Correlated electron dynamics in strong laser fields

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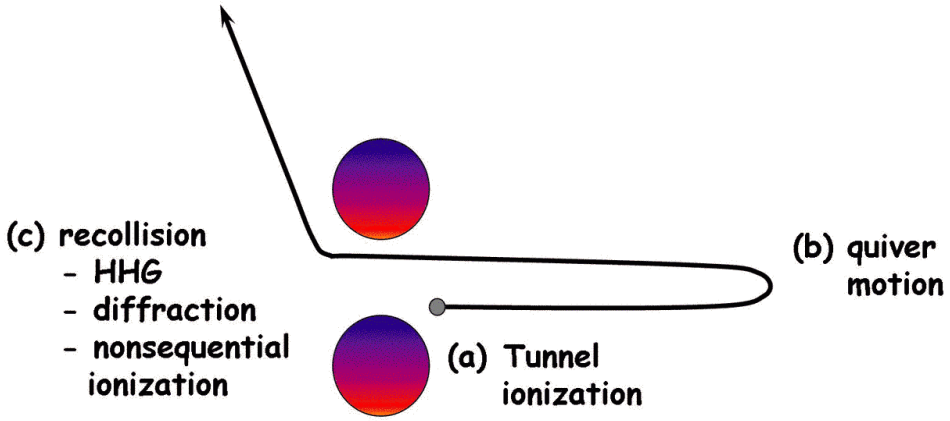
Outline

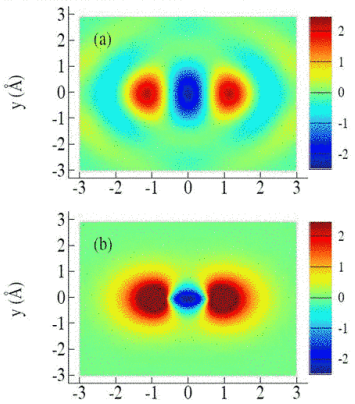
1. Quantum few body-dynamics
2. Classical many-body dynamics

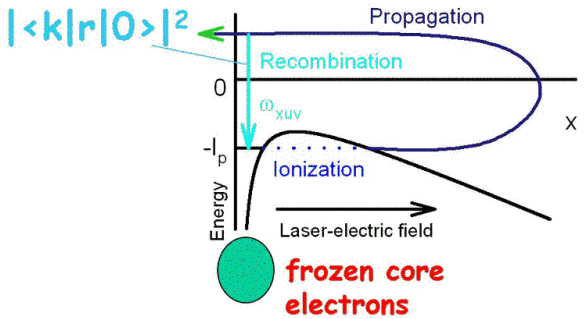
Single active electron approximation (SAE)



Noble gases and small molecules -> SAE

Molecular orbital tomography





Molecular orbital tomography

1. Rotate target molecule
2. Create HHG snapshots from the target molecule
3. Retrieve HOMO by inverse FFT and some other stuff

The Pauli exclusion principle

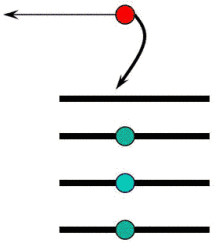
$|\langle k|r|0\rangle|^2$

$$\vec{d}(\vec{k}) = \sum_{i=1}^{n} \langle \phi(\vec{r}_1,\ldots,\vec{r}_n) \,|\, \vec{r}_i \,|\, 0(\vec{r}_1,\ldots,\vec{r}_n) \rangle$$

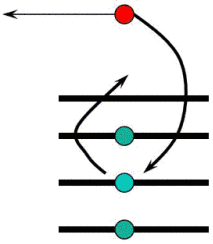
measurement of the HOMO

$$\phi = \hat{A}\left[0_+(\vec{r}_1,\ldots,\vec{r}_{n-1}) \otimes k(\vec{r}_n)\right]$$

electron recombines to HOMO

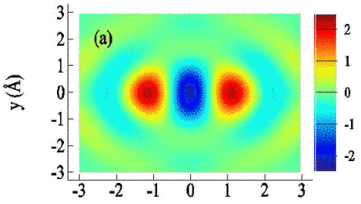


free electron recombines and exchanges position with a core electron



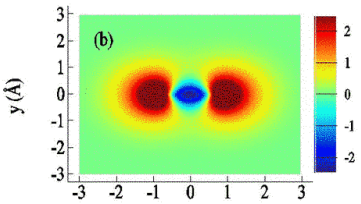
The wavefunction of N_2

N_2 HOMO reconstructed from measurement



1. HHG calculated from single-electron dipole moment
2. wavefunction retrieved from HH spectrum

= HOMO calculated by GAMESS



1. HHG calculated from multi-electron dipole moment
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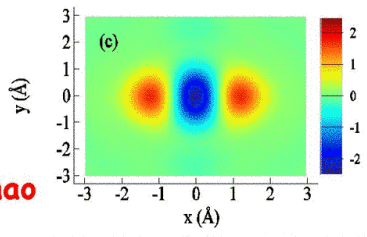


Zengxiu Zhao

Multi-electron dynamics in strong fields

Theoretical challenge:

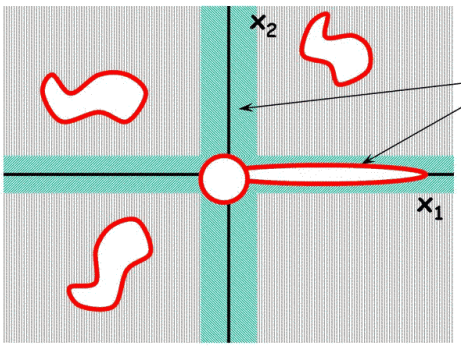
Two notoriously difficult problems
1. many-body physics
2. non-perturbative dynamics

