

ROBUST COHERENT WAVEPACKET CREATION BY MEANS OF DISSIPATION



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(QSIM19 at KITP Santa Barbara, 24.04.2019)

Overview

- Strong-field / attosecond physics in a (very tiny) nutshell.

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- Toward (interacting?) many-body systems . . .

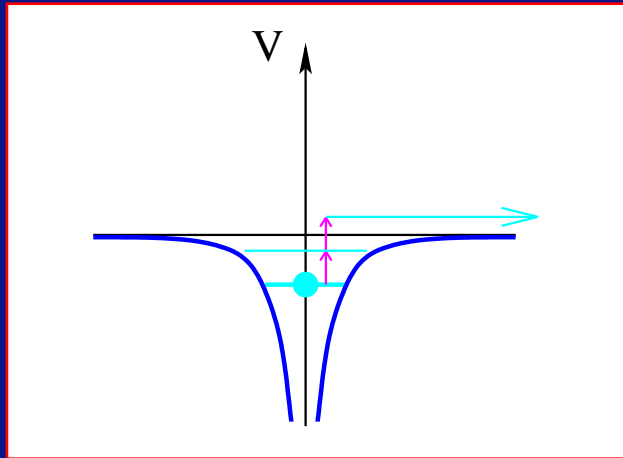
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Disclaimer:

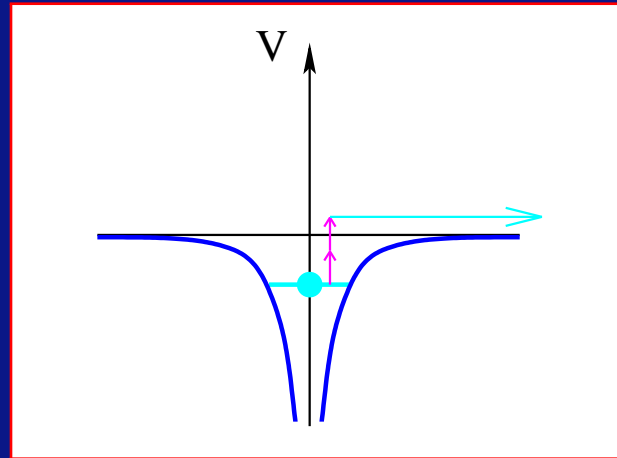
This talk tries to invoke discussions (and leaves many questions unanswered . . .).

Qualitative strong-field ionization models



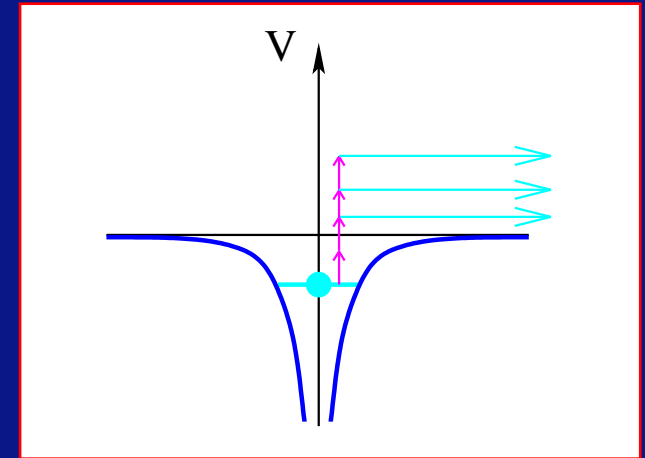
REMPI

Resonance-enhanced multiphoton ionization



Multiphoton Ionization

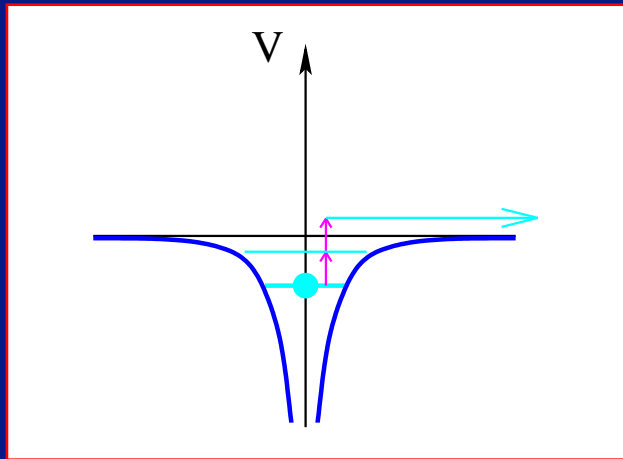
Non-resonant multiphoton ionization



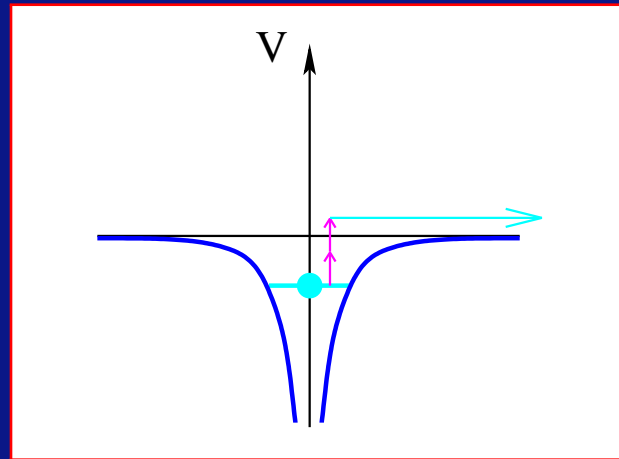
ATI

Above-threshold ionization

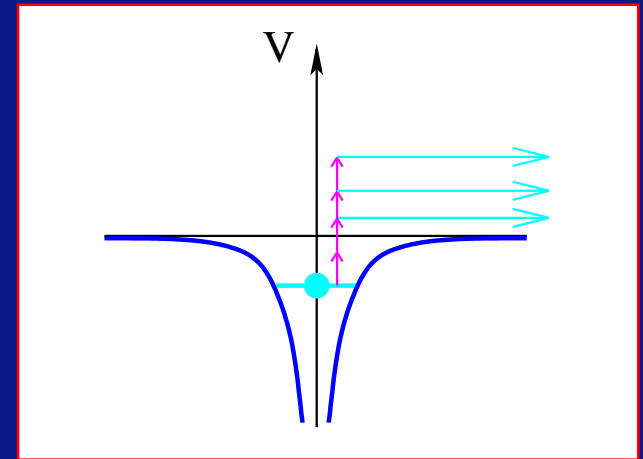
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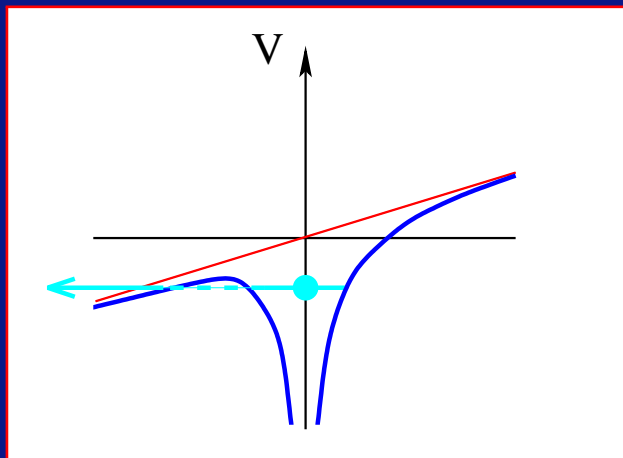
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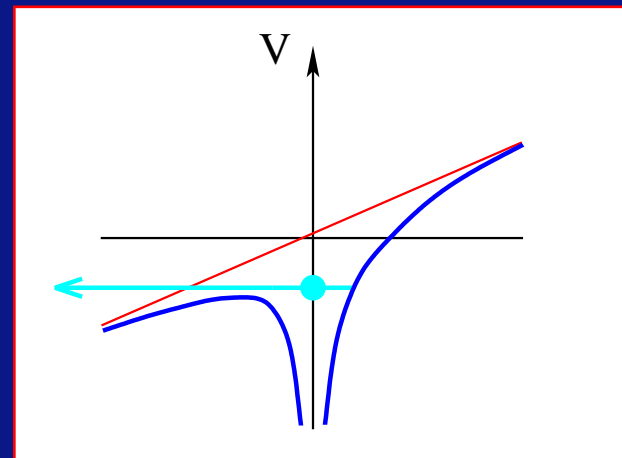
Multiphoton Ionization
Non-resonant multiphoton ionization



ATI
Above-threshold ionization



Tunnel ionization



Over-the-barrier ionization

Multiphoton regime

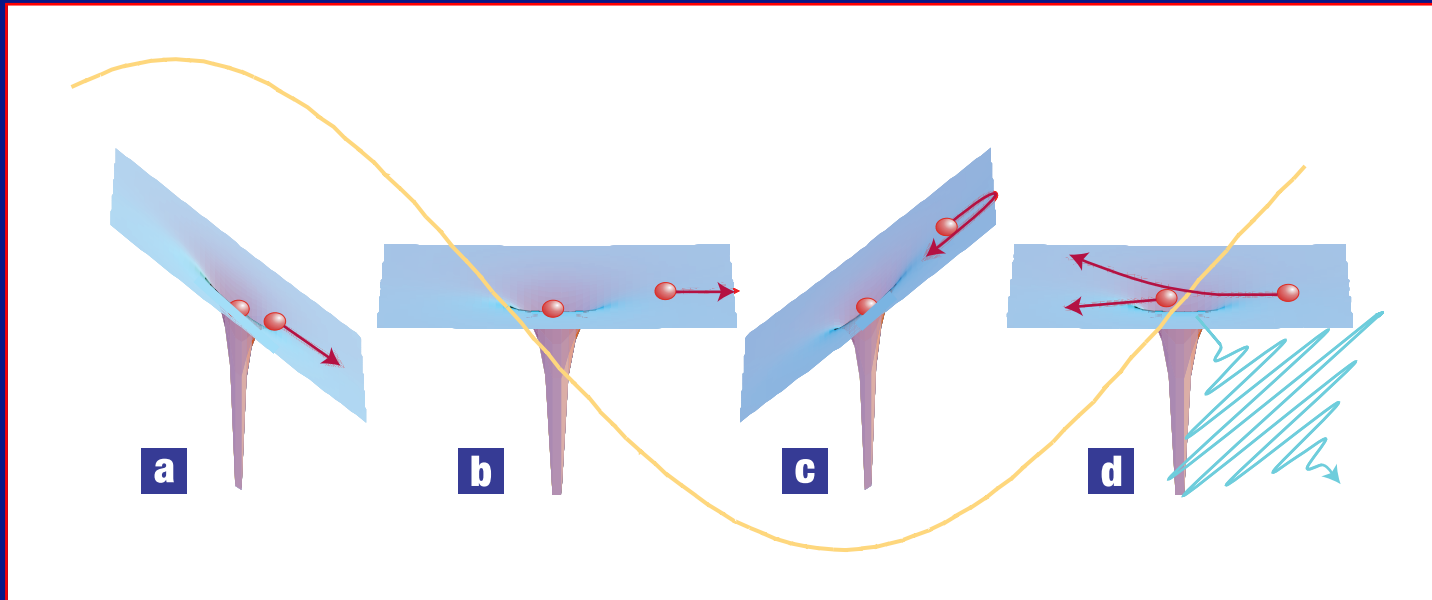
(upper row)

vs.

Quasi-static regime

(lower row)

Corkum's 3-step model:

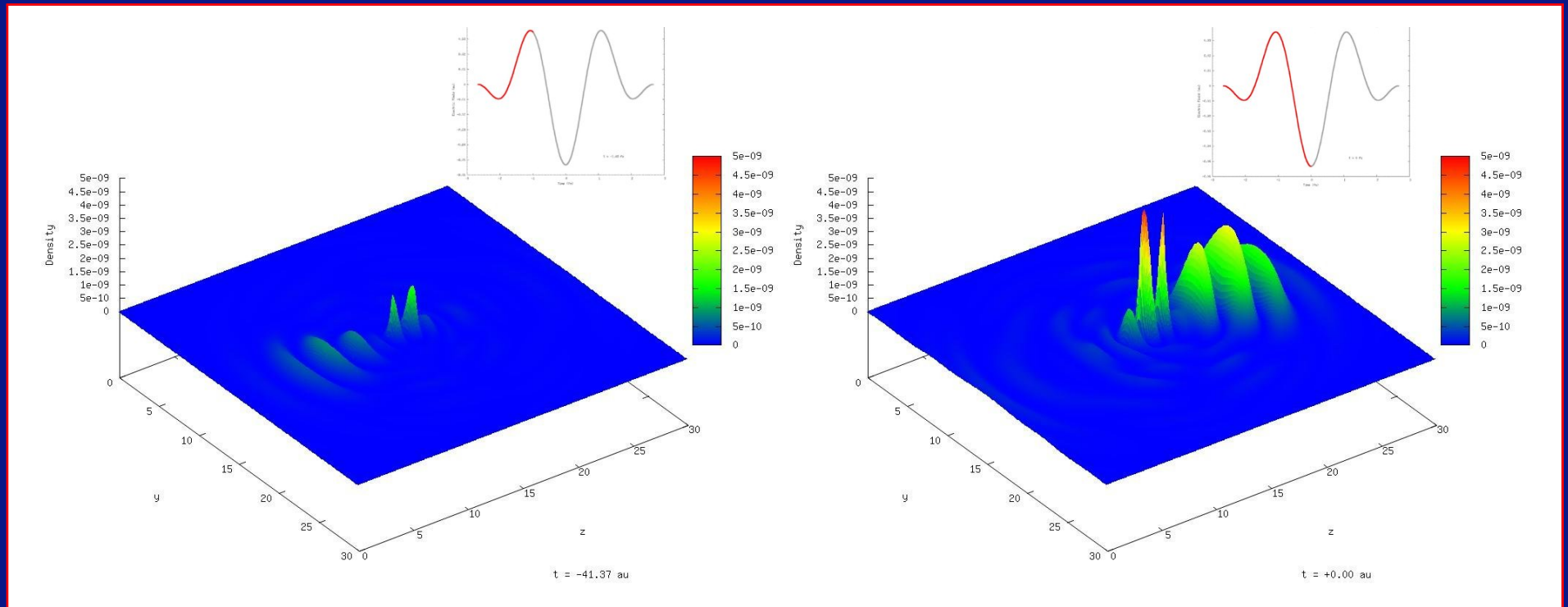


review: P. B. Corkum and F. Krausz, *Nature Phys.* **3**, 381 (2007)

1. Electron escapes through or over the electric-field lowered Coulomb potential (a).
2. Electronic wavepacket moves away until the field direction reverses (b) and is (partly) driven back to its parent ion (c).
3. The returning electron may (d)
 - scatter elastically (electron diffraction)
 - scatter inelastically (excitation, dissociation, double ionisation, . . .)
 - recombine radiatively (high-harmonic radiation).

