Slalom in complex time: semiclassical trajectories in strong-field ionization and their analytical continuations

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In memoriam

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Complex trajectories in quantum orbits

- Strong-field physics is grounded on trajectories
- Tunnelling trajectories require complex times
- First-principles trajectories require complex positions
- Complex positions change everything
- This has physical implications on the photoelectron spectra

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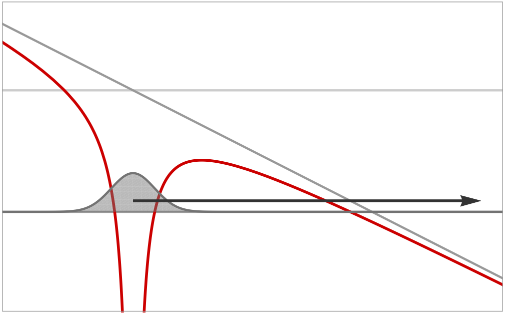
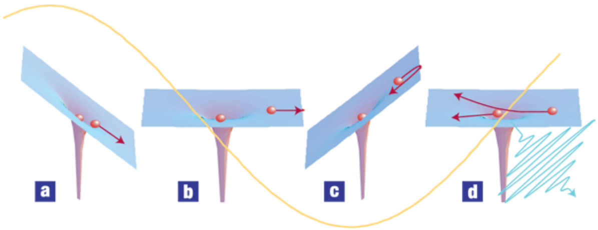
Ionization in the strong-field approximation

We want to study the ionization of atoms or molecules in a strong, long-wavelength field, in the 'optical tunnelling' regime.



Why strong fields? Lots of cool stuff!

- Quantum effects beyond the perturbative regime
- High-order harmonic generation
- High-harmonic spectroscopy
- Laser-driven electron diffraction and holography
- Probing atoms and molecules at their own timescales



Corkum & Krausz, *Nature Phys* 3, 381 (2007)

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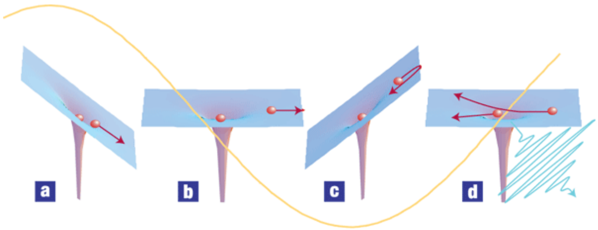

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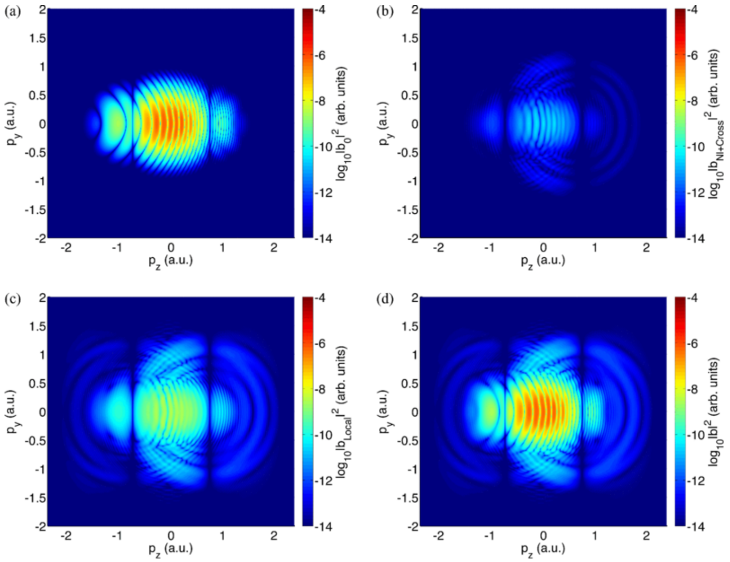

N. Suárez et al., *Phys. Rev. A* **94**, 043423 (2016)

LETTERS
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nature physics

Attoclock reveals natural coordinates of the laser-induced tunnelling current flow in atoms

Adrian N. Pfeiffer[1]*, Claudio Cirelli[1], Mathias Smolarski[1], Darko Dimitrovski[2]*, Mahmoud Abu-samha[2], Lars Bojer Madsen[2] and Ursula Keller[1]

In the research area of strong-laser-field interactions and attosecond science[1], tunnelling of an electron through the barrier formed by the electric field of the laser and the atomic potential is typically assumed to be the initial key process that triggers subsequent dynamics[1–3]. Here we use the attoclock technique[4] to obtain experimental information about the electron tunnelling geometry (the natural coordinates of the tunnelling current flow) and exit point. We confirm vanishing tunnelling delay time, show the importance of the

propagation of the liberated electron, the instant of ionization can be mapped to the angle of the final momentum of the electron in the polarization plane, measured with cold target recoil ion momentum spectroscopy[18] (Fig. 2).