Solution of few-body problem I

Exact solution

curse of dimensions

$\Psi(r_1,r_2,t)$ … 1000 points in one dimension → 10^{18} points



Hartree-Fock

$\Psi(r_1,r_2) = \Psi_1(r_1)x\Psi_2(r_2) - \Psi_2(r_1)x\Psi_1(r_2)$

Density functional

$\Psi(r_1,r_2) = \Psi_1(r_1)x\Psi_2(r_2)$

Configuration interaction

$\Psi(r_1,r_2) = A_{12}[\Psi_1(r_1)x\Psi_2(r_2) - \Psi_2(r_1)x\Psi_1(r_2)]$
$+A_{13}[\Psi_1(r_1)x\Psi_3(r_2) - \Psi_3(r_1)x\Psi_1(r_2)]$
$+A_{23}[\Psi_2(r_1)x\Psi_3(r_2) - \Psi_3(r_1)x\Psi_2(r_2)]$

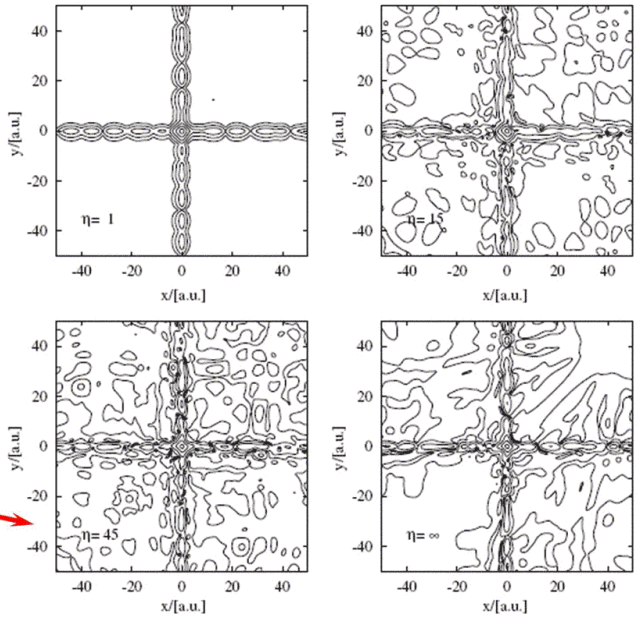
\+ = Multi-configuration time-dependent Hartree Fock (MCTDHF)

Recollision: 2e⁻ x 1D - He

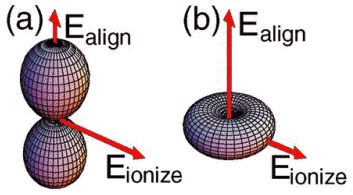
n	$\eta = \binom{n}{2}$	P_η^{2+}
2	1	0.0059
4	6	0.0246
6	15	0.0189
8	28	0.0250
10	45	0.0283
12	66	0.0273
∞	∞	0.0257

