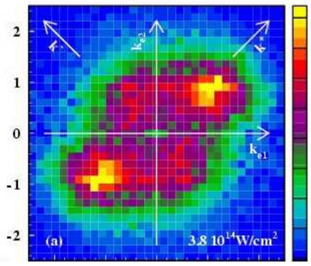
Classical and quantum effects in strong field multiple ionization

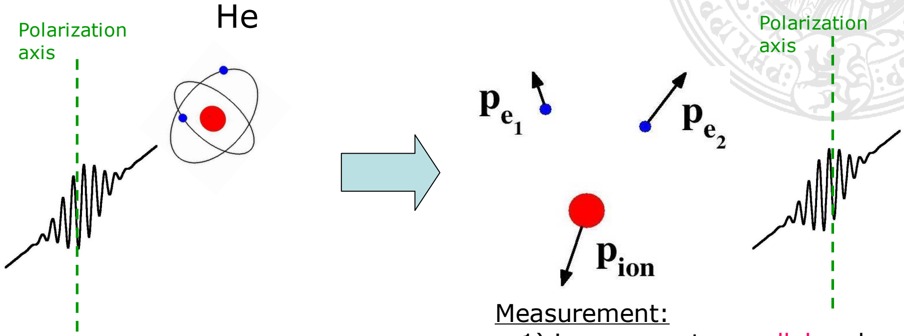


Krzysztof Sacha
Jakub Zakrzewski

Jakub S. Prauzner-Bechcicki
Bruno Eckhardt

Instytut Fizyki, Universytet Jagiellonski, Krakow
Fachbereich Physik, Philipps-Universität Marburg

Experimental situation



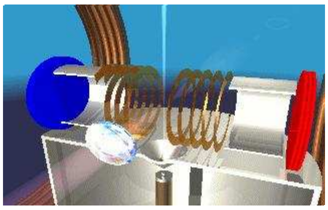
Short laser pulses linearly polarized
(220 fs, 800 nm, 10^{15} W/cm^2)

Measurement:

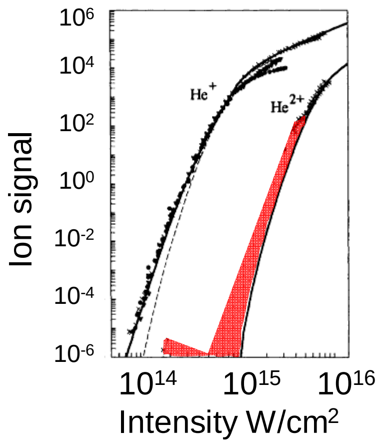
1) ion momenta parallel and perpendicular to the field axis,
2) electron momenta

$$\vec{p}_{ion} = -(\vec{p}_{e_1} + \vec{p}_{e_2})$$

The experiment



Lasers: H. Giessen
(Marburg/Stuttgart)
Detectors: R. Dörner
(Frankfurt)



Kulander et al, PRL 73 (1994) 1227

Rescattering model

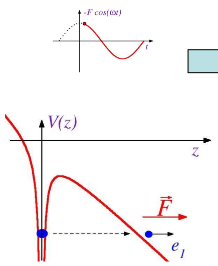
$$H=\frac{\vec{p}_1^{\,2}}{2}+\frac{\vec{p}_2^{\,2}}{2}-\frac{2}{r_1}-\frac{2}{r_2}+\frac{1}{|\vec{r}_1-\vec{r}_2|}+Ff(t)(z_1+z_2)\cos(\omega t)$$

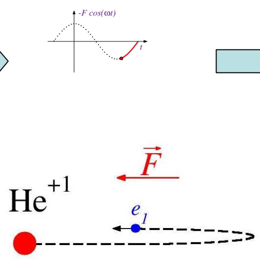
I. Tunnelling:

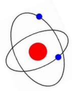
II. Rescattering:

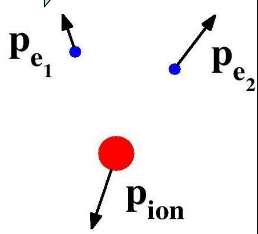
III. Highly excited complex:

IV. Decay of highly excited complex:





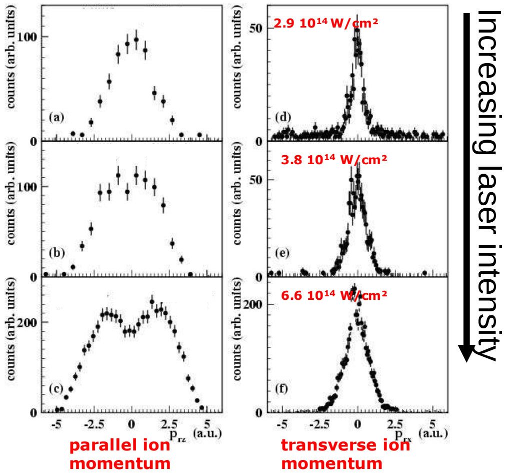




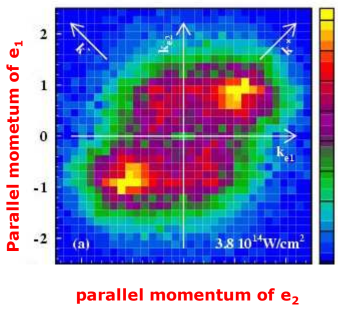
Corkum; Kulander et al 1993

Momentum distributions

Weber et al., PRL 83, 443 (2000):
Helium



Weber et al., Nature 405, 658 (2000):
Argon



Symmetric electron escape:
equal parallel momenta
opposite perpendicular momenta

Wannier´s view of double ionization

G.H. Wannier, Phys Rev 90, 817 (1953):

