The strong-field approximation for atoms and ions and for long and for short pulses

D. B. Milosevic
University of Sarajevo, Bosnia and Herzegovina

W.Becker
Max-Born Institut, Berlin, Germany

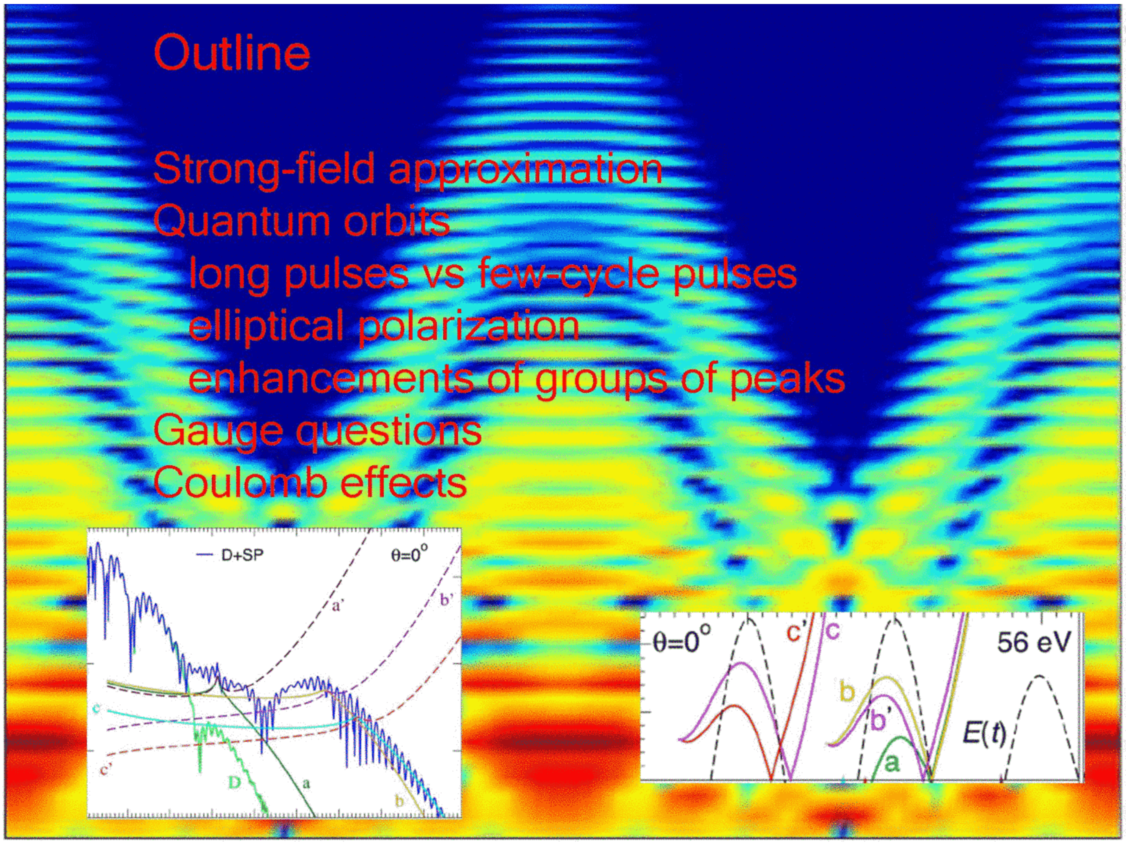
Attosecond Science, Kavli Institute for Theoretical Physics, Santa Barbara, CA, August 1 - 4, 2006

Collaborators

D. Bauer, MPI Heidelberg
G. G. Paulus, Texas A&M
R. Kopold, Siemens, Regensburg
M. Kleber, TU München

A. Gazibegovic-Busuladzic, Univ. of Sarajevo
E. Hasovic, Univ. of Sarajevo

supported by VolkswagenStiftung, Deutsche Forschungsgemeinschaft, the Ministry of Education and Science, Canton Sarajevo, and the Federal Ministry of Education and Science, Bosnia and Hercegovina



Strong-field approximation (SFA)
Keldysh–Faisal–Reiss (KFR) approximation

Matrix element for ionization into a state with drift momentum $\mathbf{p}$:

$$M_{\mathbf{p},E_0} = \lim_{t\to\infty} \langle \psi_{\mathbf{p}}(t)|\Psi(t)\rangle = \lim_{t\to\infty, t'\to-\infty} \langle \psi_{\mathbf{p}}(t)|U(t,t')|\psi_0(t')\rangle$$

$(\psi_{\mathbf{p}}, (\psi_0)$ = field-free scattering state (bound state) of the atom,
$U(t,t')$ = time-evolution operator of the system atom + field)

Dyson expansion with respect to the interaction H_I with the laser fi eld

$$U = U_0 - iUH_IU_0$$

$$M_{\mathbf{p},E_0} = -i \lim_{\substack{t\to\infty \\ t'\to-\infty}} \langle \psi_{\mathbf{p}}(t)|UH_IU_0|\psi_0(t')\rangle$$

(U_0 = atomic propagator, without the laser field
$U^{(\mathrm{Vv})}$ = propagator in the laser field, without the atom)

Volkov wave functions

Solutions of the Schrödinger equation

$$i\partial_t \psi_{\mathbf{k}}(\mathbf{r},t)^{(\mathrm{Vv})} = \left(\frac{1}{2m}\hat{\mathrm{p}}^2 - e\mathbf{r}\cdot\mathbf{E}\right)\psi_{\mathbf{k}}(\mathbf{r},t)^{(\mathrm{Vv})}$$

in the presence of a plane electromagnetic wave with the vector potential $\mathbf{A}(t)(\mathbf{E} = -\partial_t \mathbf{A})$

$$\psi_{\mathbf{k}}(\mathbf{r},t)^{(\mathrm{Vv})} = (2\pi)^{-3/2} e^{-i(\mathbf{k}-e\mathbf{A}(t))\cdot\mathbf{r}} \times \exp\left[-\frac{i}{2m}\int^t d\tau(\mathbf{k}-e\mathbf{A}(\tau))^2\right]$$

Volkov time-evolution operator

$$U^{(\mathrm{Vv})}(t,t') = -i\theta(t-t')\int d^3\mathbf{k}|\psi_{\mathbf{k}}(t)^{(\mathrm{Vv})}\rangle\langle\psi_{\mathbf{k}}(t')^{(\mathrm{Vv})}|$$

Note: $\mathbf{A}(t)$ can be a *finite* pulse

Strong-field approximation:

