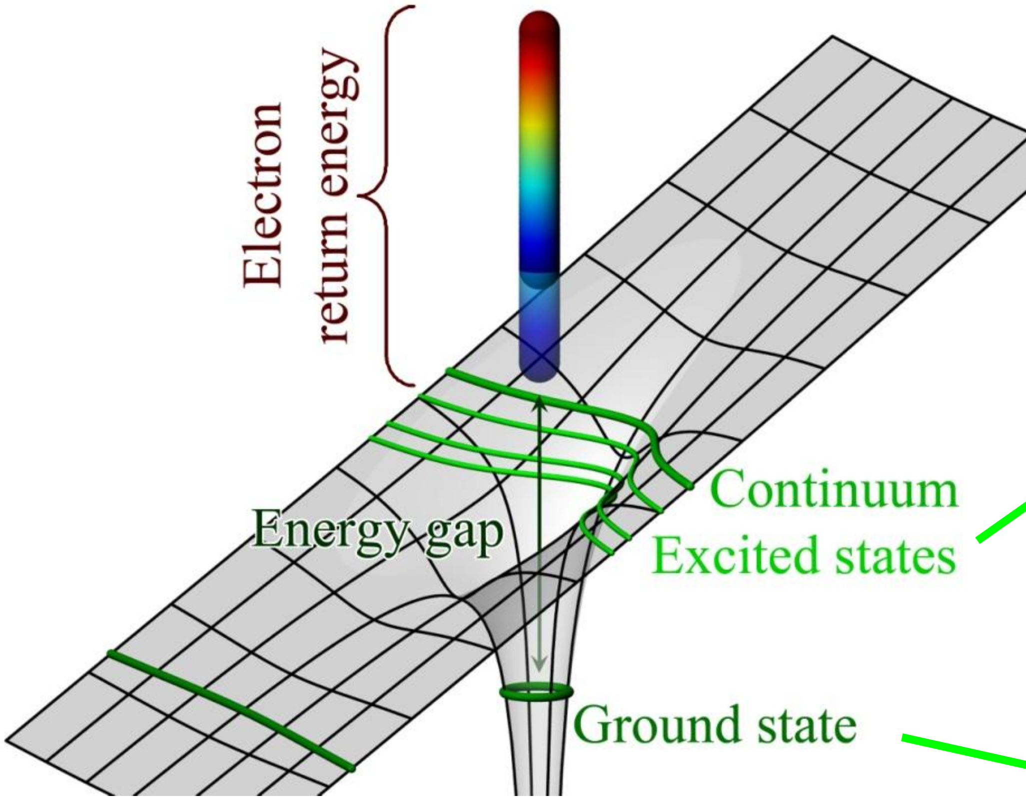
Recolliding Periodic Orbits (RPO)
Organize recollision with
circular polarization





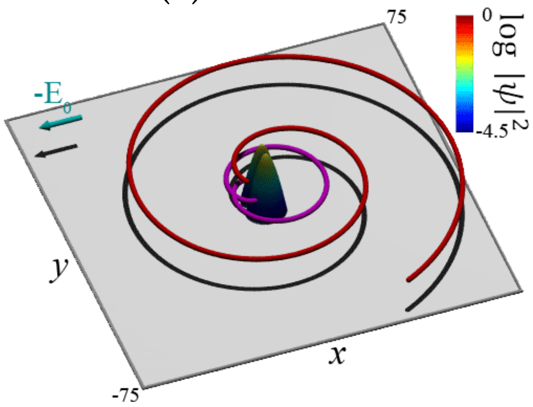
$$\widetilde{\mathcal{H}} = -\frac{\Delta}{2} + V(|\tilde{\mathbf{x}}|) + \underbrace{i\omega(\hat{\tilde{x}}\partial_{\tilde{y}} - \hat{\tilde{y}}\partial_{\tilde{x}})}_{-\omega\hat{L}_z} + E_0\hat{\tilde{x}}$$

Atomic high harmonic generation … almost there!