→ **time-resolved imaging, attosecond pulses, . . .**

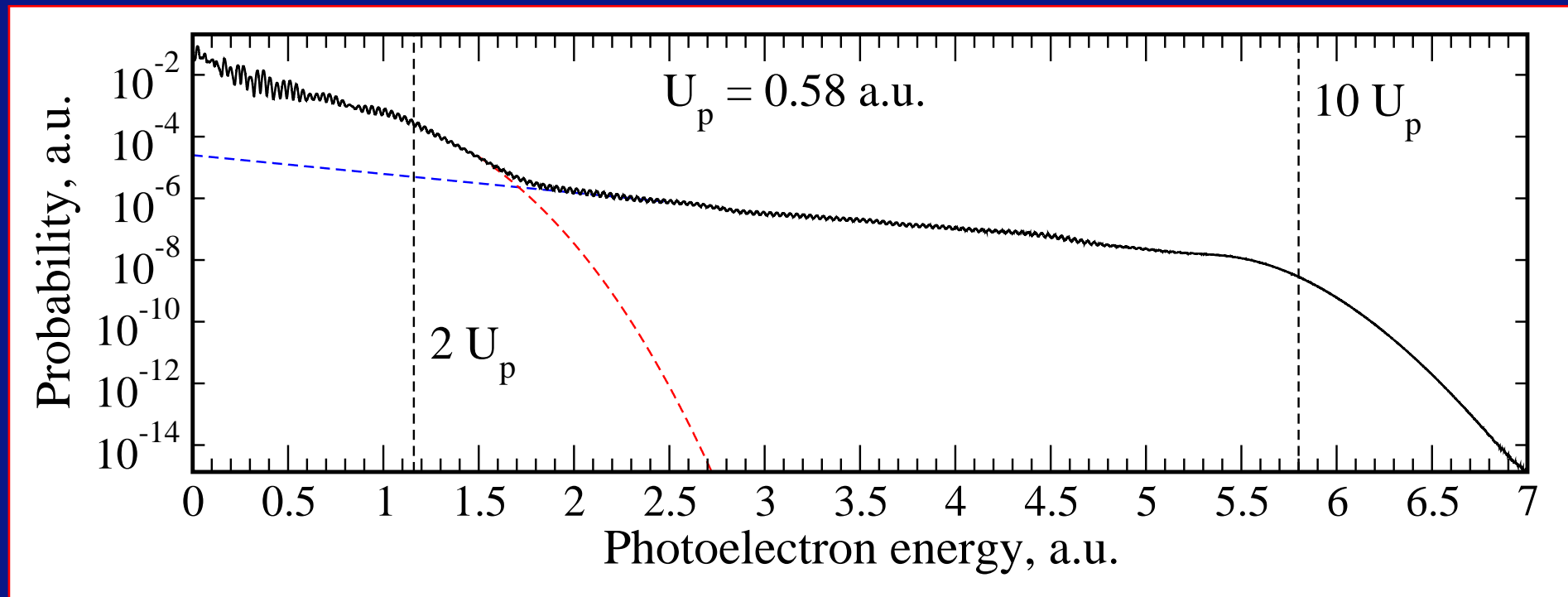
Example electronic wavepacket (H_2^+)



Electronic wavepacket at two different times within a 2-cycle laser pulse.
(Only the continuum part is shown.)

→ **strongly driven dissipative quantum system.**

Example electron spectrum (ATI)



Hydrogen atom (laser parameters: 1300 nm; 6 cycles; \cos^2 ; $I_{\max} = 10^{14}$ W/cm²).

Direct electrons: 0 to about 2 times the ponderomotive energy $U_p = I/(4\omega^2)$.

Rescattered electrons: dominate spectrum beyond $2 U_p$.

→ extremely highly non-linear process.

From atoms to molecules:

Atoms in ultrashort intense laser fields:

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Tunneling ionization rate: (see, e. g., Landau-Lifshitz)

$$\Gamma(F) \propto \exp \left[-\frac{2 (2 E_b)^{3/2}}{3F} \right]$$

with field electric strength F and electron's binding energy E_b .

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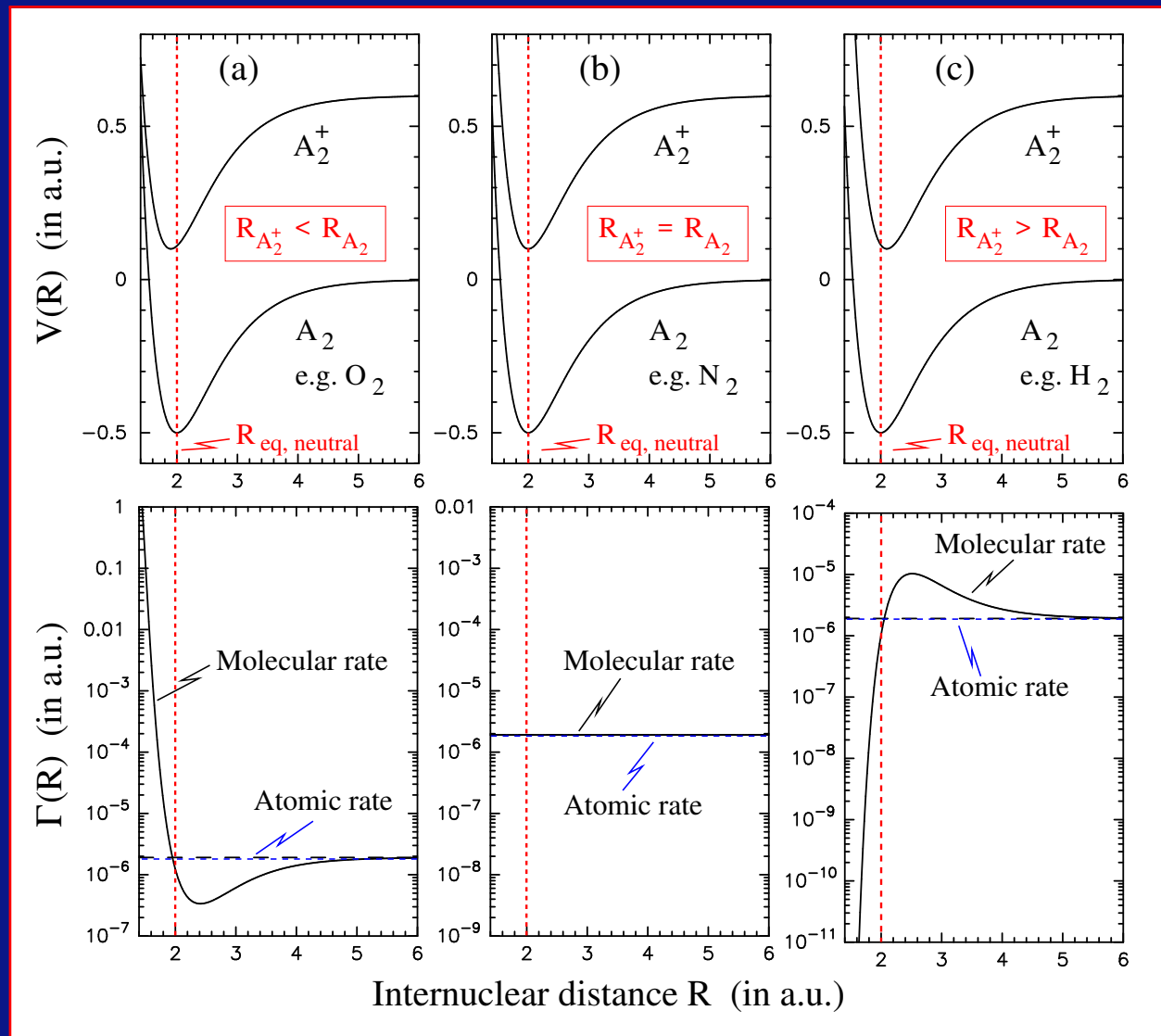
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Molecules: Nuclear-geometry dependence of tunnel ionization?

Molecular effects: R -dependence (extnd. ADK model)



ADK model:

$$\Gamma_{ADK} \propto \exp\left(-\frac{2(2I_P)^{3/2}}{3F}\right)$$

with

Γ_{ADK} : ionization rate

F : field strength

I_P : ionization potential

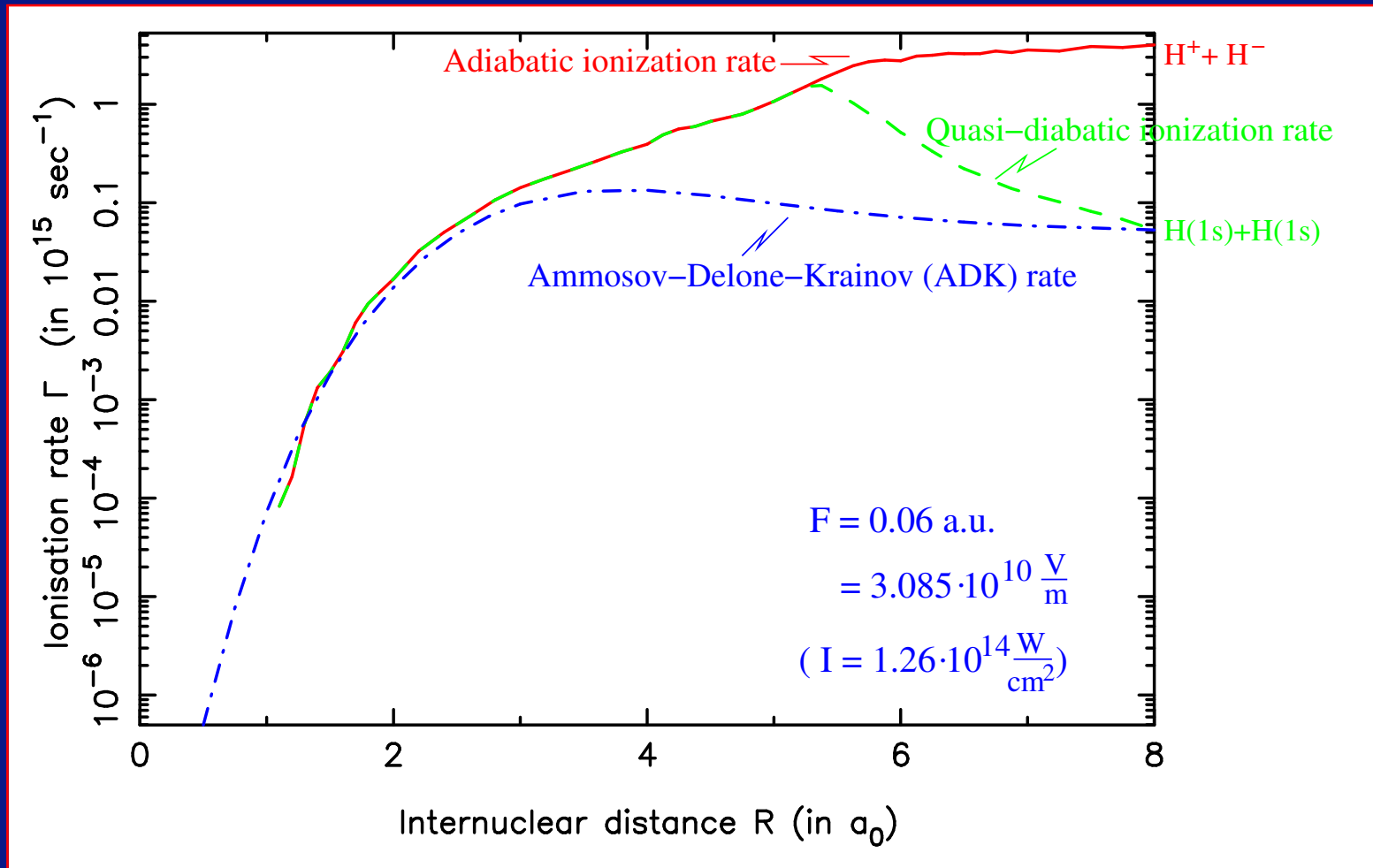
Extended ADK model:

Replace ionization potential I_P with

$$E^{A_2^+}(R) - E^{A_2}(R)$$

No Franck-Condon distribution for, e.g., H_2 or O_2 [A. S., *J. Phys. B* 33, 4365 (2000)].