(KFR, Lewenstein)

replace $U \to U^{(\mathrm{Vv})}, \ \psi_{\mathbf{p}} \to \psi_{\mathbf{p}}^{(\mathrm{Vv})}$

$$\begin{aligned} M_{\mathbf{p},E_0} &= -i \lim_{\substack{t\to\infty \\ t'\to-\infty}} \langle\psi_{\mathbf{p}}(t)|UH_I U_0|\psi_0(t')\rangle \\ &\to -i\int_{-\infty}^{\infty} d\tau \langle\psi_{\mathbf{p}}^{(\mathrm{Vv})}(\tau)|H_I(\tau)|\psi_0(\tau)\rangle \\ &= -i\int_{-\infty}^{\infty} d\tau \langle\psi_{\mathbf{p}}^{(\mathrm{Vv})}(\tau)|V(\mathbf{r})|\psi_0(\tau)\rangle \end{aligned}$$

„direct“ electrons

propagation only in the fi eld after ionization

lost modifications of the initial state prior to ionization (depletion, Stark shift) (to include, need to dress the initial state)
gauge invariance of the exact amplitude,
„soft“ Coulomb effects throughout (Coulomb refocusing),
„hard“ Coulomb effects (rescattering)

Generalized Keldysh theory: Rescattering

start from the exact expression

$$M_{\mathbf{p},E_0} = -i \lim_{t\to\infty} \int_{-\infty}^{t} d\tau \langle \psi_{\mathbf{p}}(t) | U(t,\tau) H_I(\tau) | \psi_0(\tau) \rangle$$

use the Dyson equation wrt the atomic potential V

$$U = U^{(\mathrm{Vv})} - iU^{(\mathrm{Vv})} V U$$

replace again $U \to U^{(\mathrm{Vv})}$, $\psi_{\mathbf{p}} \to \psi_{\mathbf{p}}^{(\mathrm{Vv})}$ to get

$$M_{\mathbf{p},E_0} = -i \int_{-\infty}^{\infty} dt \int_{-\infty}^{t} dt' \langle \psi_{\mathbf{p}}^{(\mathrm{Vv})}(t) | V U^{(\mathrm{Vv})}(t,t') V | \psi_0(t') \rangle$$

direct + rescattered electrons

Generalized Keldysh theory: Rescattering (cont.)

an alternative expression:

$$M_{\mathbf{p},E_0} = -i \int_{-\infty}^{\infty} dt \langle \psi_{\mathbf{p}}^{(\mathrm{Vv})(t)} | H_I(t) | \psi_0(t') \rangle \quad \text{direct electrons}$$

$$-i \int_{-\infty}^{\infty} dt \int_{-\infty}^{t} dt' \langle \psi_{\mathbf{p}}^{(\mathrm{Vv})}(t) | V U^{(\mathrm{Vv})}(t,t') H_I(t') | \psi_0(t') \rangle$$

rescattered electrons

in the last line, may replace

$V(\mathbf{r}) \to V_{\mathrm{scatt}}(\mathbf{r})$ going beyond the SAEA

restored „hard“ Coulomb effects in first-order Born approximation

KFR (direct) electron spectrum

$$M_{\mathbf{p}} \sim \int dt \langle \psi_{\mathbf{p}}^{\text{Volkov}}(t)|V(\mathbf{r})|\psi_0(t)\rangle \sim \int_{-\infty}^{\infty} dt V_{\mathbf{p},0} e^{iS_{\mathbf{p},E_0}(t)}$$

V_{p0} = <p-eA(t)|V|0> $\quad S_{\mathbf{p},E_0}(t) = |E_0|t + \frac{1}{2m}\int^t d\tau [\mathbf{p} - e\mathbf{A}(\tau)]^2$

Long periodic pulse:

$S_{\mathbf{p},E_0}(t+T) = \left(|E_0| + \frac{\mathbf{p}^2}{2m} + U_P\right)T + S_{\mathbf{p},E_0}(t)$

Recall

$$\sum_{n=-\infty}^{\infty} e^{inx} = 2\pi \sum_{n=-\infty}^{\infty} \delta(x - 2n\pi)$$

$$M_{\mathbf{p}} \sim \sum_{n=-\infty}^{\infty} \delta\left(\frac{\mathbf{p}^2}{2m} + U_P + |E_0| - n\omega\right) \int_0^T dt V_{\mathbf{p},0} e^{iS_{\mathbf{p},E_0}(t)}$$

Interference from many cycles generates discrete spectrum

Saddle-point evaluation of the remaining integral over one cycle

$$\frac{d}{dt} S_{\mathbf{p},E_0}(t) = 2m|E_0| + [\mathbf{p} - e\mathbf{A}(t)]^2 \stackrel{!}{=} 0$$

(Notice $\mathbf{p} = e\mathbf{A}(t)$ in the tunneling limit)

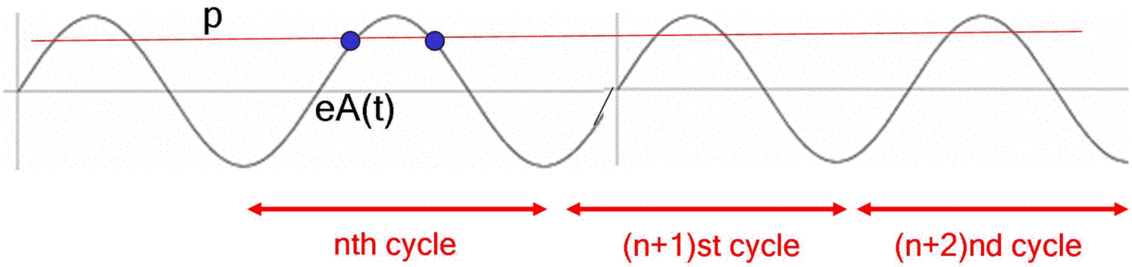
solutions $t \equiv t_s(\mathbf{p}) \quad (s = 1, 2, \ldots)$

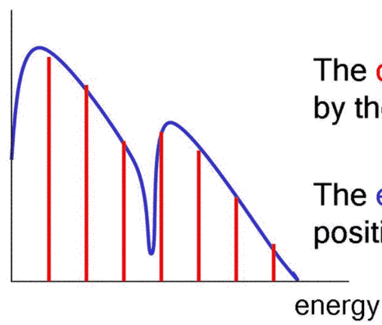
$$M_{\mathbf{p}} = \sum_n \delta\left(\frac{\mathbf{p}^2}{2m} + U_p + |E_0| - n\hbar\omega\right) \times \sum_{s,\text{one cycle}} [S''_{\mathbf{p},E_0}(t_s)]^{-1/2} e^{iS_{\mathbf{p},E_0}(t_s)}$$

- Interference from different cycles generates discrete peaks
- Interference from within one cycle generates structure of the spectral envelope