- At threshold, electrons can only escape symmetrically in opposite directions
- Deviations from symmetry are amplified
- The cross section increases like E^{α} with exponent α = 1.056 (for neutral – $2e^{-}$)
- … All as a consequence of the Coulomb interaction

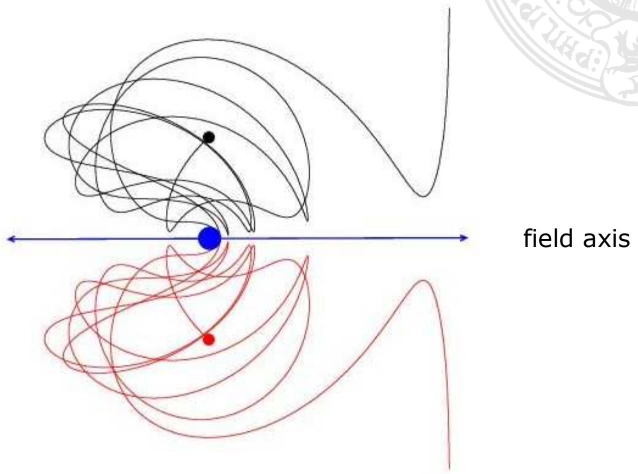
Why parallel symmetric escape???

- correlations in electronic state?
- correlations induced by push from rescattered electron?
- focussing influence of laser field?

Problem: cross sections small, direct classical simulations hopeless

Proposal: assume symmetric electron motion

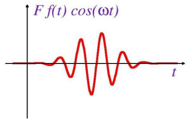
Symmetric escape

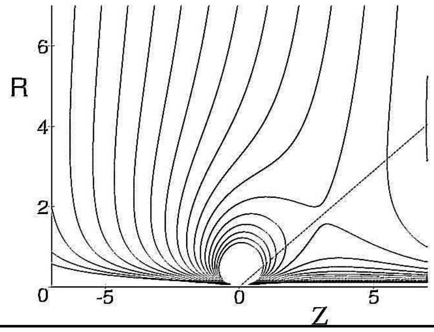


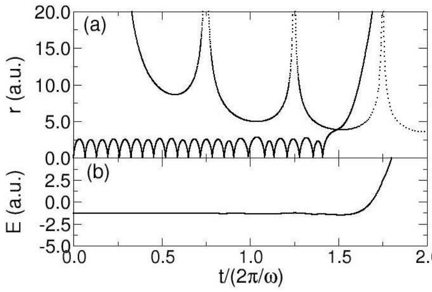
Motion in symmetric subspace

$$H = \frac{\vec{p}_R^{\,2} + \vec{p}_Z^{\,2}}{4} - \frac{4}{\sqrt{R^2 + Z^2}} + \frac{1}{2R} + Ff(t)Z\cos(\omega t)$$

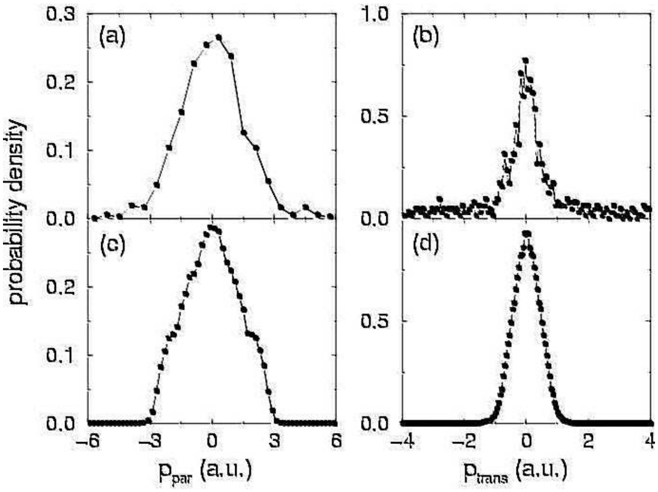
fixed time, fixed field







Comparison to experiment



parallel momentum

transverse momentum

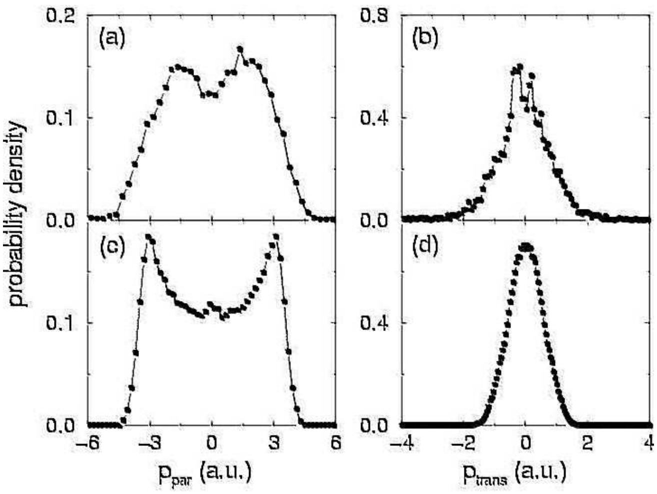
$2.9\ 10^{14}$ W/cm^2
220 fs
800 nm

experiment
Weber et al., PRL 83, 443 (2000)