Double ionization rate converges

2e⁻ continuum wf is more problematic

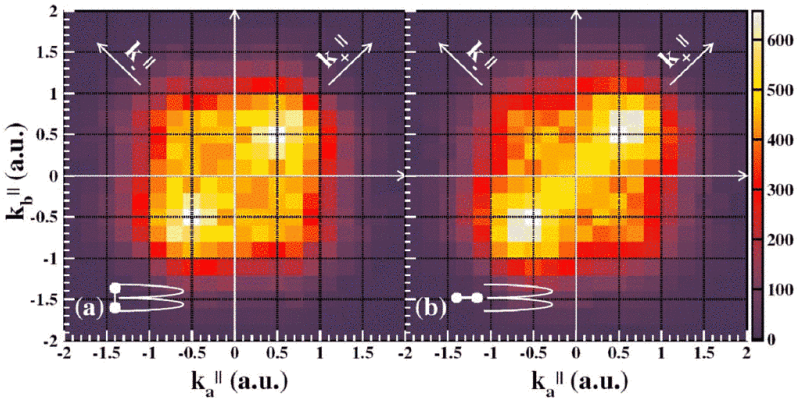


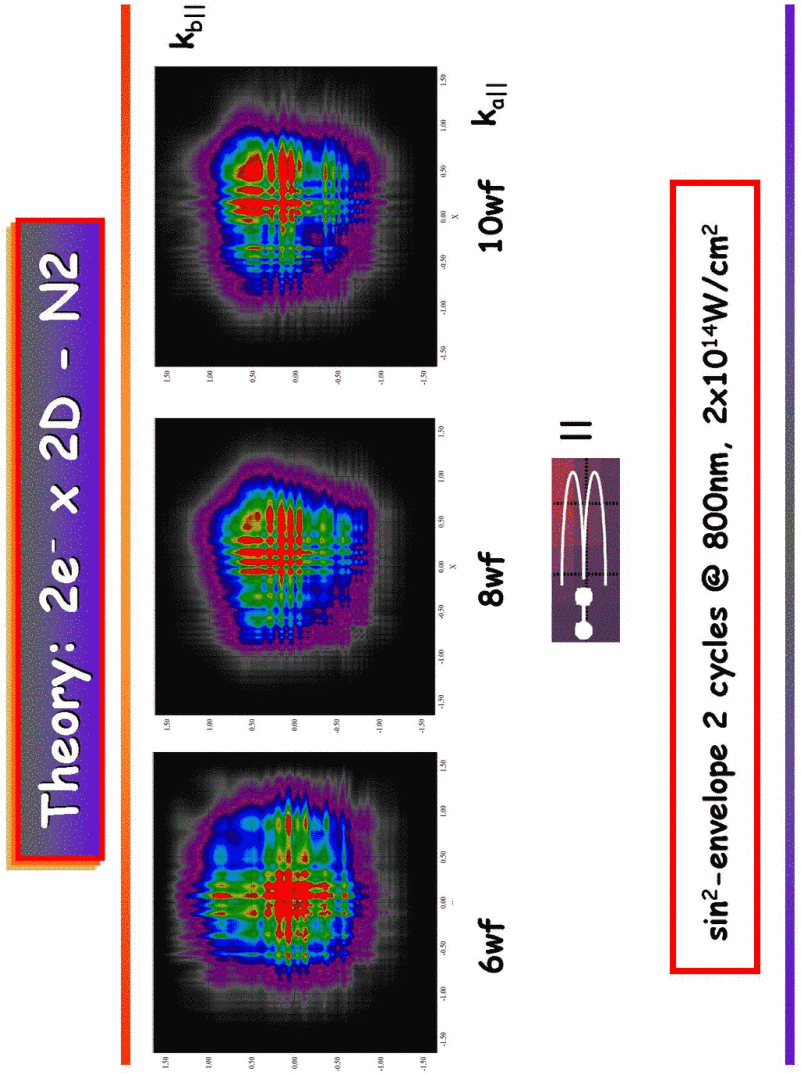
Nonsequential ionization in N_2

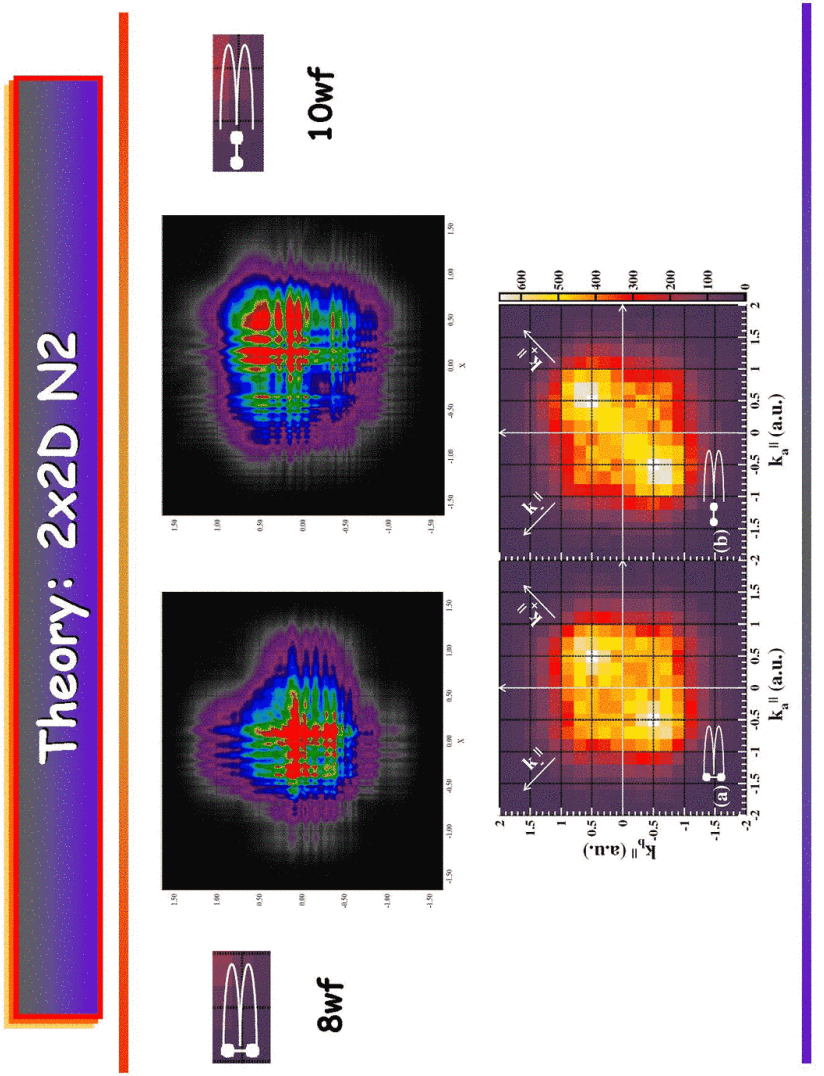


Zeidler et al., PRL 95, 203003 (2005)

40fs, 800nm, 1.3×10^{14}W/cm^2




Theory: 2e⁻ x 2D - N2
$k_{b||}$
$k_{a||}$
6wf
8wf
10wf
||
sin²-envelope 2 cycles @ 800nm, 2x10¹⁴W/cm²


Theory: 2x2D N2
8wf
10wf
(a)
(b)
$k_a^{||}$ (a.u.)
$k_b^{||}$ (a.u.)

Some facts on the code

Chris Mc Donald !!!

enemy → $\langle \varphi_{j_2}\varphi_{j_3}|\mathrm{H}_2(\boldsymbol{x}_1,\boldsymbol{x}_2)|\varphi_{l_2}\varphi_{l_3}\rangle_{\boldsymbol{x}_1,\boldsymbol{x}_2}$

Calculated by convolution
Coulomb cusp: technically no shielding is needed
Laplace: FFT

Propagation in the momentum domain

Calculation on a single 64-bit processor

How many electrons in 2D on 1 processor ? approx. 4

Can we do 3D? maybe, if code scales on an SMP structure

Tunnel ionization of complex materials

Wall of fame

1928: Oppenheimer
hydrogen

1967: Perelomov, Popov, Terentev
atoms except match coeff.

