As-few-as-possible-body physics (laser-induced nonsequential multiple ionization)

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Collaborators

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What?

Double and multiple ionization of rare-gas atoms by a short (2 – 30 cycles) low-frequency (near-infrared, Ti:Sa) intense ($I > 10^{14}$ Wcm^{-2}) laser pulse

Why?

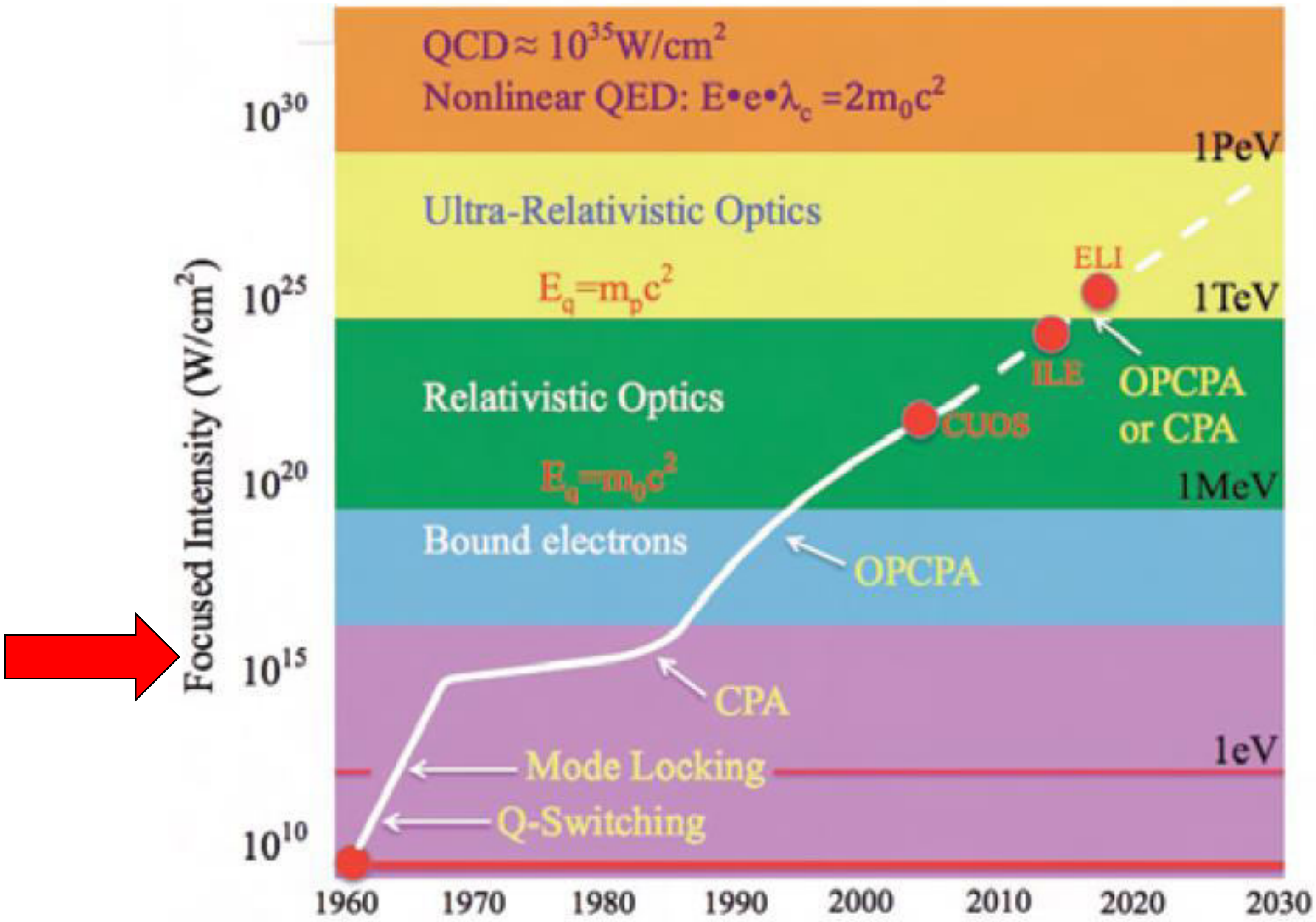
Electron-electron correlation in a few-particle system subject to an intense laser field

Extract dynamic information on the attosecond time scale

Meet the theoretical challenge

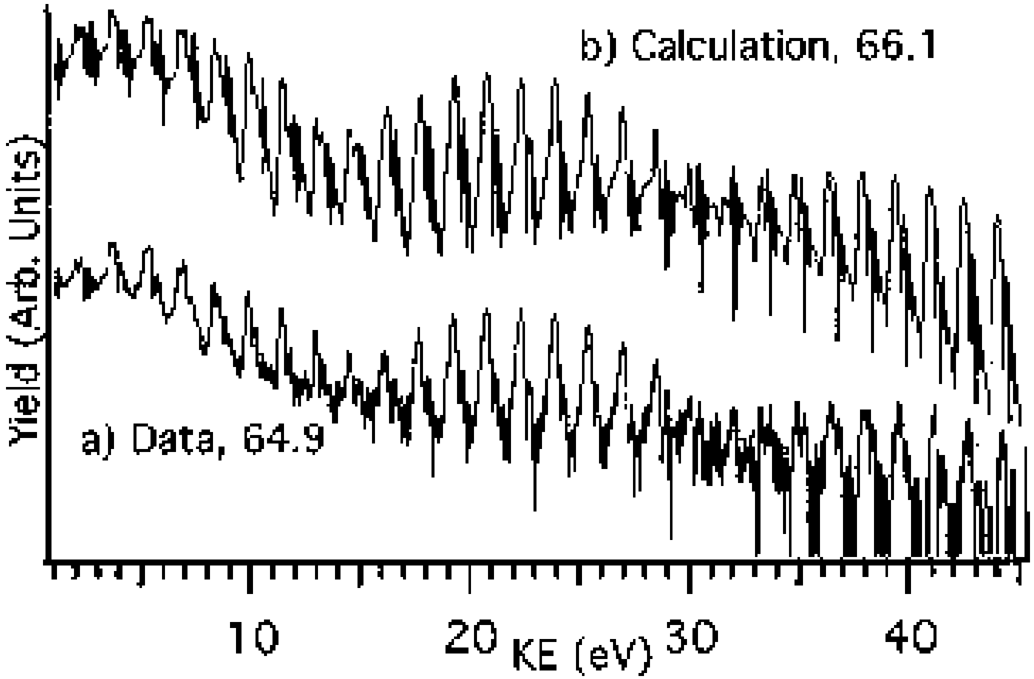
Plasma diagnostics via multiple-ion appearance intensities

Maximal laser intensities as a function of time



The amazing precision of the Single-Active-Electron Approximation

Comparison of data with one-particle 3D TDSE simulations



argon, 800 nm, 120 fs, intensity in TW/cm^2 as given

Nandor, Walker, Van Woerkom, Muller, PRA 60, R1771 (1999)

Double ionization of an atom by an intense laser pulse

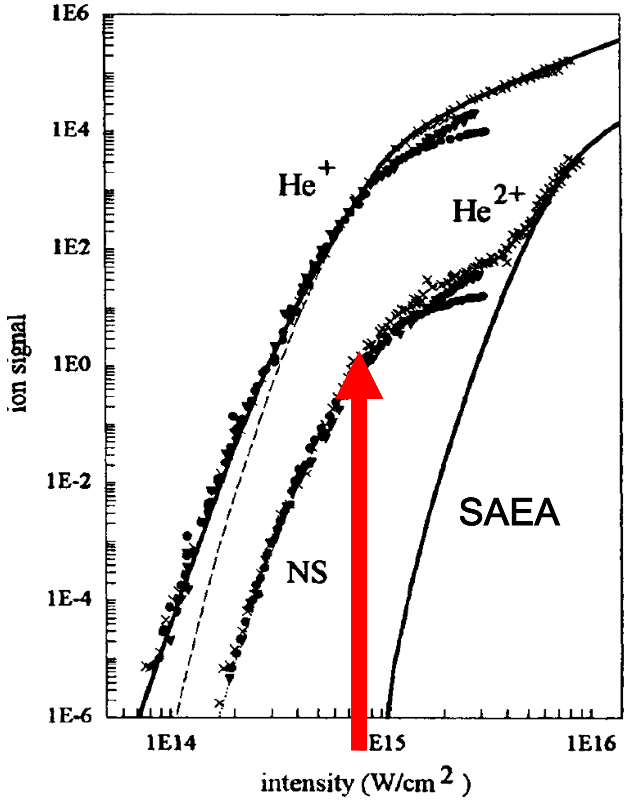
Does it require electron-electron correlation or does it not?

What are the footprints of correlation?

-- Laser-intensity dependence of the total yields
of ionized species with various charge states
-- Ion-momentum distribution
-- Correlation of electron momenta

Sequential vs. nonsequential ionization: the total rate

the „knee“



B. Walker, B. Sheehy, L.F. DiMauro, P. Agostini, K.J. Schafer, K.C. Kulander, PRL 73, 1227 (1994)

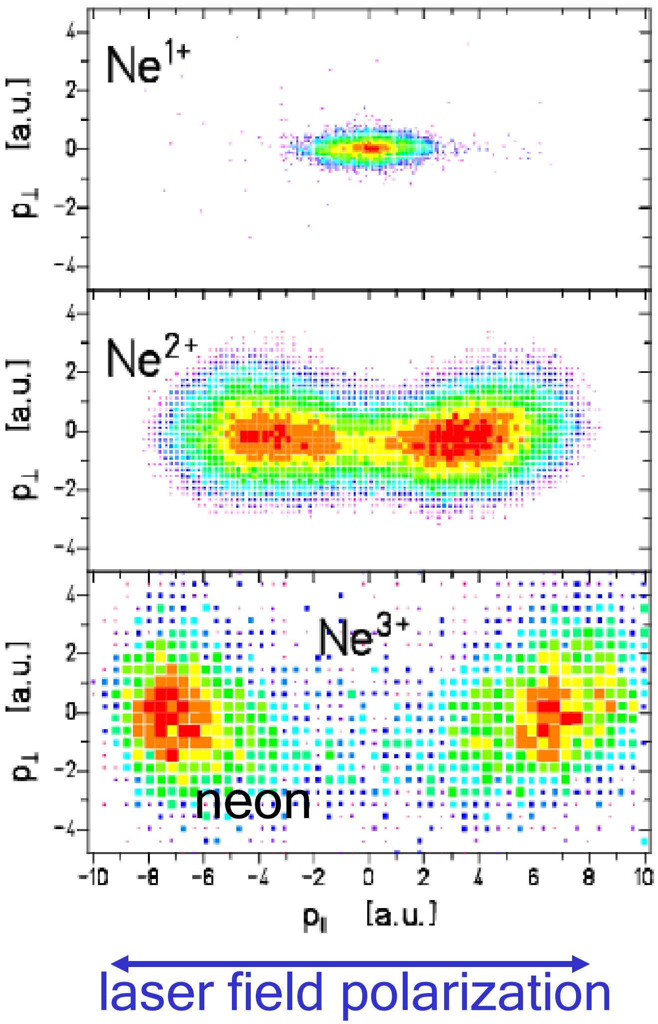
nonsequential = not sequential (requiring correlation)

first observation and identification of a nonsequential channel: A. L‘Huillier, L.A. Lompre, G. Mainfray, C. Manus, PRA 27, 2503 (1983)

The mechanism is, essentially, **rescattering**, like for high-order ATI and HHG

NB: the effect disappears for circular polarization

Nonsequential double ionization: the ion momentum

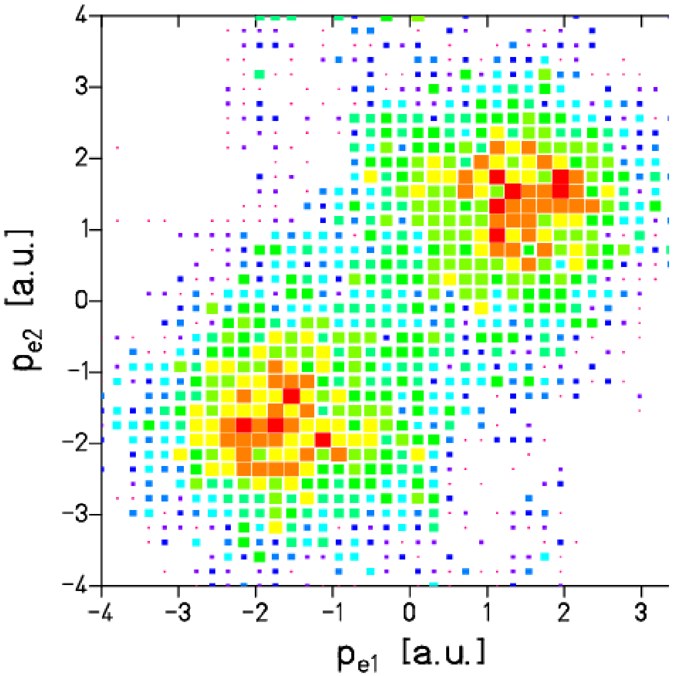


photon momentum is negligible

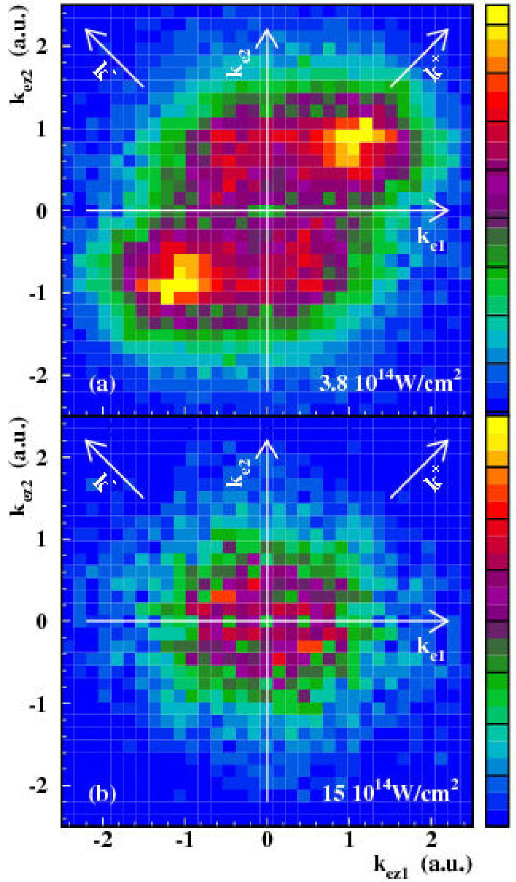
ion-momentum distribution is double-peaked

R. Moshammer, B. Feuerstein, W. Schmitt, A. Dorn, C..D. Schröter, J. Ullrich, H. Rottke, C. Trump, M. Wittmann, G. Korn, K. Hoffmann, W. Sandner, PRL 84, 447 (2000)

Nonsequential double ionization: the electron-electron correlation distribution



electrons go side by side



argon: Th. Weber, H. Giessen, M. Weckenbrock, G. Urbasch, A. Staudte, L. Spielberger, O. Jagutzki, V. Mergel, M. Vollmer, R. Dörner, Nature 405, 658 (2000)

neon: Heidelberg -MBI collaboration J. Phys. B 36, L113 (2003)

Unambiguous evidence of electron-electron correlation

In all laser-atom processes at near-infrared frequencies, a very large number of laser photons has to be absorbed

Lowest-order perturbation theory (LOPT), let alone higher orders, is impossible

What to do?

