Coulomb-corrected quantum orbits in strong-field ionization

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Contributors

Universität Rostock

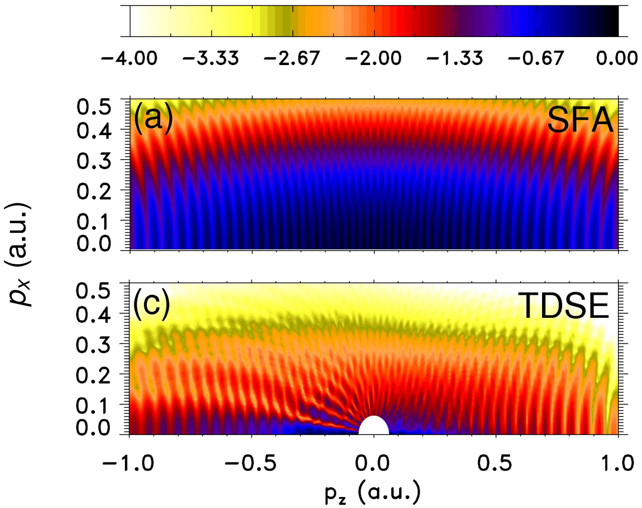
Traditio et Innovatio

SFB 652

International Max Planck Research School
Quantum Dynamics
in Physics, Chemistry and Biology

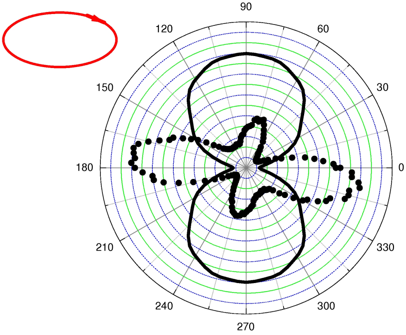
The need for Coulomb-corrected SFA

Ar, $\hat{E} = 0.05338$, $\omega = 0.0228$ (2 μm), 3 cycles, lin. pol.



The need for Coulomb-corrected SFA

Elliptical polarization: Coulomb potential breaks four-fold symmetry of angular distributions predicted by plain SFA



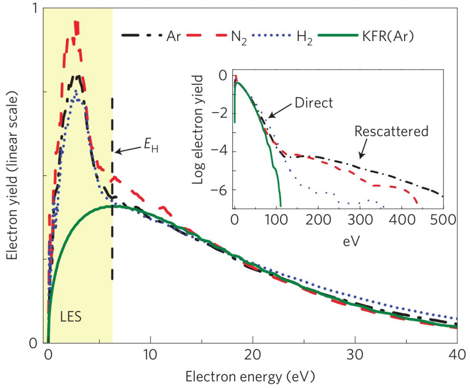
Ar,
2nd max. (energy $\epsilon \simeq 2.8\,\text{eV}$), $\lambda = 800\,\text{nm}$,
$\hbar\omega = 1.55\,\text{eV}$, $10^{14}\,\text{W/cm}^2$,
ellipticity $\xi = 0.36$

solid: SFA
points: Experiment

Data from G.G. Paulus *et al.*, Phys. Rev. Lett. **84**, 3791 (2000)

The need for Coulomb-corrected SFA

Surprise at long wavelengths: the “low energy structure” (LES)



150 TW/cm^2, $\lambda = 2\,\mu$m

C.I. Blaga *et al.*, Nature Physics **5**, 335 (2009)

Strong Field Approximation (SFA)

Differential ionization probability:

$$w(\mathcal{E}_p, \theta, \varphi) = \frac{|M(\mathbf{p})|^2 \, \mathrm{d}^3 p}{\mathrm{d}\Omega \, \mathrm{d}\mathcal{E}_p} = p|M(\mathbf{p})|^2$$

$$M^{\mathrm{SFA}}(\mathbf{p}) \quad =$$

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$$M^{\mathrm{SFA}}(\mathbf{p}) \quad = \quad \overbrace{-\mathrm{i} \int_0^{T_\mathrm{p}} \mathrm{d}t \, \langle \chi(\mathbf{p}, t) | \mathbf{r} \cdot \mathbf{E}(t) | \Psi_0(t) \rangle}^{\text{direct}}$$

with

$$|\chi(\mathbf{p}, t)\rangle = |\mathbf{p} + \mathbf{A}(t)\rangle \exp[-\mathrm{i} S(\mathbf{p}, t)], \quad S(\mathbf{p}, t) = \int \mathrm{d}t' \, \frac{[\mathbf{p} + \mathbf{A}(t')]^2}{2}$$

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with

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Saddle-point approximation

- Saddle-point approximation ($I/\hbar\omega \gg 1$)

$$\begin{aligned} M^{\mathrm{SFA}}(\mathbf{p}) &= -\mathrm{i}\int_0^{T_\mathrm{p}} \mathrm{d}t \, \langle \chi(\mathbf{p},t)|\mathbf{r}\cdot\mathbf{E}(t)|\Psi_0(t)\rangle \\ &\simeq \sum_\alpha C_\alpha(\mathbf{p},t_{s\alpha})\,\mathrm{e}^{-\mathrm{i}S(\mathbf{p},t_{s\alpha})} \end{aligned}$$

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- Saddle-point equation $(\partial S/\partial t)_{t_{s\alpha}} = 0 \Rightarrow$

$$\frac{1}{2}[\mathbf{p}+\mathbf{A}(t_{s\alpha})]^2 = -I \qquad \Longrightarrow \qquad \{t_{s\alpha}(\mathbf{p})\}$$