1986: Ammosov, Delone, Krainov (ADK)
atoms + match. coeff.

2002: Lin et al. (MO-ADK)
small molecules

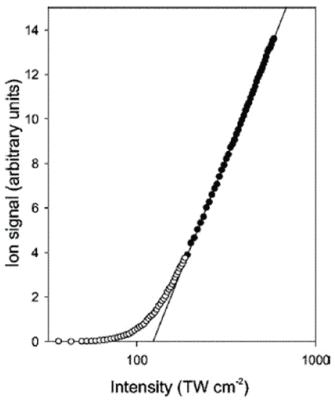
single active electron approximation

Tunnel ionization of complex systems

TABLE I. Experimental femtosecond laser ionization saturation intensities I_{sat}, Coulomb explosion onset intensities I_{CE}, and saturation intensities calculated from tunneling theory, I_{ADK}, for organic molecules and rare gas atoms.

	Ionization potential [a] (eV)	I_{sat} (10^{12} W cm^{-2})	I_{CE} (10^{12} W cm^{-2})	I_{CE}/I_{sat}	I_{ADK} (10^{12} W cm^{-2})	I_{sat}/I_{ADK}
1,3-*cyclo*-hexadiene	8.25	66	158	2.4	25	2.64
1,3,5-hexatriene	8.42	89	213	2.4	32	2.78
1,3-hexadiene	8.51	81	165	2.04	32	2.53
n-propylbenzene	8.72	59	144	2.45	30	1.97
i-propylbenzene	8.73	51	156	3.03	30	1.7
ethyl benzene	8.77	48	144	3	30	1.6
1,4-*cyclo*-hexadiene	8.82	81	161	1.99	25	3.24
toluene	8.82	56	154	2.75	32	1.75
cyclo-hexene	8.94	117	194	1.66	33	3.54
benzene	9.24	71	201	2.83	38	1.86
1-hexene	9.4	71	156	2.19	40	1.77
propene	9.73	95	208	2.19	46	2.06
cyclo-propane	9.86	110	296	2.69	48	2.29
cyclo-hexane	9.88	85	204	2.41	48	1.77
dimethylether	10.03	95	268	2.82	51	1.86
hexane	10.13	63	135	2.14	53	1.19
2-methoxyethanol	10.13	71	163	2.29	53	1.34
propyne	10.36	98	210	2.14	57	1.72
ethene	10.50	110	373	3.39	60	1.83
methanol	10.85	158	397	2.51	67	2.36
propane	10.95	112	293	2.62	69	1.62
ethane	11.52	182	400	2.20	83	2.19
xenon	12.13	112			100	1.12
neon	21.56	750			740	1.01
helium	24.58	1158			1160	1

[a]Reference [19].



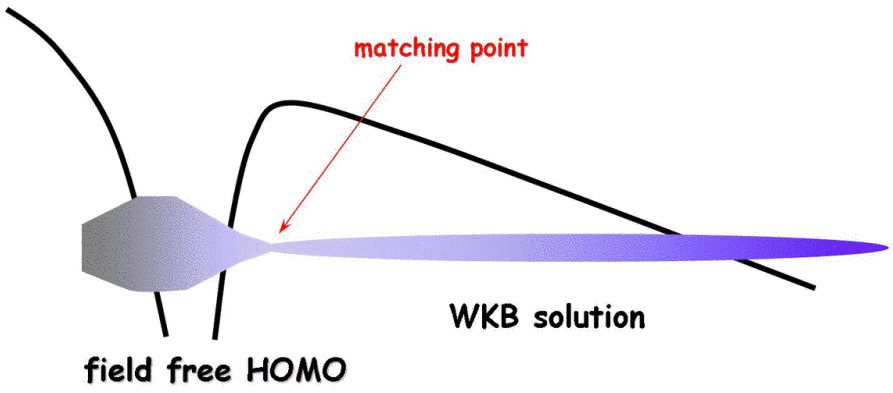
P. Corkum

Analytical analysis

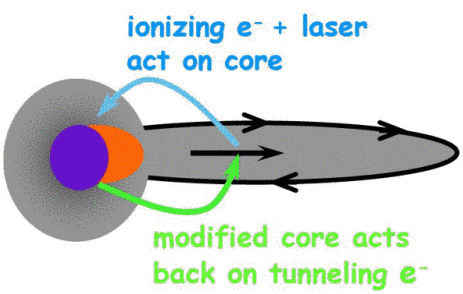
Analytic tunnel ionization theory I

noble gas: ADK
small molecules: MO-ADK

frozen core

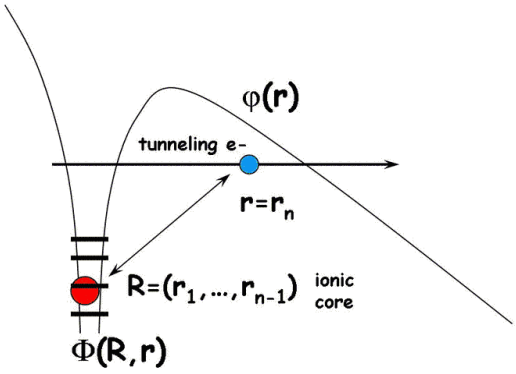


Analytic tunnel ionization theory II



Complex material

n-electron system



correlated Ansatz: $\Phi(R,r) \otimes \varphi(r)$

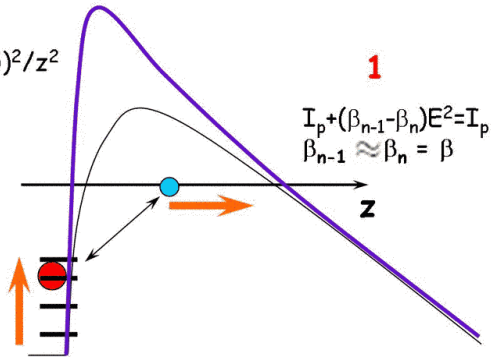
T. Brabec et al., PRL 95, 073001 (2005)