The new paradigm:
Tunneling followed by
classical propagation in the laser field

also known as the **„simple-man“ model**

An aside: the „simple-man model“

(Just one electron in the laser field
no correlation)

Tunneling ionization

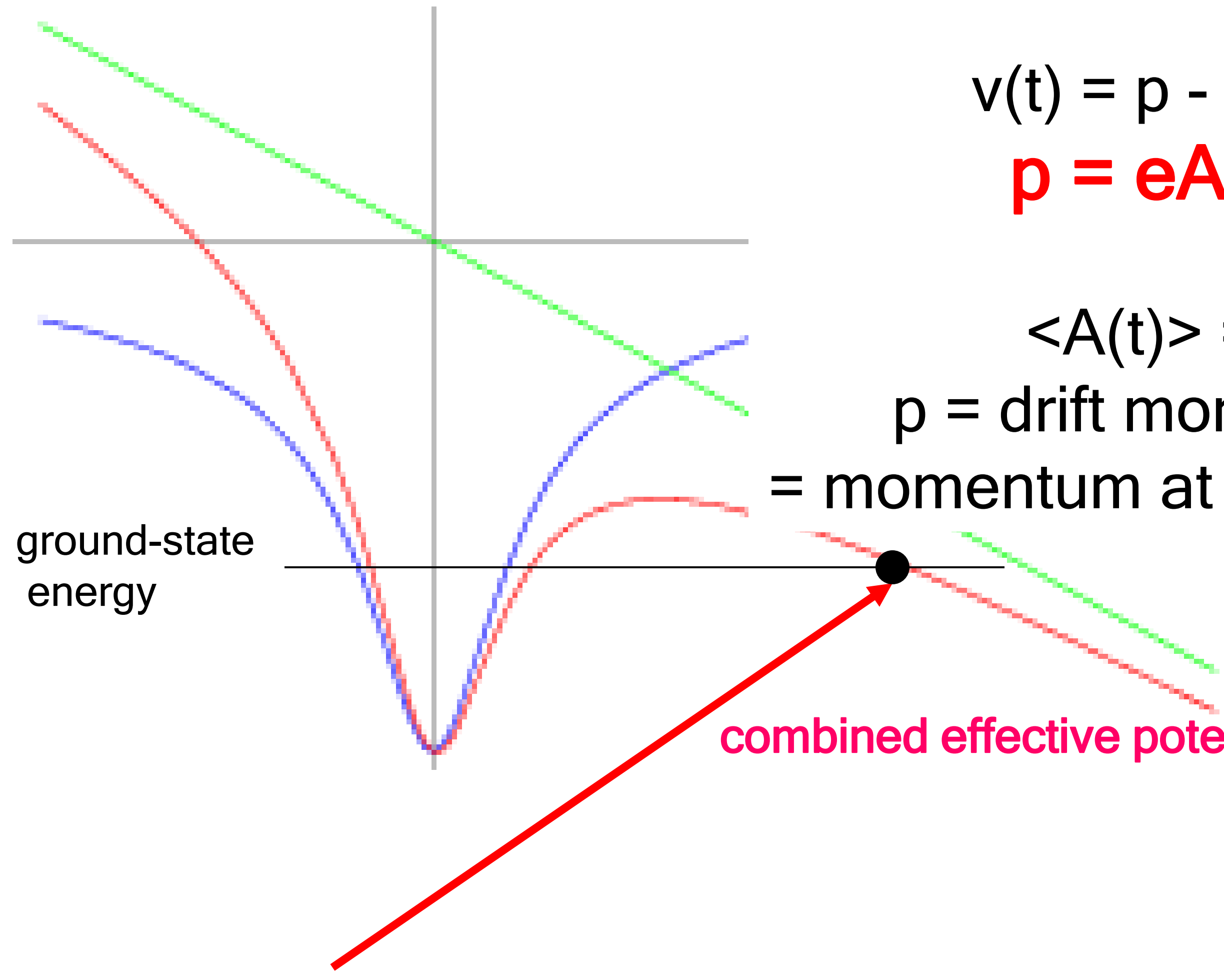
$v(t) = p - eA(t)$

$p = eA(t_0)$

$\langle A(t) \rangle = 0$

p = drift momentum

= momentum at the detector



classically: $v(t_0)=0$ at the exit of the tunnel

The „streaking condition“

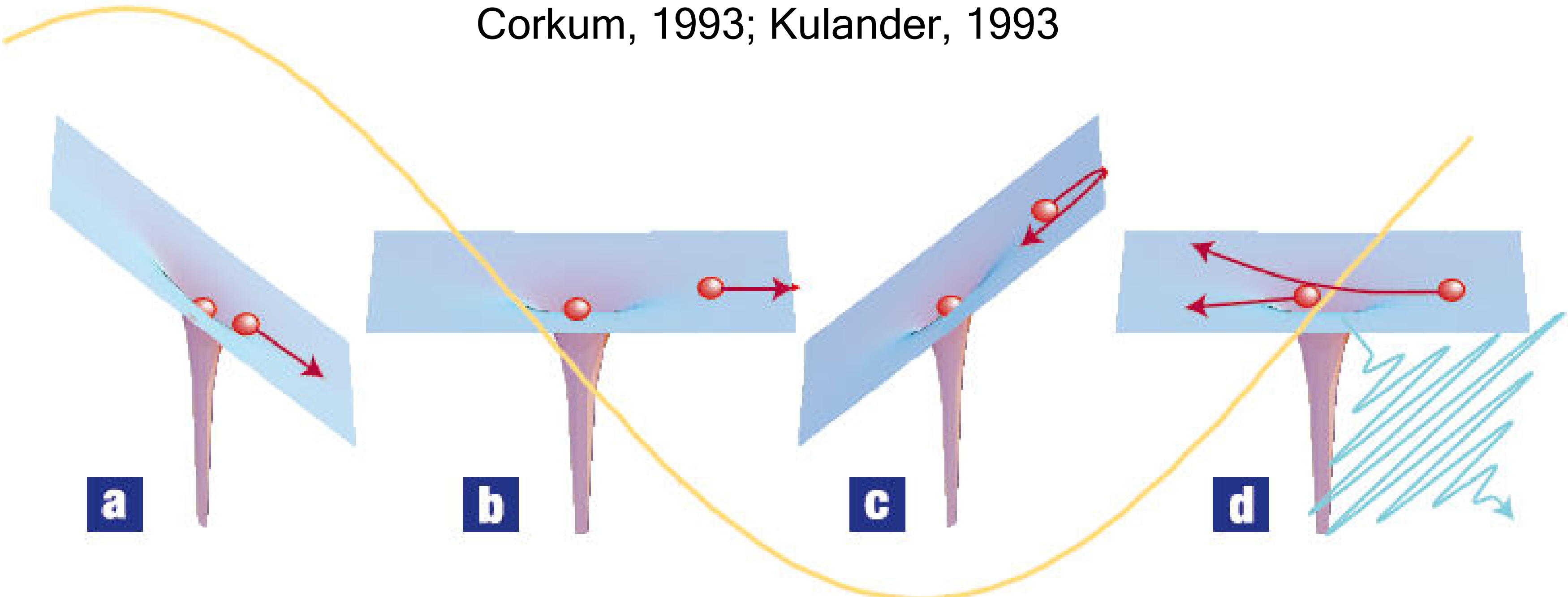
$$\mathbf{p} = e\mathbf{A}(t)$$

is at the heart of attosecond physics

p = electron momentum at the detector (outside the field)
A = vector potential of the streaking laser field
t = time of ionization

Recollision establishes correlation

Corkum, 1993; Kulander, 1993



Corkum & Krausz, Nature Phys. 3, 382 (2007)

The recolliding electron may
recombine (HHG), elastically rescatter (HATI),
inelastically rescatter (NSDI), ...

Integrating Newton's equation

$$m\ddot{x}(t) = eE(t)$$

$$m[x(t) - x(t_0)] = [\dot{x}(t_0) + eA(t_0)](t - t_0) - e\int_{t_0}^{t} d\tau A(\tau)$$

=0 (position of the atom)

=0 (initial velocity after tunneling)

= F(t) - F(t_0)

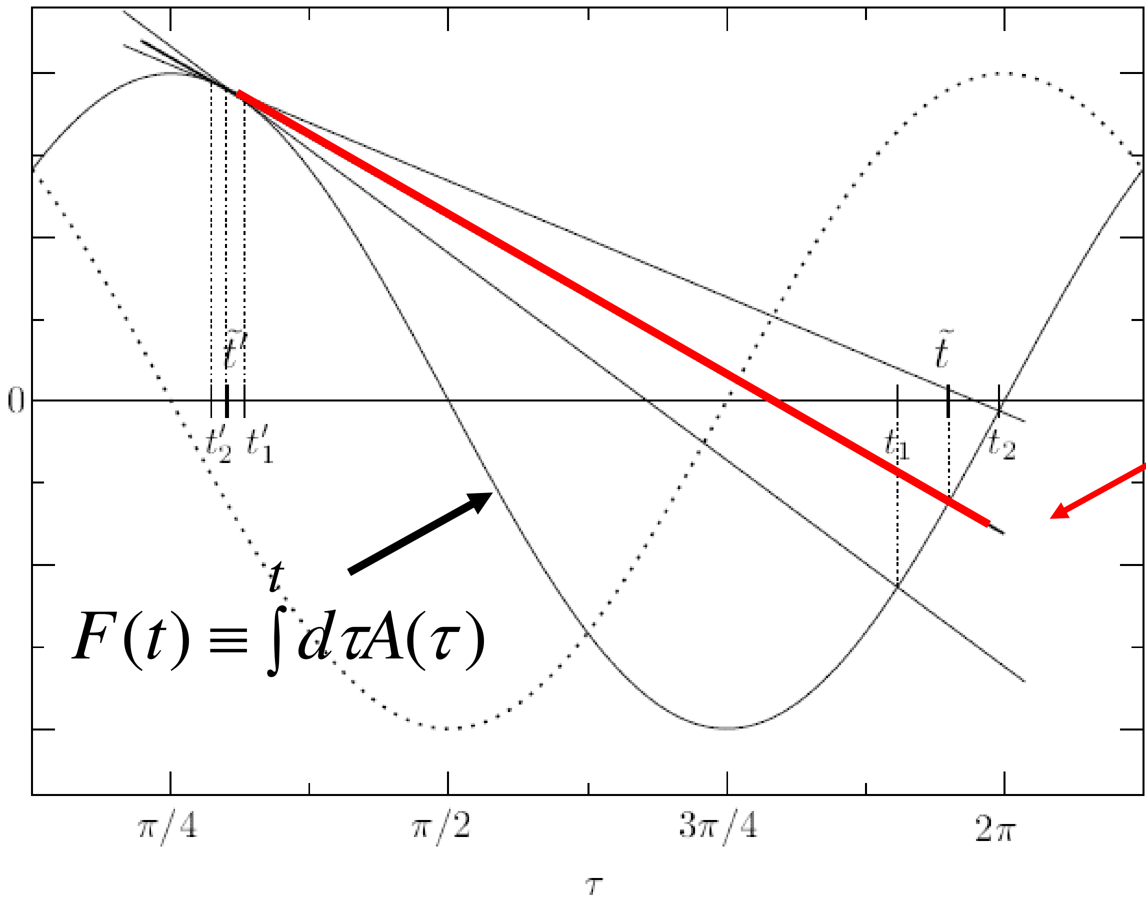
condition of recollision: $x(t) = 0$

$$F(t) = F(t_0) + (t - t_0)F'(t_0)$$

Graphical discussion of the return condition

$$F(t) = F(t') + (t - t')dF/dt\,|_{t=t'}$$

works for any field F(t)

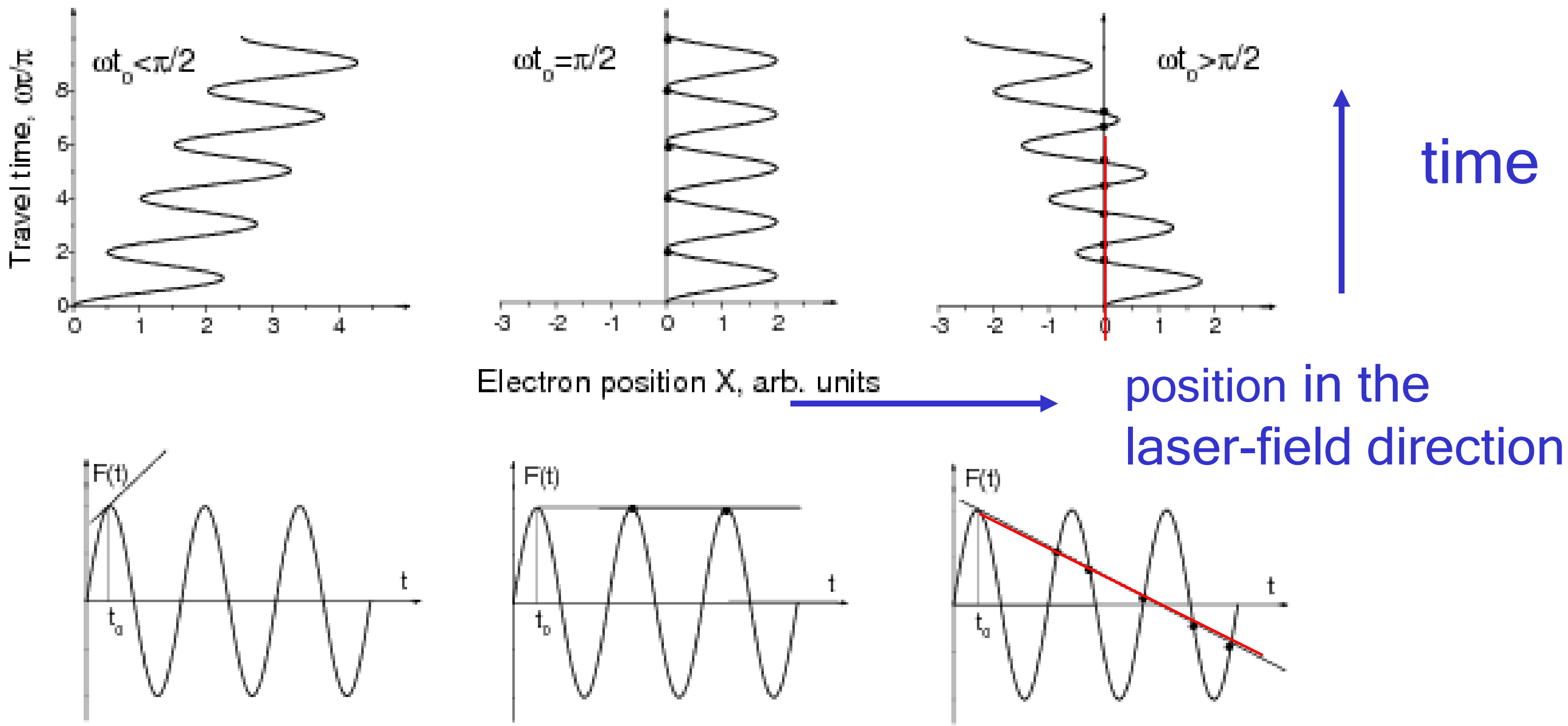


orbit with max return energy = 3.17 U_p

(U_p = pondero-motive energy)