Here, we use the attoclock to measure the offset angle θ (defined in Fig. 3) that is directly related to the complex parent ion interaction and therefore extremely sensitive to the exact tunnel geometry. The attoclock cycle, the time zero (that is, the direction of the maximum laser field vector) and the exact time evolution

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ARTICLES
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Interpreting attoclock measurements of tunnelling times

Lisa Torlina[1†], Felipe Morales[1†], Jivesh Kaushal[1], Igor Ivanov[2], Anatoli Kheifets[2], Alejandro Zielinski[3], Armin Scrinzi[3], Harm Geert Muller[1], Suren Sukiasyan[4], 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.

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PRL **119,** 023201 (2017) PHYSICAL REVIEW LETTERS week ending 14 JULY 2017

Experimental Evidence for Quantum Tunneling Time

Nicolas Camus, Enderalp Yakaboylu,* Lutz Fechner, Michael Klaiber, Martin Laux, Yonghao Mi, Karen Z. Hatsagortsyan,† Thomas Pfeifer, Christoph H. Keitel, and Robert Moshammer‡
Max-Planck-Institut für Kernphysik, Saupfercheckweg 1, 69117 Heidelberg, Germany
(Received 20 January 2017; published 14 July 2017)

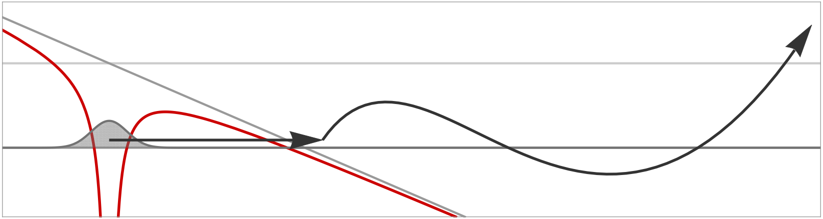
The first hundred attoseconds of the electron dynamics during strong field tunneling ionization are investigated. We quantify theoretically how the electron's classical trajectories in the continuum emerge from the tunneling process and test the results with those achieved in parallel from attoclock measurements. An especially high sensitivity on the tunneling barrier is accomplished here by comparing the momentum distributions of two atomic species of slightly deviating atomic potentials (argon and krypton) being ionized under absolutely identical conditions with near-infrared laser pulses (1300 nm). The agreement between experiment and theory provides clear evidence for a nonzero tunneling time delay and a nonvanishing longitudinal momentum of the electron at the "tunnel exit."

DOI: 10.1103/PhysRevLett.119.023201

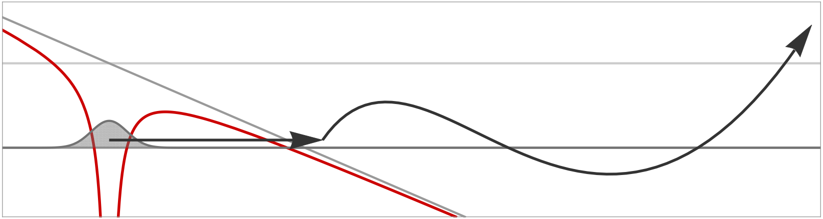
Modern short-pulse lasers generate electric fields that are comparable in strength to those that electrons experience in atoms [1]. Effectively distorting the Coulomb potential of the atomic core, these fields allow ionization of the system

the tunneling step [14,22], whether the tunneling delay time exists [23,24], and if yes, which definition of the tunneling delay time precisely predicts attoclock experiments [15,25].

In our approach the tunneling dynamics is described



- Tunnelling-plus-trajectory models work really well
- Can we provide a solid backing for them from the Schrödinger equation?



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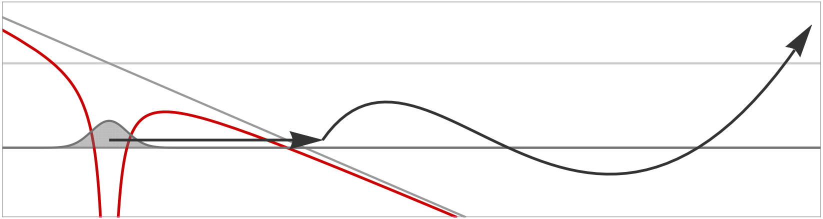

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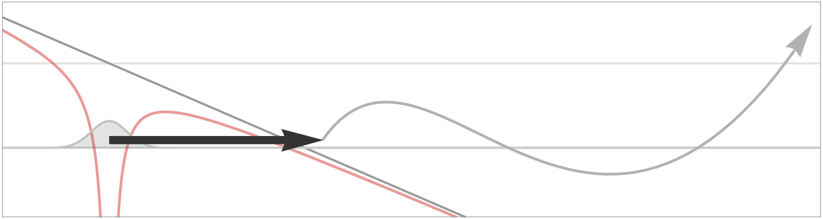
Heuristics for trajectories that inside the tunnel



- $E_{\rm kin} = E_{\rm tot} - V(\mathbf{r}) < 0$ so $v^2 < 0$
- Therefore $v = i\kappa$ is imaginary
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- So... make Δt imaginary?
- Or: how can we distil this into something that makes more sense?