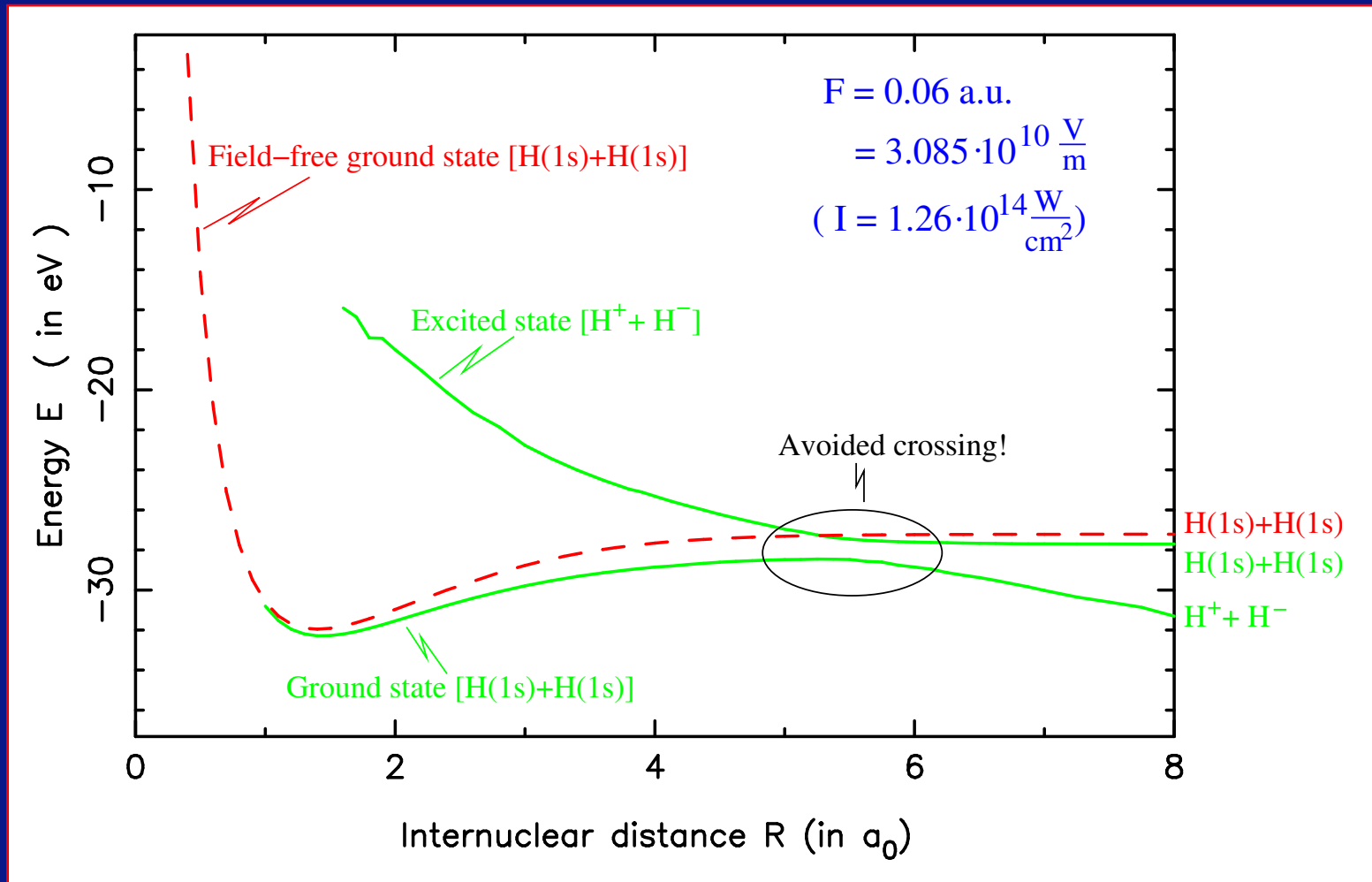
R-dependent ab initio dc ionization rate for H₂



Ab initio calculation (dc field) confirms: ionisation rate of H₂ strongly R dependent.

[A. S., *Phys. Rev. A* **61**, 051402 (R) (2000); *Phys. Rev. A* **66**, 063408 (2002).]

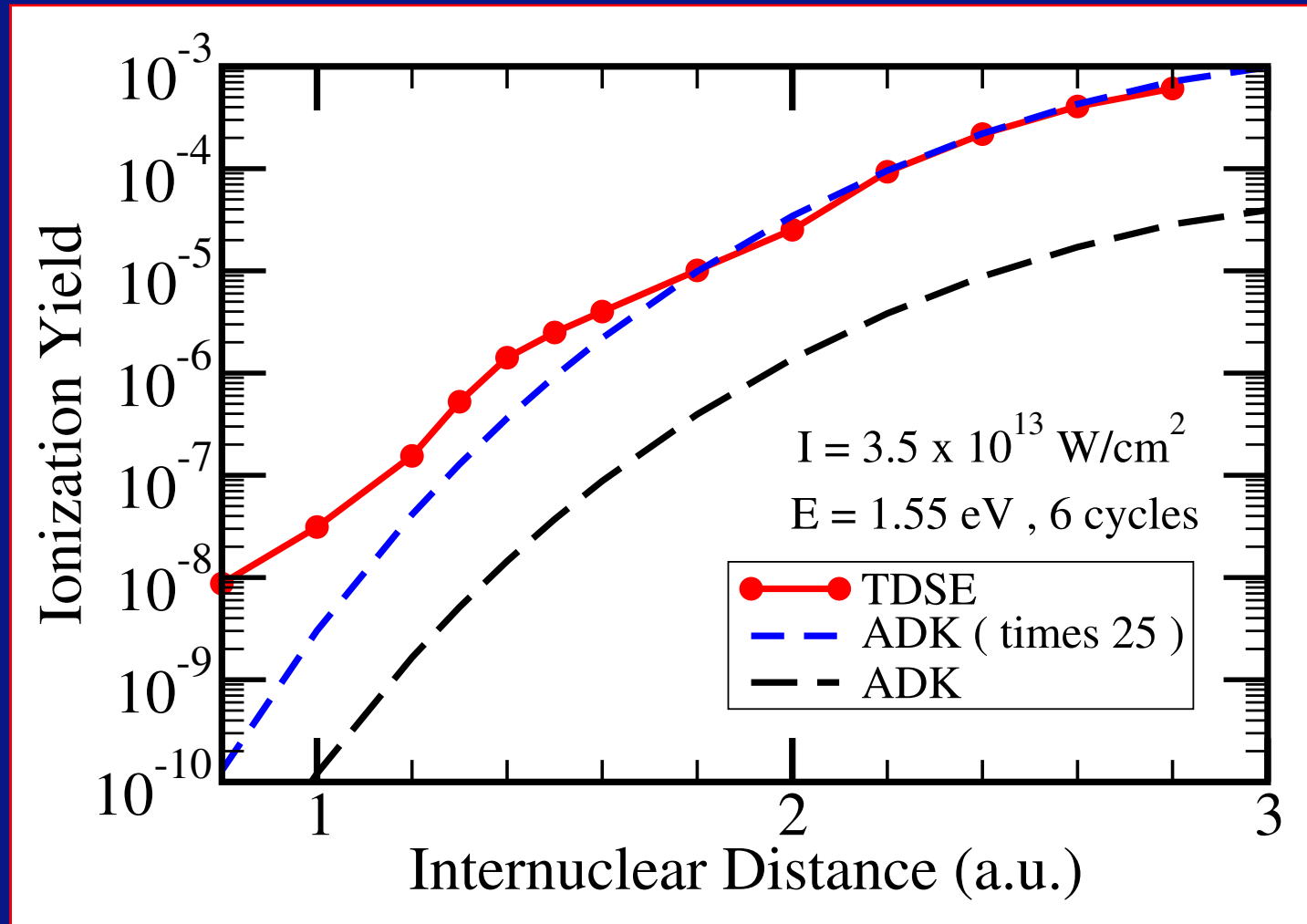
Furthermore: bond softening in neutral H₂



Ab initio complex-scaling calculation (dc field) of H₂ in an intense field.

[A. S., *Phys. Rev. A* **61**, 051402 (R) (2000); *Phys. Rev. A* **66**, 063408 (2002).]

Validity of quasi-static approximation for H₂



Full dimensional solution of TDSE: M. Awasthi, Y.V. Vanne, A. S., *J. Phys. B* **38**, 3973 (2005) [method];
M. Awasthi and A. S., *J. Phys. B:* **39**, S389 (2006) [R dependence].

Strong-Field Control: “Lochfrass” (Proposal)

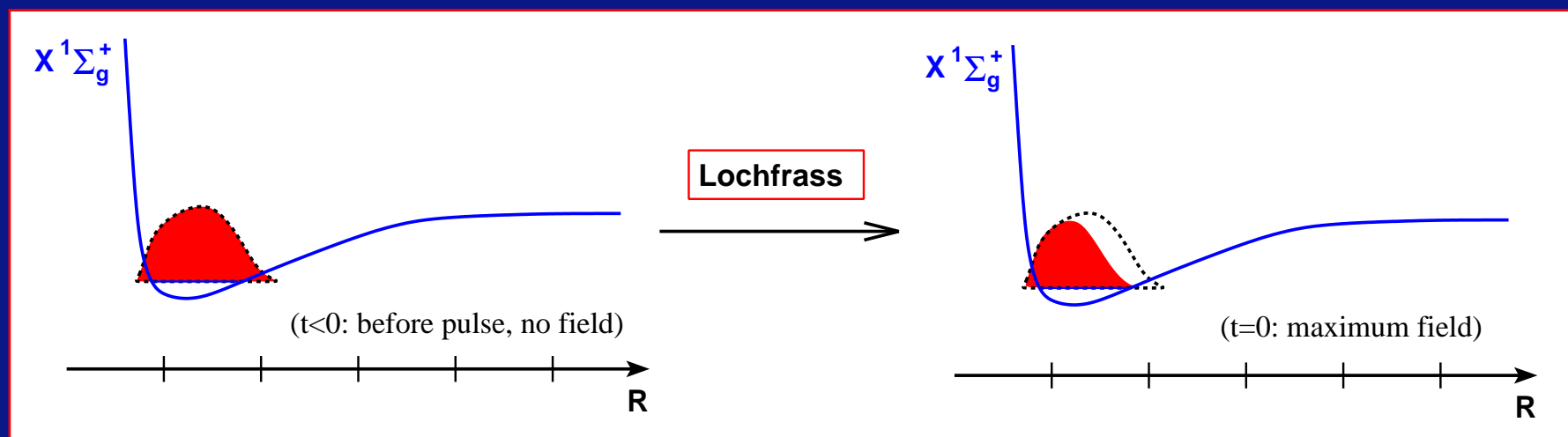
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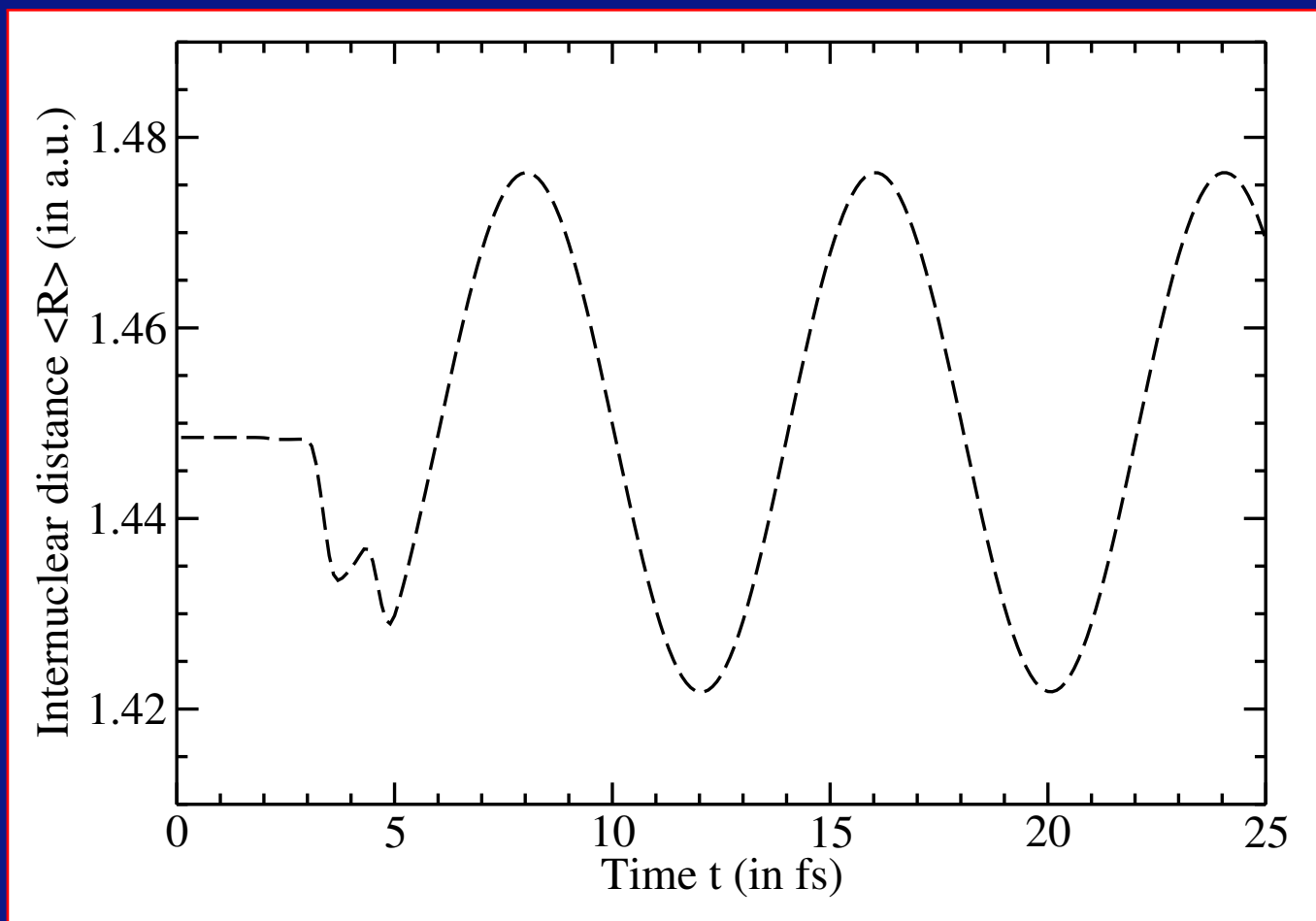
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A **superposition state** of the **ionized** and the **neutral** molecule!

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- **Purely quantum-mechanical effect:**
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- **Highly non-linear process:**
A second (probe) pulse should detect a time-dependent ionization signal.