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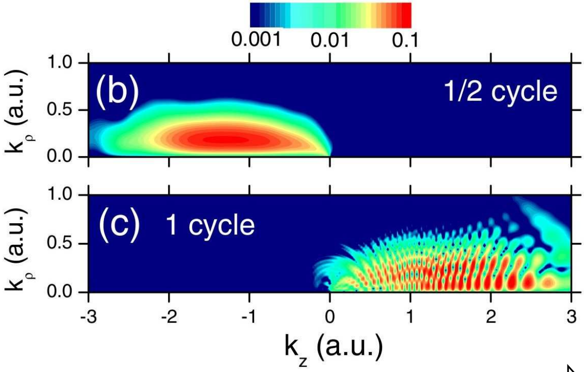
- In the plain SFA the complex saddle-point action

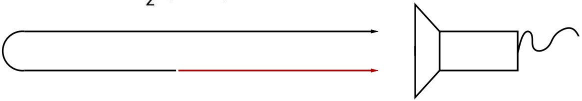
$$S[\mathbf{p},t,t_{s\alpha}(\mathbf{p})] = \frac{1}{2}\int_{t_{s\alpha}(\mathbf{p})}^{t} [\mathbf{p}+\mathbf{A}(t)]^2\,\mathrm{d}t - It_{s\alpha}(\mathbf{p})$$

can be calculated analytically (no need to calculate trajectories explicitly)

Review: D.B. Milošević *et al.*, J. Phys. B **39**, R203 (2006)

Short laser pulses: attosecond time slits



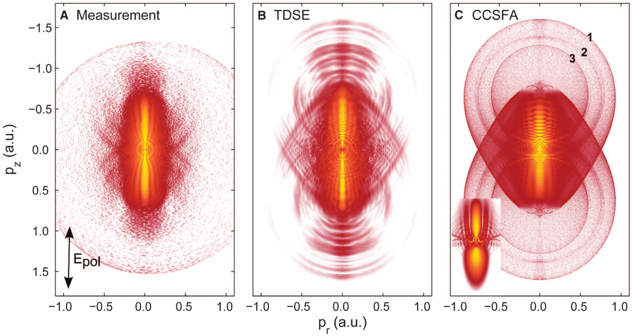


Diego G. Arbó, Emil Persson, Joachim Burgdörfer, Phys. Rev. A **74**, 063407 (2006)

Time-Resolved Holography with Photoelectrons

Y. Huismans,[1]* A. Rouzée,[1,2] A. Gijsbertsen,[1] J. H. Jungmann,[1] A. S. Smolkowska,[1] P. S. W. M. Logman,[1] F. Lépine,[3] C. Cauchy,[3] S. Zamith,[4] T. Marchenko,[5] J. M. Bakker,[6] G. Berden,[6] B. Redlich,[6] A. F. G. van der Meer,[6] H. G. Muller,[1] W. Vermin,[7] K. J. Schafer,[8] M. Spanner,[9] M. Yu. Ivanov,[10] O. Smirnova,[2] D. Bauer,[11] S. V. Popruzhenko,[12] M. J. J. Vrakking[1,2]*

Ionization is the dominant response of atoms and molecules to intense laser fields and is at the basis of several important techniques, such as the generation of attosecond pulses that allow the measurement of electron motion in real time. We present experiments in which metastable xenon atoms were ionized with intense 7-micrometer laser pulses from a free-electron laser. Holographic structures were observed that record underlying electron dynamics on a sublaser-cycle time scale, enabling photoelectron spectroscopy with a time resolution of almost two orders of magnitude higher than the duration of the ionizing pulse.



Science 331, 61 (2011)

Quantum orbits

Each of the saddle-point solutions corresponds to one orbit

Equations of motion

- $\dot{\mathbf{r}} = \nabla_{\mathbf{p}} H \qquad \Rightarrow \qquad \mathbf{v} = \dot{\mathbf{r}} = \mathbf{p} + \mathbf{A}(t)$
- $\dot{\mathbf{p}} = -\nabla_{\mathbf{r}} H \qquad \Rightarrow \qquad \dot{\mathbf{p}} = 0 \qquad \Longrightarrow \mathbf{p} = \text{const.}$

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Initial conditions (**p** is given and real)

- From

$$A(t_s) = -p_{\|} \pm \mathrm{i}\sqrt{2I + p_{\perp}^2}$$

follows $\mathrm{Re}\, A(t_s) = -p_{\|}$ and thus $\mathrm{Re}\, v_{\|}(t_s) = 0$, $\mathbf{v}_{\perp} = \mathbf{p}_{\perp}$

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- Position

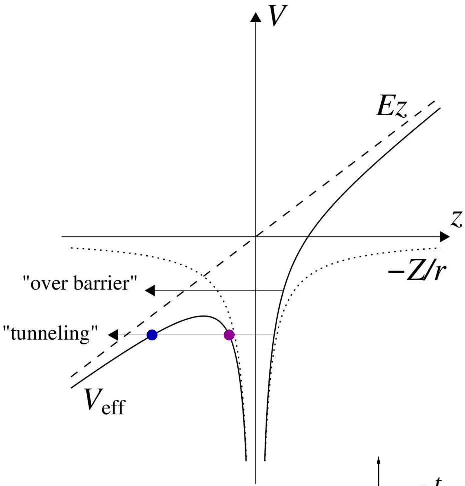
$$\mathbf{r}(\operatorname{Re} t_s) = \int_{t_s}^{\operatorname{Re} t_s} \mathrm{d}t \, [\mathbf{p} + \mathbf{A}(t)] = -\mathrm{i}\mathbf{p} \operatorname{Im} t_s + \alpha(\operatorname{Re} t_s) - \alpha(t_s) + \mathbf{r}_0$$

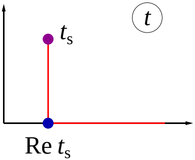
where $\alpha(t) = \int^t \mathbf{A}(t')\,\mathrm{d}t'$, and for $\operatorname{Re} \mathbf{r}(t_s) = 0 \Rightarrow \mathbf{r}_0 = \mathrm{i} \operatorname{Im} \mathbf{r}_0$

$$\Rightarrow \qquad \operatorname{Re}[\mathbf{r}(\operatorname{Re} t_s)] = \alpha(\operatorname{Re} t_s) - \operatorname{Re}[\alpha(t_s)]$$