(this yields the the maximal frequency of HHG)

Mechanism of nonsequential double ionization: Recollision of a first-ionized electron with the ion



On a revisit (the first or a later one), the first-ionized electron can free another bound electron (or several electrons) in an inelastic collision

The ponderomotive energy U_p

$$\frac{m}{2}\langle \mathbf{v}(t)^2 \rangle_T = \frac{1}{2m}\langle [\mathbf{p} - e\mathbf{A}(t)]^2 >_T = \frac{\mathbf{p}^2}{2m} + U_p$$

$$U_p = \frac{e^2}{2m}\langle \mathbf{A}(t)^2 \rangle$$ = cycle-averaged wiggling energy of a free electron in a laser field

NB: $U_p \propto \frac{I}{\omega^2}$

Recall classical cutoffs (in energy)

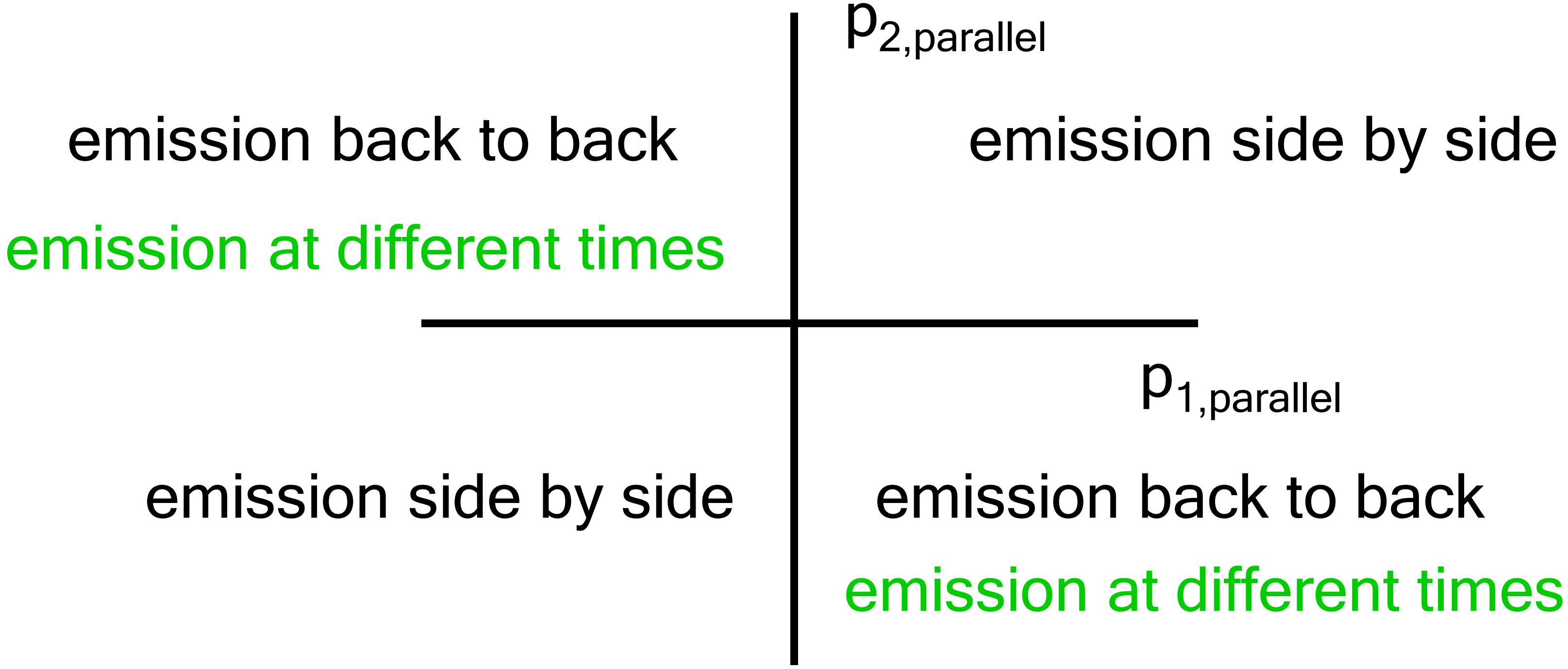
$2\ U_p$ for the drift energy of a „direct“ electron
$10.0\ U_p$ for the drift energy of a backscattered electron
$3.17\ U_p$ for the energy of the returning electron

$2\ U_p^{1/2}$ for the drift momentum of a „direct“ electron

$4.47\ U_p^{1/2}$ for the drift momentum of a backscattered electron

($0.1\ U_p$ for the drift energy of a forwardscattered electron (LES)) end of aside

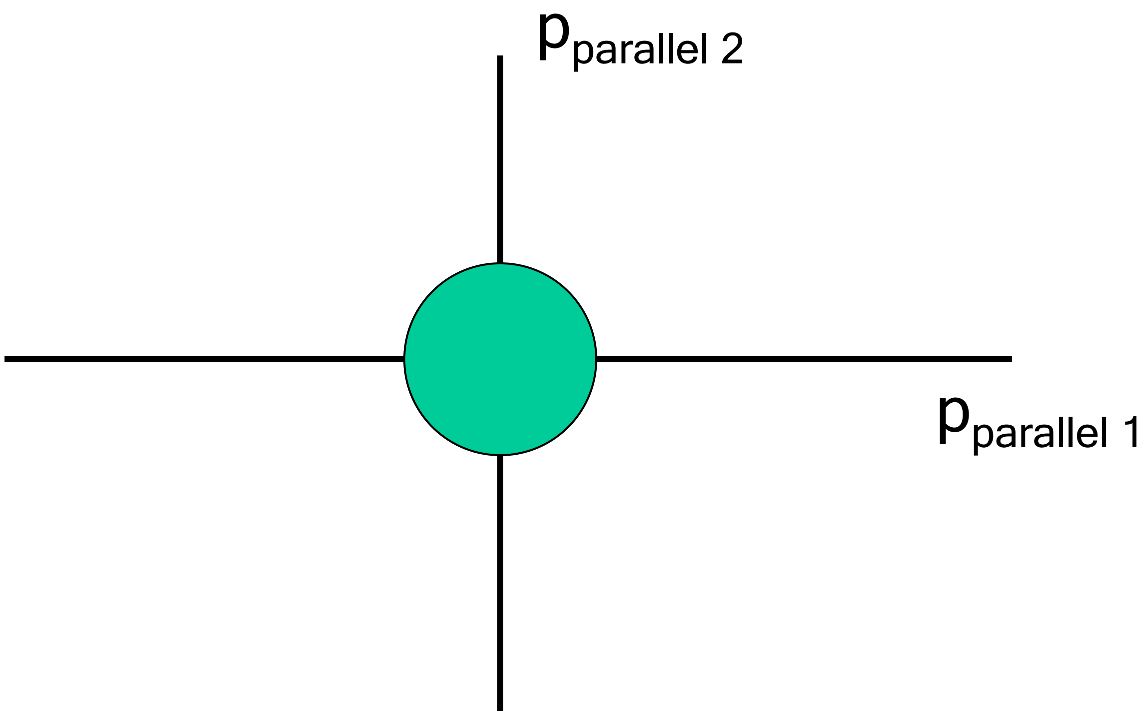
Electron-electron correlation diagram



$p_1 \longleftrightarrow p_2$ symmetry about the diagonal

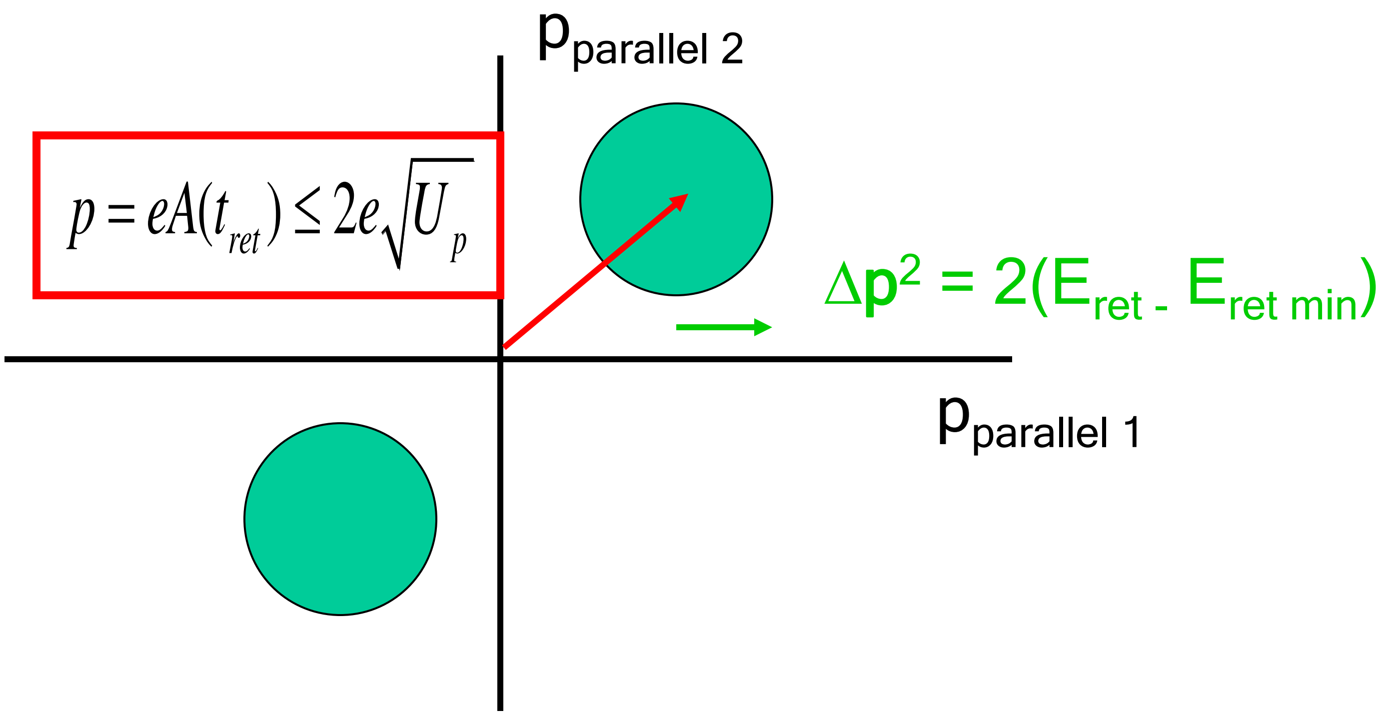
$\mathbf{p} \longleftrightarrow -\mathbf{p}$ (parity), symmetry about the off-diagonal (long pulses only)

Typical electron-electron-momentum correlation diagrams for double ionization



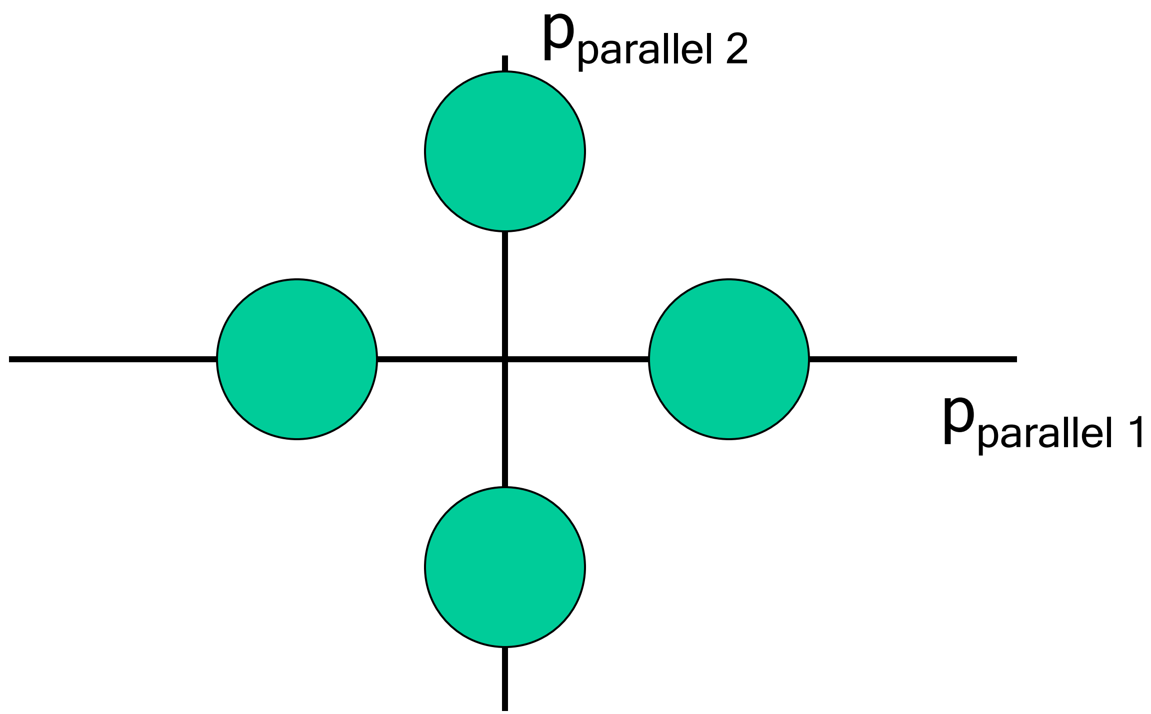
no obvious correlation

Typical electron-electron-momentum correlation diagrams for double ionization



Rescattering impact ionization (RII)