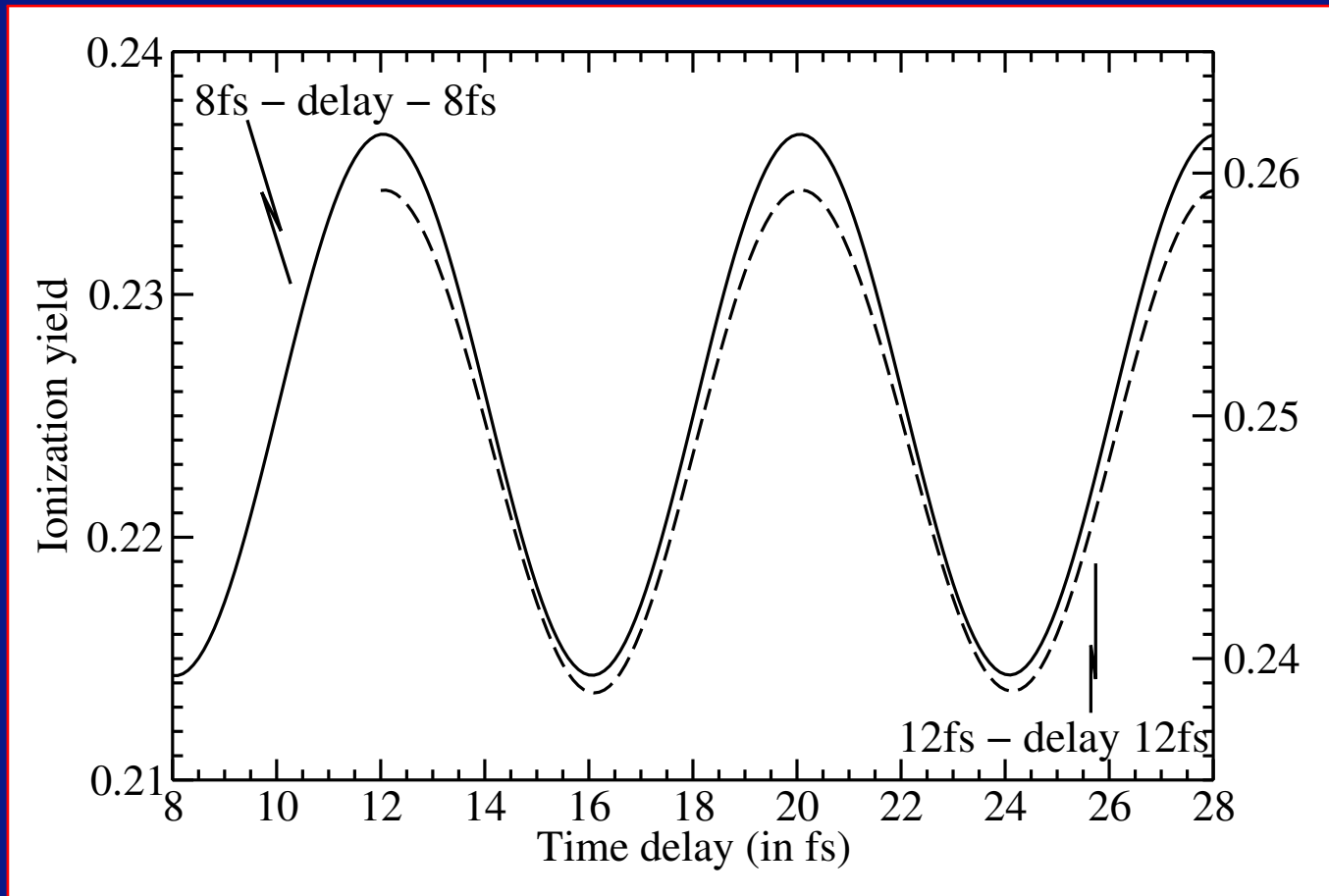
Wave-packet study (results)



Peak intensity: $I = 6 \cdot 10^{14} \text{ W/cm}^2$, Wavelength: $\lambda = 800 \text{ nm}$, Length: 8 fs.

Formation of a H_2 wavepacket by “Lochfrass” (“eating a hole”).

Wave-packet detection: Pump-probe



Identical pulses, Peak intensities: $I = 6 \cdot 10^{14} \text{ W/cm}^2$, Wavelength: $\lambda = 800 \text{ nm}$.

[E. Goll, G. Wunner, and A. S., *Phys. Rev. Lett.* **97**, 103003 (2006)]

Pump-probe experiment (MPI Heidelberg)

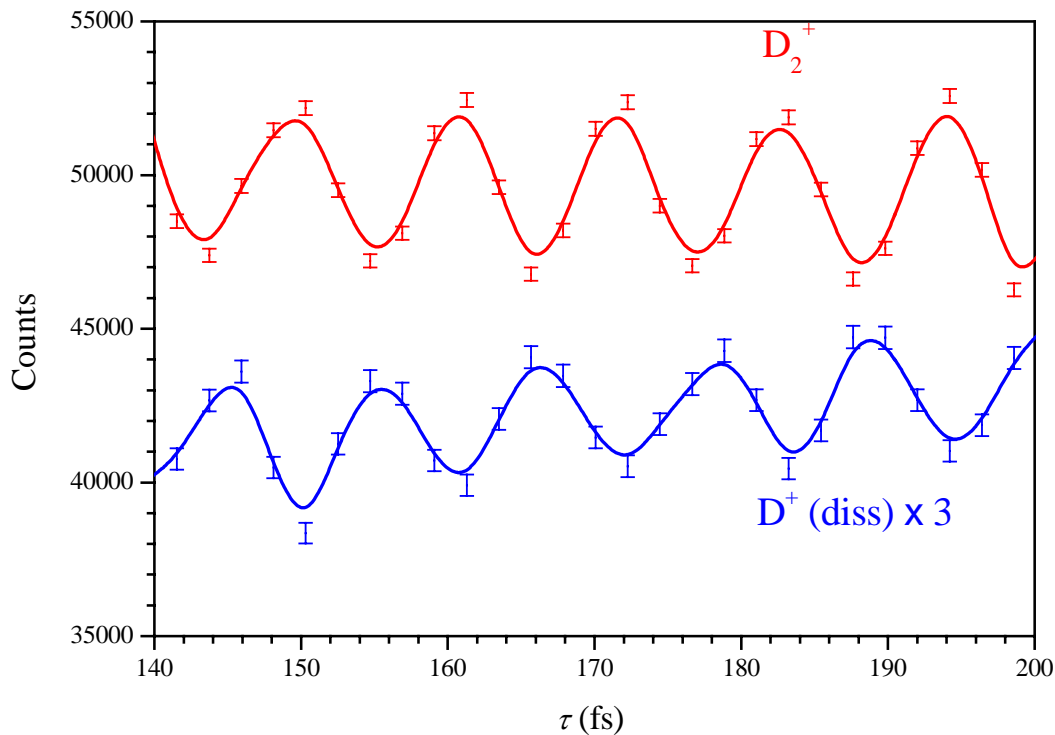


Figure 2

Parameters:

Two identical pulses,

$$I = 4(1) \cdot 10^{14} \frac{\text{W}}{\text{cm}^2},$$

$$\lambda = 795 \text{ nm},$$

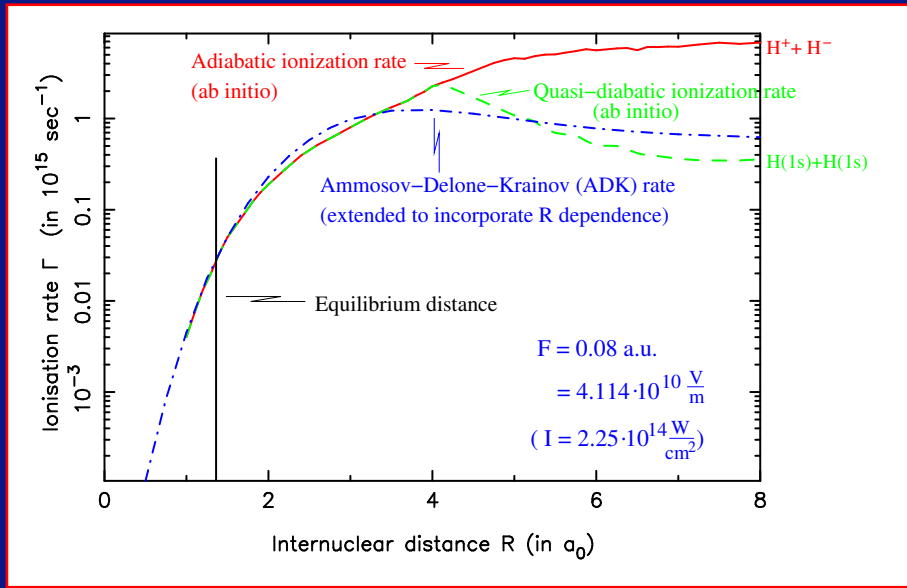
7 fs (FWHM).

[Fig. from Ergler et al.
Phys. Rev. Lett. **97**, 103004
(2006)]

→ Experiment observes the theoretically predicted oscillation!!!

[Note: expected oscillation period for D_2 : 11 fs (H_2 : 8 fs).]

Is it really Lochfraß?



“Lochfraß”

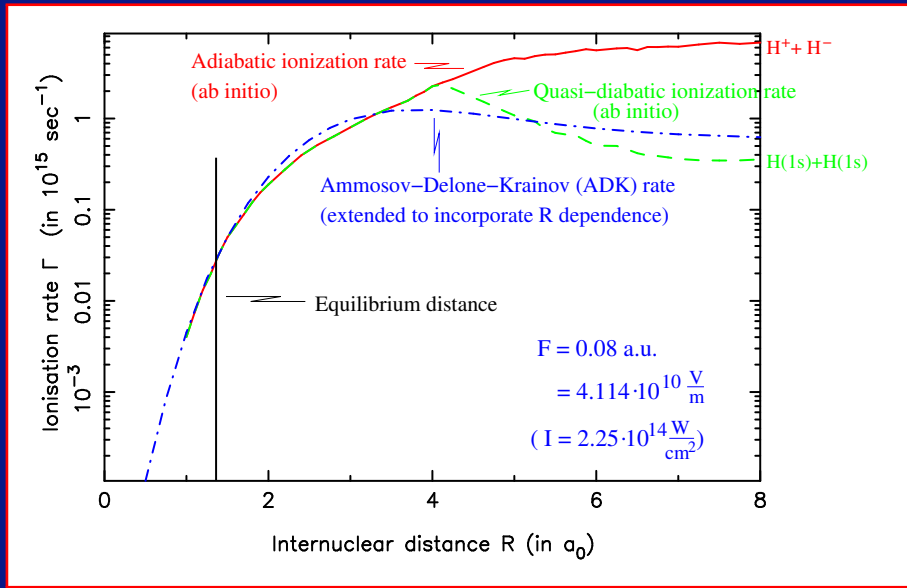
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Preferential ionization at large R :

If ionisation is fast enough,
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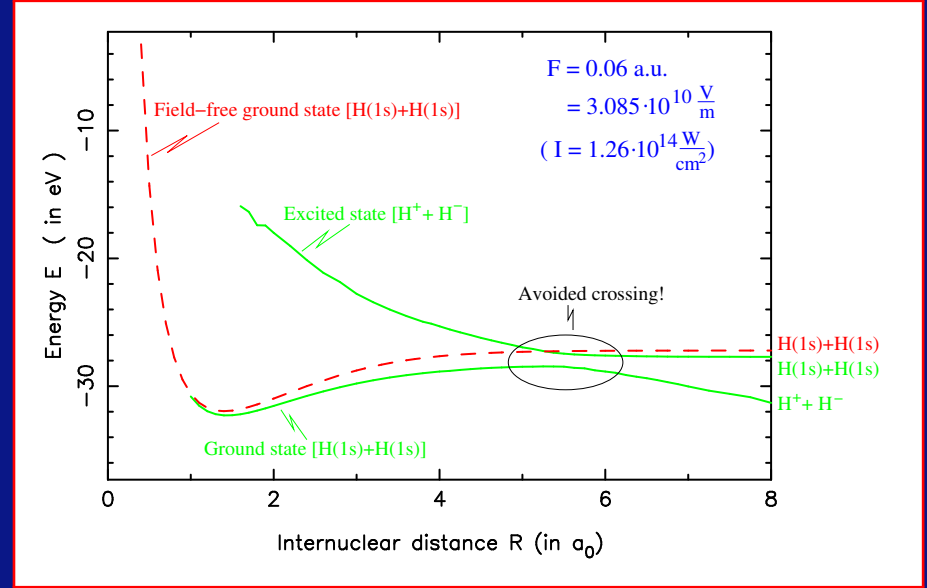
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Bond Softening

(caused by potential-curve distortion)

[A. S., *PRA* **61**, 051402(R) (2000)]

Field-induced lowering of potential curve:

The nuclear wavefunction escapes
over the suppressed barrier.

Modelling bond softening and Lochfraß

Full solution of the time-dependent Schrödinger equation:

- Beyond reach at the time of the proposal.