("tunnel exit")

Tunnel exit





Coulomb-corrected quantum trajectories

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- Shoot initial $\mathbf{p}_0 = \mathbf{p}(\mathrm{Re}\, t_s)$

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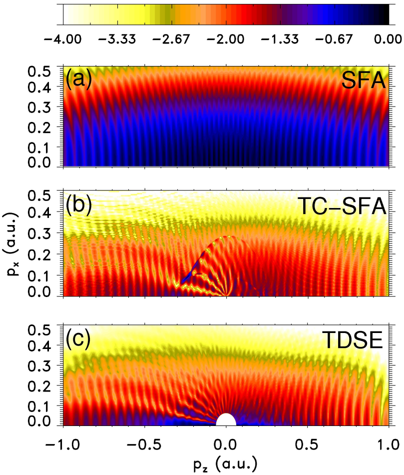
- Store result in data base
- Add all trajectories that lead to the same $\mathbf{p}(\infty)$ coherently
- The corrected saddle-point action is

$$\underbrace{S(\mathbf{p}(t), \mathrm{Re}\, t'_{s\alpha}, t'_{s\alpha}) - \int_{t'_{s\alpha}}^{\mathrm{Re}\, t'_{s\alpha}} \frac{Z\,\mathrm{d}t}{\sqrt{\mathbf{r}^2(t)}}}_{\text{"under barrier-part" affects what?}} + \underbrace{S[\mathbf{p}(t), \infty, \mathrm{Re}\, t'_{s\alpha}] - \int_{\mathrm{Re}\, t'_{s\alpha}}^{\infty} \frac{Z\,\mathrm{d}t}{|\mathbf{r}(t)|}}_{\text{purely real part affects interference pattern}}$$

Trajectory shooting

Ar, $\hat{E} = 0.05338$, $\omega = 0.0228$ (2 μm), 3 cycles

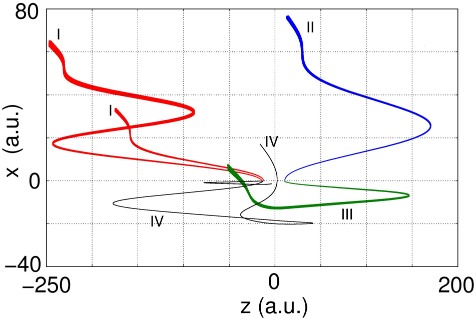


Huge improvement!

T.-M. Yan, S.V. Popruzhenko, M.J.J. Vrakking, D. Bauer, PRL 105, 253002 (2010)

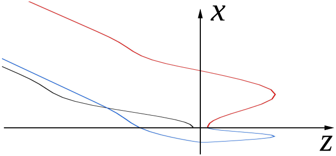
Classes of trajectories

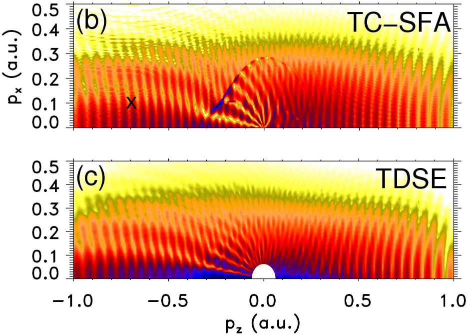
- class I: tunnelexit$\cdot p_z > 0$, $p_{0x} p_x > 0$
- class II: tunnelexit$\cdot p_z < 0$ and $p_{0x} p_x > 0$
- **class III:** tunnelexit$\cdot p_z < 0$ and $p_{0x} p_x < 0$
- class IV: tunnelexit$\cdot p_z > 0$ and $p_{0x} p_x < 0$



Classes of trajectories

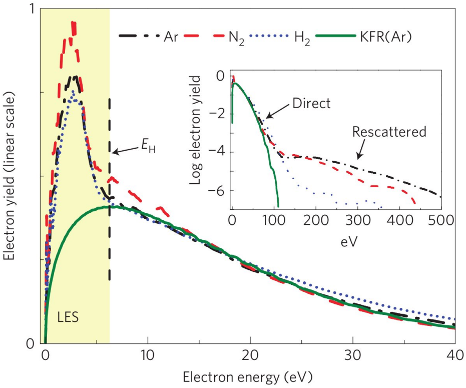
Analysis of spectral features in terms of trajectories possible → maximum insight





E.g., side lobes due to interference of class-II and class-III trajectories

The origin of the LES



150 TW/cm^2, $\lambda = 2\,\mu$m

C.I. Blaga *et al.*, Nature Physics **5**, 335 (2009)

see also W. Quan *et al.*, Phys. Rev. Lett. 103, 093001 (2009)