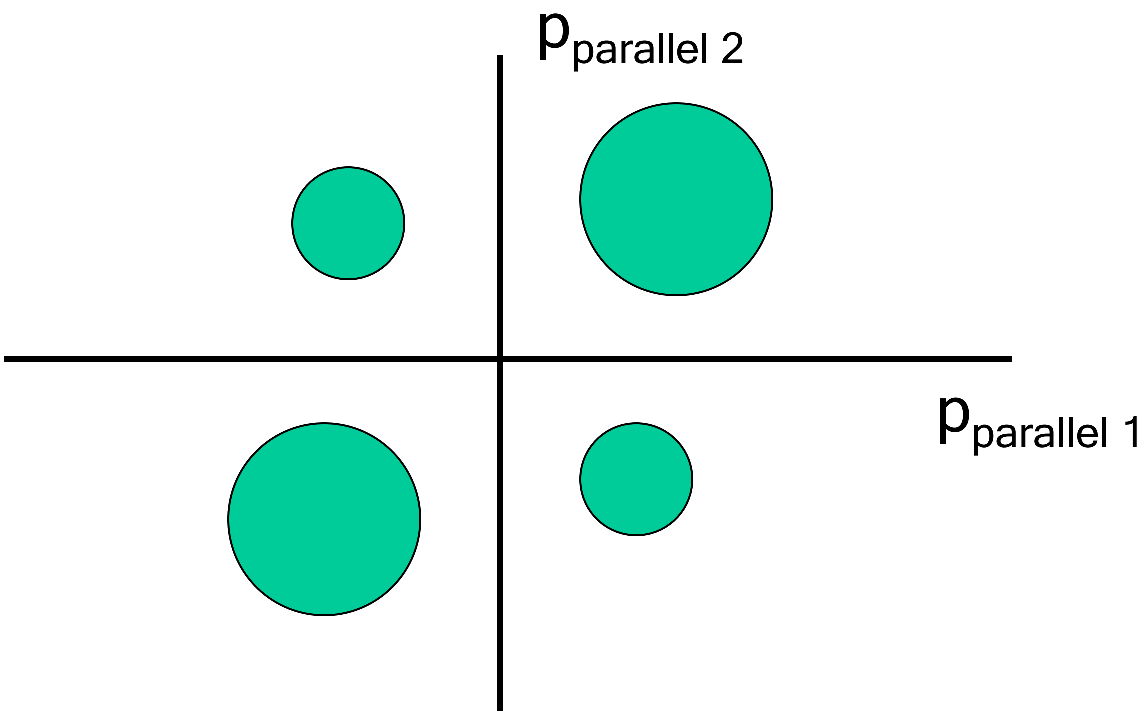
Typical electron-electron-momentum correlation diagrams for double ionization



Rescattering-excitation with subsequent ionization (RESI)

first-liberated electron returns near a zero of the field (max vector potential) and moves the bound electron to an excited state from where it tunnels out at a later time (max field, zero vector potential)

Typical electron-electron-momentum correlation diagrams for double ionization



RII + some delayed second ionization

Theoretical approaches

„All at once“:

numerical solution of the time-dependent Schrödinger equation (TDSE) (limited to helium)

(K. Taylor et al., S. X. Hu, A. Scrinzi, full dimensionality)

(A. Becker et al., center of mass one-dim.)

numerical solution of the completely classical laser-induced escape from a two-electron bound state (time-dependent Newton equations (TDNE) (J. Eberly et al.)

density-functional methods (D. Bauer)

„Step by step“:

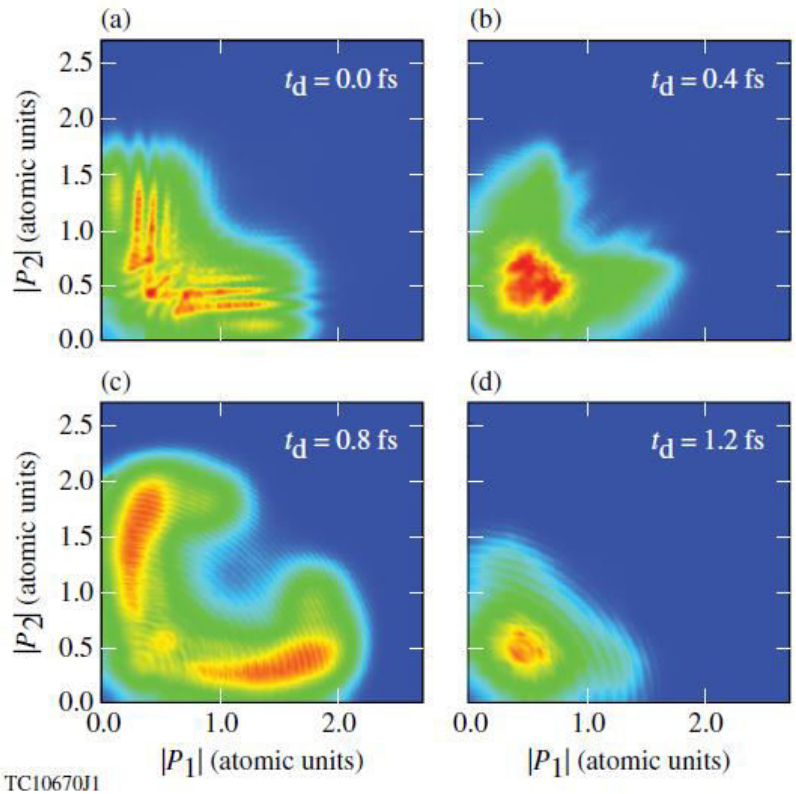
evaluation of the Feyman diagrams considered to be most important

classical-trajectory methods (with quantum-tunneling injection: L.-B. Fu, J. Liu, J. Chen, S.G. Chen, Nam et al.)

escape from a classical excited complex (Sacha, Eckhardt)

Solution of the time-dependent Schrödinger equation

800 nm laser wavelength, 2 electrons in 3 dimensions



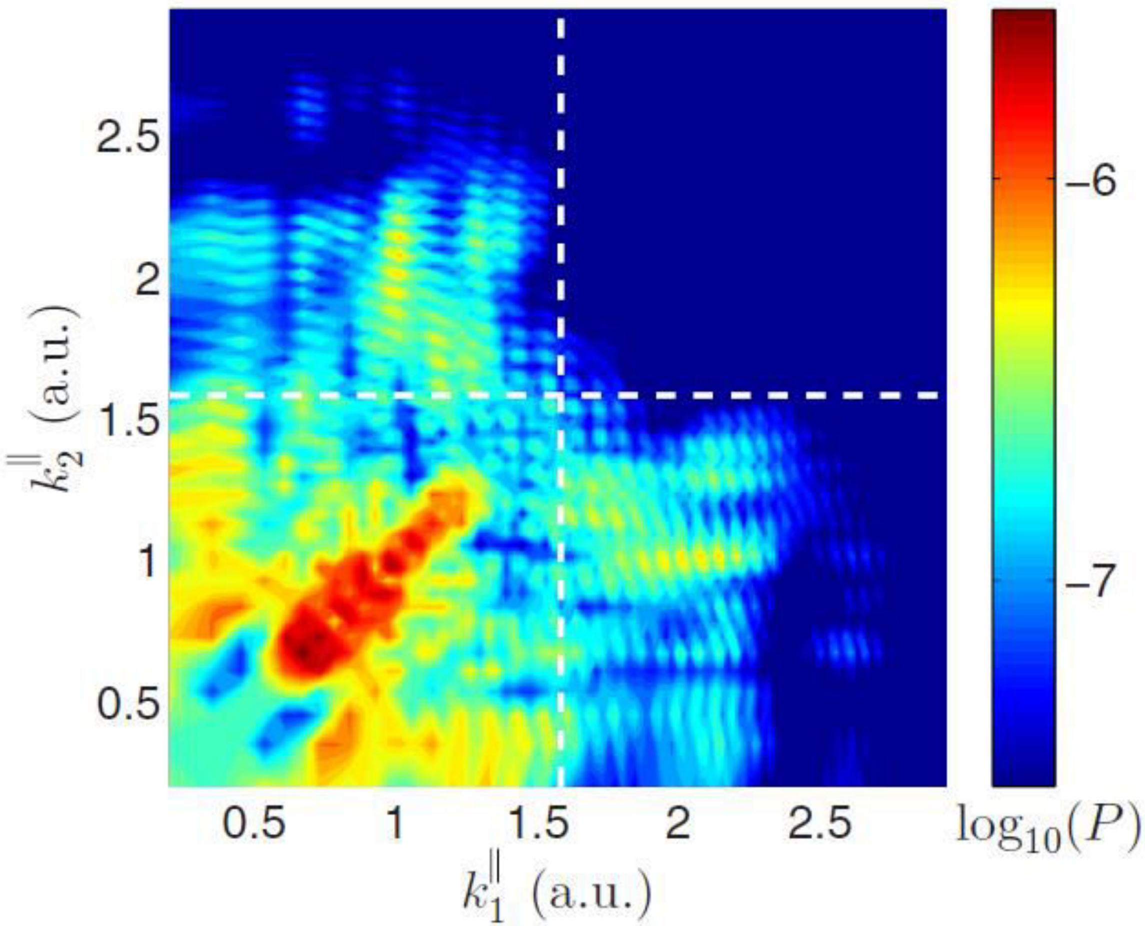
800 nm few-cycle laser pulse (5 fs) plus an attosecond XUV pulse (41 eV) delayed by the time t_d

(10,000 CPUs for 1000 hours)

S. X. Hu, PRL 111, 123003 (2013)

Solution of the time-dependent Schrödinger equation

helium, 800 nm
3 x 10^{14} W/cm^2
(U_p = 19.2 eV)



A. Zielinski, V. P. Majety, A. Scrinzi, Phys. Rev. A 93, 023406 (2016)

Theoretical approaches

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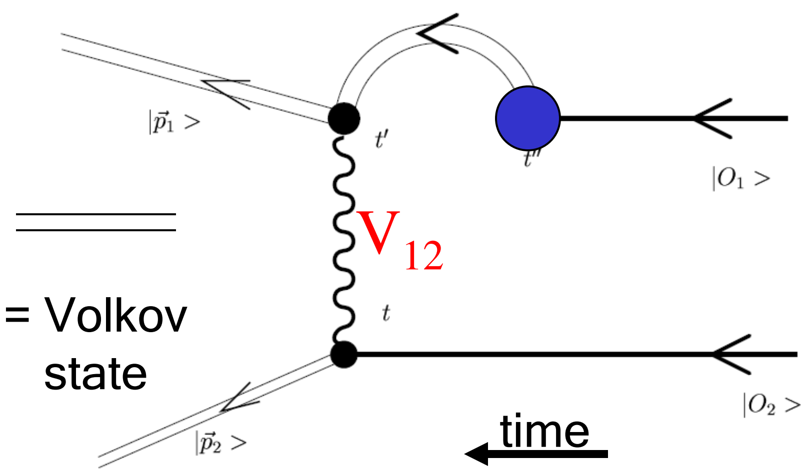
S-matrix element for nonsequential double ionization (rescattering scenario)



V(**r**,**r**') = V_{12} = electron-electron interaction

V(**r**'') = binding potential of the first electron

$$S_{\mathbf{p}_1,\mathbf{p}_2} = \int d^4x d^4x' d^4x'' \psi^{(\mathrm{Vv})}_{\mathbf{p}_1}(x)^* \psi^{(\mathrm{Vv})}_{\mathbf{p}_2}(x')^* \delta(t-t') V(\mathbf{r},\mathbf{r}')$$

$$\times \quad \psi^{(0)}_2(x') G^{(\mathrm{Vv})}(x,x'') V(\mathbf{r}'') \psi^{(0)}_1(x'') + (\mathbf{p}_1 \leftrightarrow \mathbf{p}_2)$$

A. Becker, F.H.M. Faisal, PRL 84, 3546 (2000); R. Kopold, W. Becker, H. Rottke, W. Sandner, PRL 85, 3781 (2000); S.V. Popruzhenko, S. P Goreslavski, JPB 34, L230 (2001); C. Faria, H. Schomerus, X. Liu, W. Becker, PRA 69, 043405 (2004)

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 $\mathbf{A}(t)(\mathbf{E} = -\partial_t \mathbf{A})$

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

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

Evaluation of the double-ionization amplitude

Volkov propagator $U^{(V)}(t,t') = \int d^3\mathbf{k} |\psi_{\mathbf{k}}^{(V)}(t)\rangle\langle\psi_{\mathbf{k}}^{(V)}(t')|$

$$M = -\int_{-\infty}^{\infty} dt \int_{-\infty}^{t} dt' \int d^3\mathbf{k} V_{\mathbf{pk}} V_{\mathbf{k}0} \exp[iS_{\mathbf{p}}(t,t',\mathbf{k})],$$

form factors

The integrands are strongly oscillating, hence the integrals get their most important contributions from the stationary points of the phase

$$S_{\mathbf{p}}(t,t',\mathbf{k}) = -\frac{1}{2}\left[\sum_{n=1}^{2}\int_{t}^{\infty} d\tau[\mathbf{p}_n + \mathbf{A}(\tau)]^2 + \int_{t'}^{t} d\tau[\mathbf{k} + \mathbf{A}(\tau)]^2\right] + |E_{01}|t' + |E_{02}|t.$$