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Model Hamiltonian (nuclear motion with dissipation):

$$\hat{H}(R, t) = \hat{H}_0(R) + \Delta\hat{V}(R, F(t)) - \frac{i}{2}W(R, F(t))$$

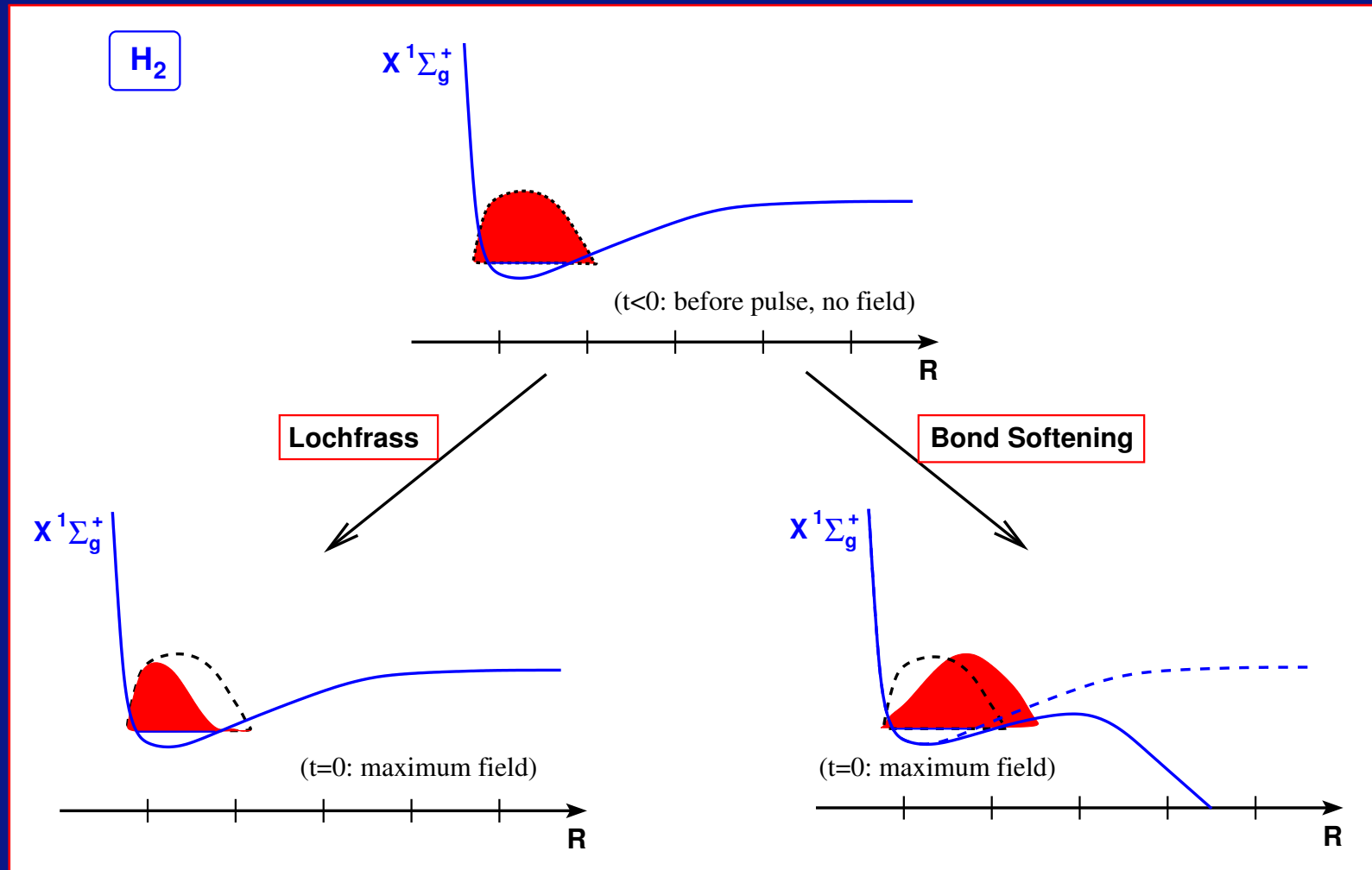
$\hat{H}_0(R)$: field-free time-independent Hamiltonian.

$\Delta\hat{V}(R, F(t))$: field-induced distortion of the potential curve.

$W(R, F(t))$: field-induced (quasi-static) ionization rate.

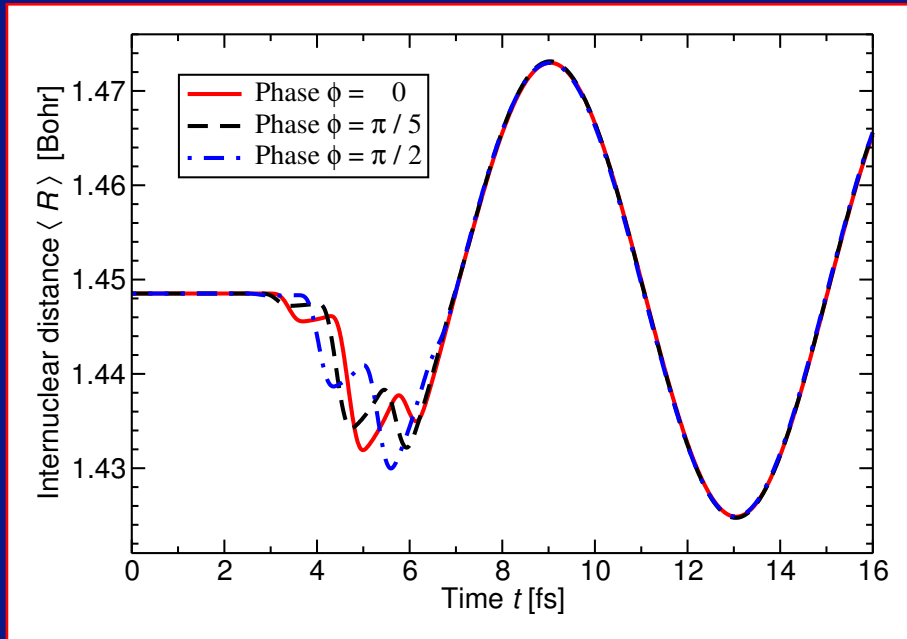
$F(t)$: time-dependent electric field component of the laser pulse.

How to experimentally determine the mechanism?

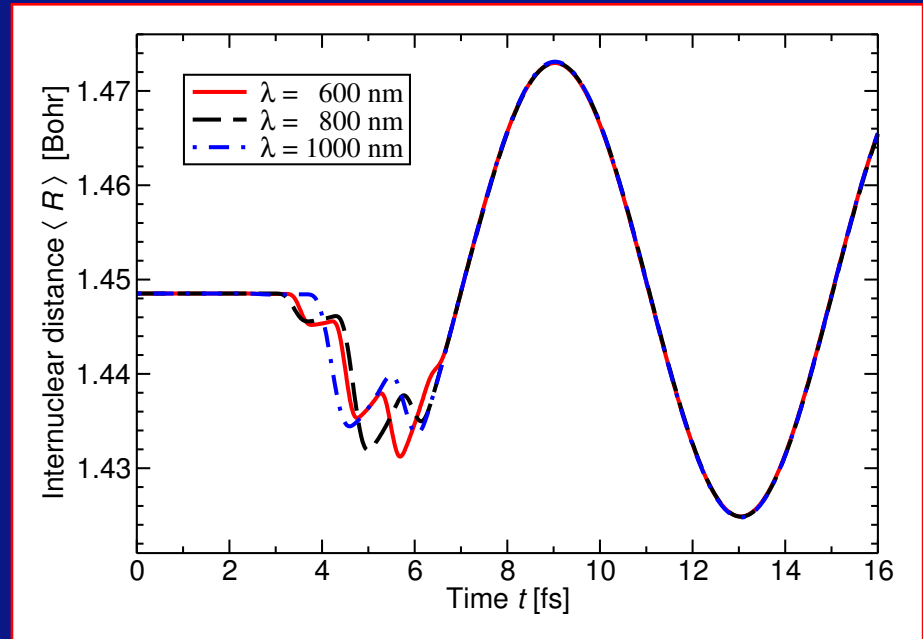


Lochfrass and Bond Softening may be distinguished by the absolute phase!!!

Robustness of Lochfraß



Variation of the **absolute** (carrier-envelope) **phase ϕ** of the ultrashort laser pulse.



Variation of the laser **wavelength λ** .

→ **Lochfraß is extremely robust!**

Determination of the mechanism

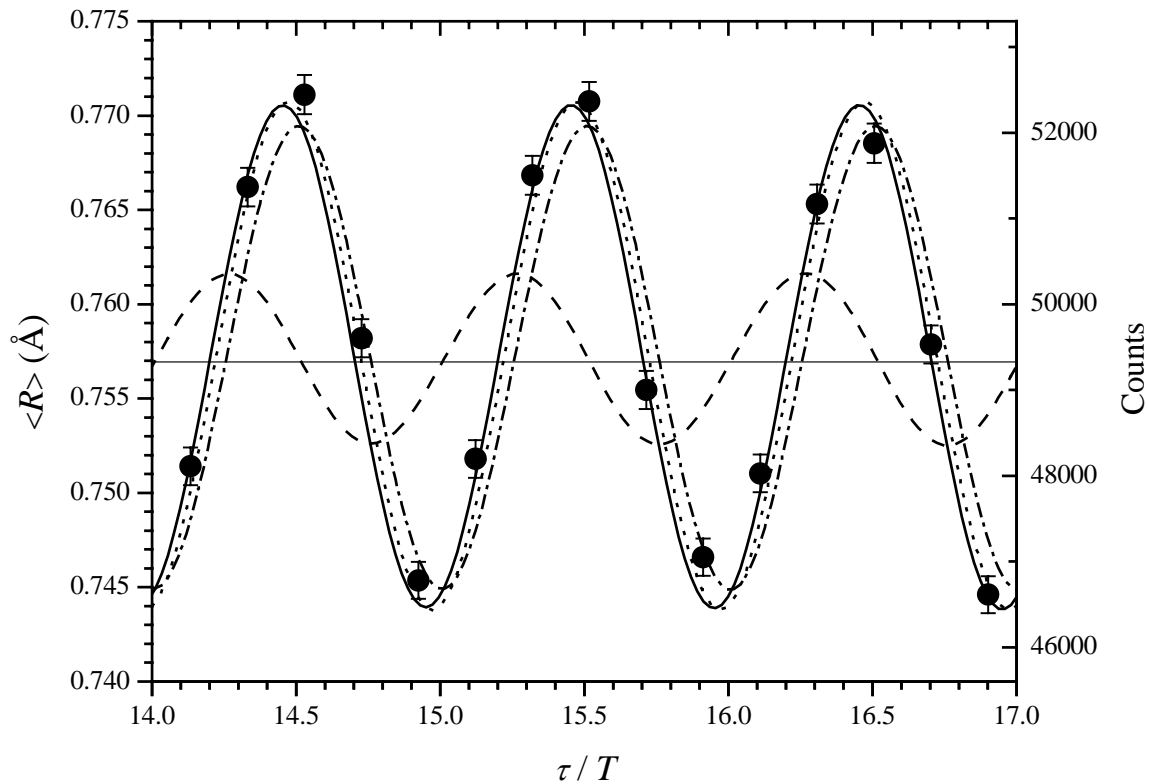


Figure 3

Dashed:

Bond Softening

Chain:

Lochfrass.

Solid:

Both effects,

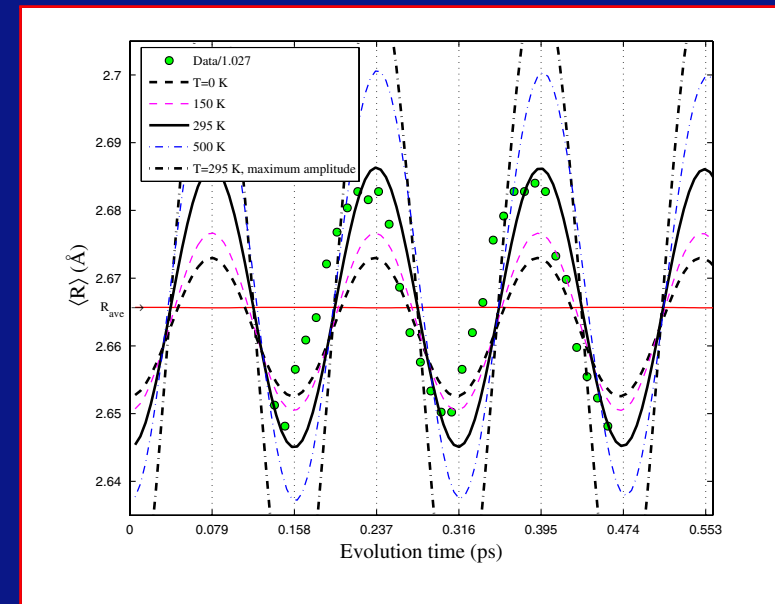
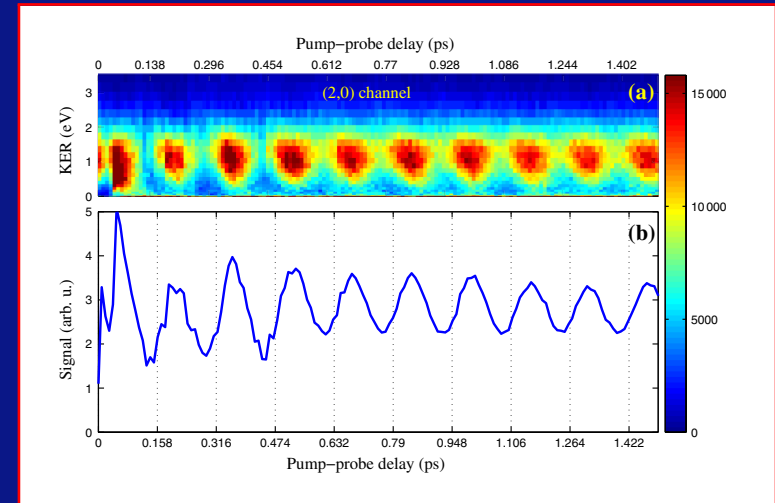
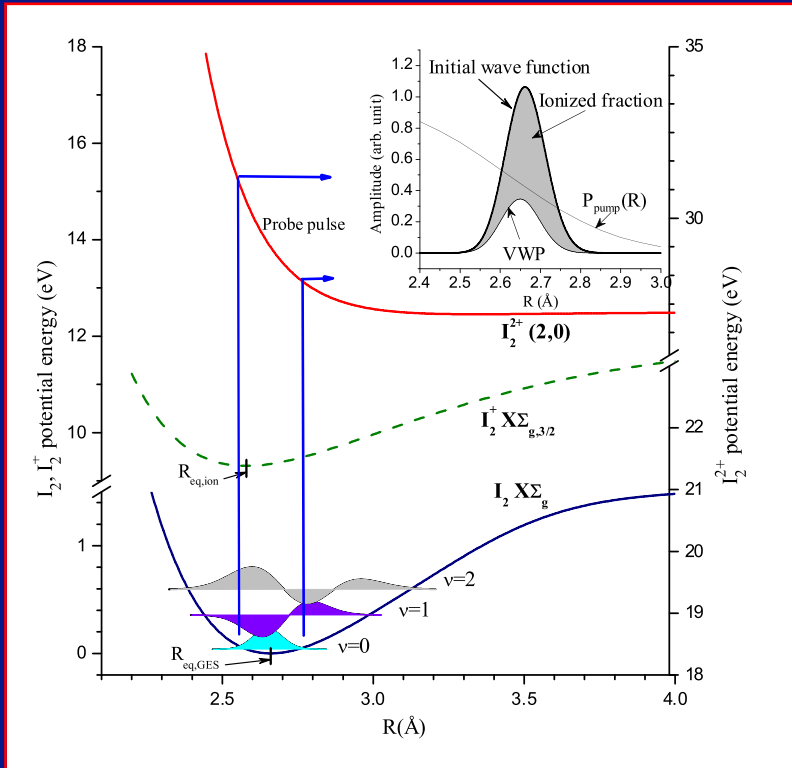
Circles:

Experiment.