The origin of the LES

- Chengpu Liu and Karen Z. Hatsagortsyan,
  Phys. Rev. Lett. 105, 113003 (2010)
- Tian-Min Yan, S. V. Popruzhenko, M. J. J. Vrakking, and D. Bauer,
  Phys. Rev. Lett. 105, 253002 (2010)
- Zhongyuan Zhou and Shih-I Chu,
  Phys. Rev. A 83, 033406 (2011)
- Dmitry A. Telnov and Shih-I Chu,
  Phys. Rev. A 83, 063406 (2011)
- A. Kästner, U. Saalmann, J.M. Rost,
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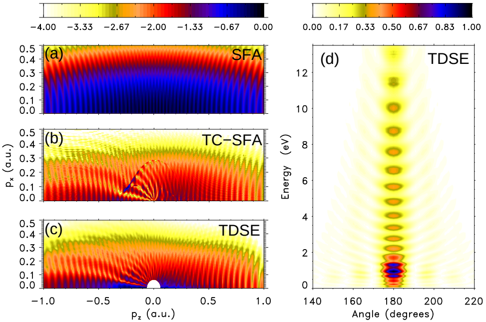
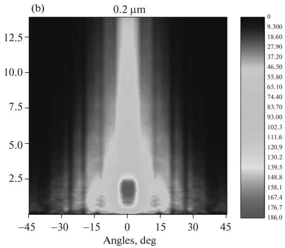
- Christoph Lemell, Konstantinos I. Dimitriou, Xiao-Min Tong, Stefan Nagele, Daniil V. Kartashov, Joachim Burgdörfer, and Stefanie Gräfe,
  Phys. Rev. A 85, 011403 (2012)
- Chengpu Liu and Karen Z. Hatsagortsyan,
  Phys. Rev. A 85, 023413 (2012)
- C. Y. Wu, Y. D. Yang, Y. Q. Liu, Q. H. Gong, M. Wu, X. Liu, X. L. Hao, W. D. Li, X. T. He, and J. Chen,
  Phys. Rev. Lett. 109, 043001 (2012)

Caustic in the TC-SFA

Ar, $\hat{E} = 0.05338$, $\omega = 0.0228$ (2 μm), 3 cycles





F. Catoire *et al.*, Laser Phys. **19**, 1574 (2009)

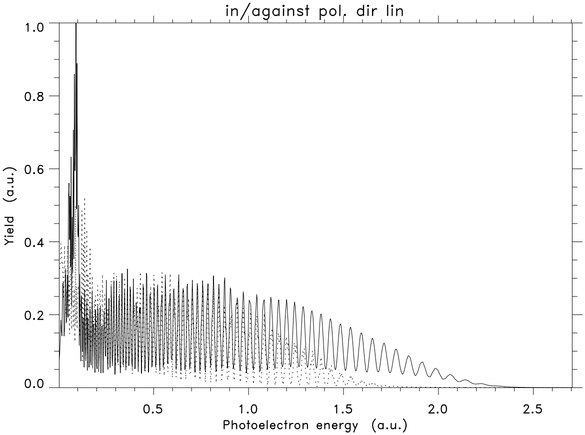
LES visible in the TDSE result

Appears as caustic structure in the TC-SFA

T.-M. Yan, S.V. Popruzhenko, M.J.J. Vrakking, D. Bauer, PRL 105, 253002 (2010)

LES at $\lambda = 3.2\,\mu$m, $\hat{E} = 0.0534$, 4-cycle pulse, Ar

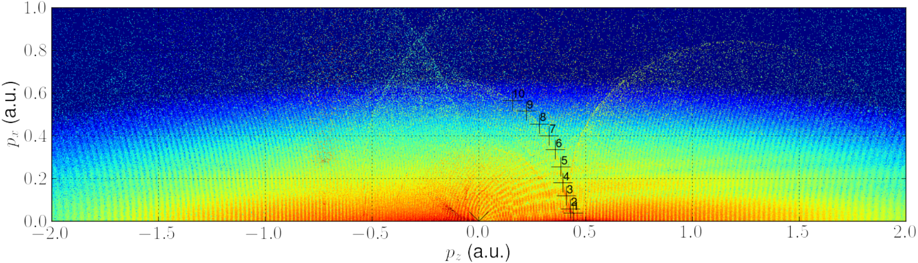
TDSE result



LES at $\mathcal{E} \simeq 0.1$, i.e., $p \simeq 0.45\,$a.u., mostly in forward direction

LES at $\lambda = 3.2\,\mu$m, $\hat{E} = 0.0534$, 4-cycle pulse, Ar

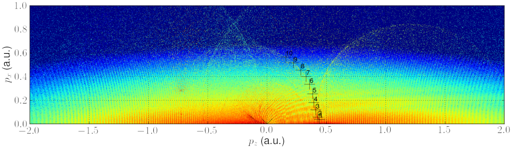
TC-SFA result

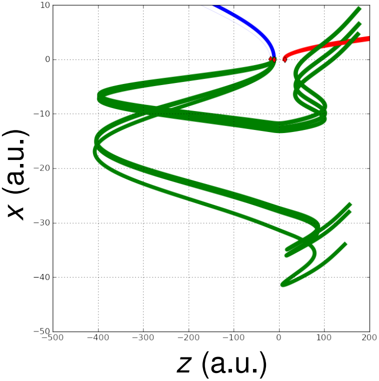


Indeed, LES at $p \simeq 0.45\,$a.u., mostly in forward direction

LES at $\lambda = 3.2\,\mu$m, $\hat{E} = 0.0534$, 4-cycle pulse, Ar

TC-SFA result — contributing quantum orbits





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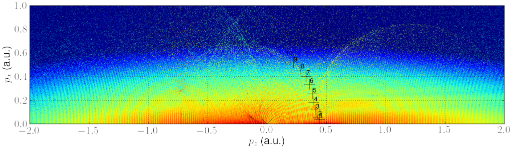
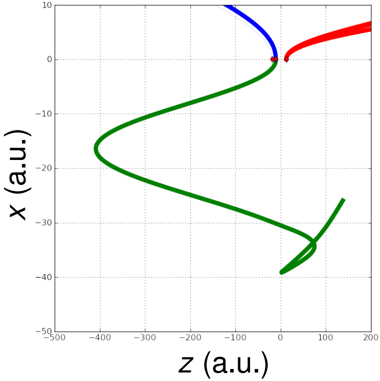
TC-SFA result — contributing quantum orbits





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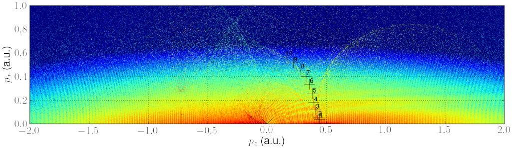