Analytic tunnel ionization theory III

2 $V(\mathrm{r}) = -\frac{Z}{|\mathrm{r}|} + \frac{2\beta E}{z^2} - \frac{\beta}{z^4}$ + $(l+0.5)^2/z^2$

I_{n-1}-βE^2-$2\beta E/z^2$+β/z^4

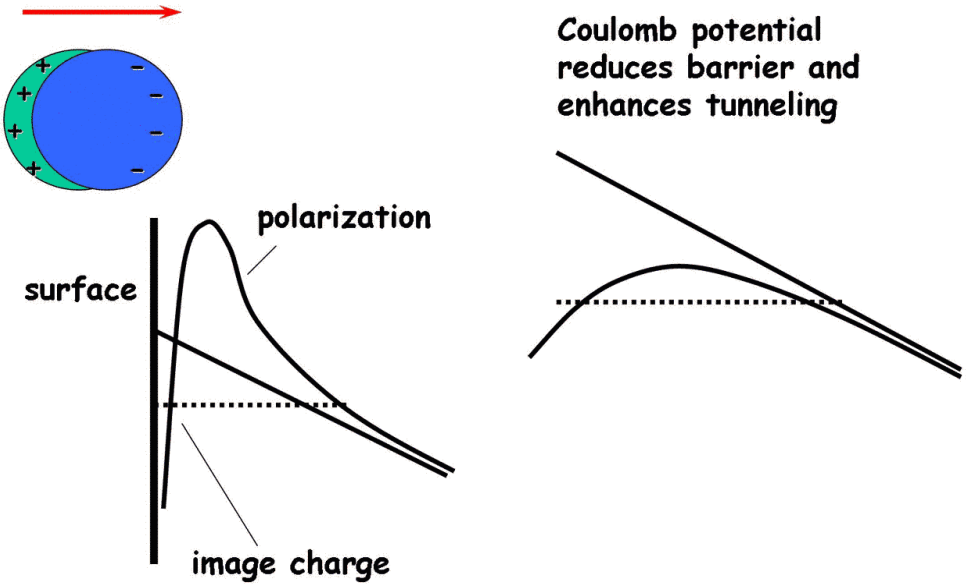
3



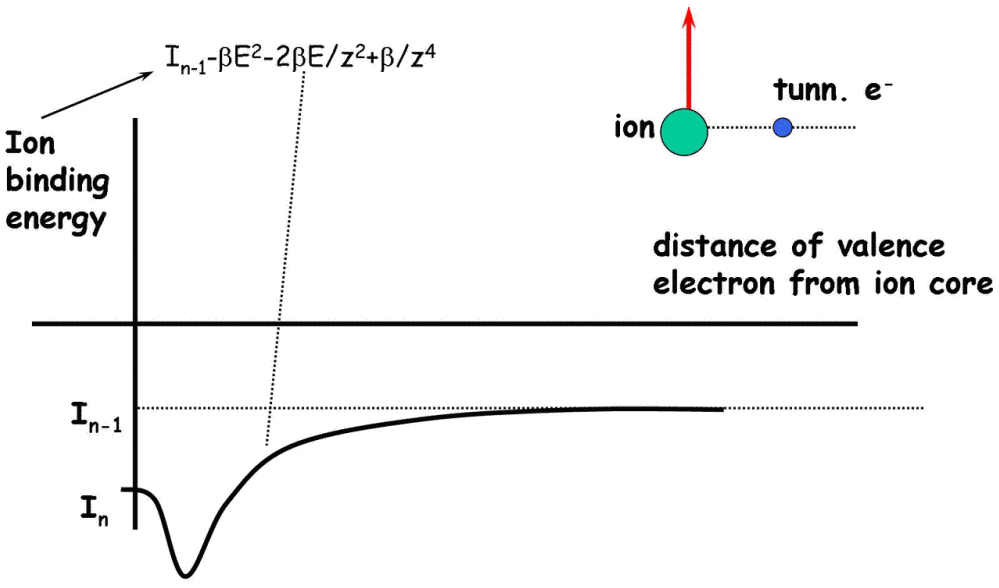
1. Stark shift negligible
2. single electron potential accounting for the interaction with the core
3. core reconfiguration during tunneling (Born-Oppenheimer approximation)