For a few-cycle pulse: no discrete peaks! Just a few contributions interfere

One cycle vs many cycles

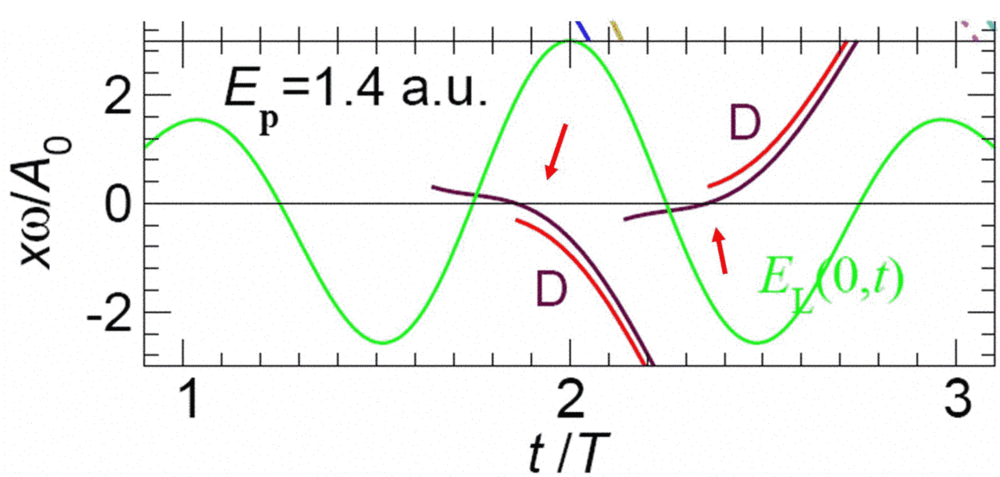




The discreteness of the spectrum is generated by the superposition of all cycles

The envelope is generated by the superposition of the two solutions within one cycle

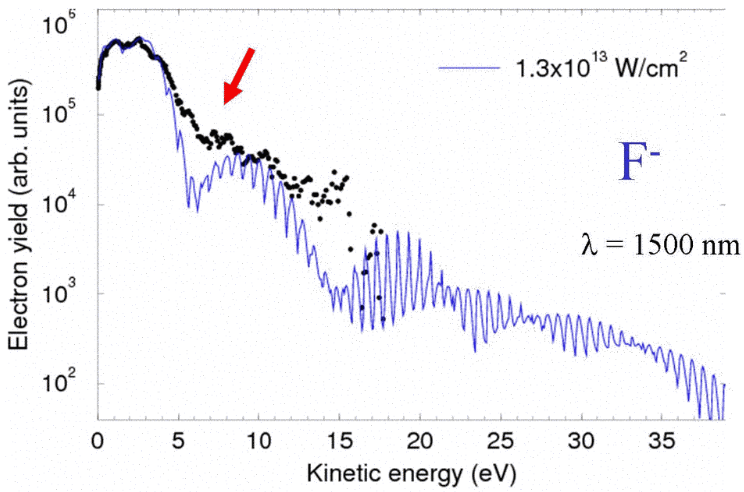
Examples of direct quantum orbits



One member of a pair of orbits experiences the Coulomb potential more than the other (see later)

Interference of the two solutions from within one cycle

(includes focal averaging)



Data: I. Yu Kiyan, H. Helm, PRL 90, 183001 (2003)
(1.1 x 10^{13} Wcm^{-2})
Theory: D.B. Milosevic et al., PRA 68, 070502(R) (2003)
(1.3 x 10^{13} Wcm^{-2})

cf. M.V. Frolov, N.M. Manakov, E.A. Pronin, A.F. Starace, JPB 36, L419 (2003)

Improved Keldysh approximation for HATI (cont.)

$$M(\mathbf{p}) \sim \int_{-\infty}^{\infty} dt \int_{-\infty}^{t} dt' \int d^3q \langle \psi_{\mathbf{p}}^{(\mathrm{Vv})}(t)|V|\psi_{\mathbf{q}}^{(\mathrm{Vv})}(t)\rangle \times \langle \psi_{\mathbf{q}}^{(\mathrm{Vv})}(t')|V|\psi_0(t')\rangle$$

using the saddle-point approximation

$$[\mathbf{q} - e\mathbf{A}(t')]^2 = -2m|E_0| \qquad \text{ionization}$$

$$(t-t')\mathbf{q} = \int_{t'}^{t} d\tau e\mathbf{A}(\tau) \qquad \text{return}$$

$$[\mathbf{q} - e\mathbf{A}(t)]^2 = [\mathbf{p} - e\mathbf{A}(t)]^2 \qquad \text{elastic scattering}$$

to replace the five-dimensional integration by a summation over the saddle points, expand in terms of "quantum orbits"

$$M(\mathbf{p}) \sim \sum_{\text{orbits } s} a_s(\mathbf{p}) \exp[iS_s(\mathbf{p})]$$

Quantum-orbit expansion of the transition amplitude

$$M(\mathrm{p}) = \sum_{\text{orbits } s} a_s(\mathrm{p}) \exp[iS_s(\mathrm{p})]$$

cf. Feynman's path integral

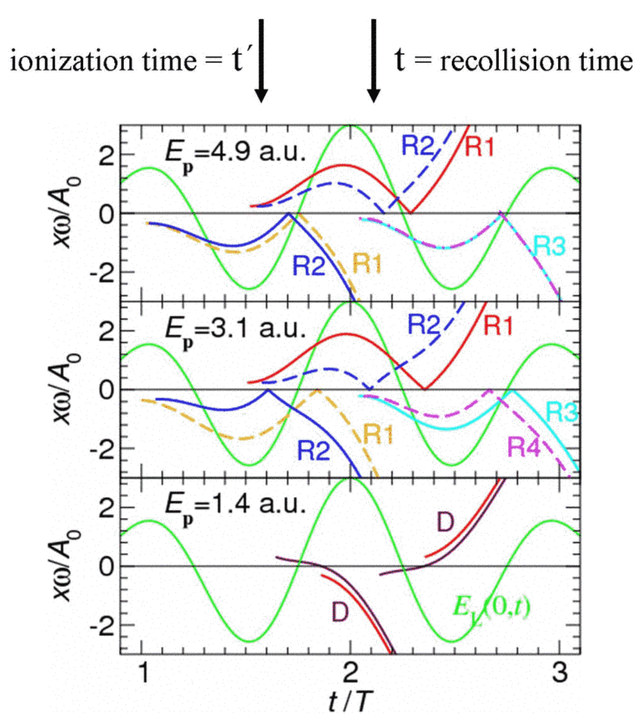
P. Salières et al., Science 292, 902 (2001)

The quantum orbits are defined by the solutions $(t_s, t'_s, \mathrm{k}_s)$ $(s = 1, 2, \ldots)$ of the saddle-point equations:

$$m\mathrm{x}(t) = \begin{cases} (t - t'_s)\mathrm{k}_s - \int_{t'_s}^{t} d\tau e\mathbf{A}(\tau), & (\mathrm{Re}\, t'_s \le t \le \mathrm{Re}\, t_s) \\ (t - t_s)\mathrm{p} - \int_{t_s}^{t} d\tau e\mathbf{A}(\tau). & (t \ge \mathrm{Re}\, t_s) \end{cases}$$