Stationary-phase saddle-point equations:
retrieving the simple-man model with rescattering

$$[\mathbf{k}+\mathbf{A}(t')]^2 = -2|E_{01}|,$$

ionization of the first electron at time t‘

$$\sum_{n=1}^{2}[\mathbf{p}_n+\mathbf{A}(t)]^2 = [\mathbf{k}+\mathbf{A}(t)]^2 - 2|E_{02}|$$

the returning first electron kicks out the second electron

$$\int_{t'}^{t} d\tau[\mathbf{k}+\mathbf{A}(\tau)] = 0$$

electron freed at time t‘ returns at time t

solutions (t_s,t_s‘) = (rescattering time, ionization time) (s = 1,2,...) are complex

$$M^{(\mathrm{SPA})} = \sum_s A_s \exp(iS_s)$$

$$S_s = S_{\mathbf{p}}(t_s, t_s', \mathbf{k}_s) \qquad A_s = (2\pi i)^{5/2}\frac{V_{\mathbf{p}\mathbf{k}_s}V_{\mathbf{k}_s 0}}{\sqrt{\det S_{\mathbf{p}}''(t,t',\mathbf{k})|_s}}$$

(Effective) electron-electron interactions

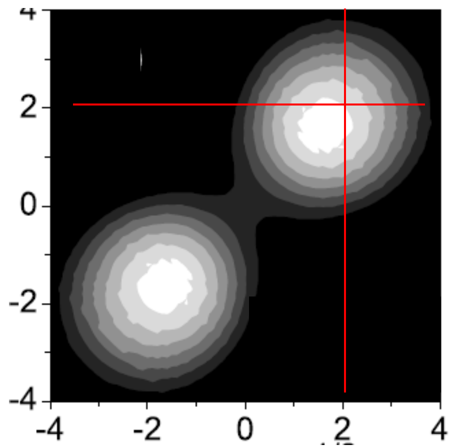
$V(\mathbf{r}_1,\mathbf{r}_2) =$

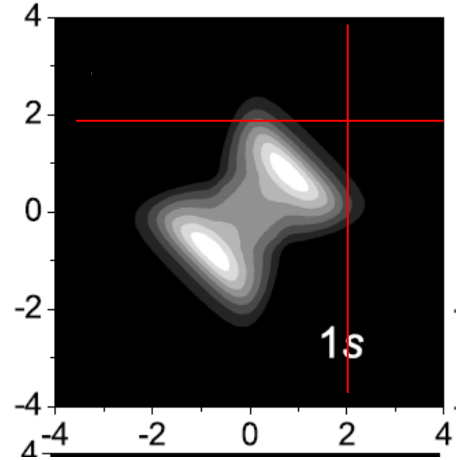
$\|\mathbf{r}_1 - \mathbf{r}_2\|^{-1}$	e-e Coulomb ⇐
$\delta(\mathbf{r}_1 - \mathbf{r}_2)\ \delta(\mathbf{r}_2)$	three-body contact ⇐
$\|\mathbf{r}_1 - \mathbf{r}_2\|^{-1}\ \delta(\mathbf{r}_2)$	Coulomb with nuclear recoil
$\delta(\mathbf{r}_1 - \mathbf{r}_2)$	e-e contact

In principle, the total effective interaction could be the sum of several of the above

e-e correlation distributions generated by different effective interactions

$\delta(\mathbf{r}_1 - \mathbf{r}_2)\delta(\mathbf{r}_2)$

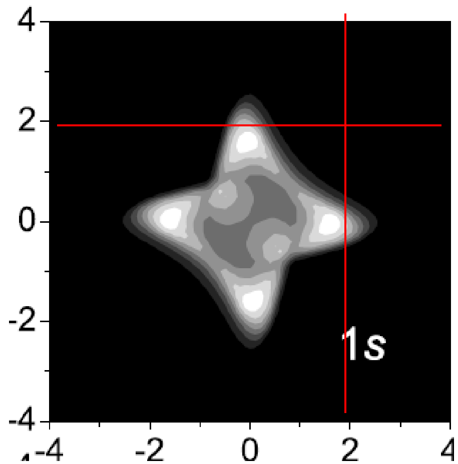


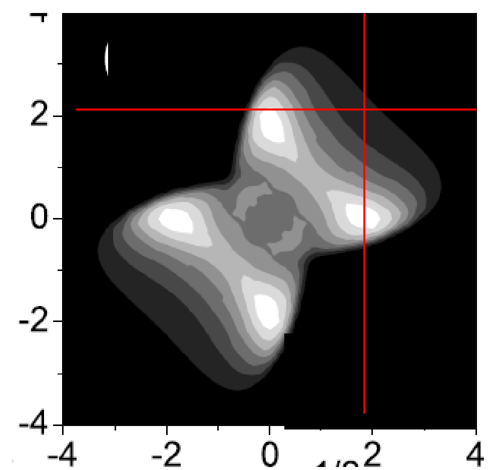


$\delta(\mathbf{r}_1 - \mathbf{r}_2)$

plots of $p_{2||}$ vs. $p_{1||}$ scaled by $U_p^{1/2}$

$|\mathbf{r}_1 - \mathbf{r}_2|^{-1}$





$|\mathbf{r}_1 - \mathbf{r}_2|^{-1}\,\delta(\mathbf{r}_2)$

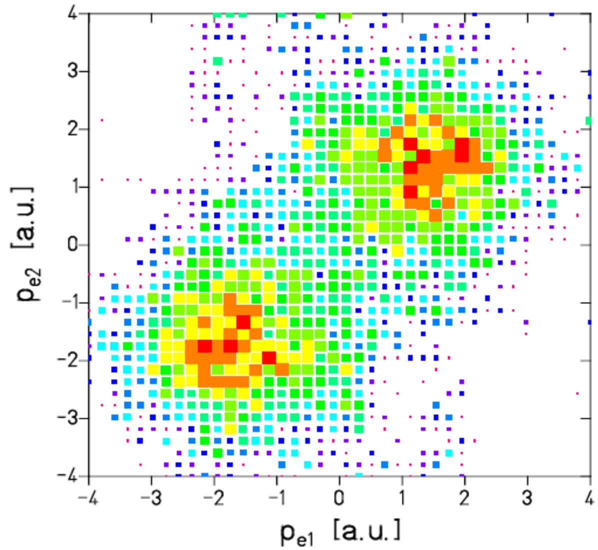
Ti:Sa, neon, 8 x 10^{14} Wcm^{-2}

red lines: $p = 2\,U_p^{1/2}$

Comparison with neon data

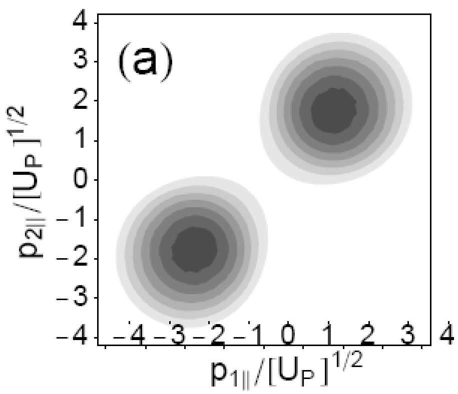
10^{15} Wcm^{-2}

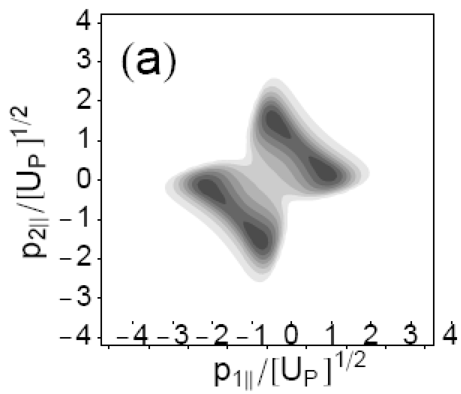
Rottke, Moshammer et al. (2003)



$\mathcal{S}$-matrix theory:

V_{12} = 3-body contact

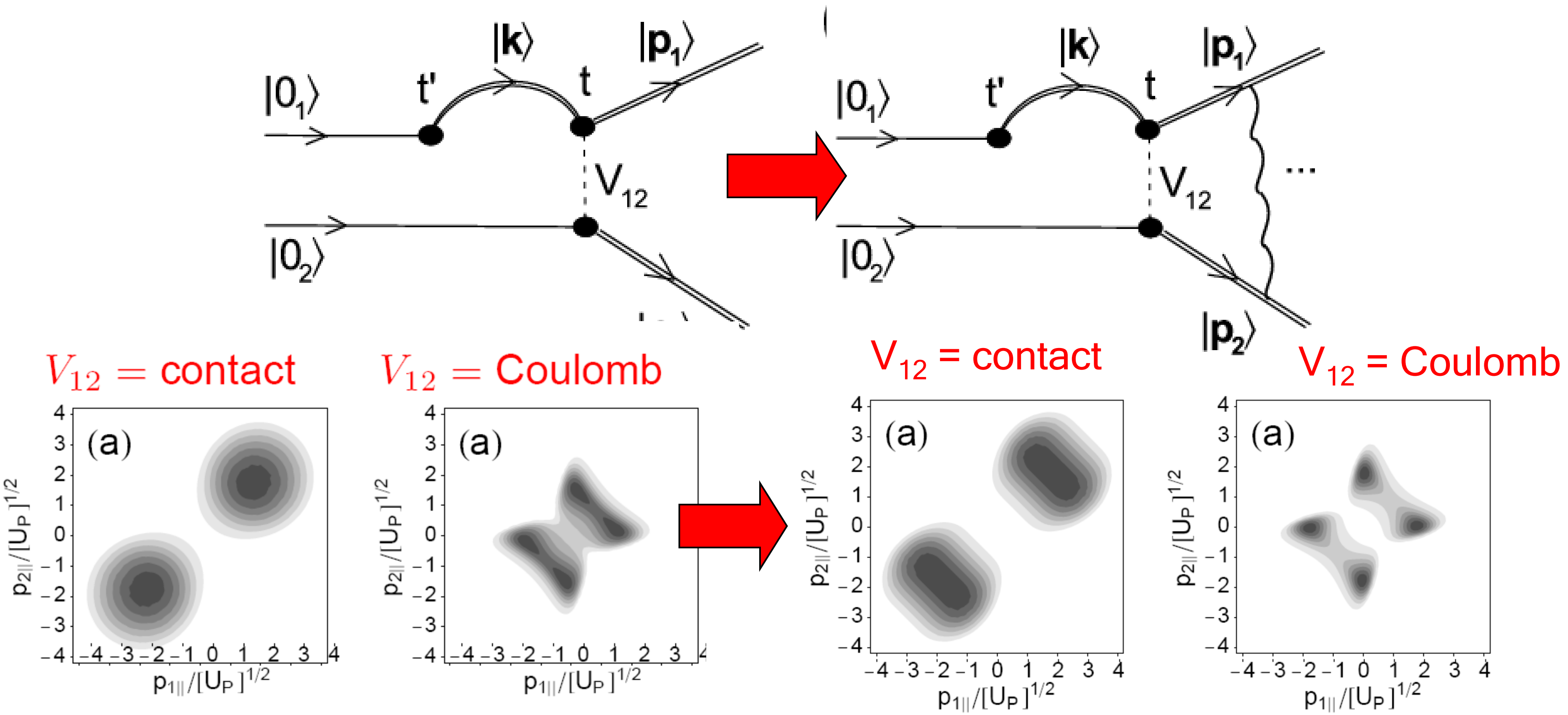




V_{12} = Coulomb

Coulomb interaction does not reproduce the data

Introducing the Coulomb repulsion between the two electrons in the final state ...



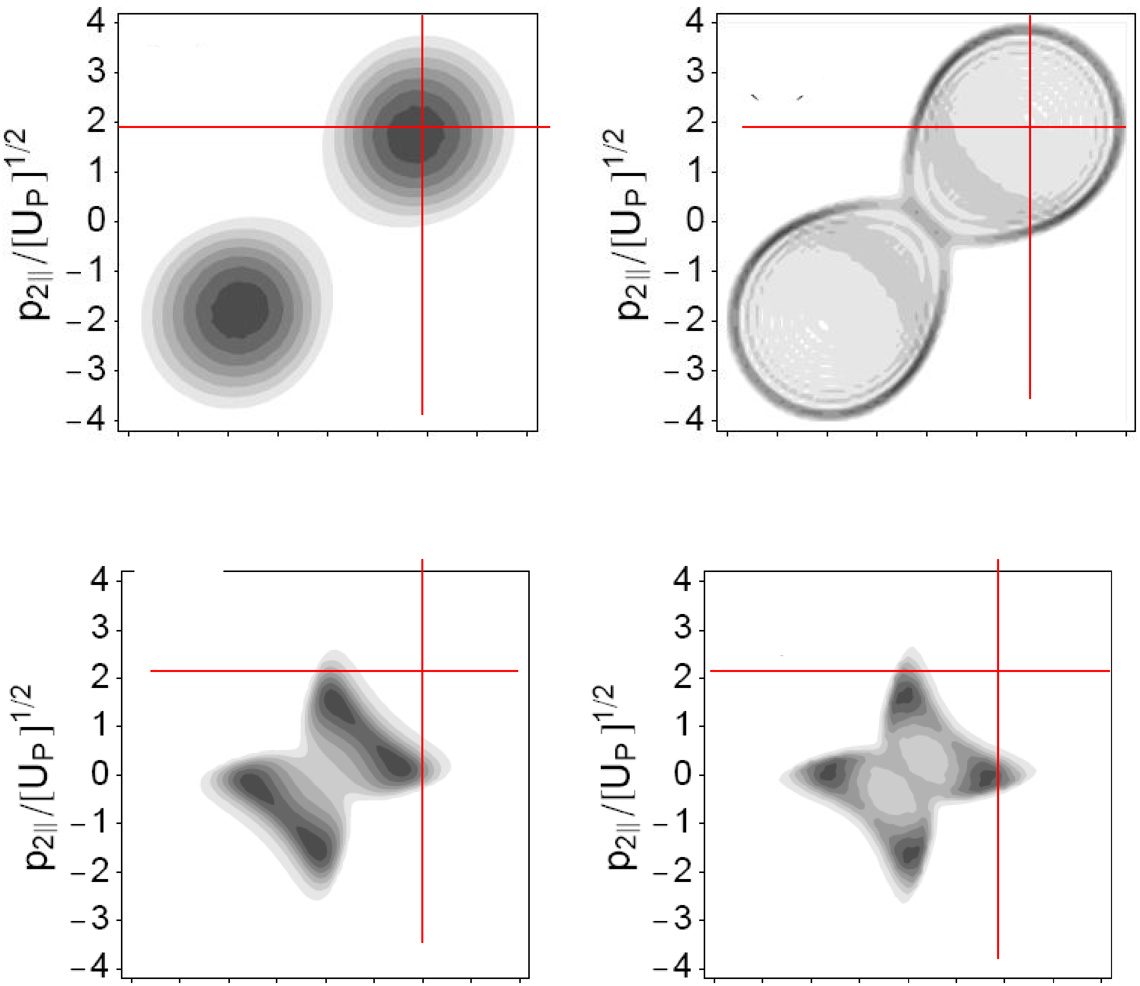
... does not improve the agreement with the data

Restricting the transverse momenta

p_{1T}, p_{2T} integrated | $|p_{iT}| < 0.1\ U_p^{1/2}$

3-body contact

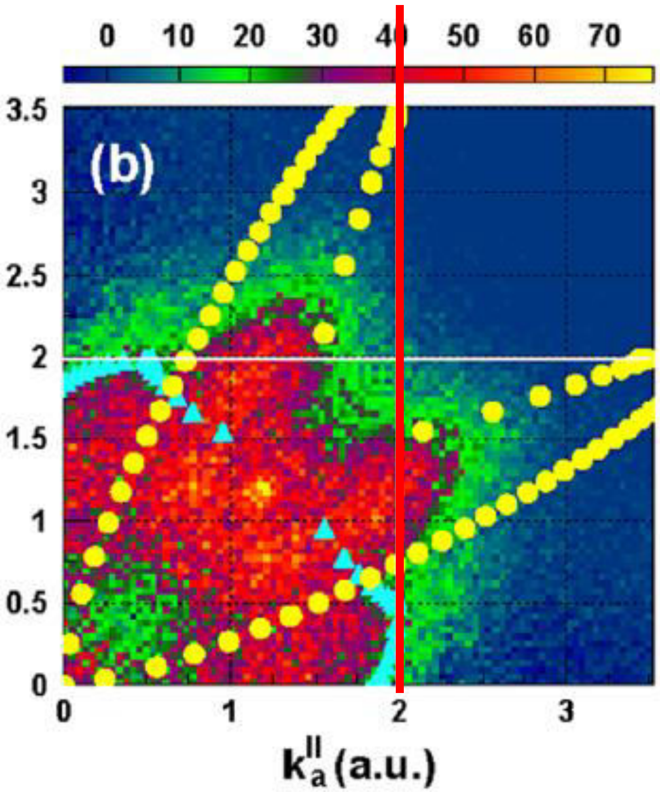
Coulomb



neon
$\omega = 0.551$ a.u.
$U_p = 1.2$ a.u.