T. Brabec et al., PRL 95, 073001 (2005)

Why does polarization reduce tunneling

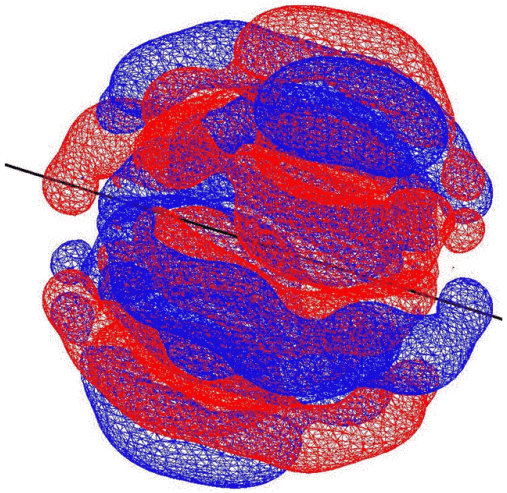


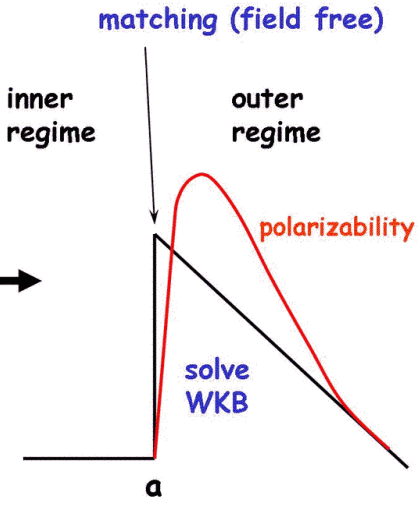
How does the ion reconfigure during tunnel ionization



Ionization of complex materials

C_{60}^{Z+} ... DFT β=80A³ a=3.5A





Quasi-analytical theory

Generalized ADK/MO-ADK theory

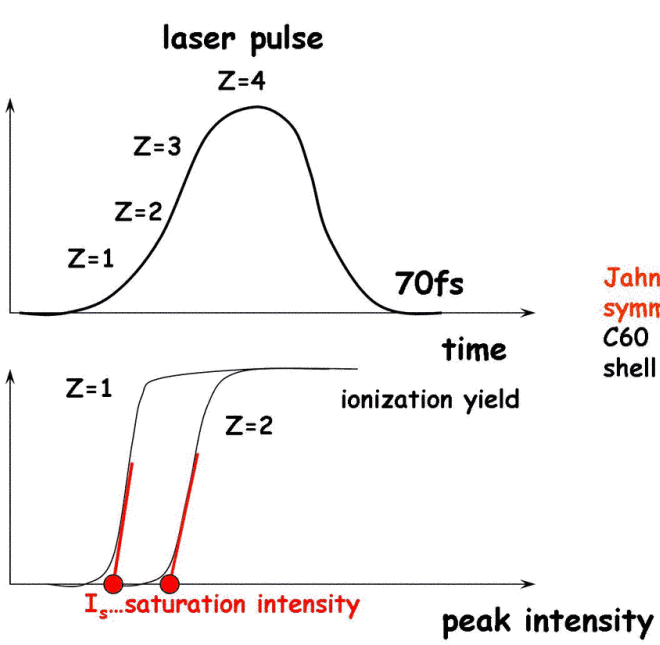
$$w_m = w_o \left(\frac{E}{2\kappa^2}\right)^{(l+1/2)^2 E/\kappa^3} \left(\frac{E}{2\kappa^2}\right)^{2\beta E^2/\kappa^3} \left(\frac{E}{2\kappa^2}\right)^{-\frac{5E^3}{2\kappa^7}}$$

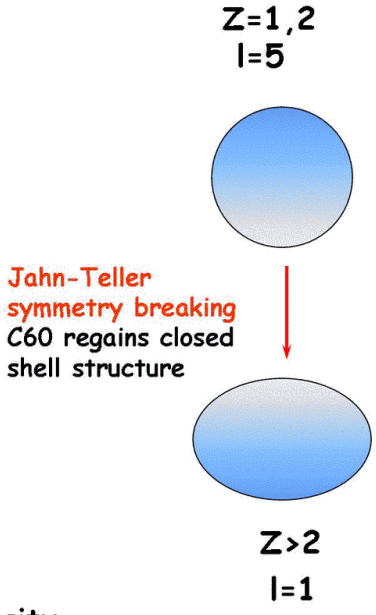
l-barrier laser polarization image charge

$$w_o = \frac{|C_{lm}|^2}{2^{|m|}|m|!\kappa^{2\mathcal{Z}/\kappa-1}} \left(\frac{2\kappa^3}{E}\right)^{\frac{2\mathcal{Z}}{\kappa}-|m|-1} e^{-2\kappa^3/3E}$$