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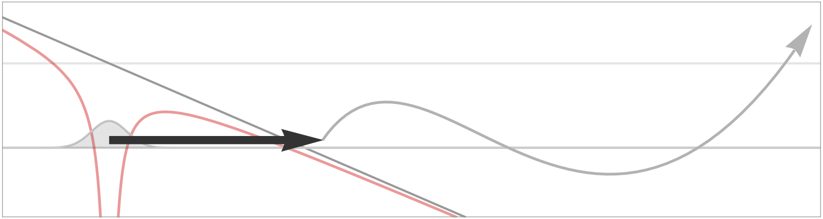

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The Strong-Field Approximation

- The simplest approach is to take a single ground state $|g\rangle$ ionizing into a laser-driven continuum:

$$|\psi(t)\rangle = \underbrace{a(t)\,|g\rangle}_{\text{ground state}} + \underbrace{\int b(\mathbf{p},t)e^{-\frac{i}{2}\int_\infty^t(\mathbf{p}+\mathbf{A}(\tau))^2\mathrm{d}\tau}\,|\mathbf{p}+\mathbf{A}(t)\rangle\mathrm{d}\mathbf{p}}_{\text{continuum}}.$$

- This gives an ionization amplitude in terms of an oscillatory integral.

$$\langle\mathbf{p}|\psi(T)\rangle = \int_{-\infty}^{T} e^{iI_p t-\frac{i}{2}\int_t^\infty(\mathbf{p}+\mathbf{A}(\tau))^2\mathrm{d}\tau}\,\langle\mathbf{p}+\mathbf{A}(t)|V_L|g\rangle\,\mathrm{d}t$$

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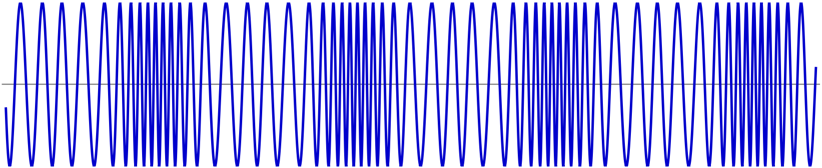
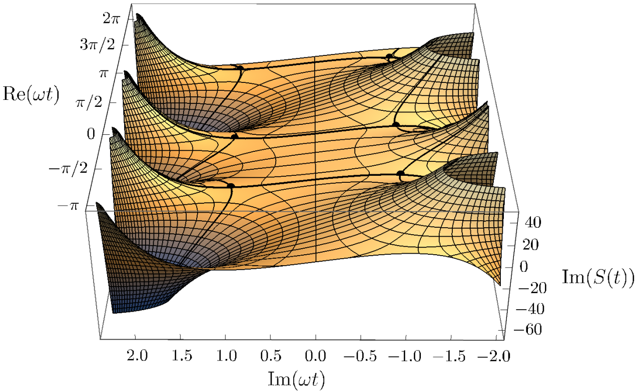


To solve this we shift the integration path into the complex plane



Then we localize the integral to the contributions from the saddle points

Trajectories in the Strong-Field Approximation

- This is called the saddle-point approximation. This gives contributions from a discrete set of saddle points:

$$\langle \mathbf{p}|\psi(T)\rangle = \sum_{t_s} \sqrt{\frac{2\pi}{iS''(t_s)}}\,\langle \mathbf{p}+\mathbf{A}(t_s)|V_L|g\rangle\, e^{iI_p t_s - \frac{i}{2}\int_{t_s}^{\infty}(\mathbf{p}+\mathbf{A}(\tau))^2 \mathrm{d}\tau}.$$

- Each contribution represents a trajectory with kinetic action $S = \frac{1}{2}\int_{t_s}^{\infty}(\mathbf{p}+\mathbf{A}(\tau))^2\mathrm{d}\tau$, ionizing at time t_s.
- The starting time t_s is complex:

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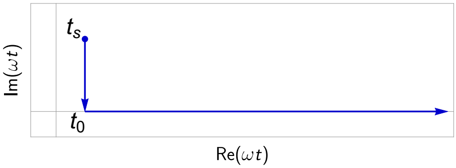

How can we include the Coulomb potential into this description?

- It would be nice to expand this description to include the Coulomb interaction with the nucleus. Something like

$$\langle \mathbf{p}|\psi(T)\rangle \propto e^{iI_p t_s - iS_C(\mathbf{p},t_s)}\ ?$$

- This is known as the Coulomb-Corrected SFA. The action splits in two:

$$\langle \mathbf{p}|\psi(T)\rangle \propto \underbrace{e^{iI_p t_s - \frac{i}{2}\int_{t_s}^{t_0}(\mathbf{p}+\mathbf{A}(\tau))^2 \mathrm{d}\tau}}_{\text{tunnelling}} \underbrace{e^{-iS_C(\mathbf{p},t_0)}}_{\text{action on exact trajectory}}.$$

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Is there a first-principles way to arrive at this description?