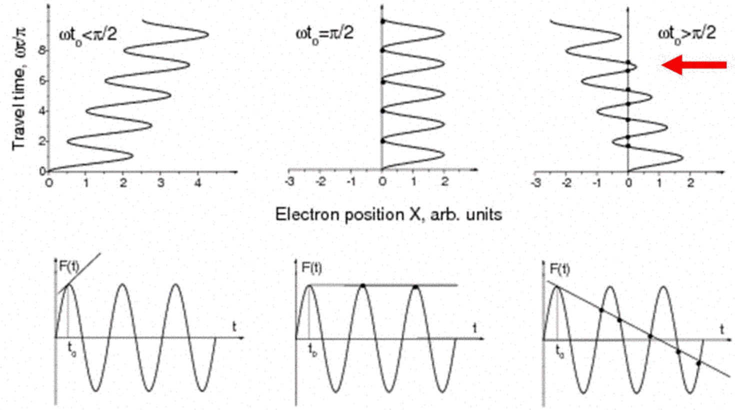
x(t=t_s') = 0, but Re [x(Re t_s')] different from 0

Rescattered quantum orbits in space and time



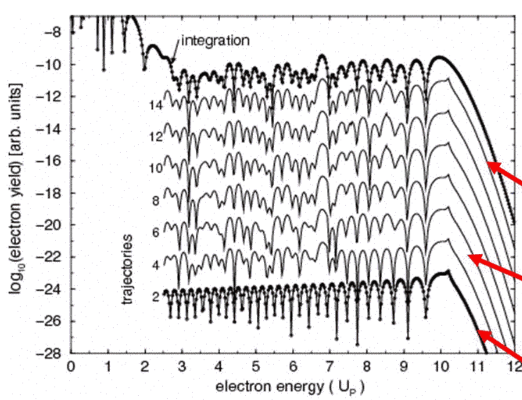
„Long orbits“ or late returns

The electron may **revisit** many times and **rescatter** upon a late return



What is the effect of the late returns?

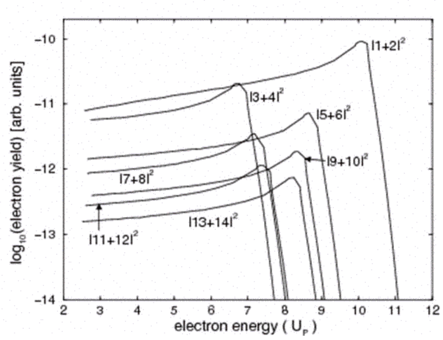
Building up the ATI spectrum from quantum orbits



shortest 14 orbits

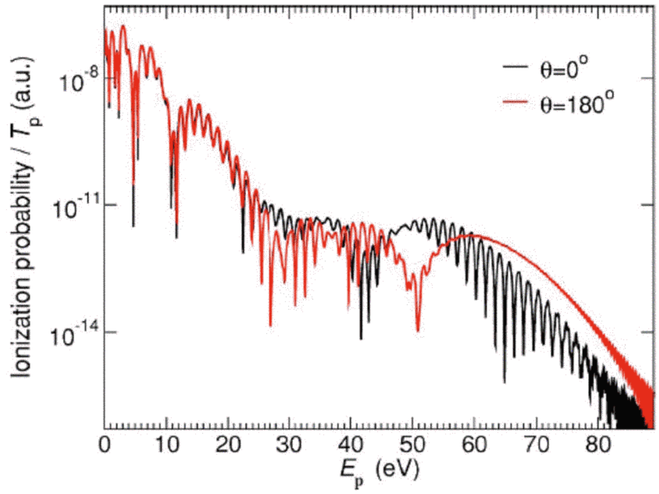
shortest six orbits 1 +...+ 6

shortest two orbits 1+2



Magnitude of the contributions of the various pairs of orbits

Few-cycle-pulse ATI spectrum: violation of backward-forward symmetry



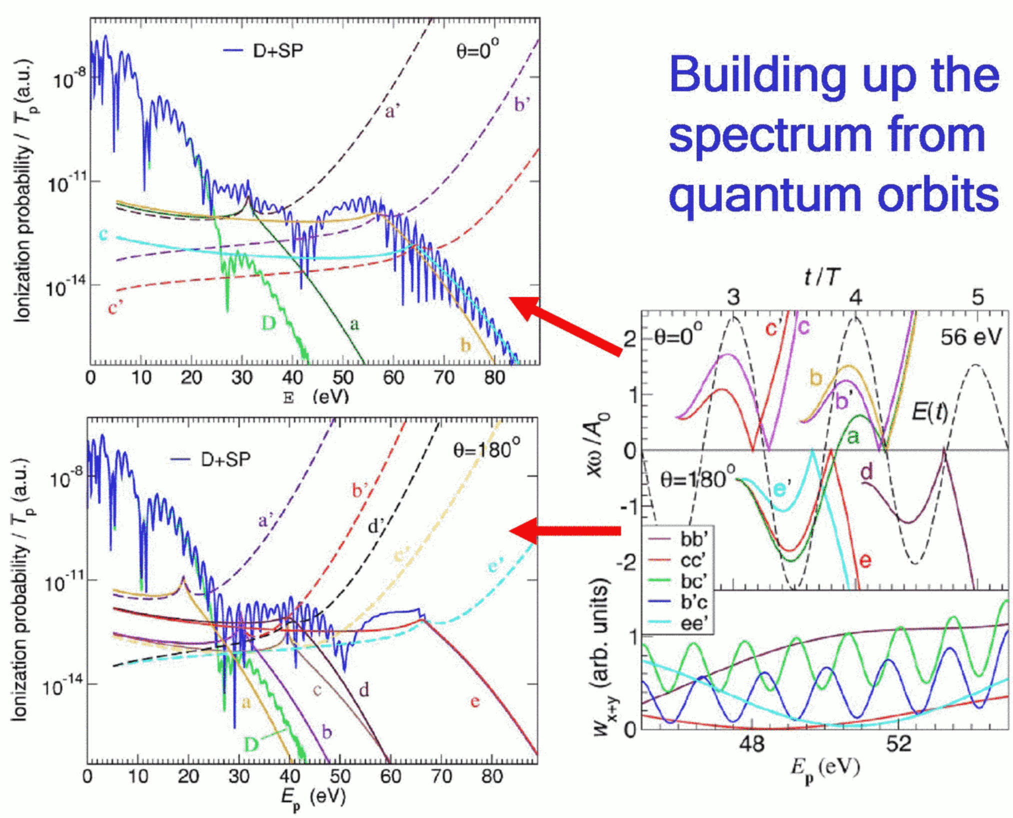
argon, 800 nm
7-cycle duration
sine square envelope
cosine pulse, CEP = 0
10^{14} Wcm^{-2}