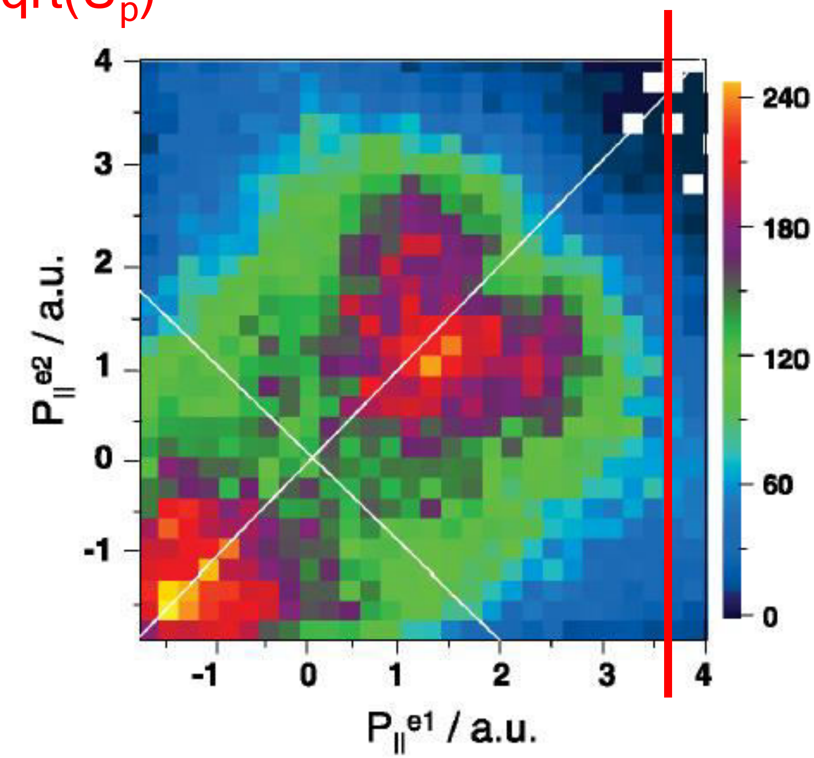
Evidence of Coulomb repulsion of the liberated electrons

$p = 2\ \mathrm{sqrt}(U_p)$



helium, 40 fs, 800 nm
4.5 x 10^{14} W/cm^2 (U_p = 27 eV)

A. Staudte et al., PRL 99, 263002 (2007)



helium, 25 fs, 800 nm
1.5 x10^{15} W/cm^2 (U_p = 89 eV)

A. Rudenko et al., PRL 99, 263003 (2007)

A classical model

Injection of the electron into the continuum at time t‘
at the rate R(t‘)
The rest is classical:
The electron returns at time t=t(t‘) with energy $E_{ret}(t)$
Energy conservation in the ensuing recollision

$$|S_{\mathbf{p}_1,\mathbf{p}_2 \mathrm{class}}|^2 \sim \int dt' R(t') \delta \left(E_{\mathrm{ret}}(t) - \frac{1}{2m}[\mathbf{p}_1 - e\mathbf{A}(t)]^2 - \frac{1}{2m}[\mathbf{p}_2 - e\mathbf{A}(t)]^2 - |E_{02}| \right)$$

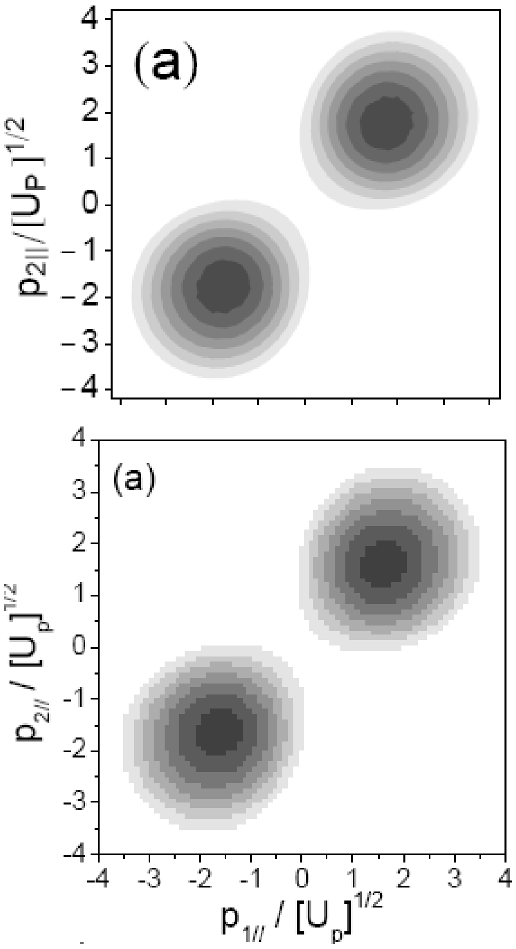
All phase space, no specific dynamics

Cf. statistical models in chemistry, nuclear, and particle physics

Comparison: quantum vs classical model

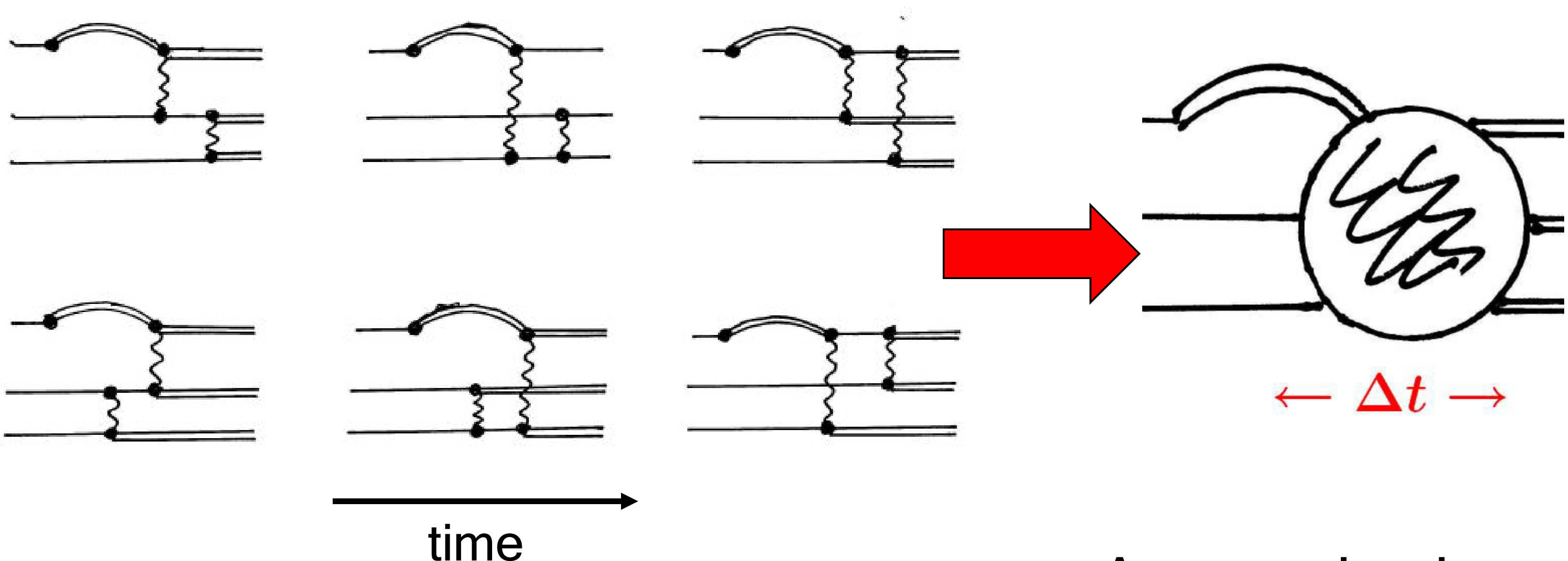
quantum

classical



sufficiently high above threshold,
the classical model works as well
as the full quantum model

Nonsequential triple ionization



Assume it takes a time Δt for the electrons to „thermalize“

NB: one internal propagator
→ 4 additional integrations

Nonsequential N-fold ionization via a thermalized N-electron ensemble

fully differential N-electron distribution:

$$F(\mathbf{p}_1, \mathbf{p}_2, \ldots, \mathbf{p}_N) = \int dt' R(t')$$

$$\times \delta \left(E_{\text{ret}}(t) - E_0^{(N)} - (1/2m) \sum_{n=1}^{N} [\mathbf{p}_n - e\mathbf{A}(t + \Delta t)]^2 \right)$$

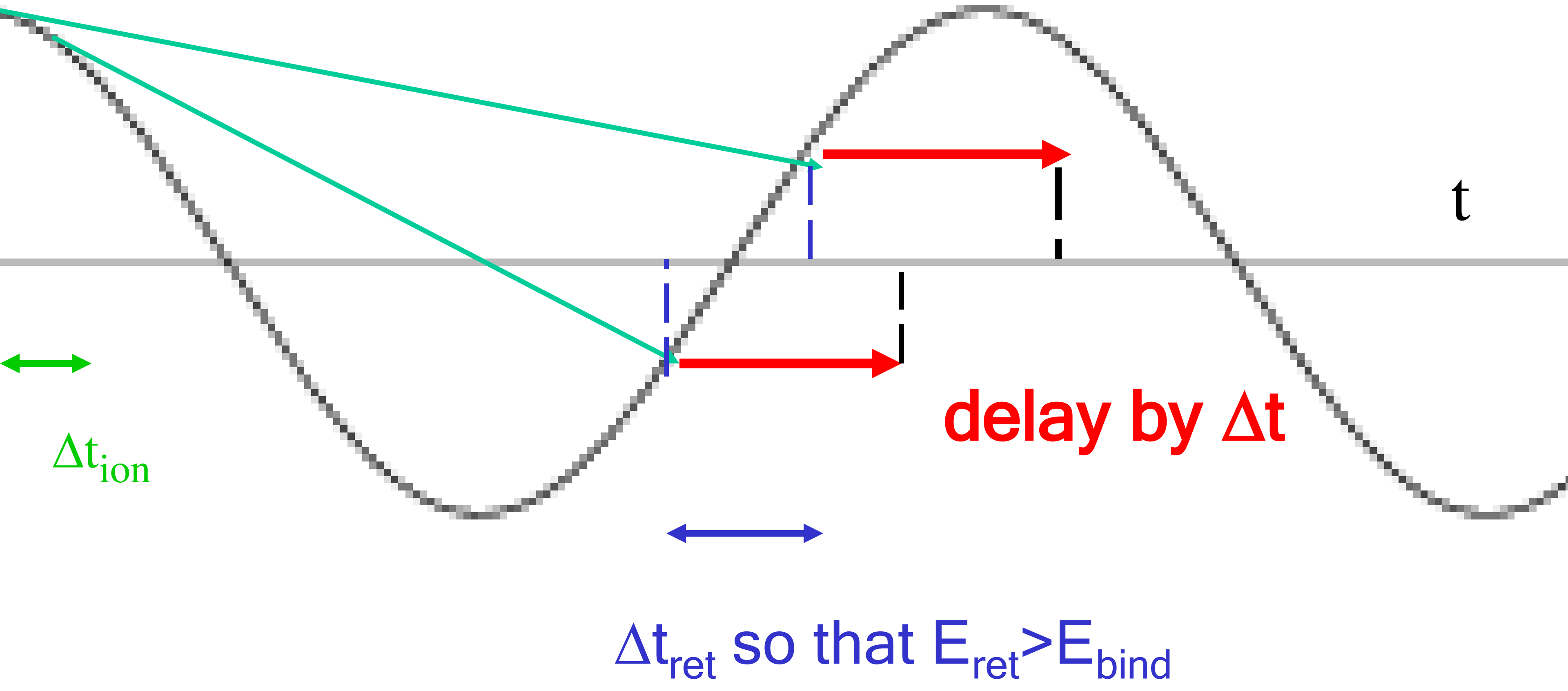
$= m\mathbf{v}(t+\Delta t)$

Ion-momentum distribution:

$$F_{\text{ion}}(\mathbf{P}) \equiv \int \prod_{n=1}^{N} d^3\mathbf{p}_n \delta \left(\mathbf{P} + \sum_{n=1}^{N} \mathbf{p}_n \right) F(\mathbf{p}_1, \mathbf{p}_2, \ldots, \mathbf{p}_N)$$