- The action $e^{iI_p t_s - \frac{i}{2}\int_{t_s}^{\infty}(\mathbf{p}+\mathbf{A}(\tau))^2 \mathrm{d}\tau}$ comes from the continuum wavefunction. If we want to modify the continuum dynamics, we should do it at this level.
- Semiclassical perturbation theory, in the exponent, gives the eikonal-Volkov wavefunctions:

$$\left\langle \mathbf{r} \middle| \mathbf{p}^{(\mathrm{EV})}(t) \right\rangle \propto \underbrace{e^{i(\mathbf{p}+\mathbf{A}(t))\cdot\mathbf{r}}}_{\text{plane wave}} \underbrace{e^{-\frac{i}{2}\int_{\infty}^{t}(\mathbf{p}+\mathbf{A}(\tau))^2 \mathrm{d}\tau}}_{\text{kinetic action}} \underbrace{e^{-i\int_{\infty}^{t} V(\mathbf{r}_{\mathrm{L}}(\tau;\mathbf{r},\mathbf{p},t))\mathrm{d}\tau}}_{\text{Coulomb correction}}$$

- Here $\mathbf{r}_{\mathrm{L}}(\tau; \mathbf{r}, \mathbf{k}, t) = \mathbf{r} + \int_t^{\tau}(\mathbf{p} + \mathbf{A}(\tau'))\mathrm{d}\tau'$ is the laser-driven trajectory that starts at $\mathbf{r}$ and has asymptotic momentum $\mathbf{p}$.

Smirnova, Spanner & Ivanov, *Phys. Rev. A* **77**, 033407 (2008)

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$$\left\langle \mathbf{r} \middle| \mathbf{p}^{(\mathrm{EV})}(t) \right\rangle \propto \underbrace{e^{i(\mathbf{p}+\mathbf{A}(t))\cdot\mathbf{r}}}_{\text{plane wave}} \underbrace{e^{-\frac{i}{2}\int_{\infty}^{t}(\mathbf{p}+\mathbf{A}(\tau))^2 \mathrm{d}\tau}}_{\text{kinetic action}} \underbrace{e^{-i\int_{\infty}^{t} V(\mathbf{r}_{\mathrm{L}}(\tau;\mathbf{r},\mathbf{p},t))\mathrm{d}\tau}}_{\text{Coulomb correction}}$$

- Here $\mathbf{r}_{\mathrm{L}}(\tau; \mathbf{r}, \mathbf{k}, t) = \mathbf{r} + \int_t^{\tau}(\mathbf{p} + \mathbf{A}(\tau'))\mathrm{d}\tau'$ is the laser-driven trajectory that starts at $\mathbf{r}$ and has asymptotic momentum $\mathbf{p}$.

Smirnova, Spanner & Ivanov, *Phys. Rev. A* **77**, 033407 (2008)

Fencing off the Coulomb singularity

- The eikonal wavefunctions are perturbative in the Coulomb potential so they can't get too close to the singularity at $\mathbf{r} = 0$.
- To handle this we fence off the continuum using an artificial boundary.

- Known as Analytical R-Matrix theory.

Torlina & Smirnova, PRA **86**, 043408 (2012)

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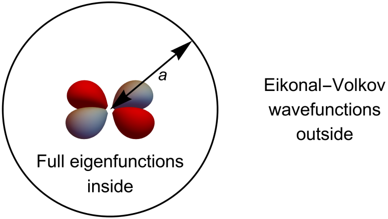

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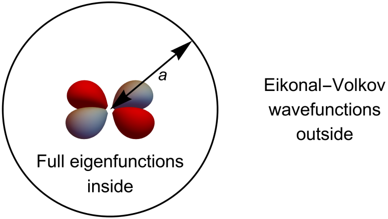

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$$\langle \mathbf{p}|\psi(T)\rangle \propto e^{iI_p t_s + \frac{i}{2}\int_T^{t_s}(\mathbf{p}+\mathbf{A}(\tau))^2 \mathrm{d}\tau} e^{-i\int_{t_\kappa}^{T} U\left(\int_{t_s}^{\tau} \mathbf{p}+\mathbf{A}(\tau')\,\mathrm{d}\tau'\right)\mathrm{d}\tau}$$

SFA component Coulomb correction

- Two major differences:
  - Trajectory is only laser-driven.

$$\mathbf{r}_\mathrm{L}(t) = \int_{t_s}^{t} (\mathbf{p}+\mathbf{A}(\tau))\mathrm{d}\tau$$

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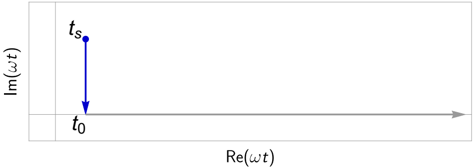
full action →

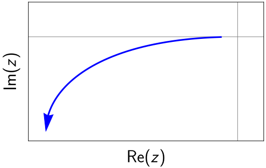
first principles ↓

SFA	CC-SFA
ARM	

What does this mean for the trajectories?