Different cutoffs

Peaks vs no peaks

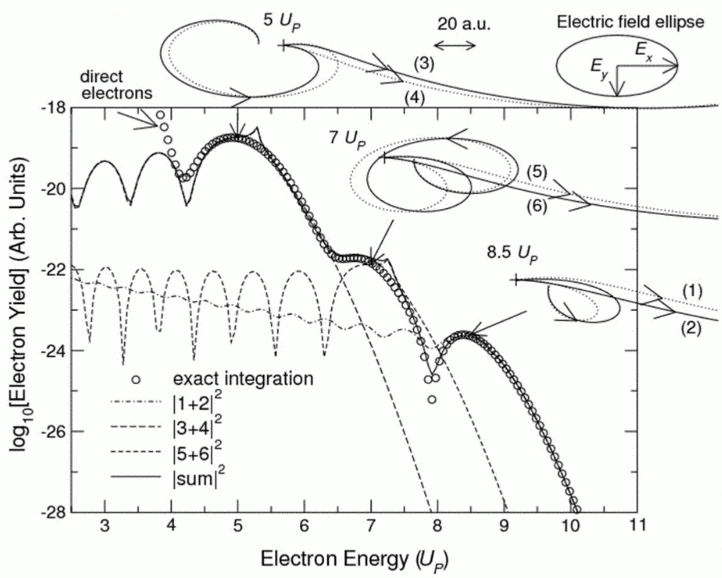
D. B. Milosevic, G. G. Paulus, WB, PRA 71, 061404 (2005)

Building up the spectrum from quantum orbits



2-dimensional quantum orbits: elliptical polarization

Different orbits dominate different parts of the spectrum



$\xi = 0.5$

ω = 1.59 eV
I_p = 24.5 eV
I = 5 x 10^{14} Wcm^{-2}

Note: the shortest orbit is weak

Kopold, Milosevic, and WB, PRL 84, 3831 (2000)

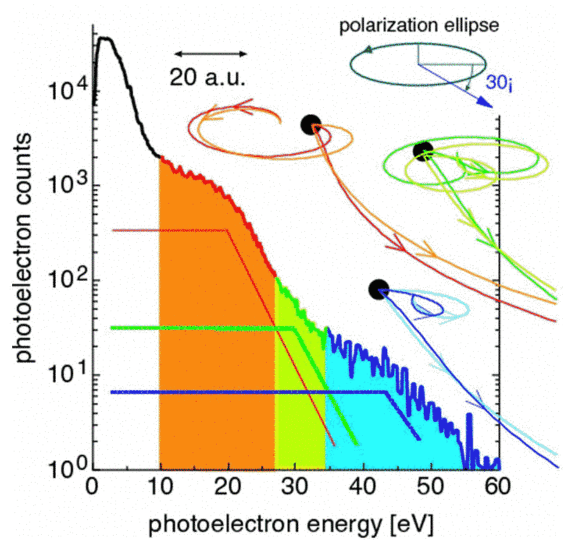
Quantum orbits for elliptical polarization: Experiment vs. theory

$\xi = 0.36$

xenon at 0.77 x $10^{14}$$Wcm^{-2}$

The plateau becomes a staircase

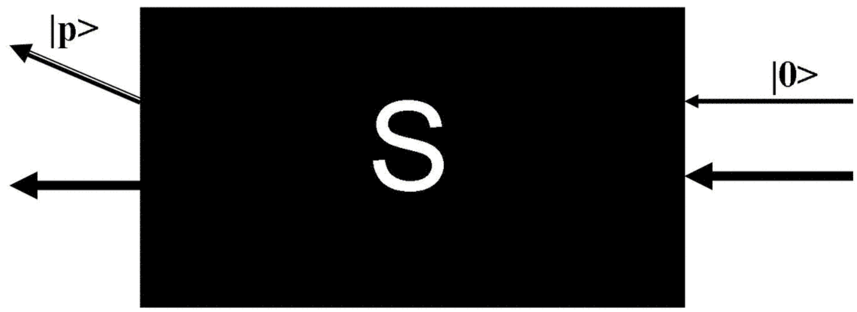
The shortest orbits are not always the dominant orbits



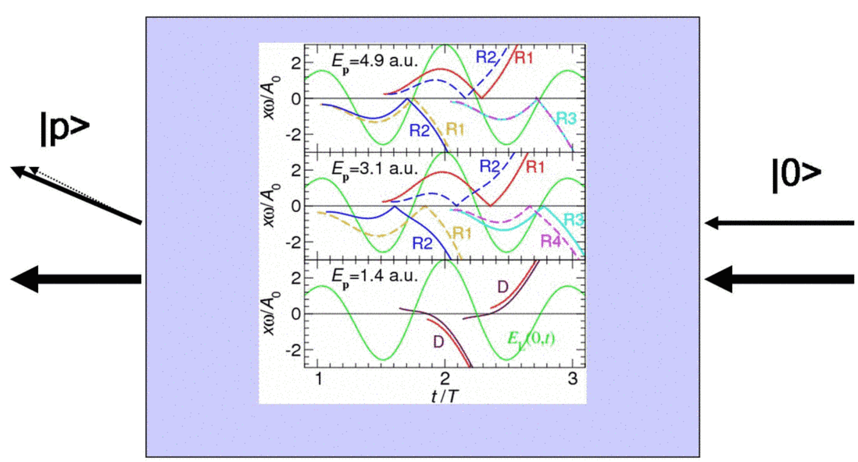
Salieres, Carre, Le Deroff, Grasbon, Paulus, Walther, Kopold, Becker, Milosevic, Sanpera, Lewenstein, Science 292, 902 (2001)

The black box of S-matrix theory ...