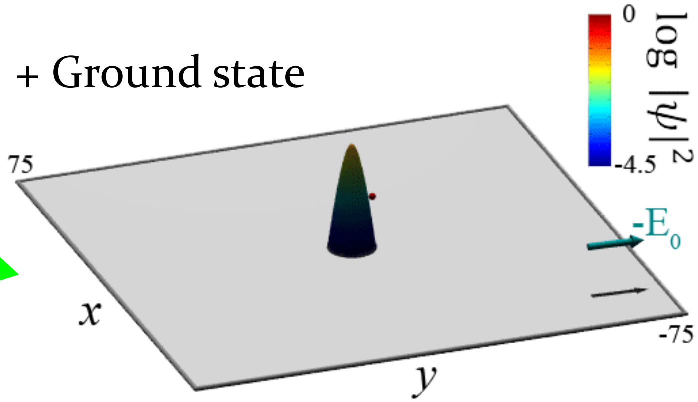


- Recipe for HHG

  Excited state(s)



\+ Ground state

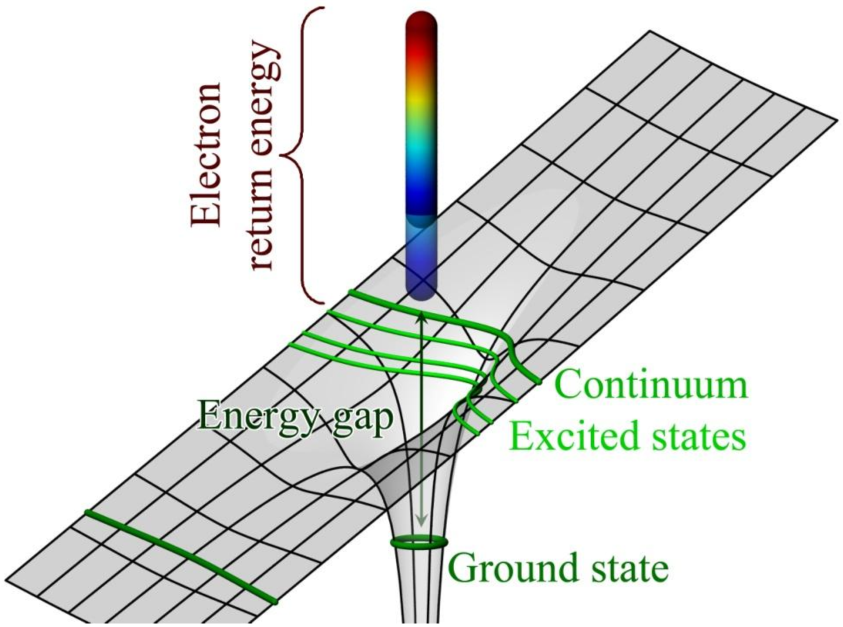


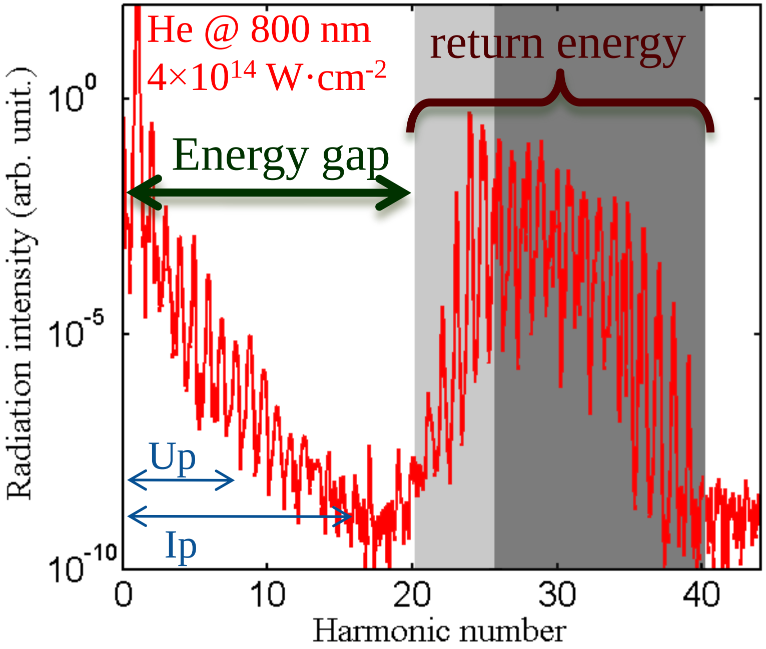
= High Harmonic Generation

- Excited states for recollision
- Ground state to recollide with something

What recolliding periodic orbits tell us

- Specific energy return conditions
- Specific HHG emission



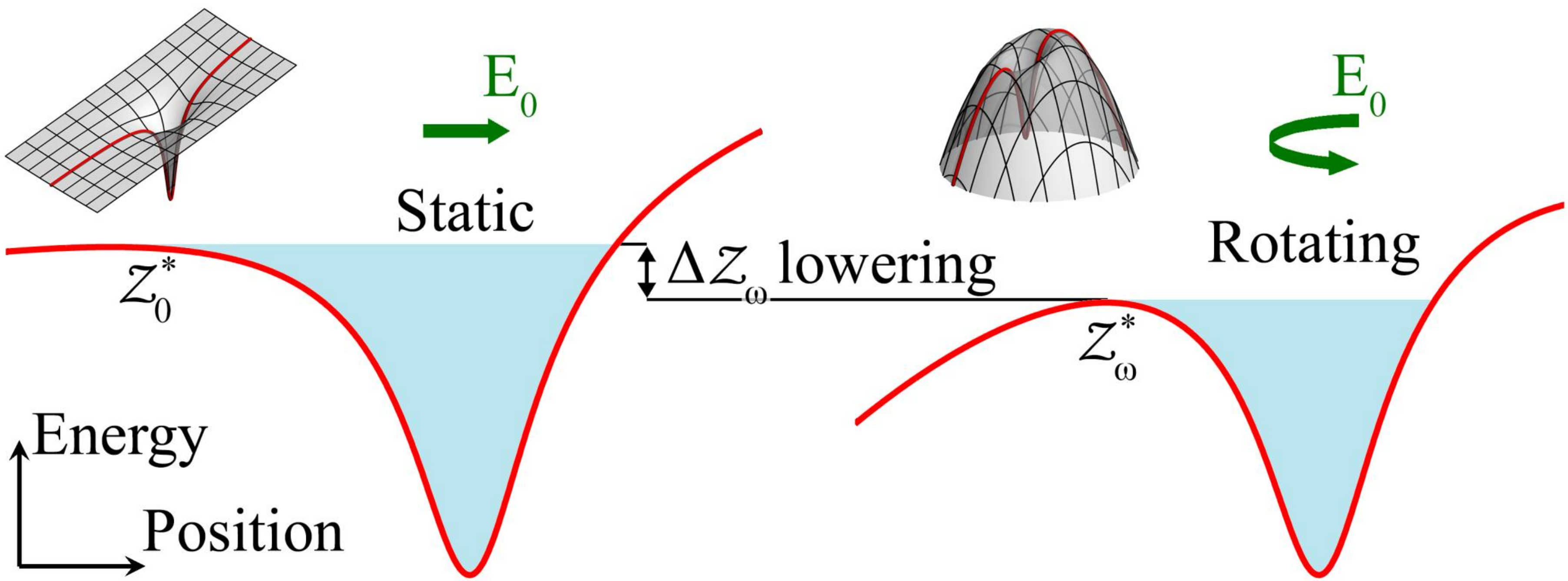


Our references – Video abstract & LabTalk: http://iopscience.iop.org/0953-4075/labtalk-article/56671

- Recollisions and Correlated double ionization with circularly polarized light - PRL **105**, 083002 (2010)
- How Key Periodic Orbits Drive Recollisions in a Circularly Polarized Laser Field - PRL **110**, 253002 (2013)
- Quantum-classical correspondence in circularly polarized high harmonic generation – JPB **47**, 041001 (2014)

Ionization beyond the adiabatic approximation

- In the adiabatic approximation, ionization rates with circular polarization should be independent of the laser frequency