calculated data
E=-0.6 a.u.
t_0=0.3 T_d

Physica Scripta **T90** (2001) 185
Phys Rev A **63** (2001) 043414

Comparison to experiment



parallel momentum   transverse momentum

$6.6\ 10^{14}\ W/cm^2$
220 fs
800 nm

experiment
Weber et al., PRL 83, 443 (2000)

calculated data
E=-0.4 a.u.
$t_0=0.3\ T_d$

Physica Scripta **T90** (2001) 185
Phys Rev A **63** (2001) 043414

Symmetric escape !!!

- Formation of highly excited complex after collision
- Rapid decay with almost frozen field
- Dominance of symmetric subspace as a consequence of dynamical instability: deviations from symmetry are amplified and one electron will be pushed back

Coulomb repulsion between electrons enforces symmetric escape

Threshold behaviour

- Fix initial energy and field strength
- consider cross section as function of excess energy above saddle
- exponential escape along reaction coordinate with rate λ_R

$$p_R = e^{\lambda_R t}$$

- exponential separation perpendicular to it with rate $\lambda_\perp$

$$p_\perp = e^{\lambda_\perp t}$$

- momenta that lead to double ionization satisfy:

$$|p_\perp| < |p_R|^{\lambda_\perp / \lambda_R}$$

- cross section behaviour

$$\sigma(E) \propto E^\alpha$$

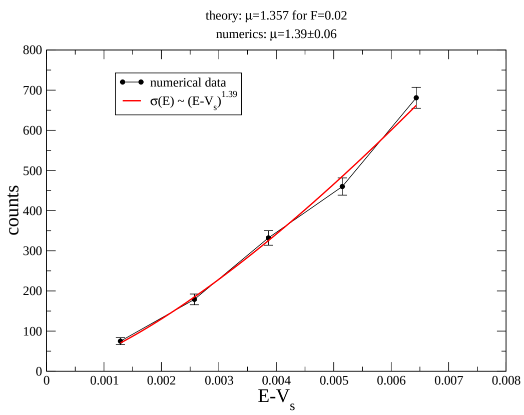
$$\alpha = \frac{\lambda_\perp}{\lambda_R} \approx 1.2918$$

Threshold behaviour

Confirmation in a 1+1 dimensional model

$$\mu_{theory} = 1.357$$

$$\mu_{simulation} = 1.39$$



EPL 56, 651 (2001)

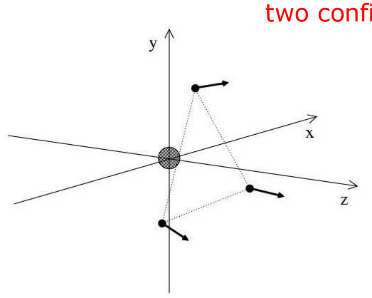
J Phys B 39, 3865 (2006

Summary so far:

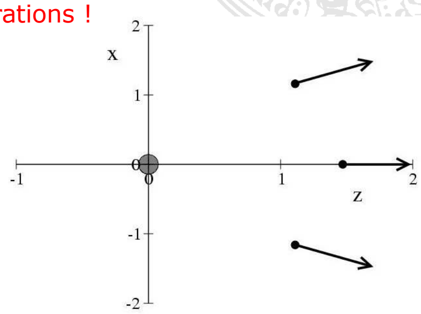
- Double ionization while field is on
- Dynamics in symmetric subspace reproduces observations
- Two-electron configuration in steady field guards two electron channel
- Algebraic variation of cross section with excess energy

Triple ionization

Three electrons :
two configurations !



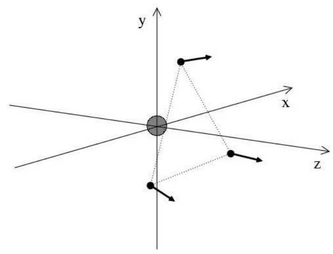
Three equivalent electrons



Two equivalent
+ one extra
electron

Triple ionization

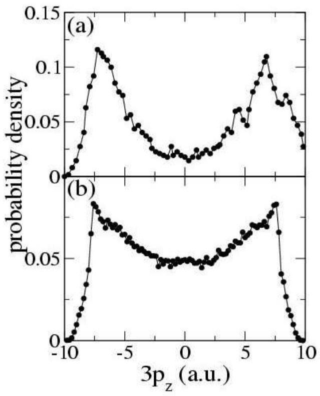
Dominant configuration:



Three equivalent electrons !