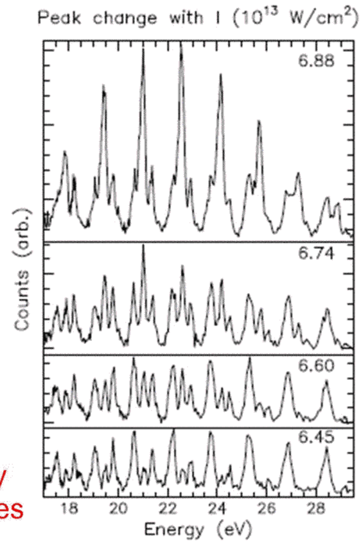
|out> = S|in>



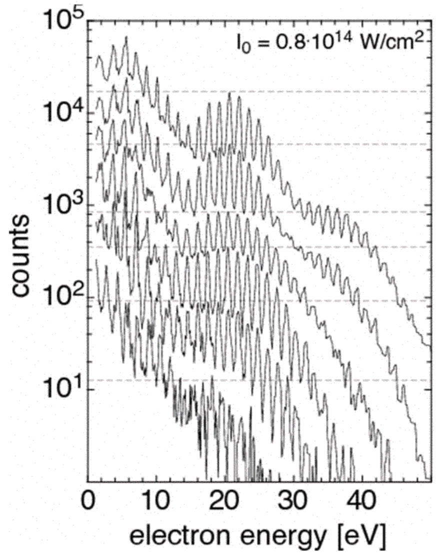
... has been made transparent



Intensity-dependent enhancements



intensity increases by 6%

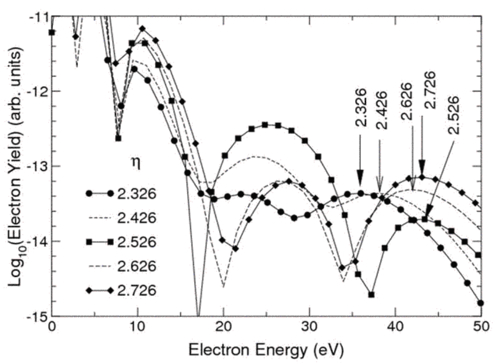


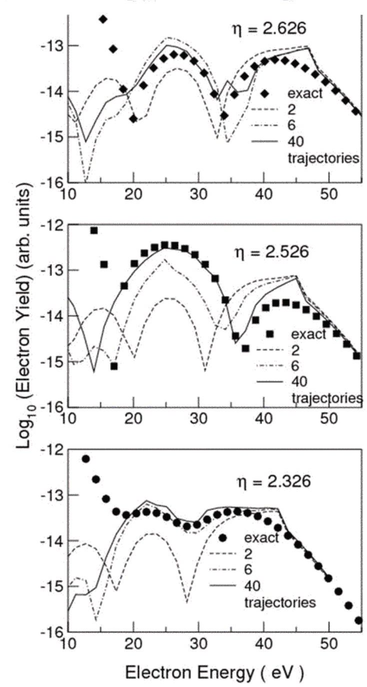
Hertlein, Bucksbaum, Muller, JPB 30, L197 (1997)

Paulus et al., PRA 64, 021401 (2001)

see, also, Hansch, Walker, van Woerkom, PRA 55, R2535 (1997)

Intensity-dependent enhancements: SFA-type theory



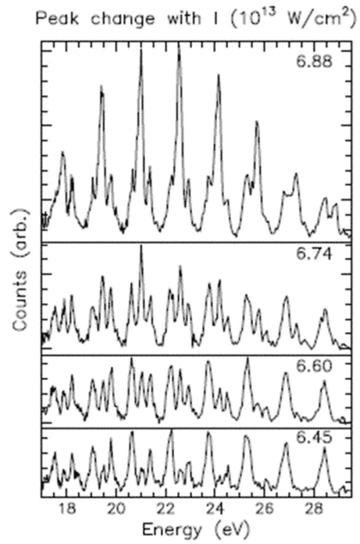


Enhancements occur at channel closings

where $U_p + I_p = N\omega$

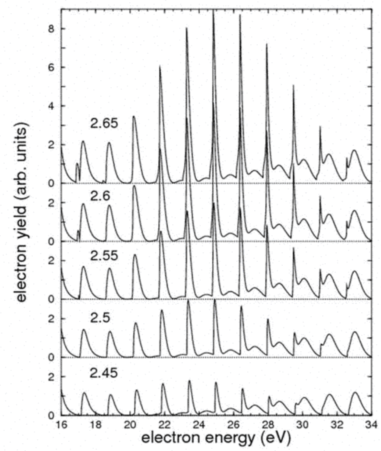
At the enhancement intensity, many more orbits are needed for good convergence than at other intensities

Enhancements: SFA-type theory vs experiment



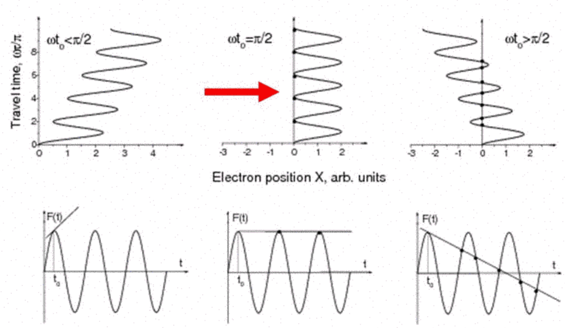
argon spectra,
6.45 10^{13} Wcm^{-2} < I < 6.88 10^{13} Wcm^{-2}
Hertlein, Bucksbaum, and Muller, JPB 30, L197 (1997)

Focal-averaged zero-range potential
SFA simulation



6.39 10^{13} Wcm^{-2} < I < 6.91 10^{13} Wcm^{-2}
Kopold, Becker, Kleber, Paulus,
JPB 38, 217 (2002)

Physical origin of the enhancements



For zero drift momentum, p = 0, the electron revisits infinitely often

$$p^2/(2m) = N\omega - I_p - U_p = 0$$

$$I_p + U_p = N\omega$$
„Channel Closing“

Constructive interference of long quantum orbits

Quantum effect!!!

Analytical proof:
S.V. Popruzhenko, P.A. Korneev, S.P. Goreslavskii, WB, PRL 89, 023001 (2002)
D.B. Milosevic, WB, PRA 66, 063417 (2002)

Alternative explanations of the intensity-dependent enhancements

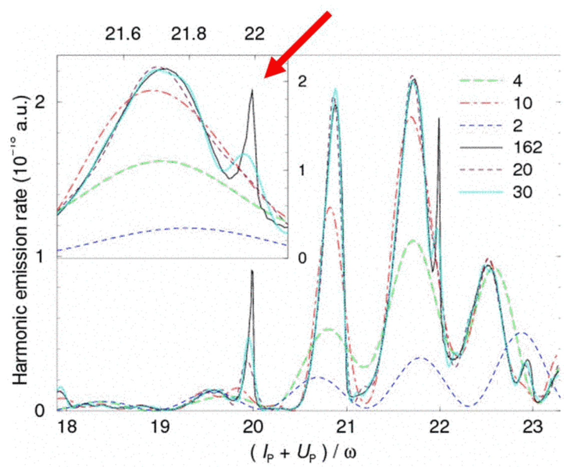
Threshold cusps a la Wigner/Baz:

B. Borca, M.V. Frolov, N.L. Manakov, A.F. Starace, PRL 88, 193001 (2002)

Multiphoton resonance with ponderomotively upshifted Rydberg state:

H.G. Muller, F.C. Kooiman, PRL 81, 1207 (1998)
H.G. Muller, PRL 83, 3158 (1999)
J. Wassaf, V. Veniard, R. Taieb, A. Maquet, PRL 90, 013003 (2003)