- On the downwards leg to the real time axis, the trajectory becomes complex, through $\int_{t_s}^{t_i}(\mathbf{p}+\mathbf{A}(\tau))\mathrm{d}\tau$.





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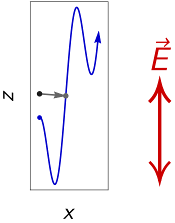

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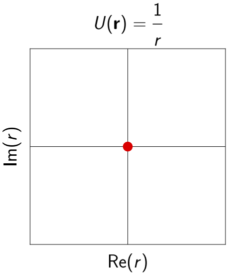

- **Question:** where is this singularity in the complex plane, and do we need to be careful to avoid it?

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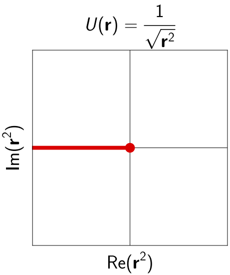

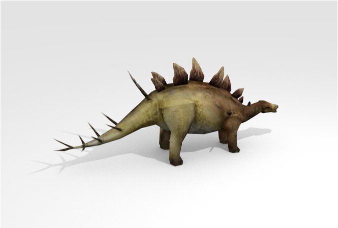

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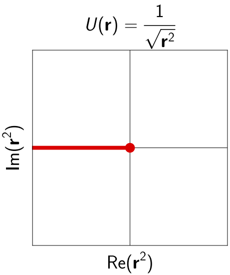

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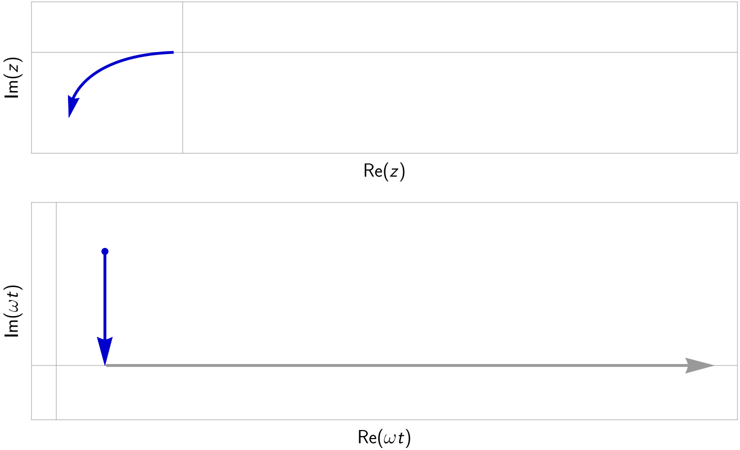
Where is the singularity?

$$z_{\text{L}}(t) = \int_{t_s}^{t} (p_z + A(\tau))\text{d}\tau, \quad \sqrt{\mathbf{r}^2} = \sqrt{x^2 + y^2 + z^2}.$$



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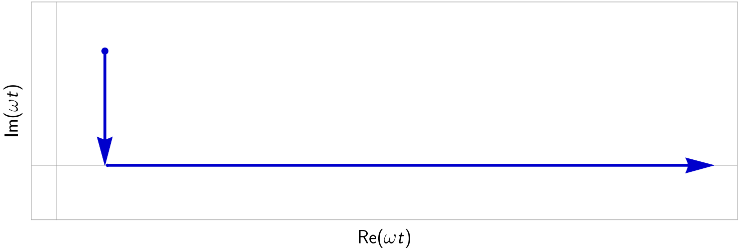

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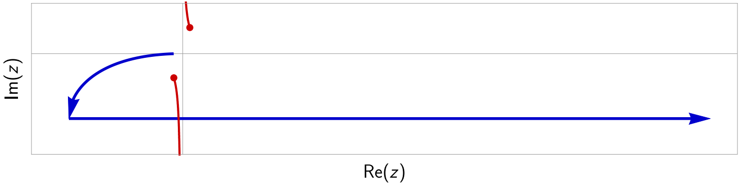