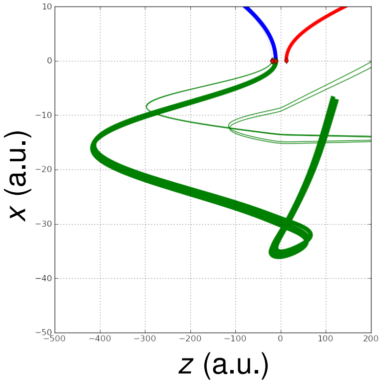

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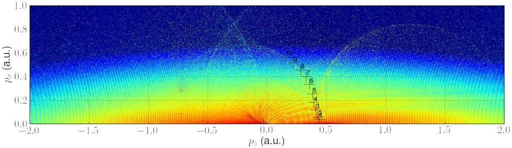
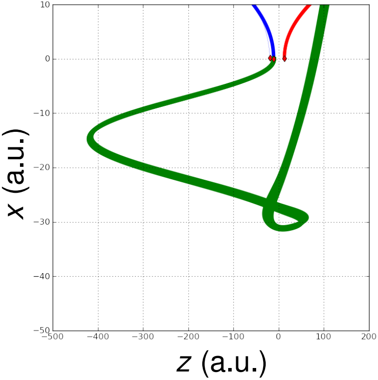
TC-SFA result — contributing quantum orbits





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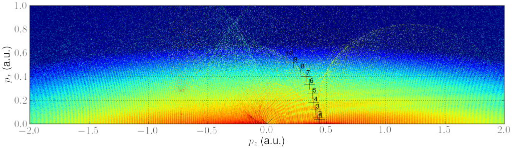

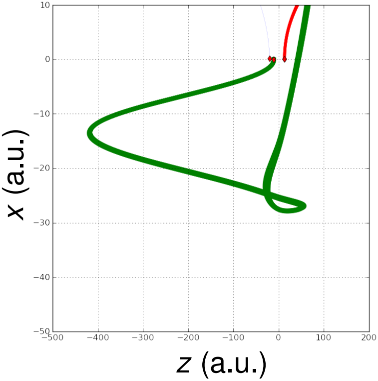

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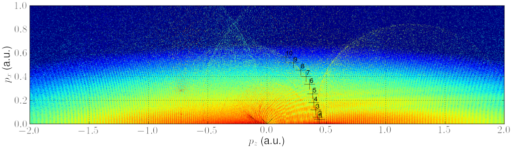

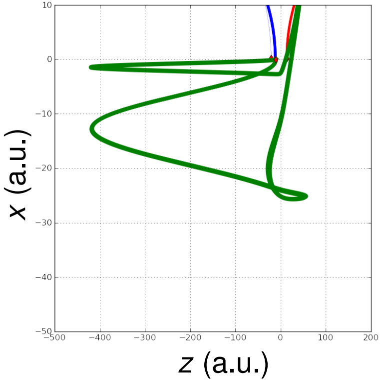

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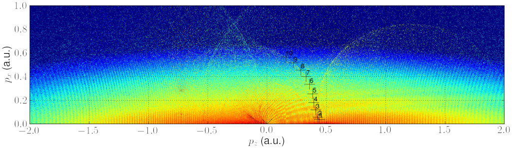

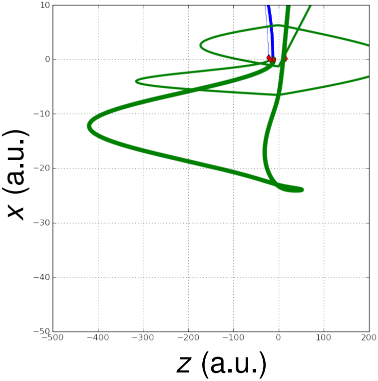

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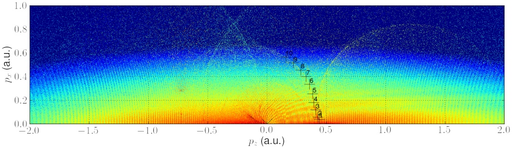

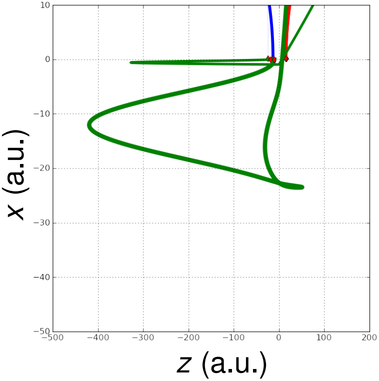

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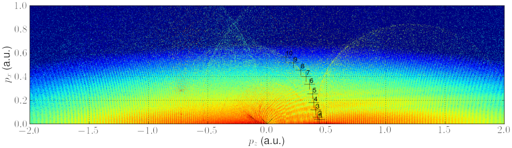

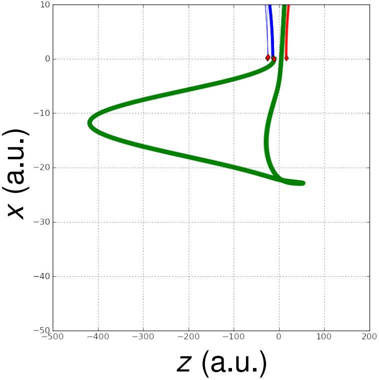

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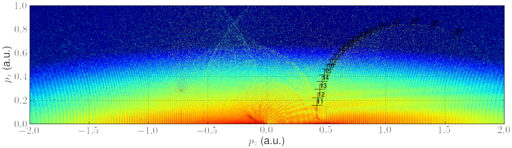