HHG channel closings: an extreme example



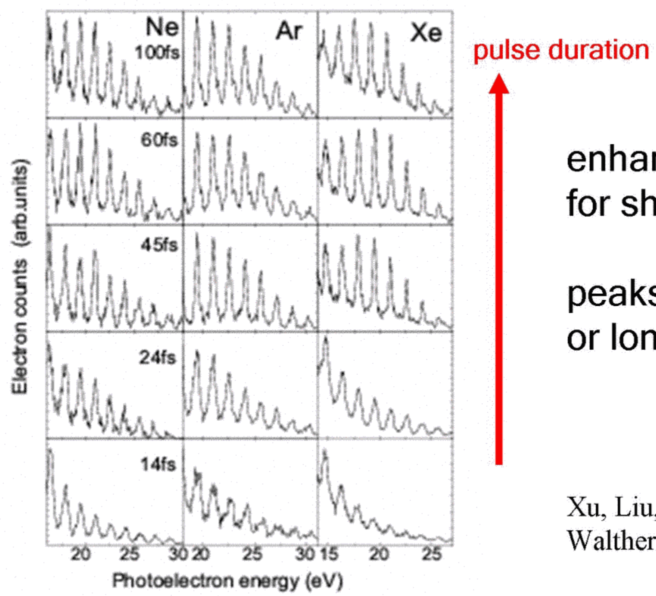
H25 as a function of intensity $(I_p + U_p)/\omega$

need more than 100 orbits (25 cycles) to reproduce the spike at the N = 22 channel closing

ω = 1.17 eV, I_p = 13.6 eV

D. B. Milosevic, WB, PRA 66, 063417 (2002)

Enhancements disappear for short pulses



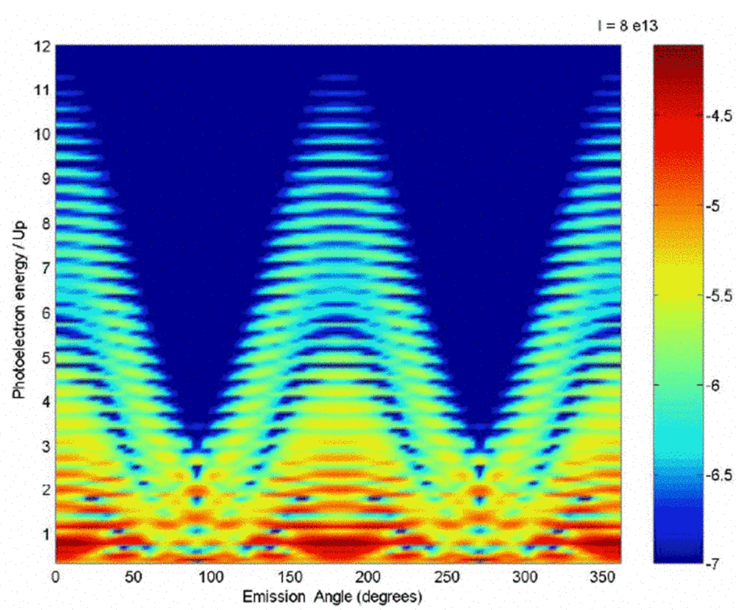
enhancements disappear for short pulses

peaks become narrower or long pulses

Xu, Liu, Kolomenskii, Schuessler, Walther, Paulus, to be published

Focal-averaged angular-resolved ATI energy spectrum

xenon, 760 nm, 8×10^{13} Wcm^{-2}



contains the effects of

channel closings (several)

cutoff (rainbow) enhancements

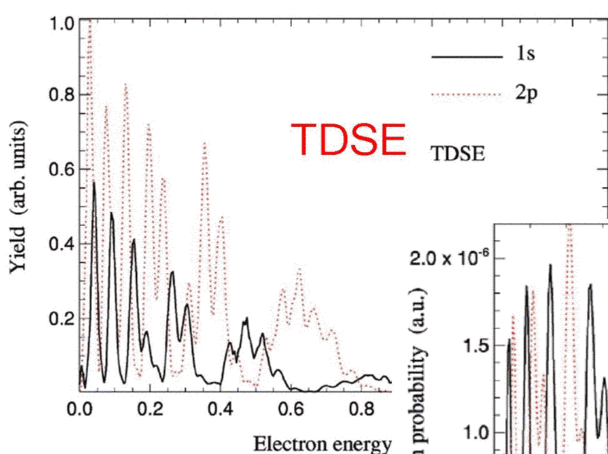
con-/destructive interferences of pairs of quantum orbits and/or the two members of each pair

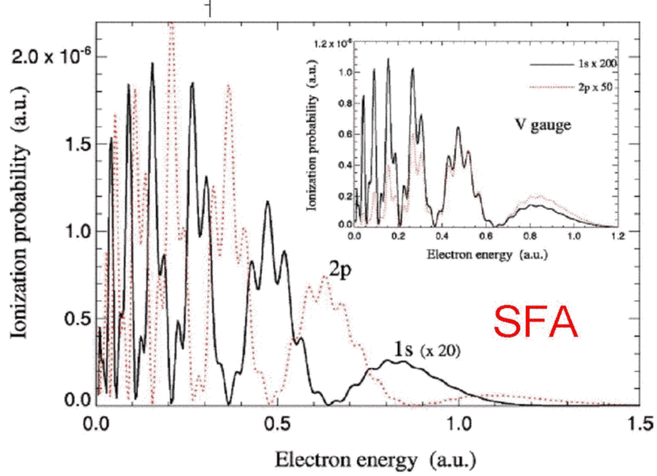
Which gauge is better for which problem?

Ionization of negative ions: angle-resolved energy spectrum which gauge works better?

ω = 0.056 a.u.
E_0 = 0.0834 a.u.
4-cycle sine-square sine pulse

D. Bauer, D.B. Milosevic, WB
PRA 72, 023415 (2005)





TDSE agrees with length-gauge SFA

Gauge vs initial-state wave function

length gauge velocity gauge

asymptotic wave function

$$\frac{1}{r}\exp(-\kappa r)Y_{lm}(\hat{r})$$

HF wave function

$$\sum_i C_i N_i r^{n_i-1}\times \exp(-\zeta_i r)Y_{lm}(\hat{r})$$

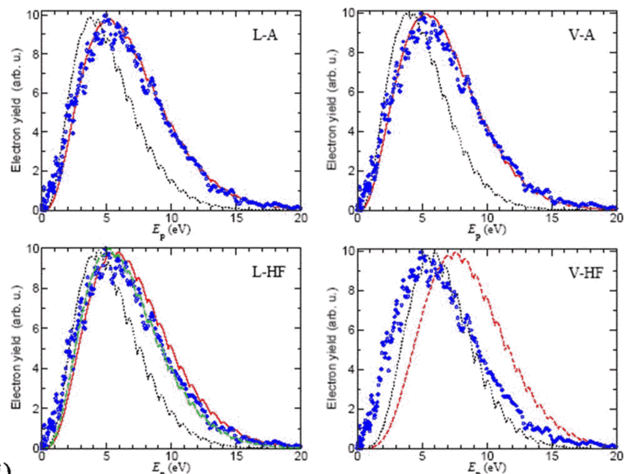


Figure 1. Comparison of the experimental data, presented by circles, for the detachment yield of F$^-$ by a circularly polarized laser field having the wavelength 1510 nm, with theoretical simulations. Left-hand panels: length gauge. Right-hand panels: velocity gauge. Upper row: asymptotic wave function (9). Lower row: Hartree-Fock wave function (10). The peak intensities underlying the calculations are for the black-dotted curve $I_{max} = 2.6\times10^{13}$ Wcm^{-2} (this is the intensity estimated for the experiment), for the solid (red) curve 3.8×10^{13} Wcm^{-2} and 3.4×10^{13} Wcm^{-2} for the dashed (green) curve (lower left panel only).