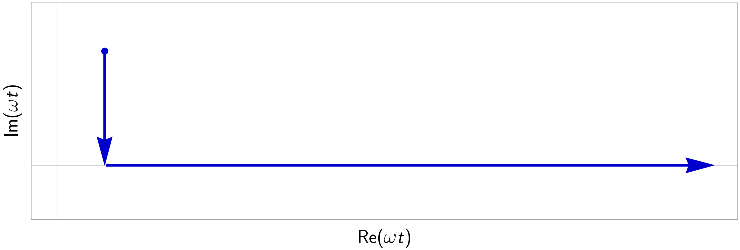

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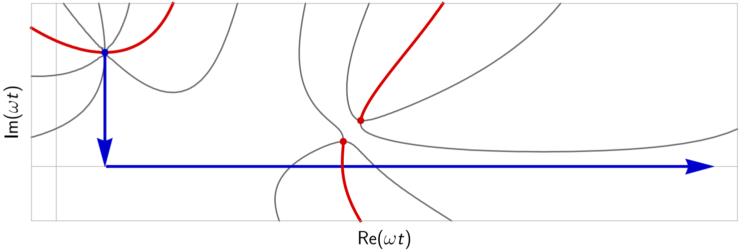

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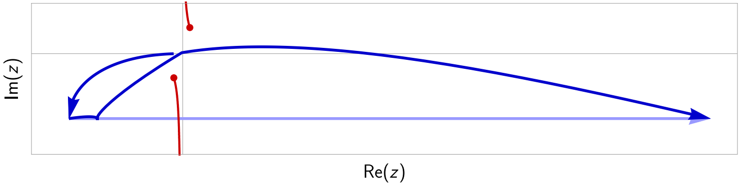

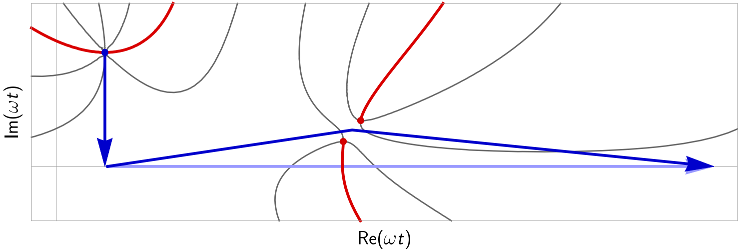

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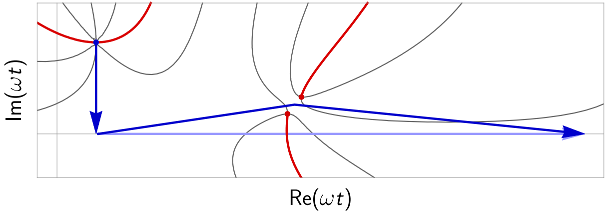

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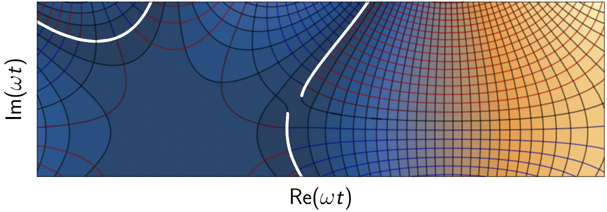
Contours of $\sqrt{\mathbf{r}_{\mathrm{L}}(t)^2}$



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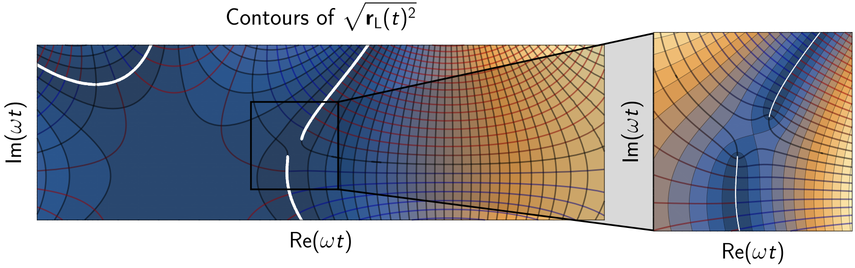

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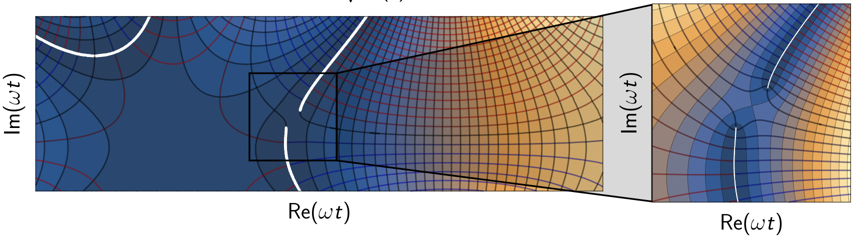

- This is a saddle point of $\sqrt{\mathbf{r}_\mathrm{L}(t)^2} \longleftrightarrow$ also of $\mathbf{r}_\mathrm{L}(t)^2$.

$$\frac{\mathrm{d}}{\mathrm{d}t}\mathbf{r}_\mathrm{L}(t)^2 = 0 \iff \mathbf{r}_\mathrm{L}(t)\cdot\mathbf{v}(t) = 0 \longleftrightarrow$$

- These are the times of closest approach to the ion

(…in complex space.)