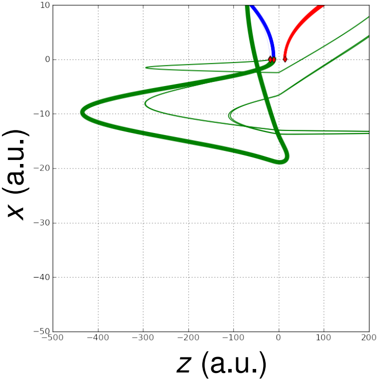

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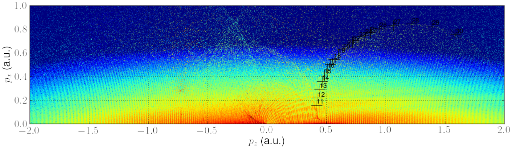

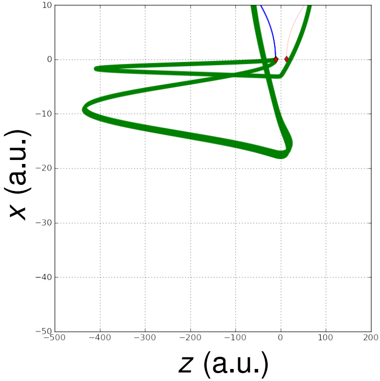

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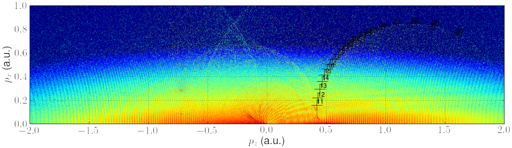

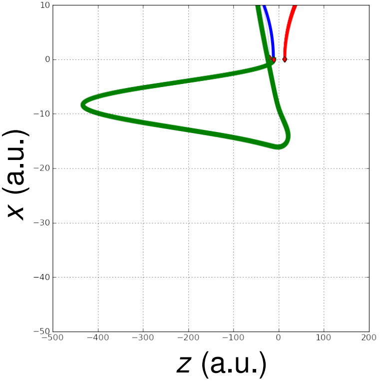

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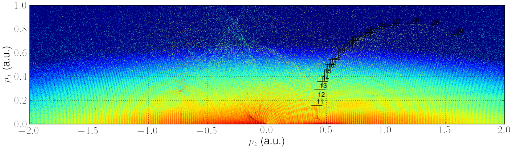

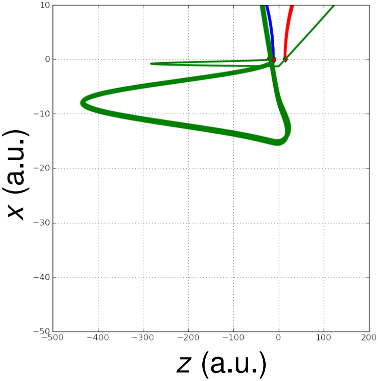

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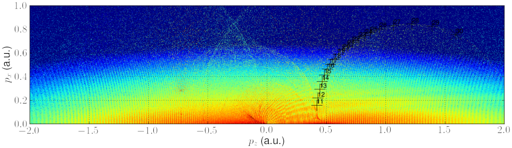

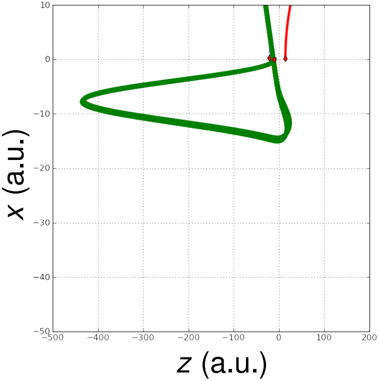

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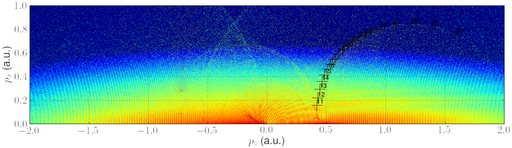

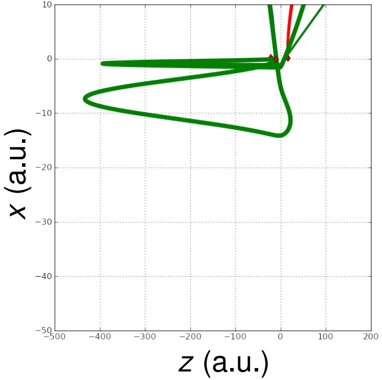

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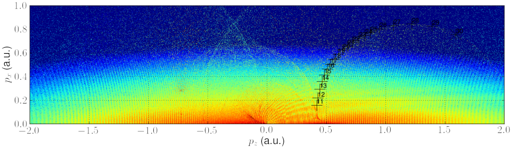

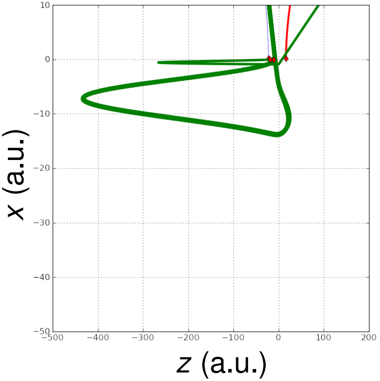

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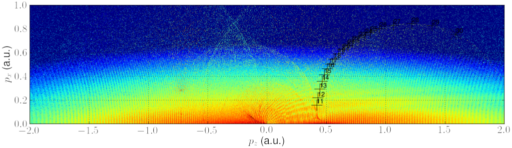