[Fig. from Ergler et al.]

→ Lochfrass is the clearly dominating mechanism!!!

Lochfrass in I_2

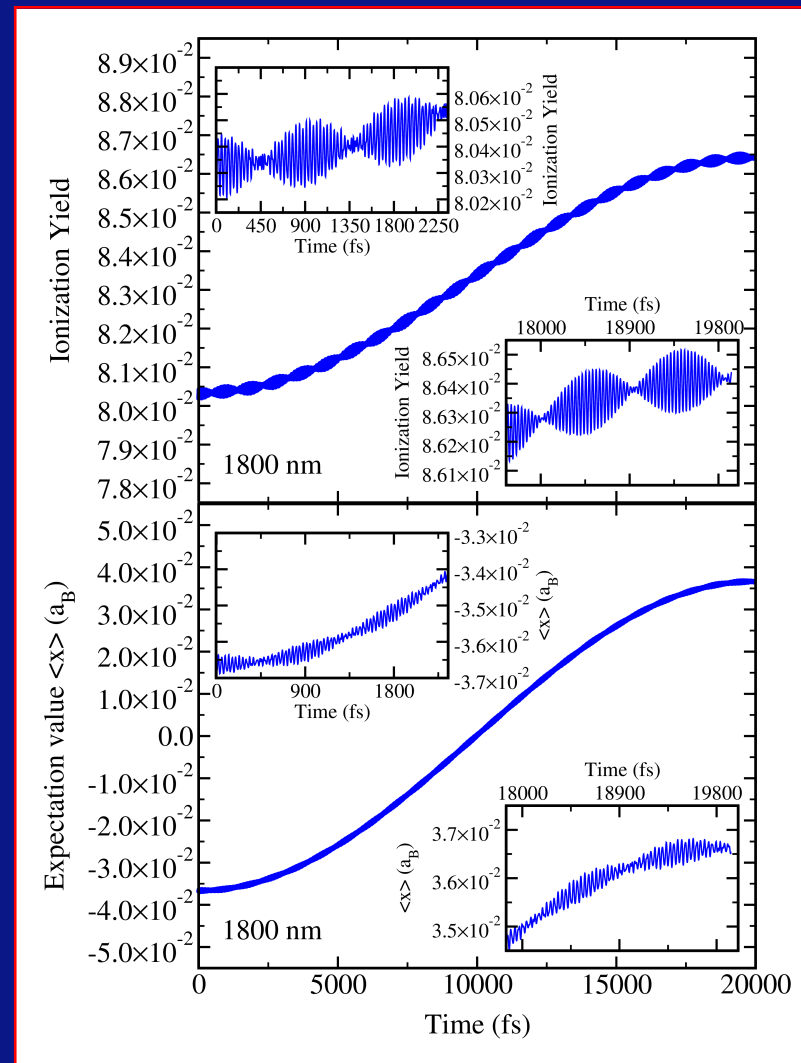
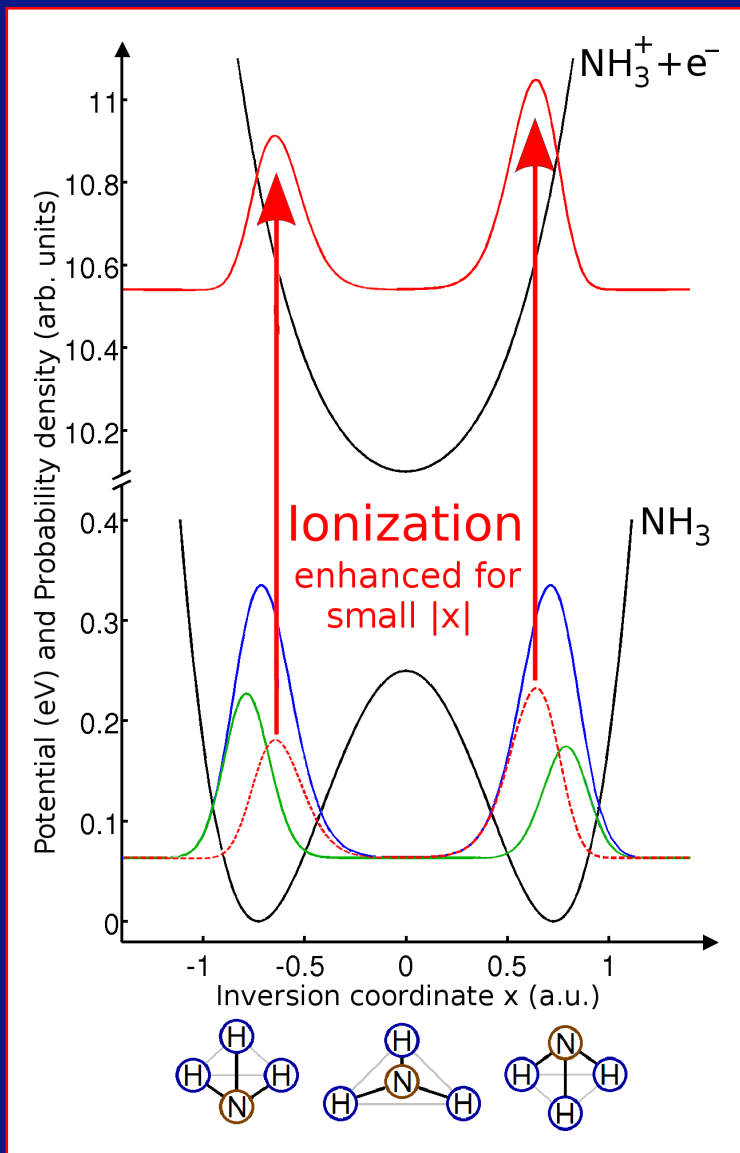


Lochfrass is again seen.

More incoherence in initial state improves coherent control scheme!

[L. Fang and G. N. Gibson,
Phys. Rev. Lett. **100**, 103003 (2008)]

Beyond diatomics: Lochfraß in ammonia (NH_3)



2 cycles (pump+probe), 1800 nm, 10^{14} W/cm^2

Real-time imaging of nuclear motion and tunneling possible [Förster et al., *Phys. Rev. A* **94**, 043405 (2016)].

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Why did the *Lochfraß* experiment work at all?

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Note: All results in perfect agreement with theoretical simulation!

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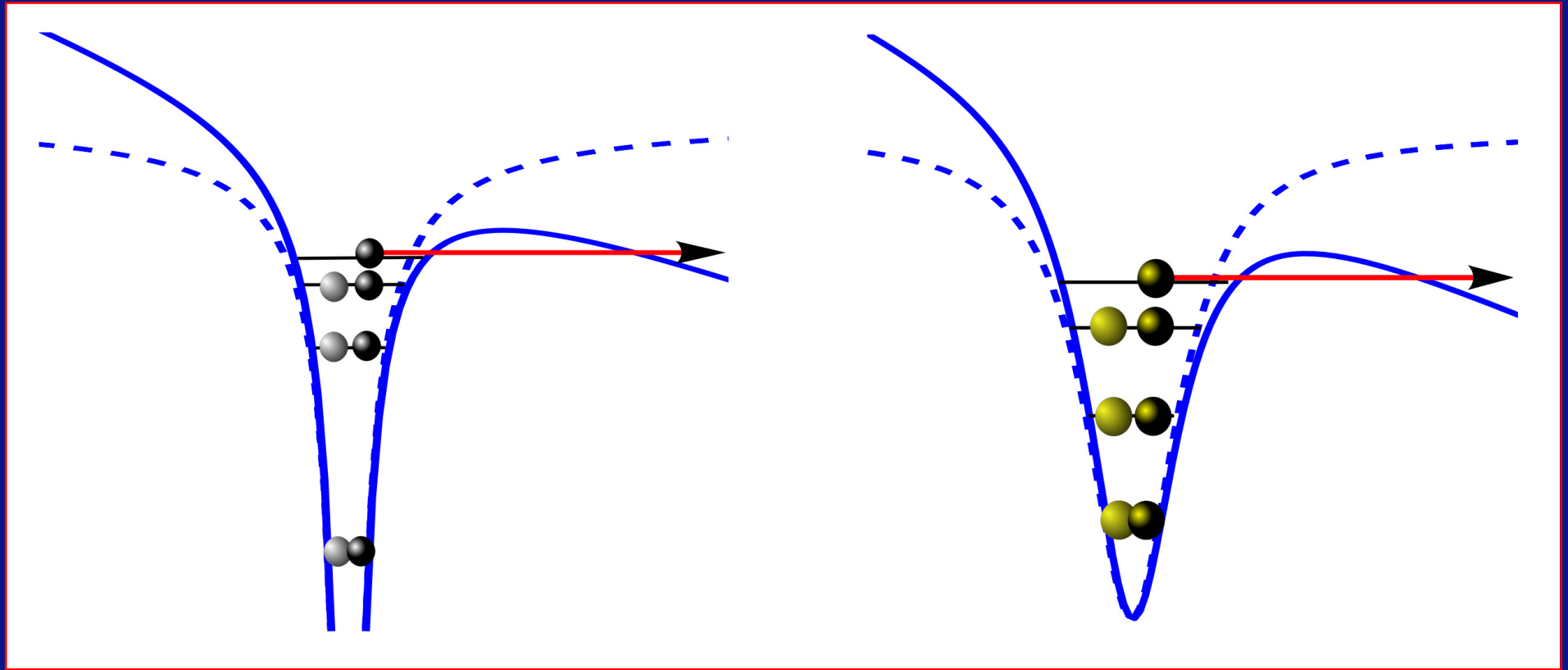
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- **This is QSIM19: where is the quantum simulator?**

Quantum-simulator for attosecond physics (I)



Atom in electric field

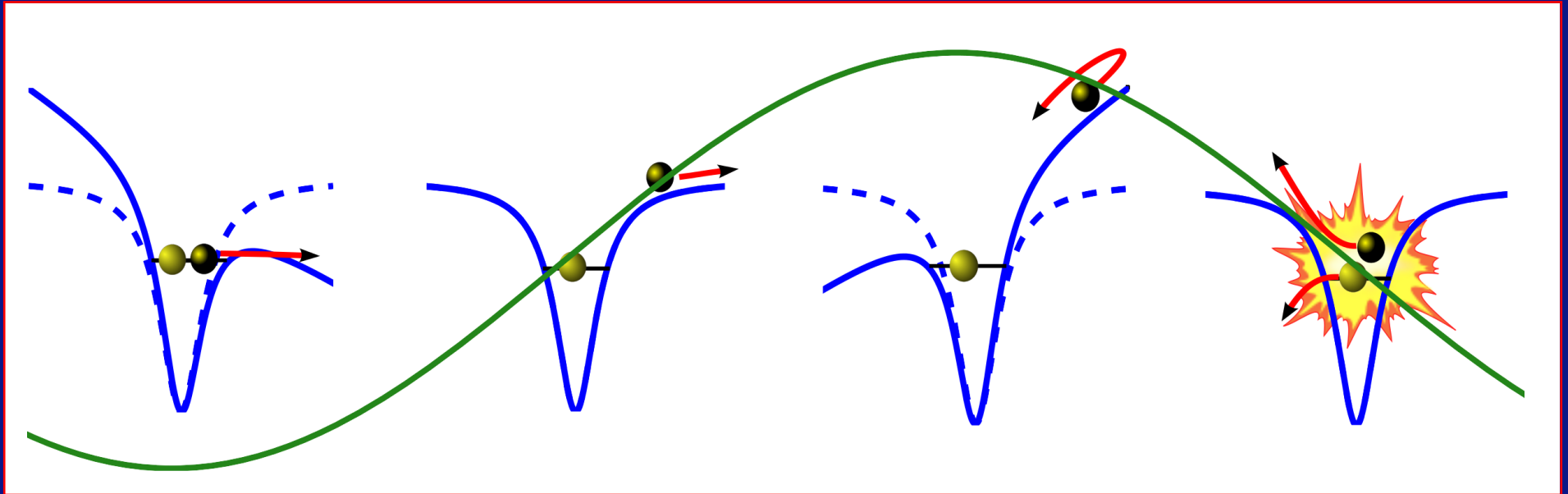
$$\hat{H}^{\text{LG}}(t) = \hat{H}_0 + \sum_{i=1}^N \mathbf{r}_i \cdot e\mathbf{E}$$

Atoms in dipole trap

$$\hat{\mathcal{H}}^{\text{LG}}(t) = \hat{\mathcal{H}}_0 + \sum_{i=1}^N \mathbf{r}_i \cdot \mu\mathcal{B}'$$

Mapping of electric field \mathbf{E} on magnetic-field gradient \mathcal{B}' .