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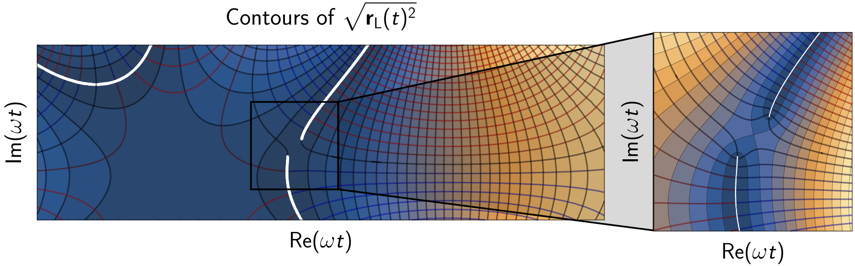

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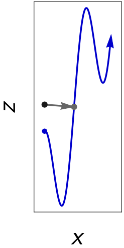

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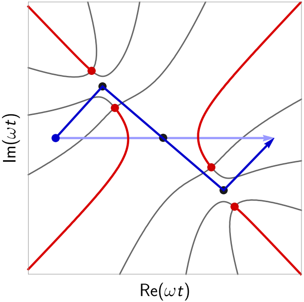

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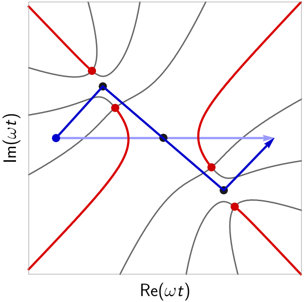
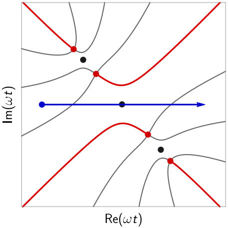
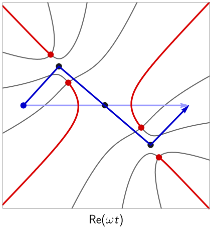
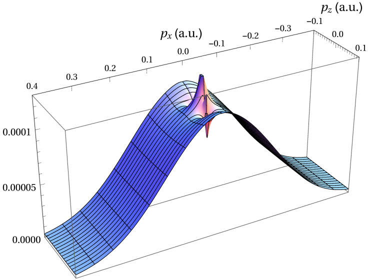


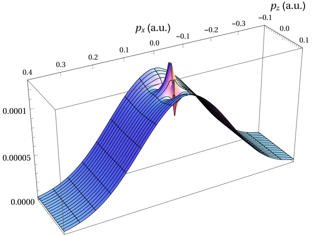
This directly impacts the photoelectron spectrum

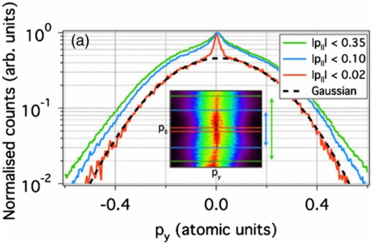


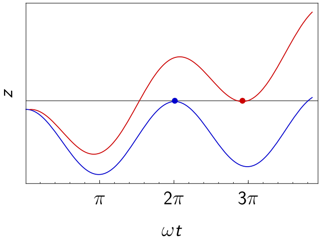




These are low-energy structures



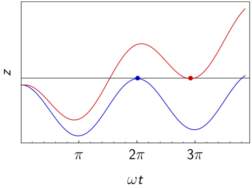




Pullen et al, *J Phys B* **47**, 204010 (2014)

What does this tell us about Near-Zero Energy structures?

- There are two mirror-image families of soft-recollision trajectories



- They should both have similar effects on the photoelectron spectrum
- They scale very different with intensity and wavelength:

$$p_z \sim \frac{2z_{\rm quiv}}{\frac{3}{2}T} \sim \frac{F}{\omega} \qquad \text{vs} \qquad p_z \sim \frac{z_{\rm exit}}{T} \sim \frac{I_p\omega}{F}$$

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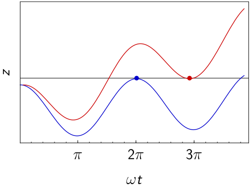

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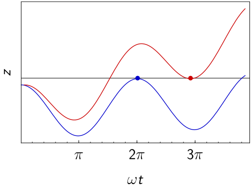

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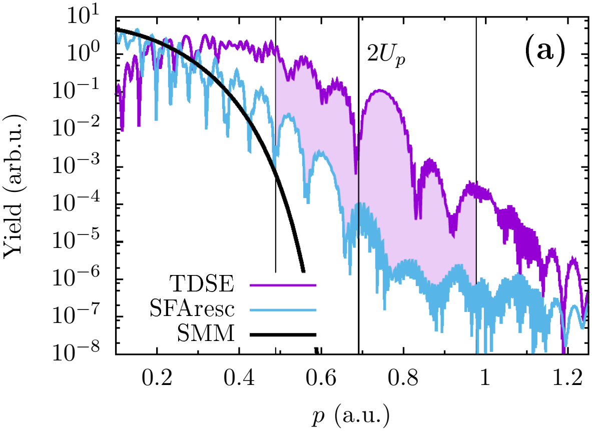
Other uses: enhancement at fast recollisions