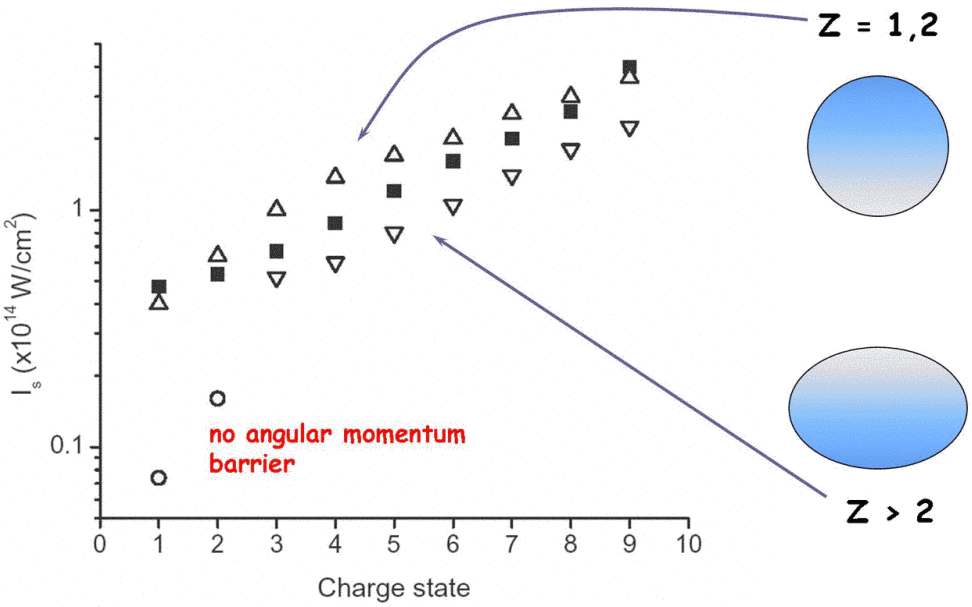
Comparison experiment-theory

C_{60} – miscellaneous



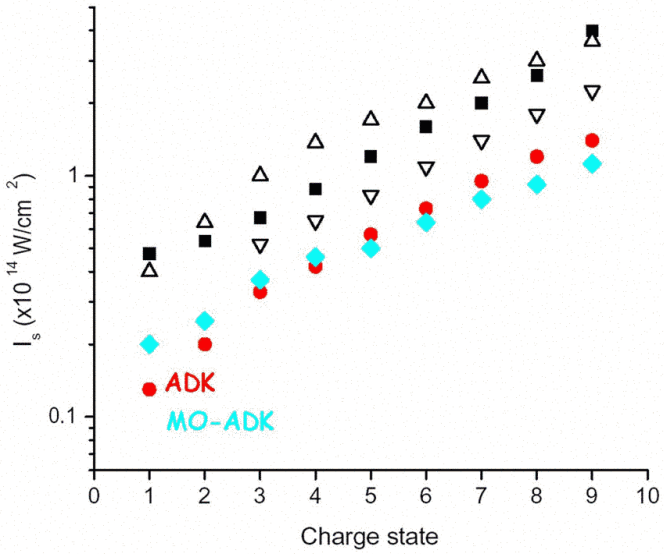


Ionization of complex materials



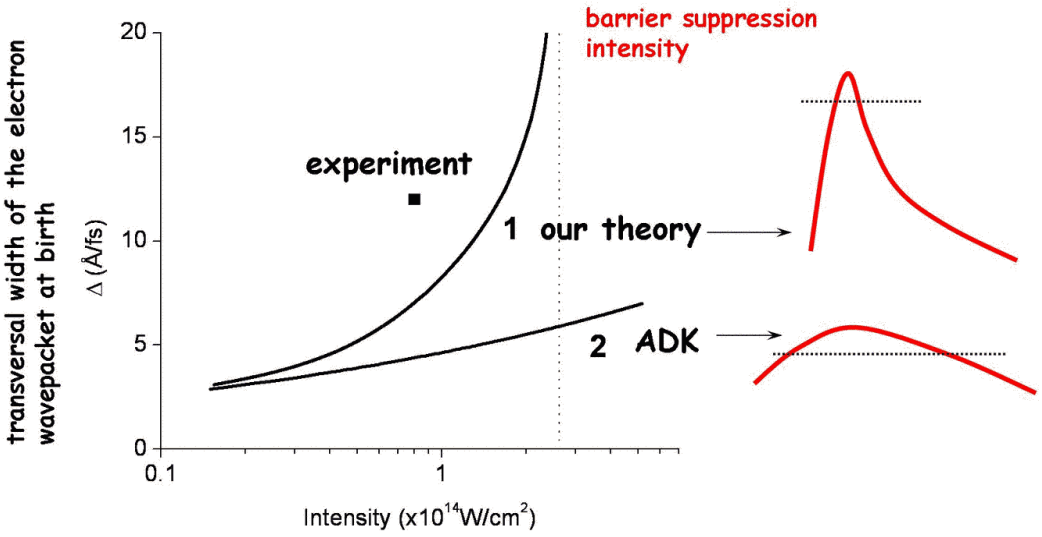
Experiment: V.R.Bhardwaj et al., PRL 91, 203004 (2003).

Comparison with ADK/MO-ADK



benzene and related systems work even better

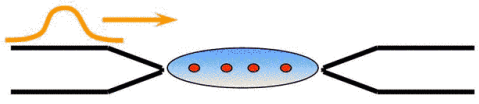
Transversal momentum distribution



Experiment: V.R.Bhardwaj et al., PRL 91, 203004 (2003).