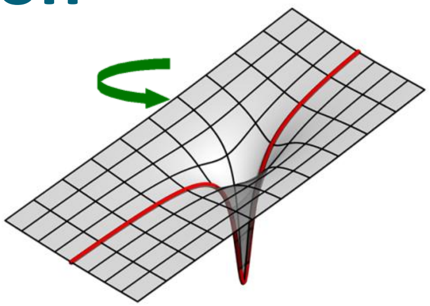
$$\hat{\mathcal{H}} = -\frac{\Delta}{2} + V(|\mathbf{x}|) + E_0\left(\hat{x}\cos\omega t + \hat{y}\sin\omega t\right)$$

$$\hat{\tilde{\mathcal{H}}} = -\frac{\Delta}{2} + V(|\tilde{\mathbf{x}}|) + \underbrace{i\omega\left(\hat{\tilde{x}}\partial_{\tilde{y}} - \hat{\tilde{y}}\partial_{\tilde{x}}\right)}_{-\omega\hat{L}_z} + E_0\hat{\tilde{x}}$$

Nonadiabatic barrier lowering computation

- Static (lab) frame Hamiltonian

$$\mathcal{H}(\boldsymbol{x},\boldsymbol{p},t,\xi) = \frac{|\boldsymbol{p}|^2}{2} + V(|\boldsymbol{x}|) + E_0\,(x\cos\omega t + y\sin\omega t) + \xi$$



- Rotating frame change of variables

$$\tilde{\boldsymbol{x}} = \boldsymbol{\Omega}(t)\boldsymbol{x} \qquad \tilde{t} = t$$

$$\tilde{\boldsymbol{p}} = \boldsymbol{\Omega}(t)\boldsymbol{p} \qquad \tilde{\xi} = \xi + \omega(xp_y - yp_x)$$

$$\boldsymbol{\Omega}(\mathrm{t}) = \begin{pmatrix} \cos\omega t & \sin\omega t \\ -\sin\omega t & \cos\omega t \end{pmatrix}$$

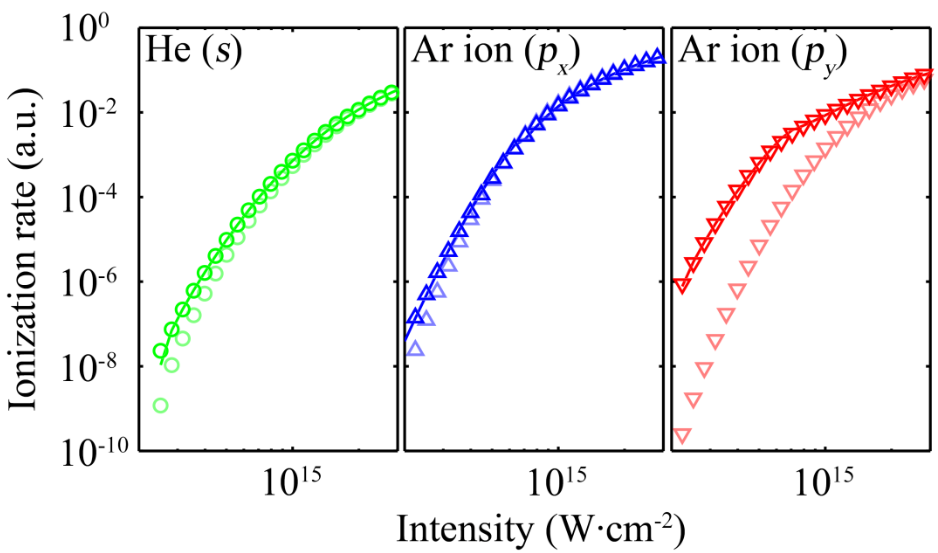
$$\tilde{\mathcal{H}}(\tilde{\boldsymbol{x}},\tilde{\boldsymbol{p}}) = \frac{|\tilde{\boldsymbol{p}}|^2}{2} + V(|\tilde{\boldsymbol{x}}|) - \underbrace{\omega(\tilde{x}\tilde{p}_y - \tilde{y}\tilde{p}_x)}_{\omega\mathcal{L}_z} + E_0\tilde{x}$$