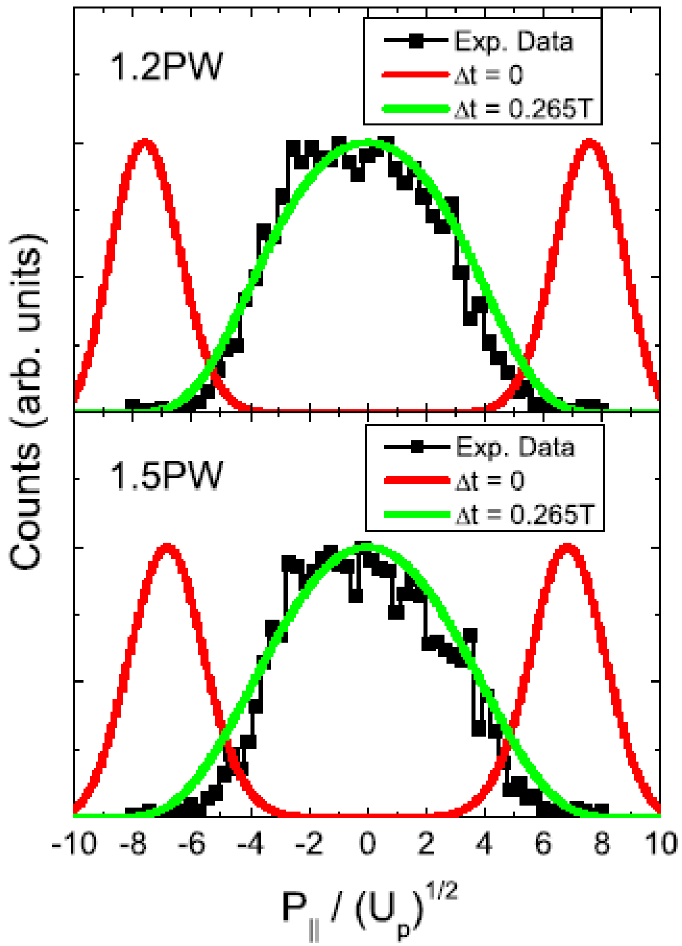
integrate over unobserved momentum components

Delay between recollision and final ionization



Higher sensitivity to the value of Δt by reducing Δt_{ret}

Nonsequential quadruple ionization of argon



$I = 1.2 \times 10^{15}$ W/cm^2
$U_p = 76.3$ eV
$\Sigma I_p = 128$ eV

$I = 1.5 \times 10^{15}$ W/cm^2
$U_p = 95.3$ eV
$\Sigma I_p = 128$ eV

Data: K. Zrost, A. Rudenko, Th. Ergler, B. Feuerstein, V. L. B. de Jesus, C. D. Schröter, R. Moshammer, and J. Ullrich, J. Phys. B 39, S371 (2007)
Theory: X. Liu, C. Figueira de Morisson Faria, W. Becker, New J. Phys. 10, 025010 (2008)

Theoretical approaches

„All at once“:

numerical solution of the time-dependent Schrödinger equation (TDSE) (limited to helium)

(K. Taylor et al., S. X. Hu, A. Scrinzi, full dimensionality)

(A. Becker et al., center of mass one-dim.)

numerical solution of the completely classical laser-induced escape from a two-electron bound state (time-dependent Newton equations (TDNE) (J. Eberly et al.)

density-functional methods (D. Bauer)

„Step by step“:

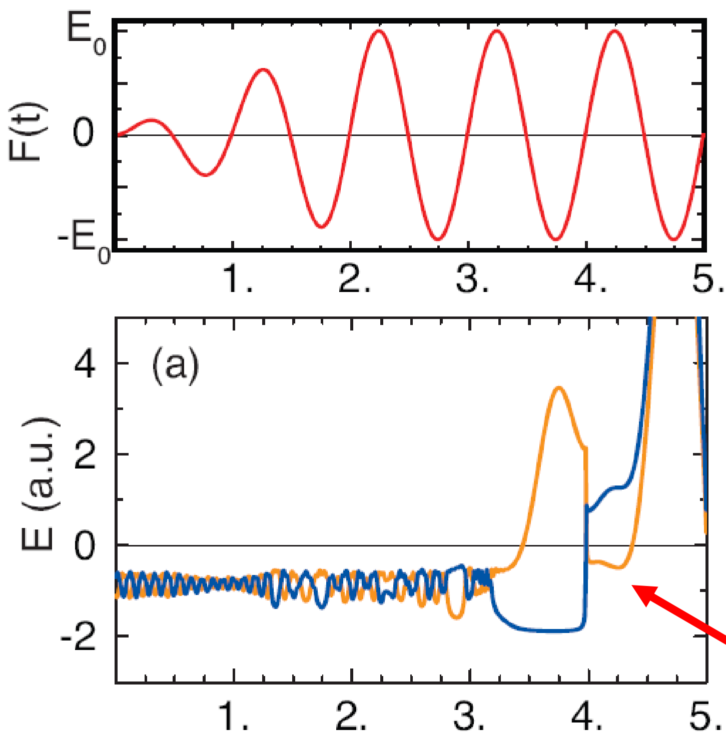
evaluation of the Feyman diagrams considered to be most important

classical-trajectory methods (with quantum-tunneling injection: L.-B. Fu, J. Liu, J. Chen, S.G. Chen, Nam et al.)

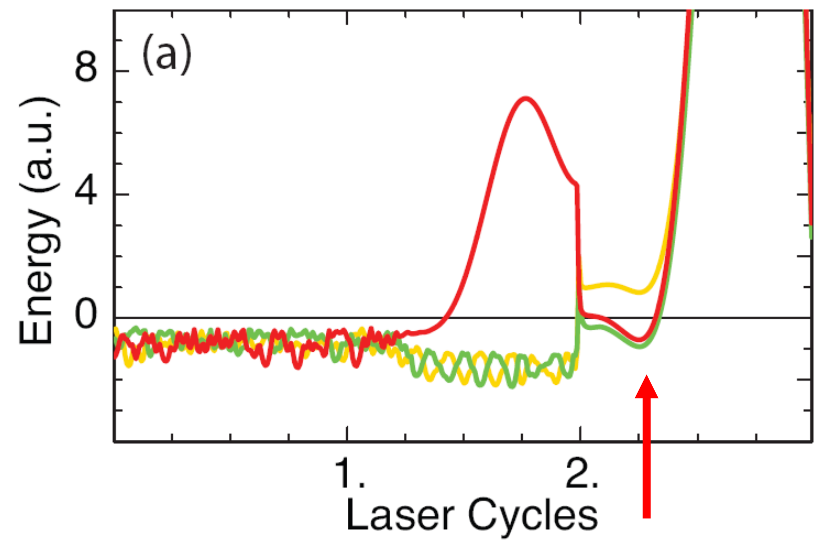
escape from a classical excited complex (Sacha, Eckhardt)

Completely classical double-ionization trajectories

NSDI



NSTI

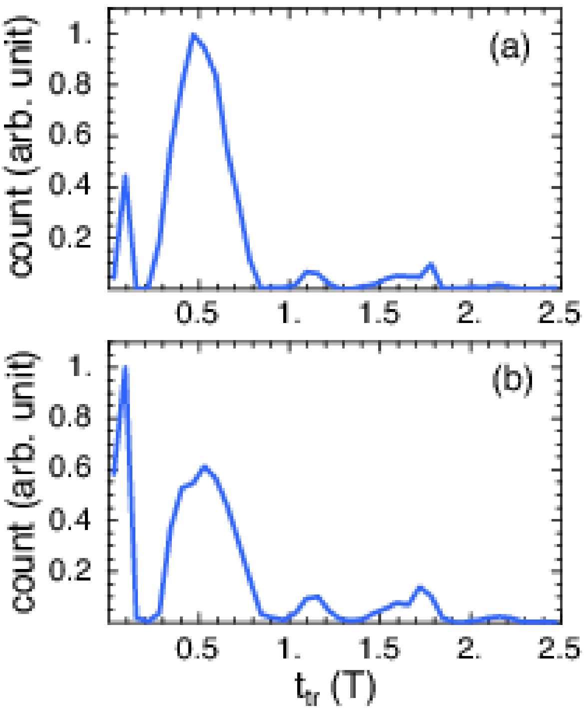


thermalization time?

can identify single-ionization time, recollision time, and double ionization time

P.J. Ho, X. Liu, WB (submitted); P.J. Ho, J.H. Eberly, Opt. Express 15, 1845 (2007)

How „real" is the „travel time"?



V_{12} = Coulomb

V_{12} = Yukawa

travel-time distribution

travel time = t_{si} - t_{recol}

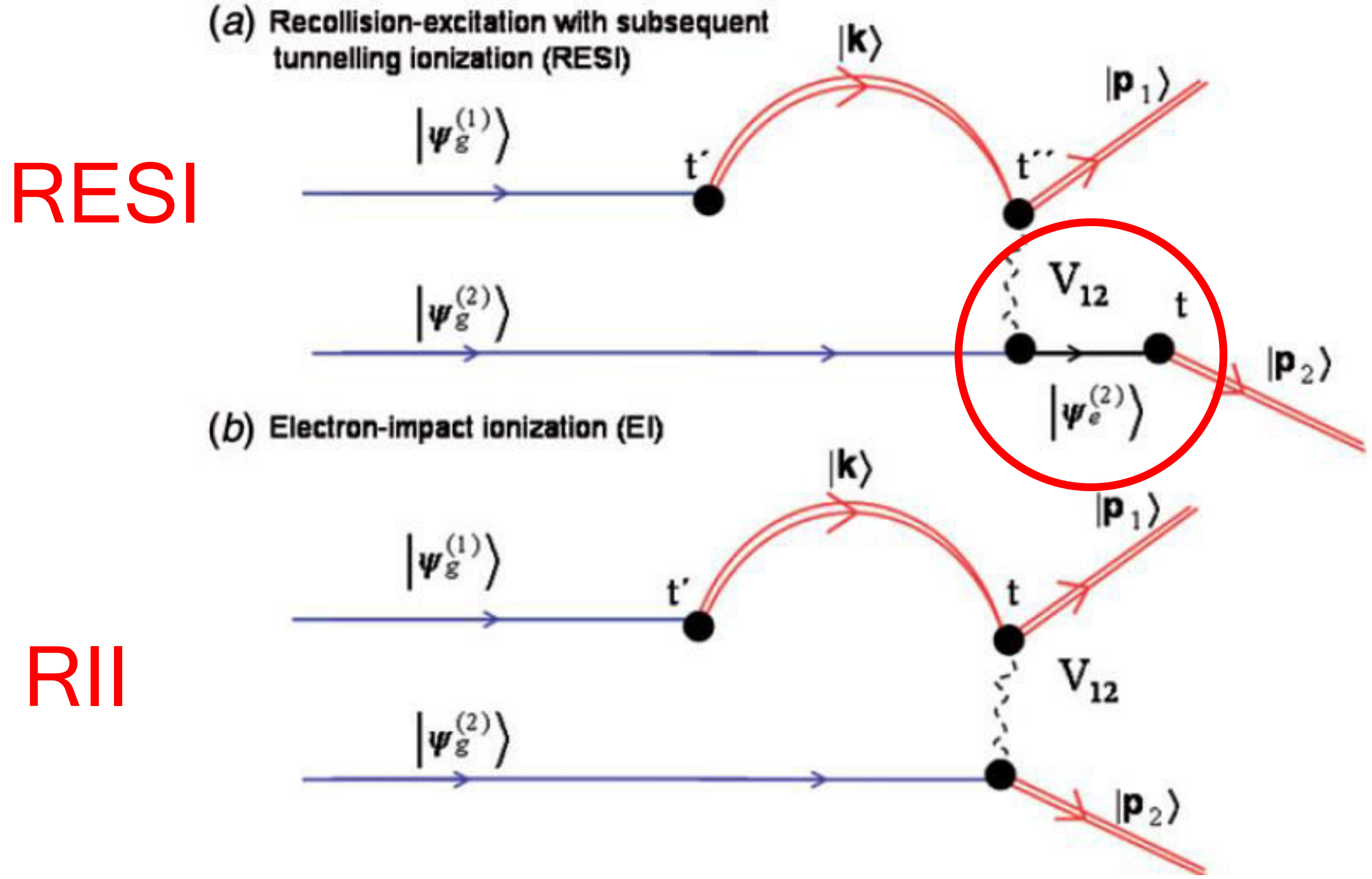
t_{si} = time when E_{kin} > interaction energies

t_{recol} = time of closest approach between the two electrons

peaks agree with the simple-man model

Phay J. Ho, X. Liu, WB (unpublished)

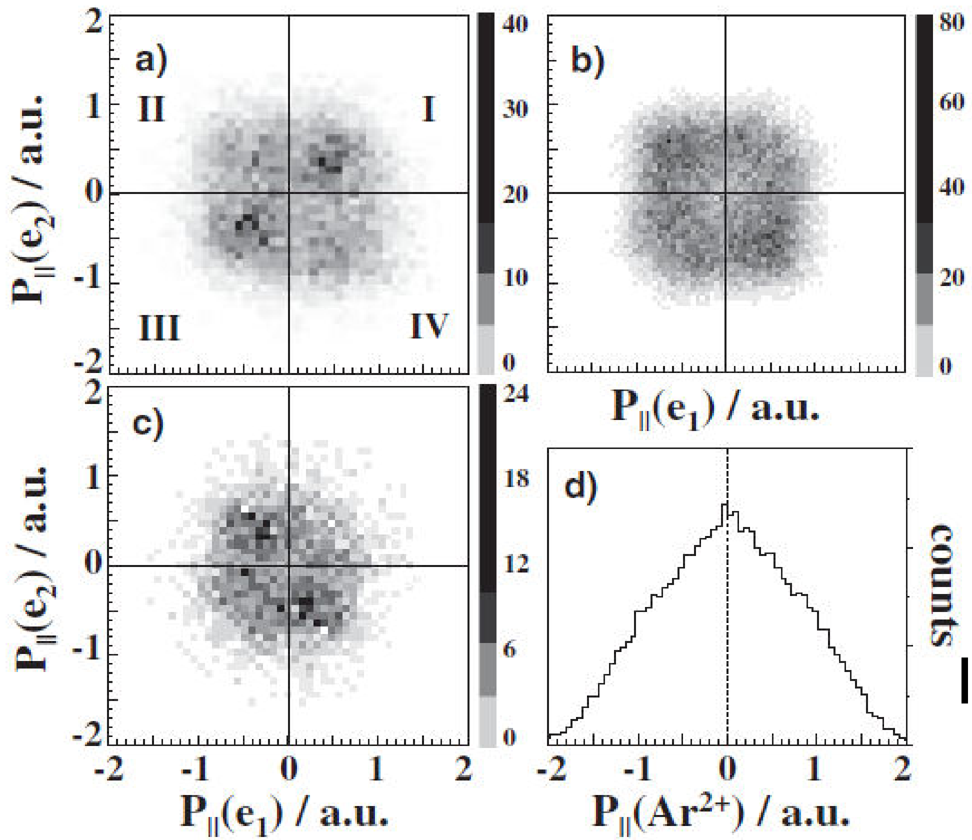
Feynman diagrams of Rescattering-impact ionization (RII) vs Recollision-excitation with subsequent ionization (RESI)



from X. Liu and C. Faria, J. Mod. Opt. 58, 1076 (2011)