Neon

($15\ 10^{14}$ W/cm^2, 30 fs, 795 nm)



parallel ion momentum

experiment

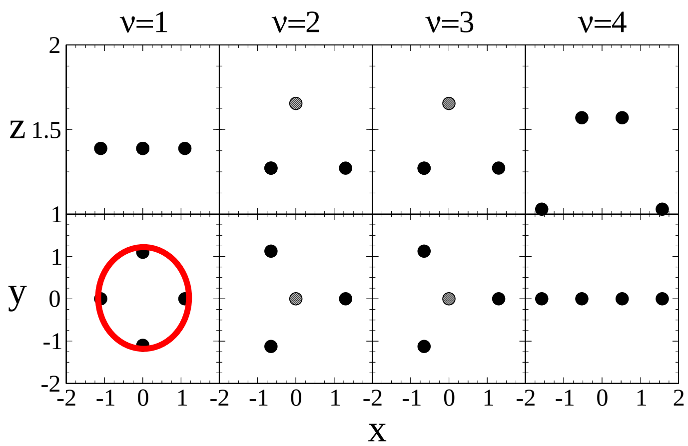
Moshammer et al.,
PRL 84, 447 (2000)

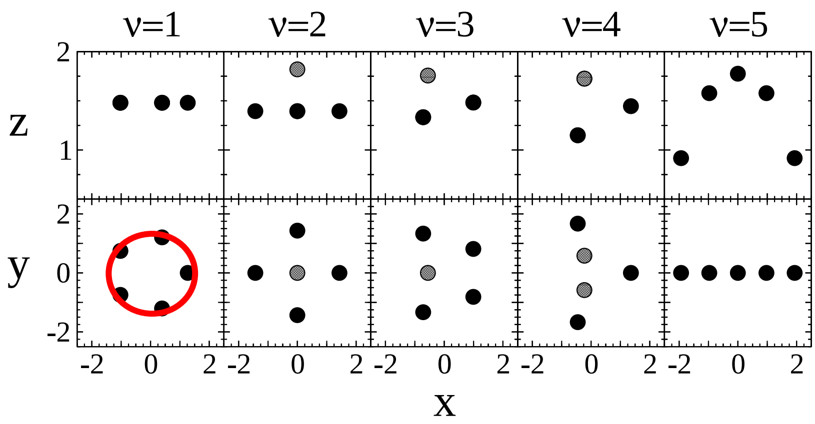
calculated data

E=-1 a.u.,

$t_0=0.3\ T_0$

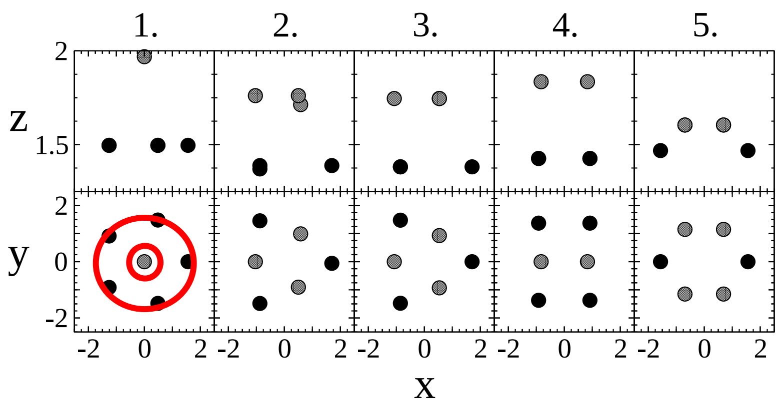
Many electron configurations: N=4

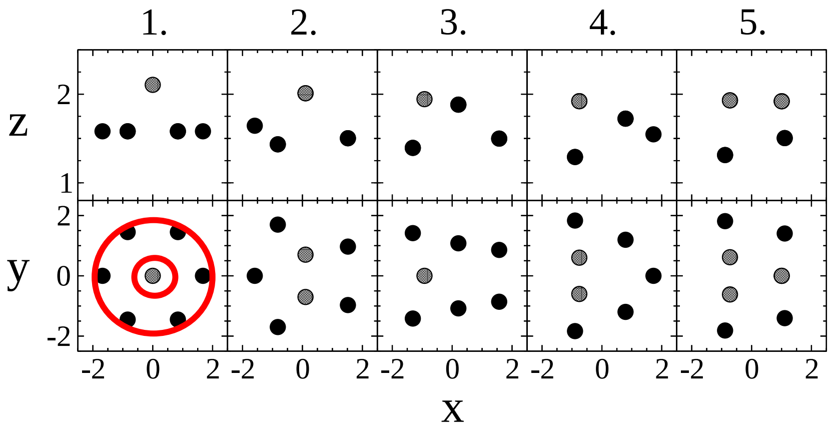


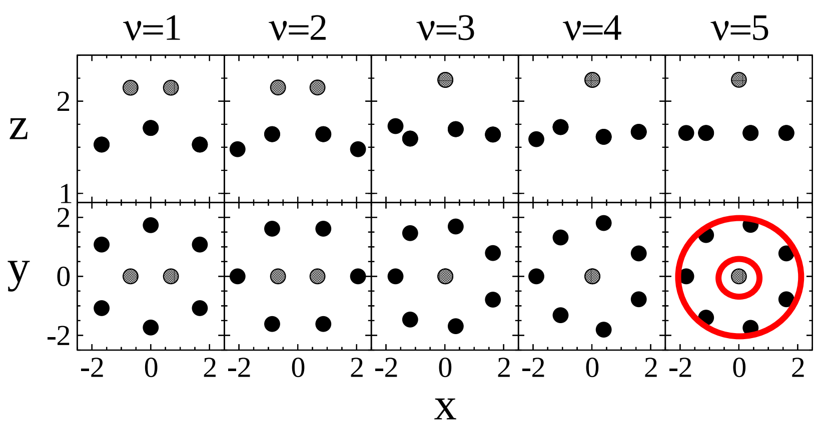


Many electron configurations: N=6

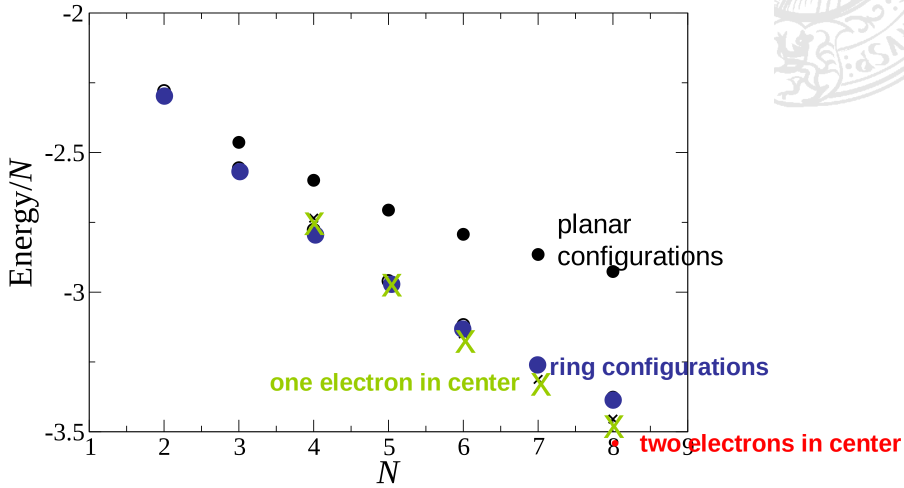
▪




1.
2.
3.
4.
5.
z
y
x


ν=1
ν=2
ν=3
ν=4
ν=5
z
y
x

Lowest transition states



Many electron transition states

N	number of saddles	E	exponent
2	1	-4.55	1.29
3	2	-7.66	2.62
4	4	-11.10	4.09
5	5	-14.80	5.73
6	11	-18.89	7.20
7	14	-23.18	8.89
8	26	-27.65	10.65

Dominant Mode:

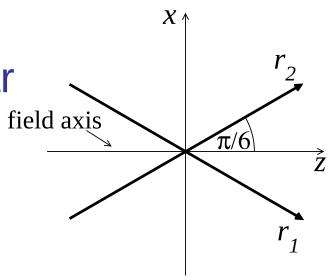
↑ equivalent electrons (N = 2–5)

↓ non-equiv. electrons (N = 6–8)

Quantum models

- Original: 3+3 degrees of freedom
- Reduction to planar 2+2 conceivable … but still too big
- Minimal model: 1+1 … with a twist:

Electron motion not collinear but at some angle:



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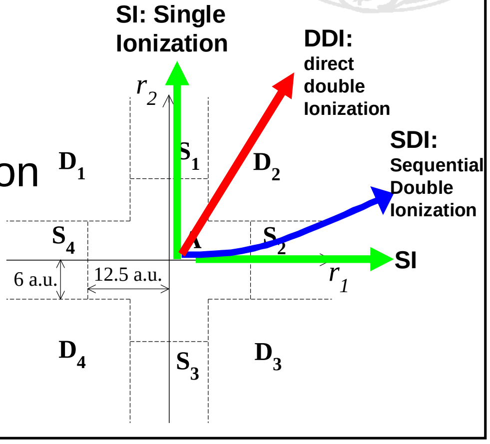
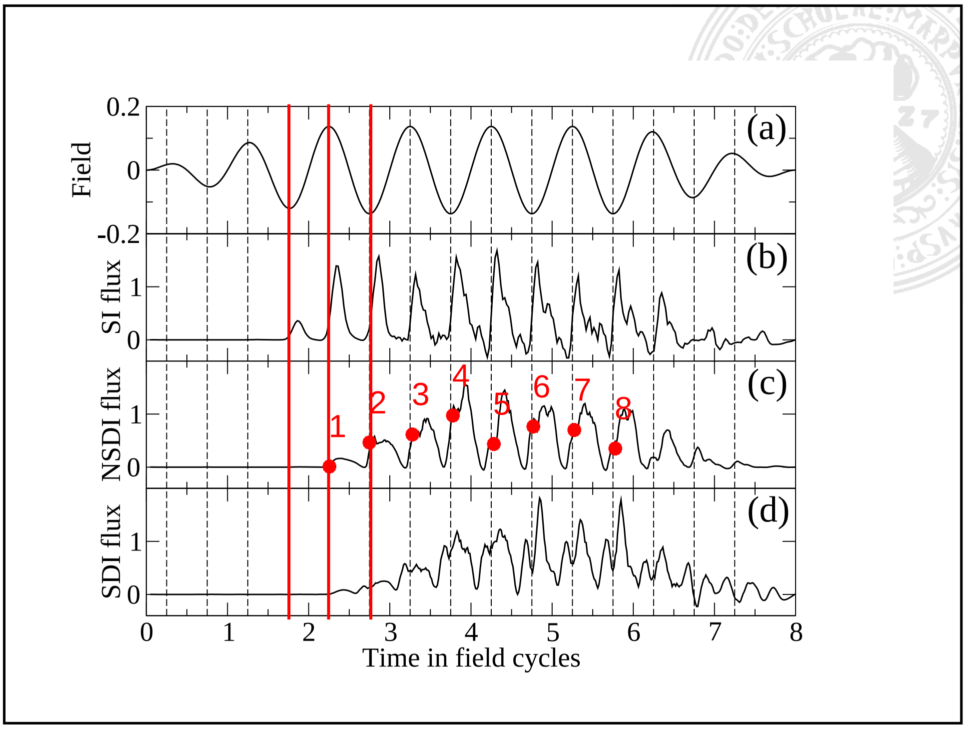
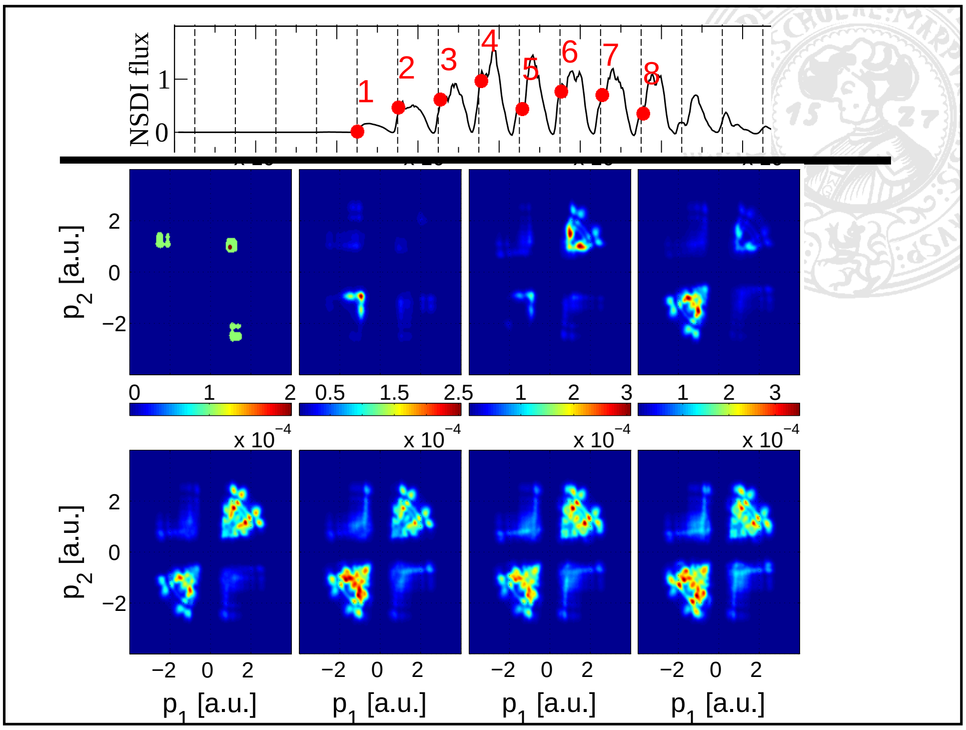
Quantum simulations:

- Start with groundstate
- Apply pulses
- Follow wave packet
- Calculate final momentum distribution
- Analyze results



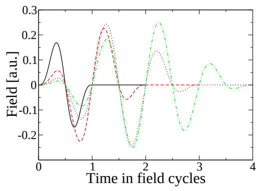
arXives:physics/0607035
PRL 98, 203002 (2007)

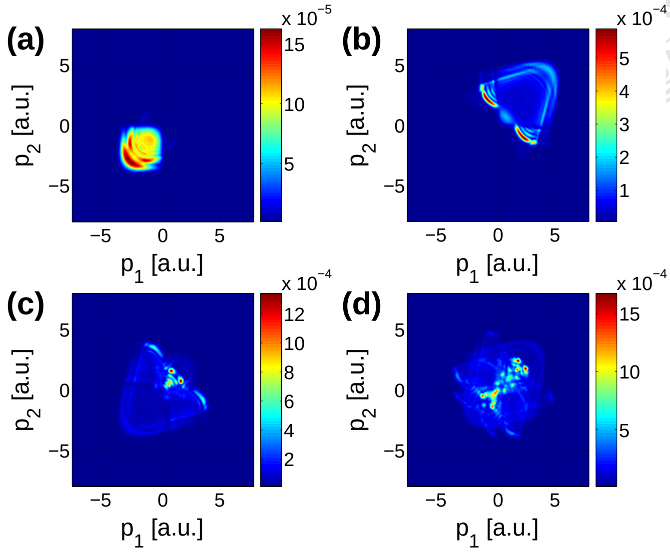

0.2
0
-0.2
Field
(a)
SI flux
(b)
NSDI flux
(c)
SDI flux
(d)
1 2 3 4 5 6 7 8
0 1 2 3 4 5 6 7 8
Time in field cycles