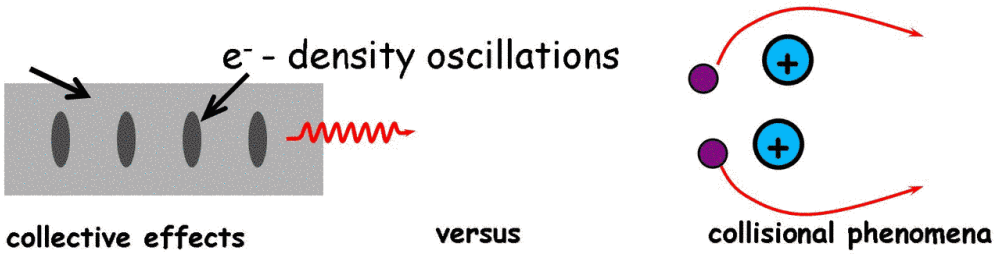
Future directions

Electron transport in single molecule transistors



transport on fs to sub-fs time scale

Collective versus collisional



Universal property of many-body systems

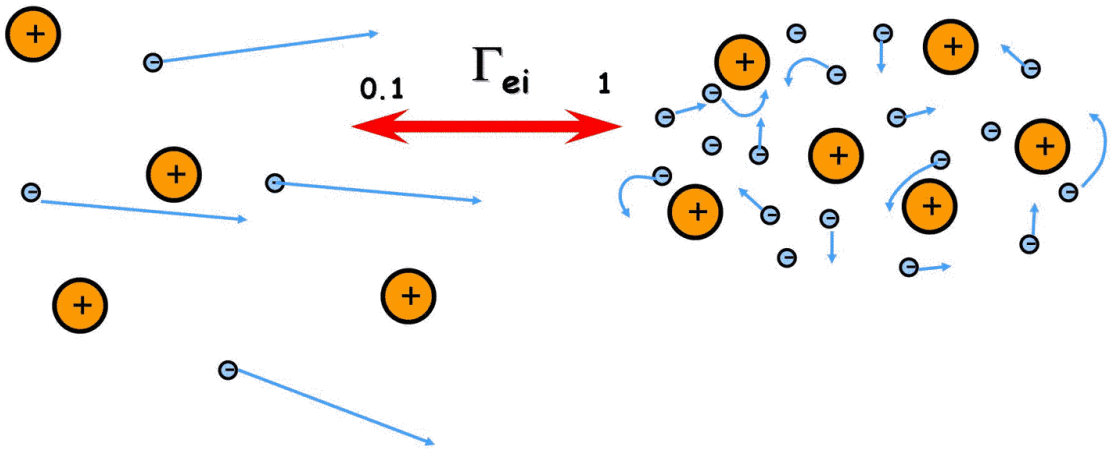
plasma physics and condensed matter → plasma oscillation
clusters → plasmons
nuclear physics → giant resonances
atomic physics → shape resonances
large molecules → doorway states

Weakly coupled plasma

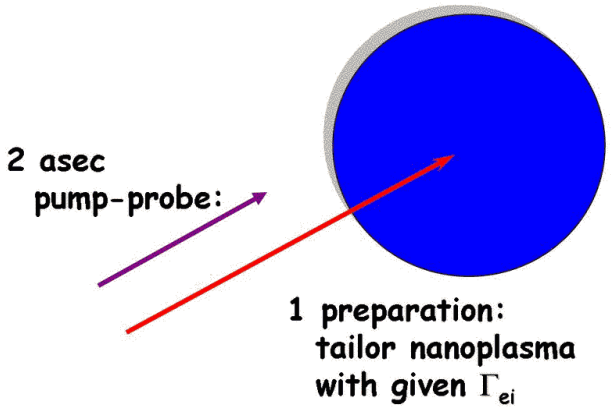
- hot and dilute
- collective mean field dominate
- conventional plasma theory

Strongly coupled plasma

- cold and dense
- many collisions
- kinetic theories break down



Attosecond plasma physics

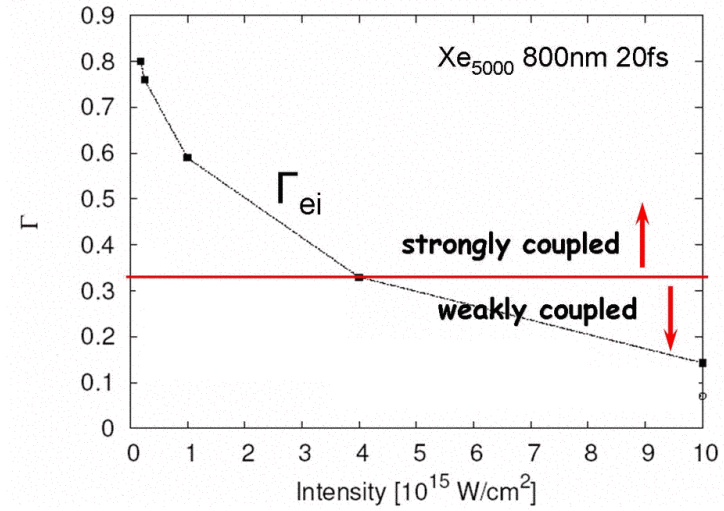


Cluster laser interaction

Tree code + rates for quantum processes

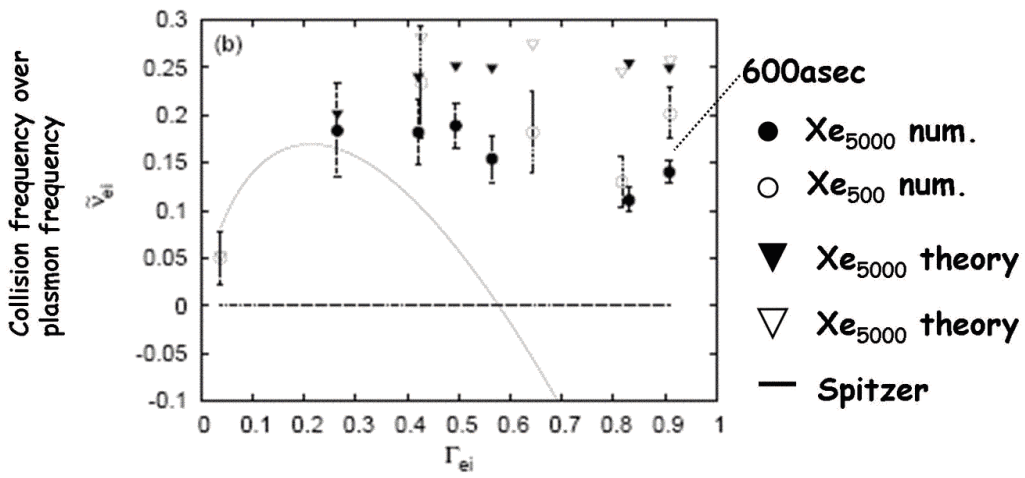
Coupling parameters vs laser intensity

quantum degenerate plasma



spans the range from weakly to strongly coupled electron plasmas leaving the plasma frequency approx. unchanged

Attosecond plasmon dephasing



1. Collision frequency is surface independent – connection to plasma physics
2. Decay is exactly exponential

HHG: Lewenstein versus exact