Going below the threshold: correlated --> anticorrelated

9 x 10^{13}
(U_p = 5.7 eV)



7 x 10^{13}
(U_p = 4.4 eV)

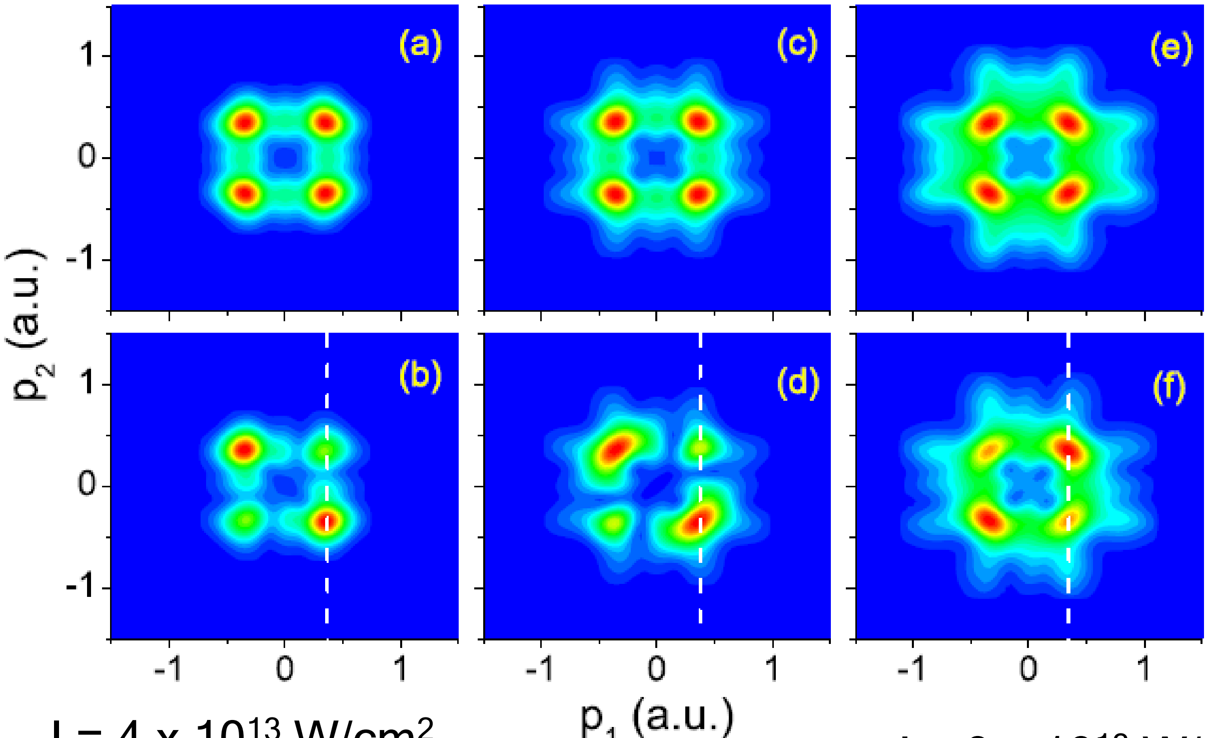
4 x 10^{13} W/cm^2
(U_p = 2.5 eV)

argon, 800 nm
$I_p(Ar^+)$ = 27.6 eV

Y. Liu, S. Tschuch, A. Rudenko, M. Dürr, M. Siegel, U. Morgner, Q. Gong, R. Moshammer, and J. Ullrich, PRL 101, 053001 (2008)

Interference of paths via different excited states of argon$^+$

Configs $3s3p^6$ (I_p = 14.1 eV), $3s^23p^4(^3P)3d$ (I_p = 11.2 eV), $3s^23p^4(^3P)4d$ (I_p = 4.9 eV)



without interference

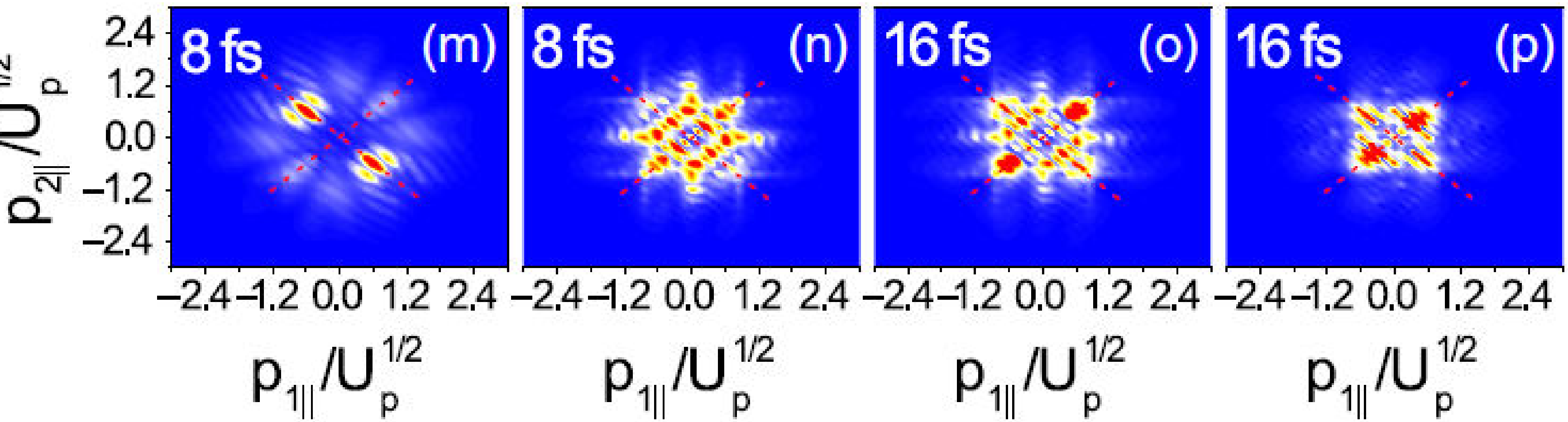
with interference

$I = 4 \times 10^{13}$ W/cm^2 $I = 7 \times 10^{13}$ W/cm^2 $I = 9 \times 10^{13}$ W/cm^2

With increasing intensity, the distribution moves from the 2/4 (back to back) to the 1/3 (side by side) quadrants

XL. Hao, J. Chen, WD. Li, B. Wang, X. Wang, W. Becker, PRL 112, 073002 (2014)

More results (same theory, more channels, more phases)



0.8×10^{14} W/cm^2 $\quad$ 1.2×10^{14} W/cm^2 $\quad$ 1.4×10^{14} W/cm^2

A. S. Maxwell and C. Figueira de Morisson Faria, PRL 116, 143001 (2016);
PRA 92, 023421 (2015)

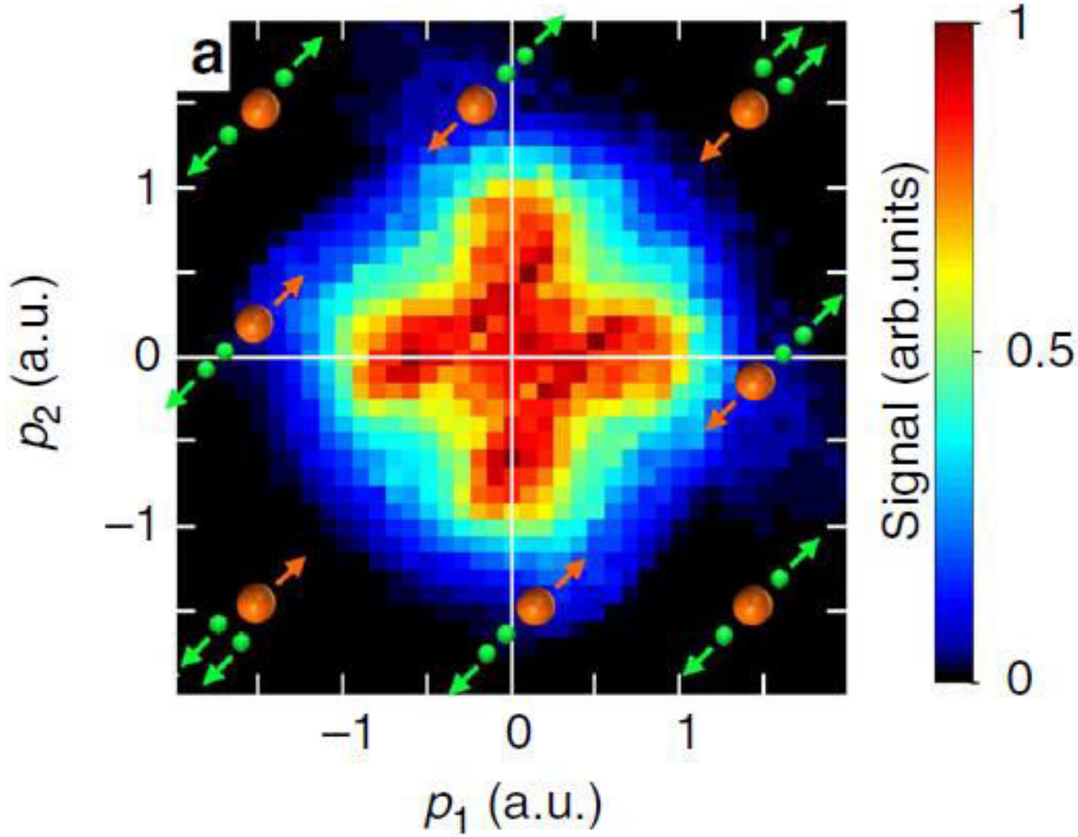
Essentially, the first quantum effect in NSDI

Cleanest realization of the RESI mechanism

argon, 800 nm
3 x 10^{14} W/cm^2

near-single-cycle
4 fs laser pulse

carrier-envelope
phase averaged

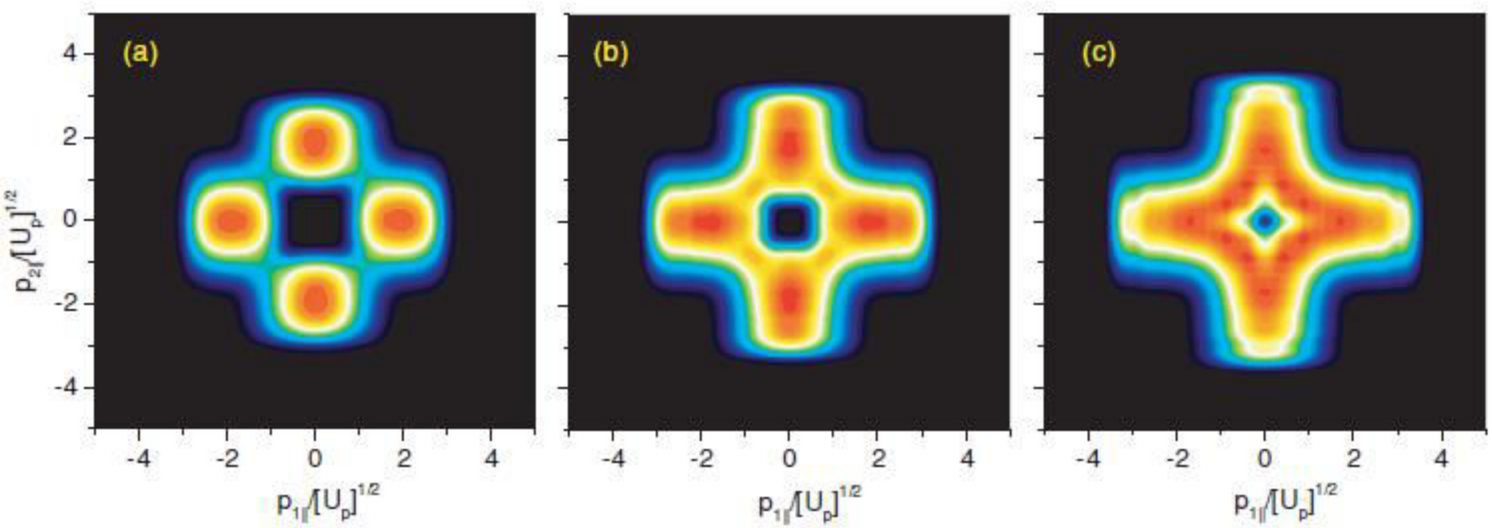


The pulse is so short as to allow for just one recollision

B. Bergues et al., Nat. Commun. 3, 813 (2012)

RESI-mechanism S-matrix calculation

helium, 800 nm, Ip(He^+) = 54.4 eV, Ip(He^{+*}) = 13.6 eV



I = 2.2 x 10^{14} W/cm^2 (U_p = 14 eV)

I = 2.5 x 10^{14} W/cm^2 (U_p = 16 eV)

I = 3.0 x 10^{14} W/cm^2 (U_p = 19 eV)

Just above or above the RESI threshold

Just above the RII threshold

T. Sharaan, M. T. Nygren, C. Figueira de Morisson Faria, PRA 81, 063413 (2010)

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escape from a classical excited complex (Sacha, Eckhardt)

Conclusions and outlook

High enough above threshold, NSDI and NSMI appear to be largely classical

Many details are poorly understood

Are there qualitative quantum effects?

Methods for nonsequential multiple ionization?

Some reviews

A. Becker, R. Dörner, and R. Moshammer,
J. Phys. B 38, S753 (2005)
W. Becker and H. Rottke, Contemp. Phys. 49, 199 (2008)
C. Figueira de Morisson Faria and X. Liu
J. Mod. Opt. 58, 1076 (2011)
W. Becker, X. Liu, P. J. Ho, and J. H. Eberly
Rev. Mod. Phys. 84, 1011 (2012)
B. Bergues, M. Kübel, N. G. Kling, C. Burger, and M. F. Kling
IEEE J. Sel. Top. Quantum Electronics 21, 8701009 (2015)