NSDI flux
1 2 3 4 5 6 7 8
p2 [a.u.]
p1 [a.u.]
x 10−4

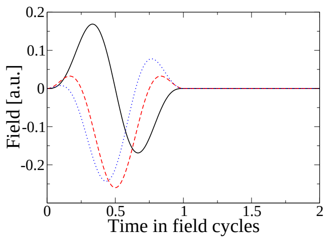
Variations with duration of pulse

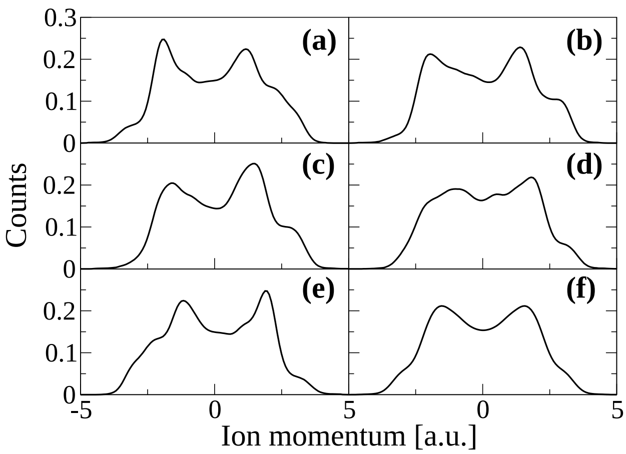
F=0.08





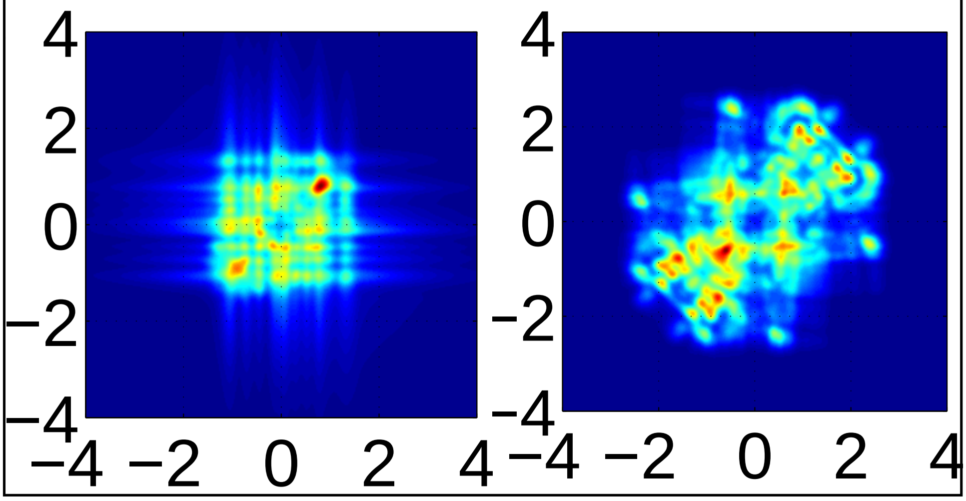
Variations with phase



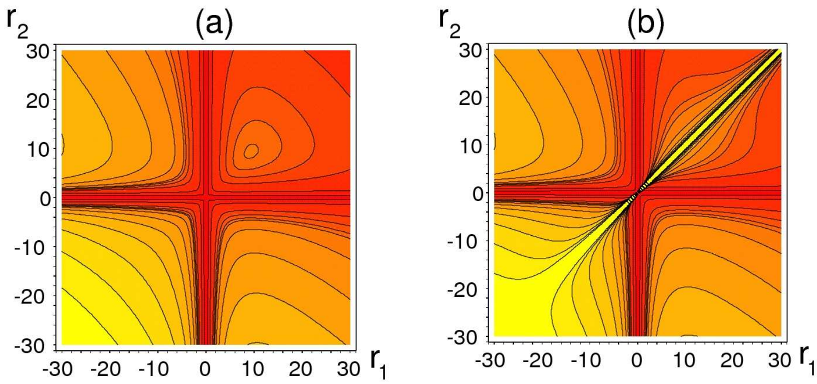


Phase differences
0, 3, 5, 8, 10 * π/10
(f): averaged over
all phases

Quantum interferences



Model potentials

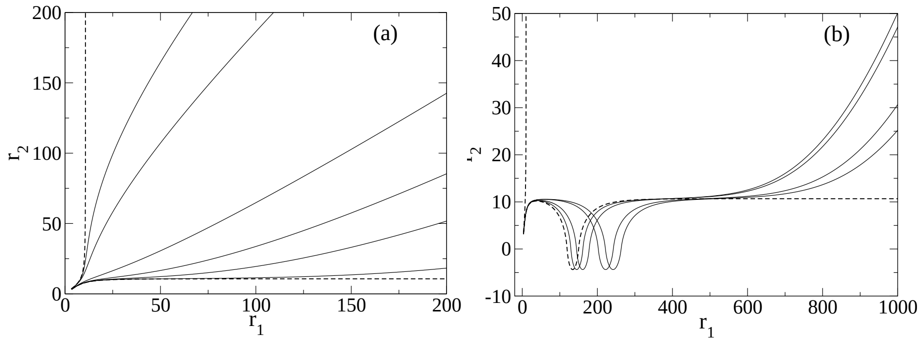


skewed tracks   aligned tracks

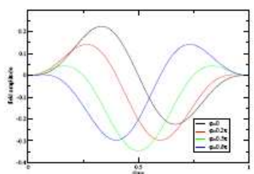
Semiclassical origin (?)