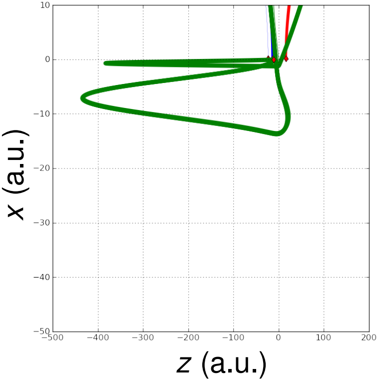

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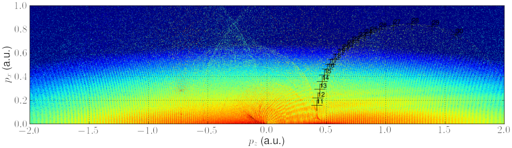

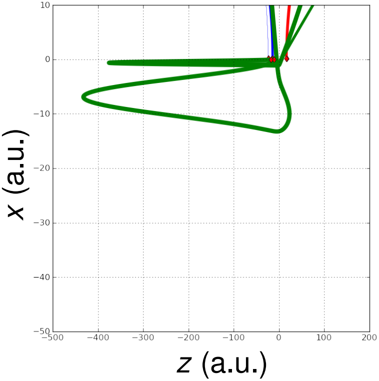

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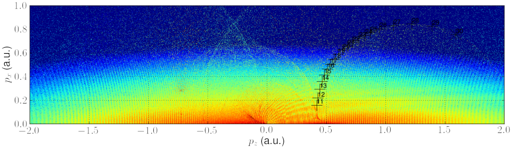

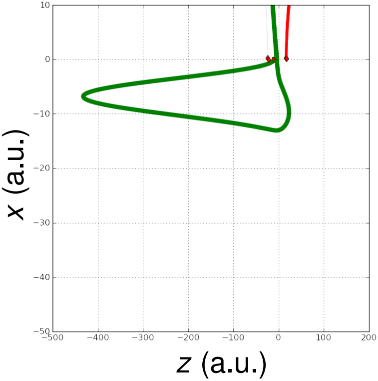

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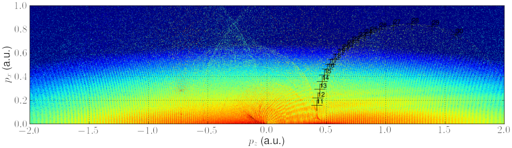
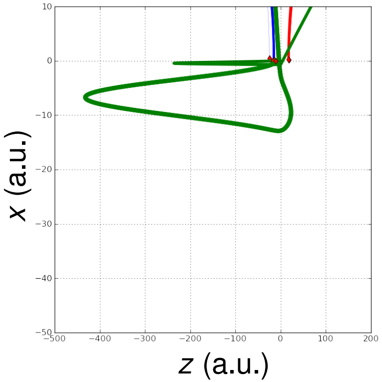
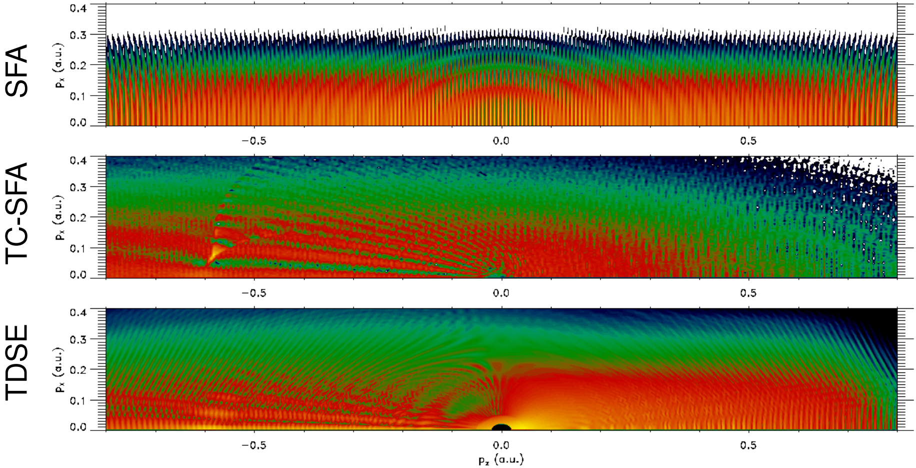




LES at even longer wavelengths

$\lambda = 4\,\mu$m, $\hat{E} = 0.0534$, $N = 3$, H(1s)



Sub-barrier Coulomb-correction

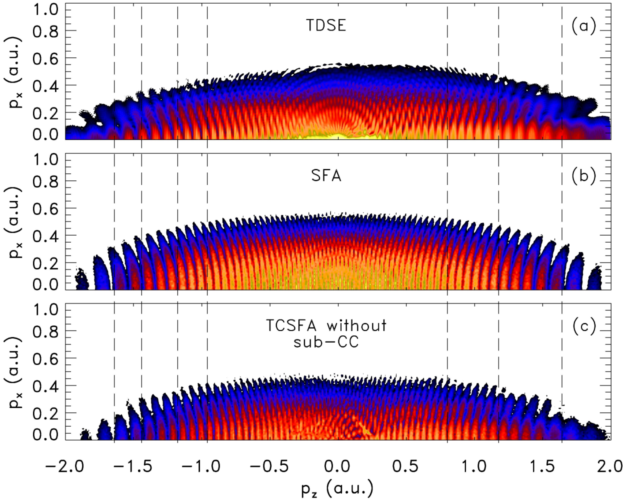
- Remember:

$$\underbrace{S(\mathbf{p}(t), \mathrm{Re}\, t'_{s\alpha}, t'_{s\alpha}) - \int_{t'_{s\alpha}}^{\mathrm{Re}\, t'_{s\alpha}} \frac{Z\, \mathrm{d}t}{\sqrt{\mathbf{r}^2(t)}}}_{\text{"under barrier-part" affects what?}} + \underbrace{S[\mathbf{p}(t), \infty, \mathrm{Re}\, t'_{s\alpha}] - \int_{\mathrm{Re}\, t'_{s\alpha}}^{\infty} \frac{Z\, \mathrm{d}t}{|\mathbf{r}(t)|}}_{\text{purely real part affects interference pattern}}$$

- So far, the Coulomb potential in the sub-barrier part has been neglected
- Difference between tunneling through triangular barrier and Coulomb barrier → **tunneling ionization formulas** (via **imaginary part of** S for most probable orbit)
- Does the **real part of** S under the barrier has an effect?