Keil, Popruzhenko & Bauer, *Phys. Rev. Lett.* **117**, 243003 (2016)

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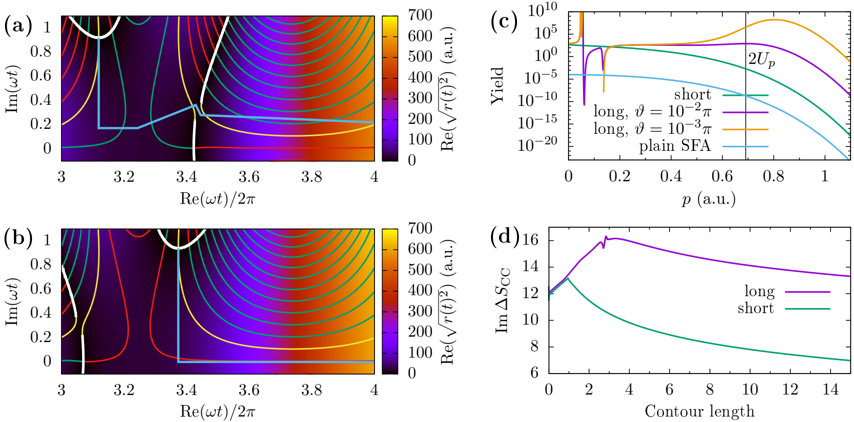

Keil, Popruzhenko & Bauer, *Phys. Rev. Lett.* **117**, 243003 (2016)

What does this tell us about trajectories after tunnelling?

- You can indeed ground the trajectory models in the Schrödinger equation.
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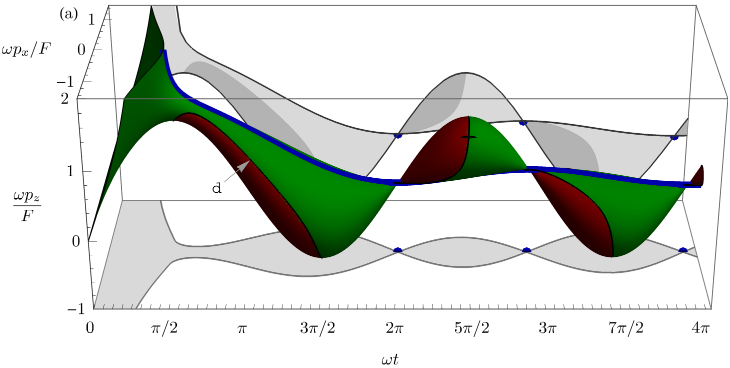
Thank you!

Saddle-point geometry



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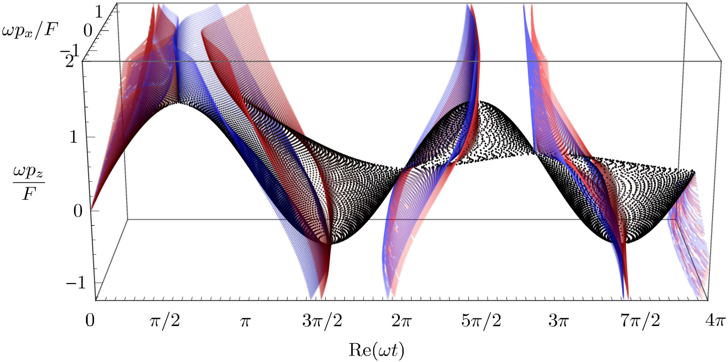

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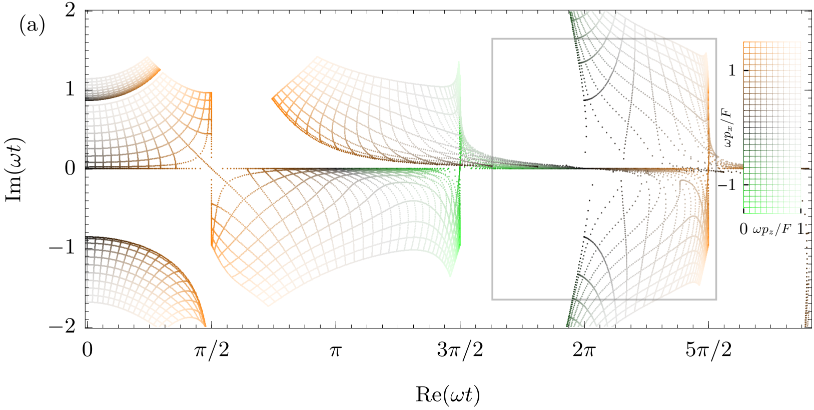

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