F^-

circular pol.
1510 nm
2.6 x 10^{13} Wcm^{-2}
(exp. est.)

Bergues, Ni, Helm
PRL 95, 263002 (2005)

theory:

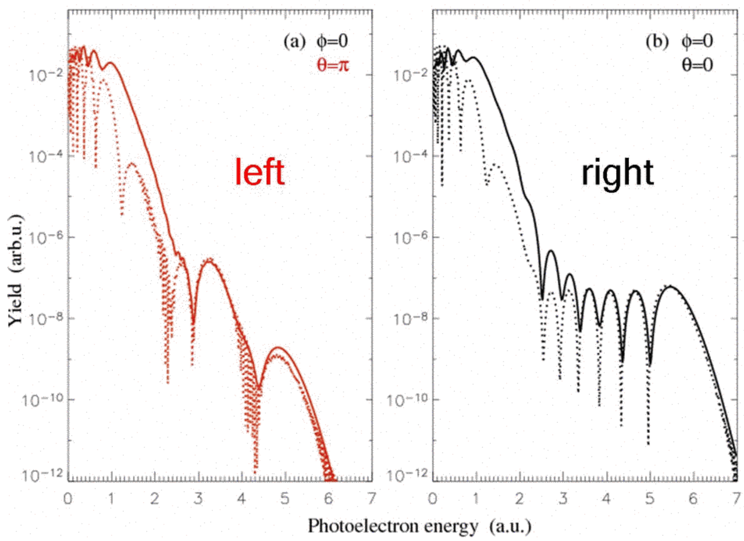
2.6 x 10^{13} Wcm^{-2}
3.8 x 10^{13} Wcm^{-2}
3.4 x 10^{13} Wcm^{-2}

all but V gauge plus HF wave function work

Outside the SFA:

Coulomb effects

Hydrogen H(1s) ATI spectra via TDSE and SFA



solid: TDSE;
dashed: SFA

ω = 0.056 a.u.
E_0 = 0.834 a.u.
4-cycle sine-square sine pulse

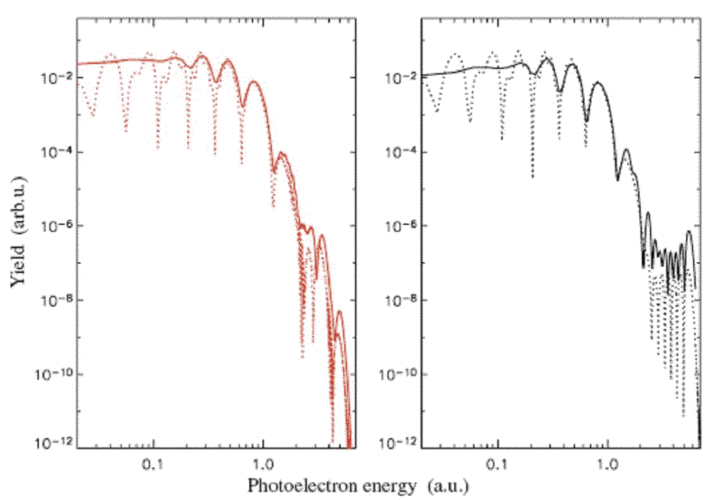
Problem areas:

very low energies

transition region between $2U_p$ and $5U_p$

D. Bauer, D.B. Milosevic, WB, JMO 53, 135 (2006)

Origin of interferences: short-range potential



solid: TDSE
dashed: SFA

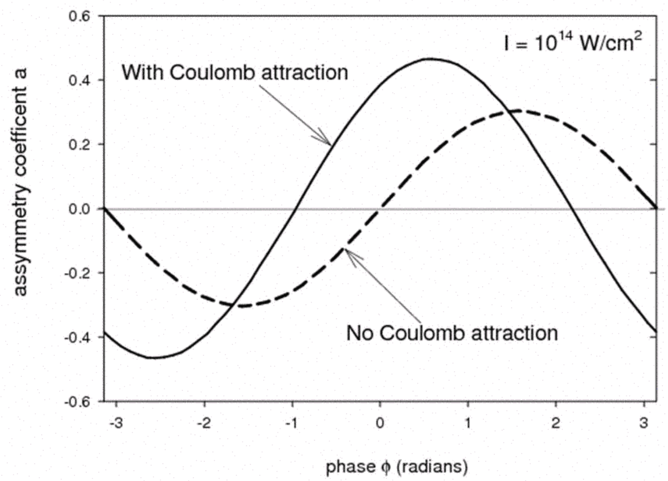
TDSE:
Coulomb potential cut at r_c = 2 a.u.

SFA:
Yukawa wave function

Interferences are not an artifact of the SFA

Backward-forward asymmetry as a function of the absolute phase

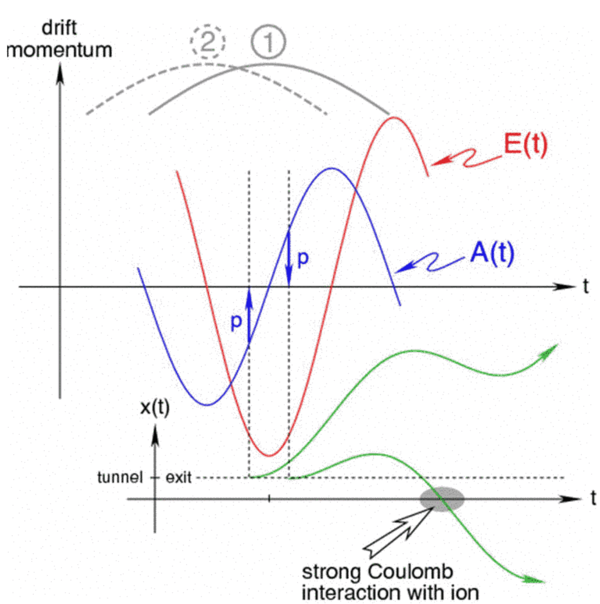
R = [W(left)-W(right)]/[W(left)+W(right)]



SFA predicts R = 0 for $\phi = 0$,
TDSE for $\phi = -0.3$

Chelkowski and Bandrauk
PRA 71, 053815 (2005)

Physical consequences of the Coulomb field



If the Coulomb field is ignored, envelope 1 yields backward-forward symmetry.

Due to Coulomb refocusing. the later orbit is preferred, violating b-f symmetry

The envelope 2 weakens the contribution of the later orbit and restores b-f symmetry.

Conclusions

The SFA is incredibly good

The SFA provides a benchmark

The Coulomb field is (sometimes) important