The lack of π

$\lambda = 2\,\mu$m, 10^{14} Wcm^{-2}, 4-cycle sin^2, H(1s)



Opposite intra-cycle fringe pattern!

Sub-barrier Coulomb-correction

- Remember:

$$\underbrace{S(\mathbf{p}(t), \mathrm{Re}\, t'_{s\alpha}, t'_{s\alpha}) - \int_{t'_{s\alpha}}^{\mathrm{Re}\, t'_{s\alpha}} \frac{Z\,\mathrm{d}t}{\sqrt{\mathbf{r}^2(t)}}}_{\text{"under barrier-part" affects what?}} + \underbrace{S[\mathbf{p}(t), \infty, \mathrm{Re}\, t'_{s\alpha}] - \int_{\mathrm{Re}\, t'_{s\alpha}}^{\infty} \frac{Z\,\mathrm{d}t}{|\mathbf{r}(t)|}}_{\text{purely real part affects interference pattern}}$$

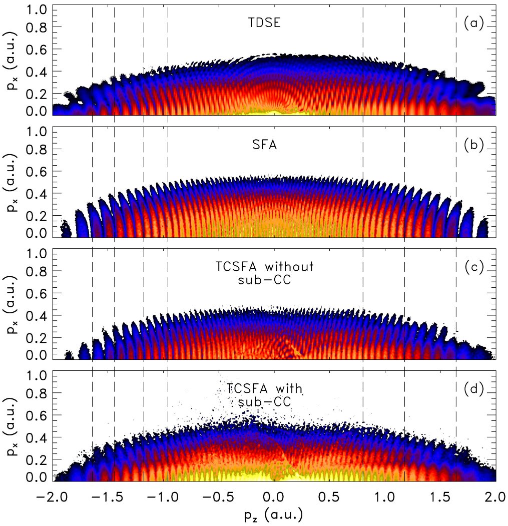
- Take $\mathbf{r}(t)$ from plain SFA to evaluate it
- One can show **analytically** that for long wavelengths the phase difference between long and short trajectory in polarization direction introduced by this Coulomb integral is

$$\Delta\phi = \frac{Z\pi}{\sqrt{2I}}.$$

T.-M. Yan, D. Bauer, (submitted) [arXiv:1209.0704]

Sub-CC: Providing the missing π

$\lambda = 2\,\mu$m, 10^{14} Wcm^{-2}, 4-cycle $\sin^2$, H(1s)



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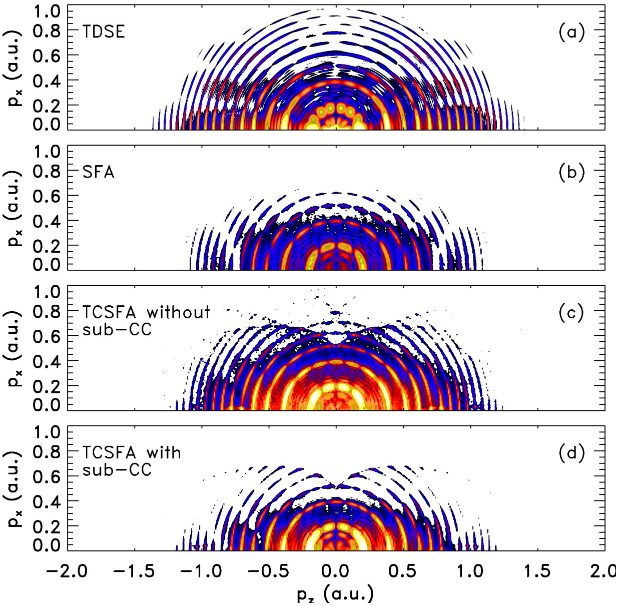
Sub-CC effect on ATI-rings

H(1s), $\lambda = 800\,\mathrm{nm}$, (2,5,2)-trapezoidal pulse, $10^{14}\ \mathrm{Wcm}^{-2}$



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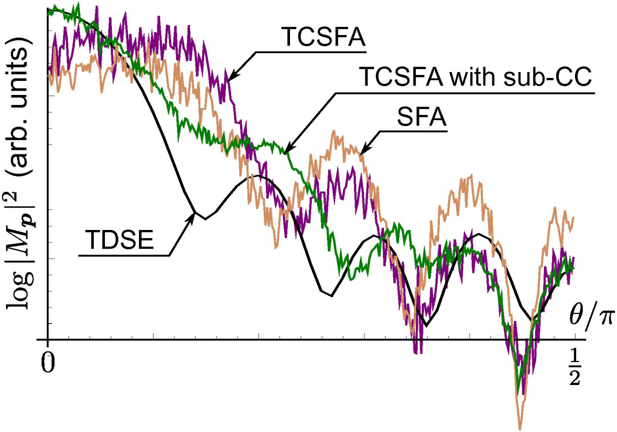

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Summary

- Mathematical "trick" (saddle-point approximation) $\rightarrow$ quantum trajectories
- If Coulomb-corrected and $I/\hbar\omega \gg 1$ $\rightarrow$ very much improved agreement with exact TDSE results
- Sub-barrier Coulomb correction $\rightarrow$ modified intra-cycle interference fringes and low-order ATI ring structures
- All spectral features can be analyzed in terms of interfering quantum orbits $\rightarrow$ maximum insight into quantum dynamics
- Trajectory-based Coulomb-corrected SFA works best where solving the TDSE becomes prohibitive