Quantum-simulator for attosecond physics (II)



Atom in electric field

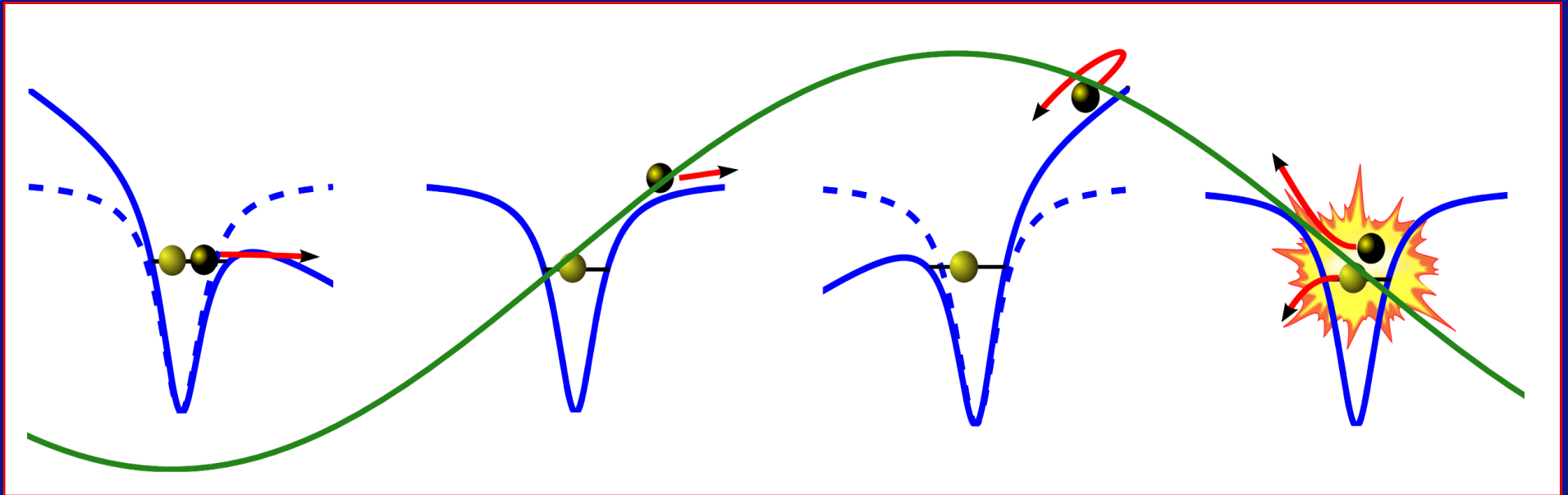
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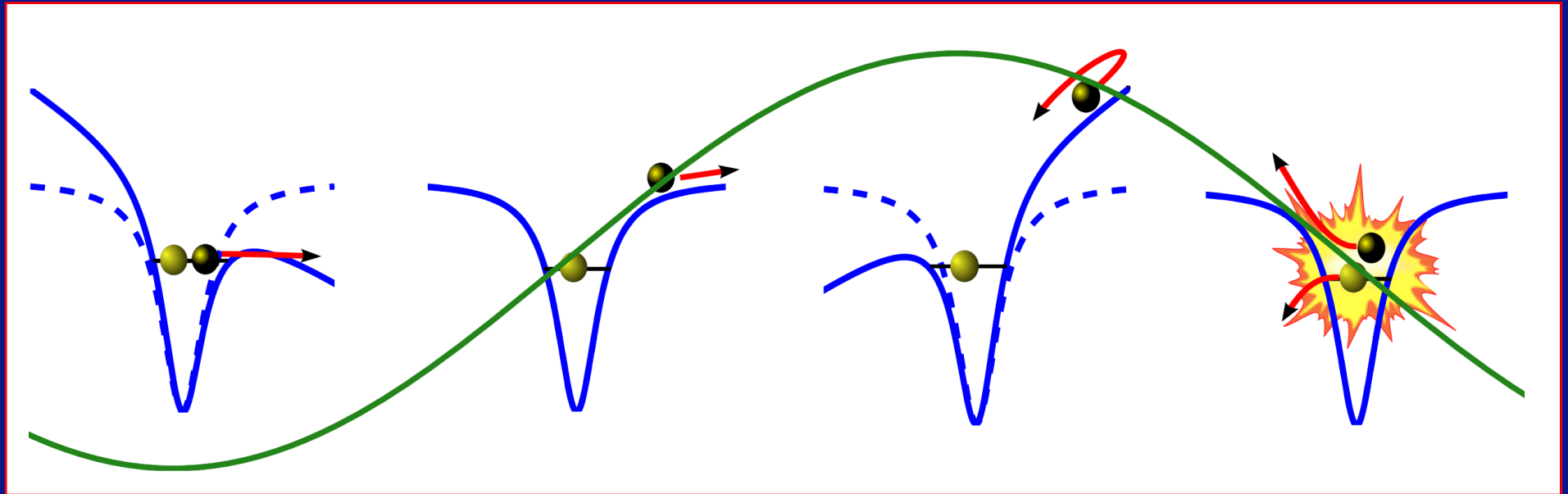
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→ attoscience in slow motion!

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Key question: does it work with realistic experimental parameters?

Possible experimental realization (cf. S. Jochim's set-up)

The experiment uses fermionic Li atoms.

The optical trap potential is **effectively one-dimensional**: aspect ratio 10:1.

Potential:

$$\mathcal{V}_L(z) = \alpha \mathcal{V}_0 \left[1 - \frac{1}{1+(z/z_r)^2} \right]$$

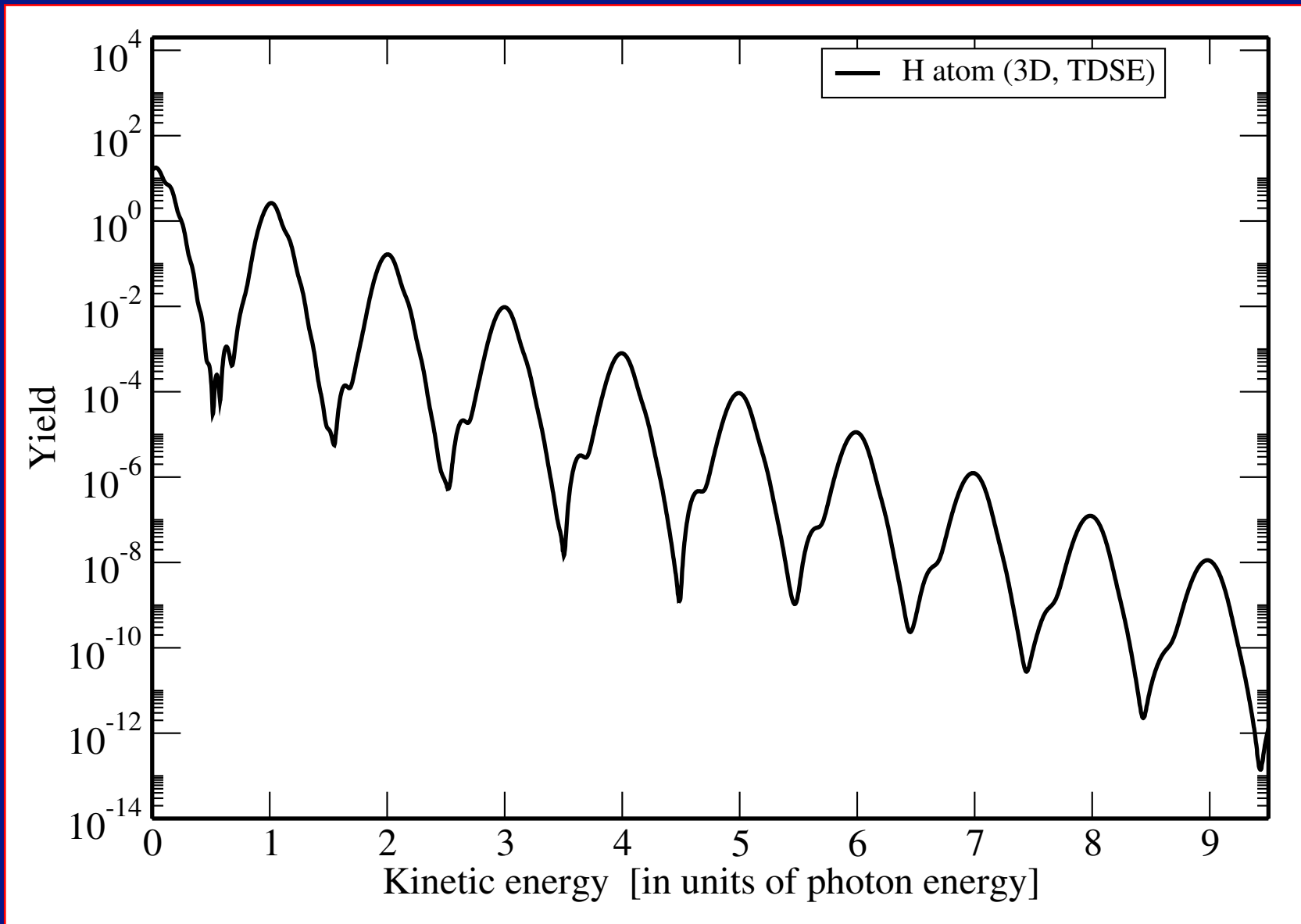
with variable parameter α , basic trap depth $\mathcal{V}_0/k_b = 3.33 \mu\text{K}$ (Boltzmann constant k_b), and the Rayleigh length $z_r = \pi w_0^2/\lambda$ ($\lambda = 1064 \text{ nm}$).

Mapping (equal Keldysh parameters and binding energies):

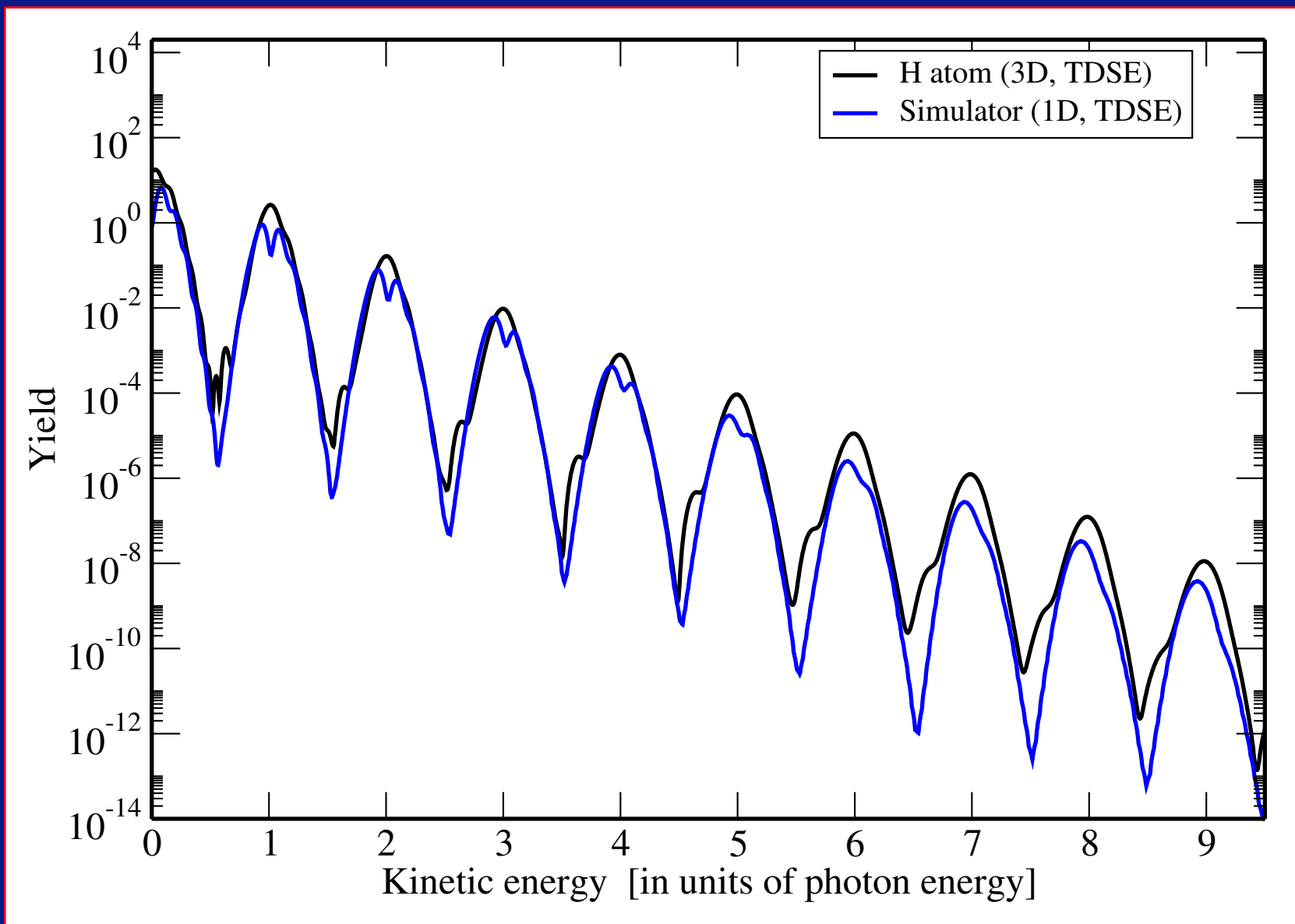
$$\gamma_e := \omega_e \frac{\sqrt{2m_e I_p}}{eE_0} = \omega \frac{\sqrt{2m_a E_b}}{\mu \mathcal{B}'_0} =: \gamma_a \quad \beta_e := \frac{I_p}{\hbar\omega_e} = \frac{E_b}{\hbar\omega} =: \beta_a \quad .$$

where I_p and E_b are the binding energies of the ground states of the field-free Hamiltonians.