- Zero velocity surface

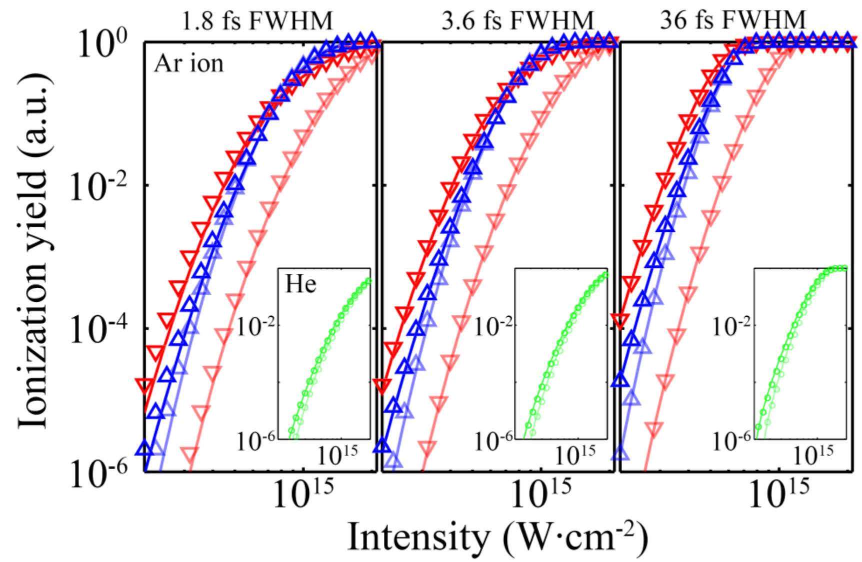
$$\mathcal{H}(\tilde{\boldsymbol{x}},\dot{\tilde{\boldsymbol{x}}} = \boldsymbol{0}) = -\frac{\omega^2}{2}|\tilde{\boldsymbol{x}}|^2 + V(|x|) + E_0\tilde{x}$$

Nonadiabatic ionization yield and rate

- Ionization rate

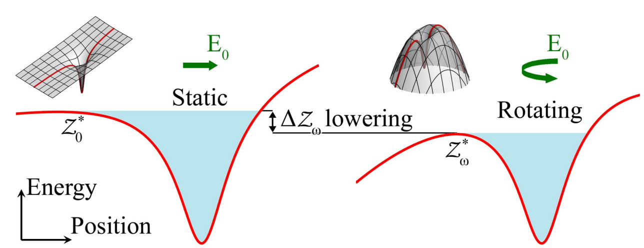


- Ionization yield



- Factorized ionization rate

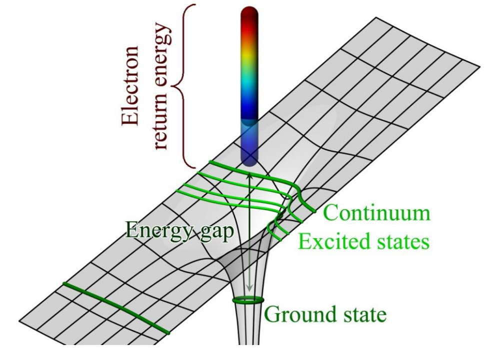
$$\Gamma_{\omega(E_0)} \approx \Gamma_0(E_0)\exp(\beta\,\Delta Z_\omega^\alpha)$$



Reference

- Electronic dynamics and frequency effects in circularly polarized strong-field physics - JPB, to appear arXiv:1406.0105 (2014)

Conclusion



- Nonlinear dynamics
  - We care about periodic orbits in classical mechanics
  - … and for the quantum analysis as well
- Circular polarization
  - Recollision with circular polarization organized by Recolliding Periodic Orbits
  - Recollision with circular polarization leads to High Harmonic Generation
  - Dynamics matters … nonadiabatic effects in ionization rates and yields
- Selected references — www.fmauger.com/
  - Recollisions with circular polarization: PRL 105, 083002 (2010)
  - Recolliding periodic orbits: PRL 110, 253002 (2013)
  - Atomic HHG with circular polarization: JPB 47, 041001 (2014)
  - Nonadiabatic effects in ionization: JPB, to appear arXiv:1406.0105 (2014)
- Thanks

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