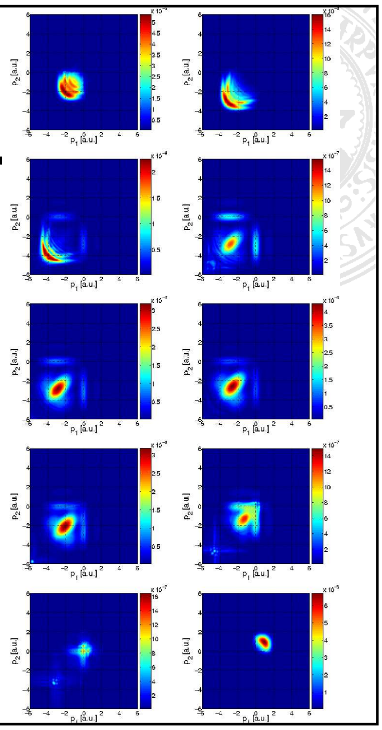
- Direct paths to double ionization…

… and indirect ones



Phase senstivity of interferences

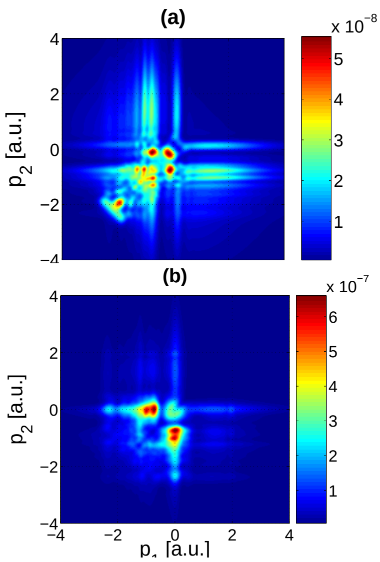




Quantum correlations

- Pauli principle:
  no two electrons in the same state
- In momentum space: different momenta
- … or, in cylindrical coordinates:
  no electrons with equal
  parallel momenta

Quantum correlations in simulation



ting from:

mmetric ground state:
agonal populated

tisymmetric first
excited state:
diagonal empty

Summary:

- Correlated escape because of Coulomb repulsion
- Ionization behaviour dominated by unstable equilibria
- Many equilibria for many electrons
- Quantum 1+1-dimensional model captures all qualitative features

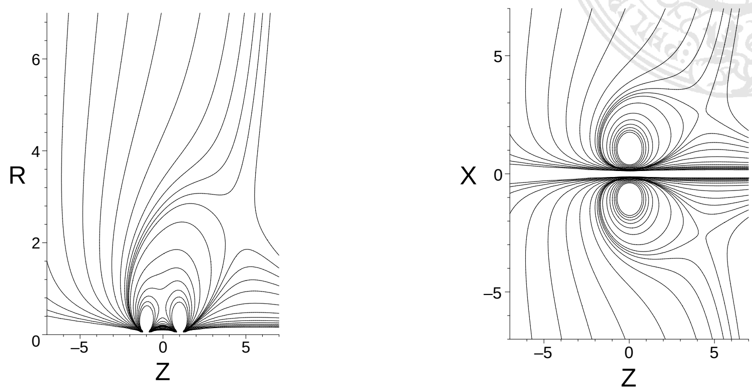
Summary:

- Quantum interferences in outgoing channels
- Strong effects from Pauli principle for antisymmetric initial state

Outlook:

- Multiple ionization of molecules
- Other observables (perpendicular momenta, angles etc.)
- Field and frequency dependence

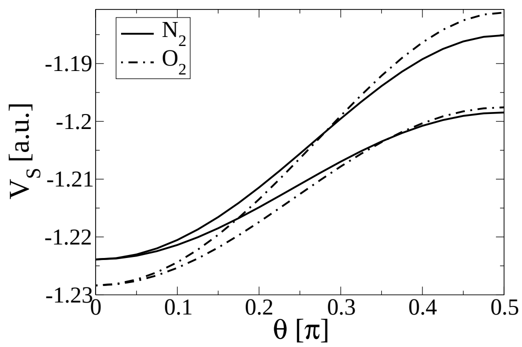
Multiple Ionization of Molecules



orientation parallel to field axis perpendicular

Multiple Ionization of Molecules

Motion of saddle energy with orientation:



lowest saddle for parallel orientation

References

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- Physica Scripta **T90** (2001) 185
- Phys Rev A **63** (2001) 043414
- Phys Rev A **64** (2001) 053401
- Europhys Lett **56** (2001) 651
- Journal of Physics B **36** (2003) 3923
- Phys Rev A **71** (2005) 033407
- J Phys B **39** (2006) 3865
- Phys Rev Lett **98** (2007) 203002
- arXives:physics/