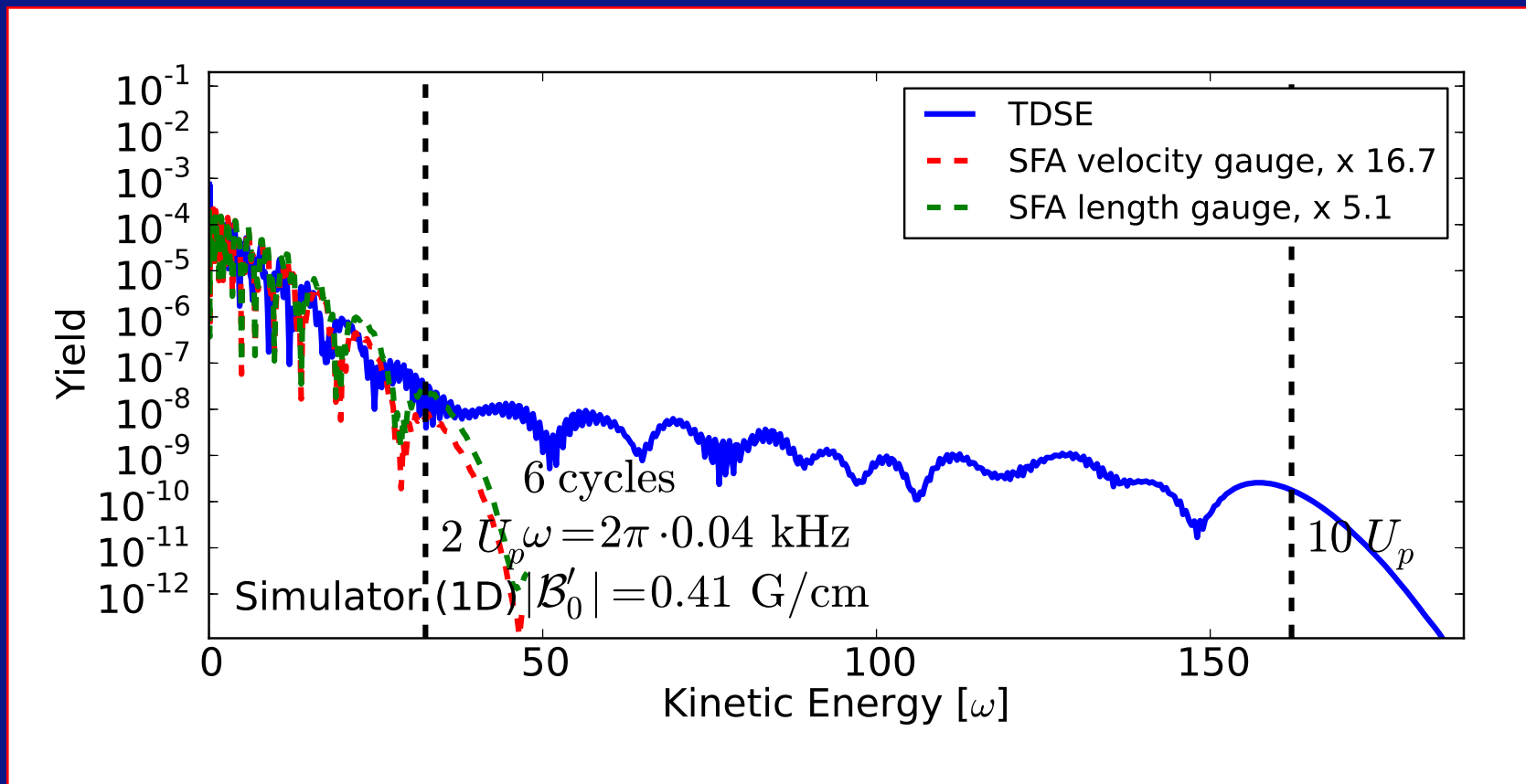
Quantum simulator in multiphoton regime (I)



Quantum simulator in multiphoton regime (II)



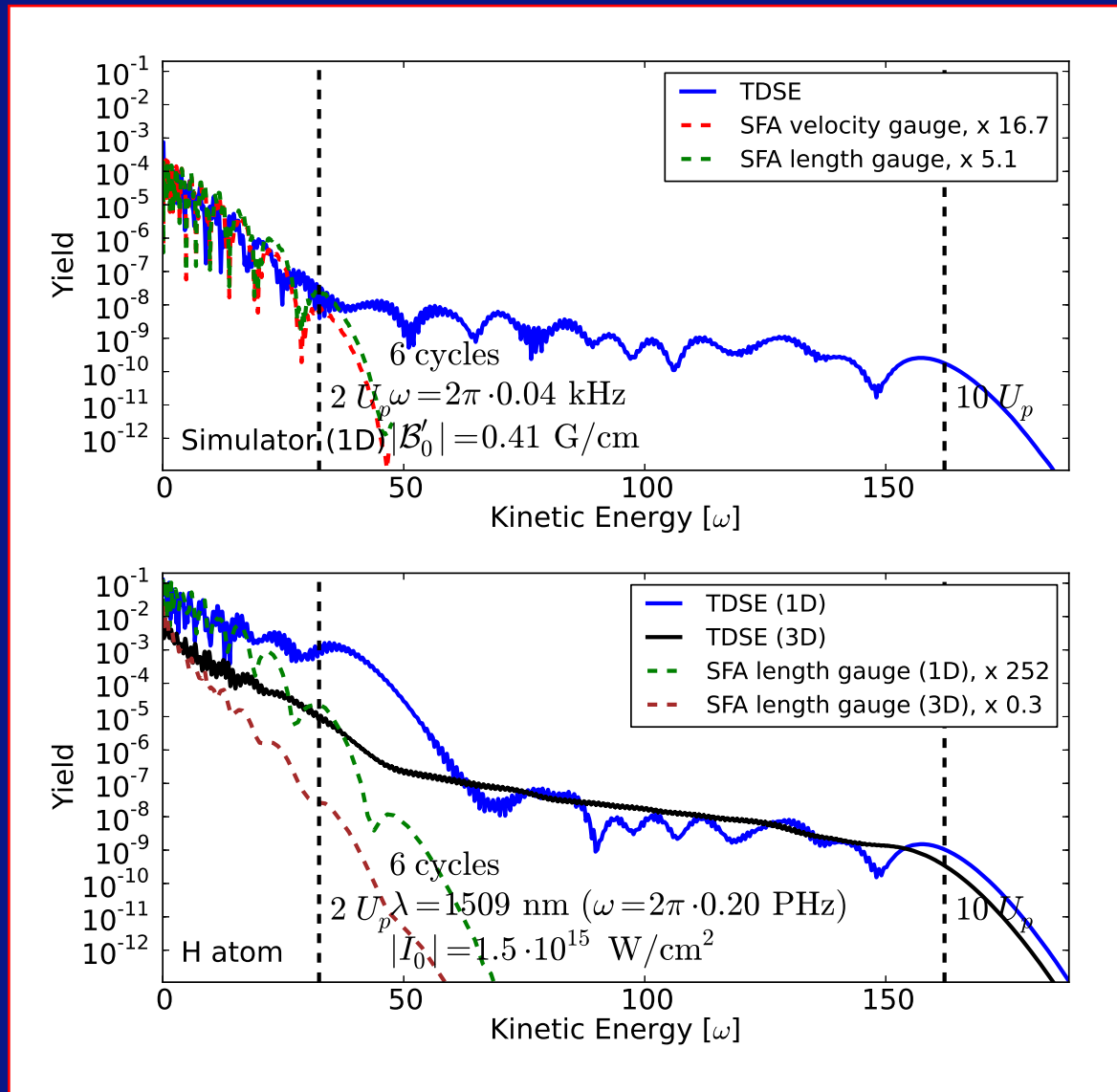
Quantum simulator in quasi-static regime (I)



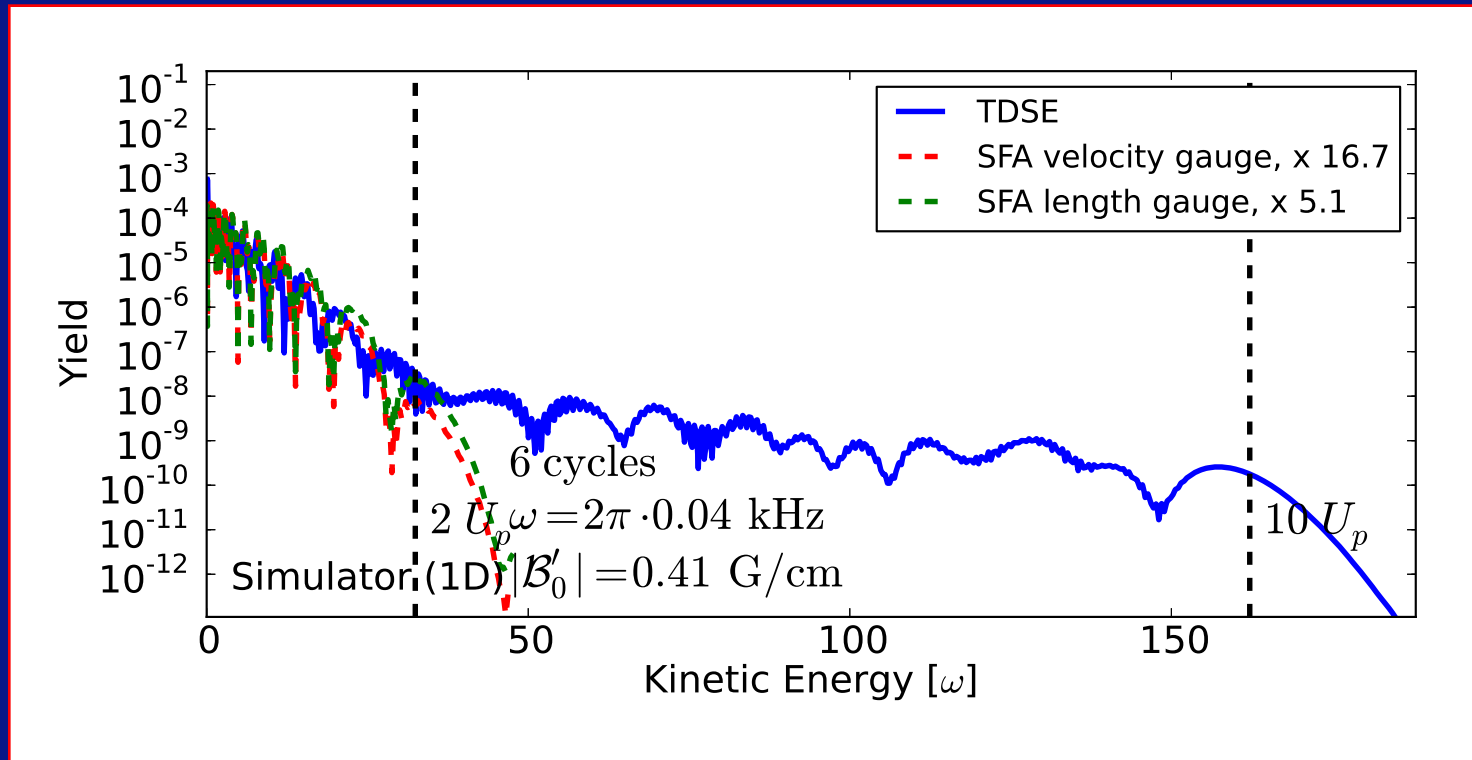
Characteristic features: direct emission ($< 2U_p$), plateau between 2 and $10 U_p$.

SFA (strong-field approximation): very popular, **long-range Coulomb interaction between electron and remaining ion is ignored!**

Quantum simulator in quasi-static regime (II)

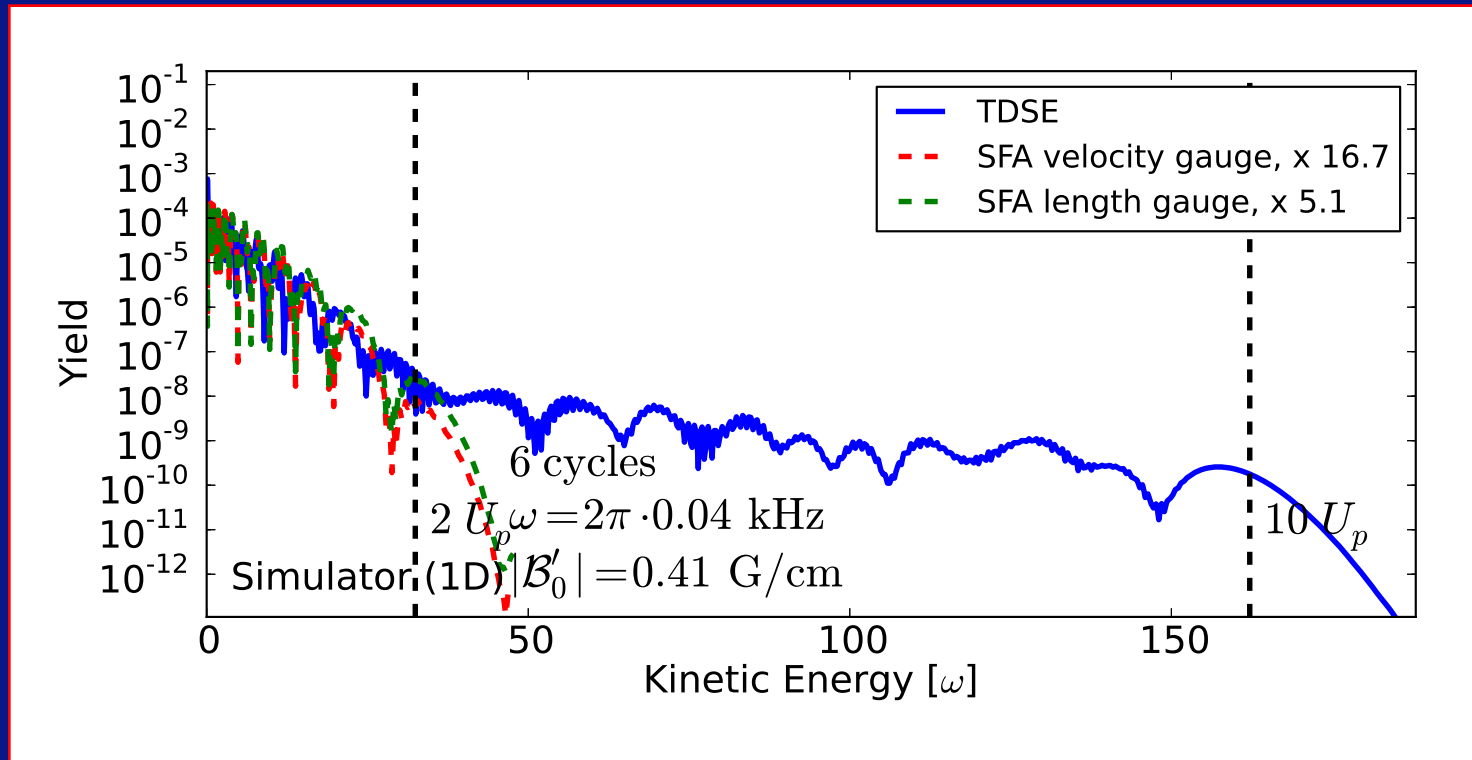


Measurement issues



Problem: in view of the **statistics** such **energy-resolved “ATI” spectra** are **hard to measure with few atoms**.

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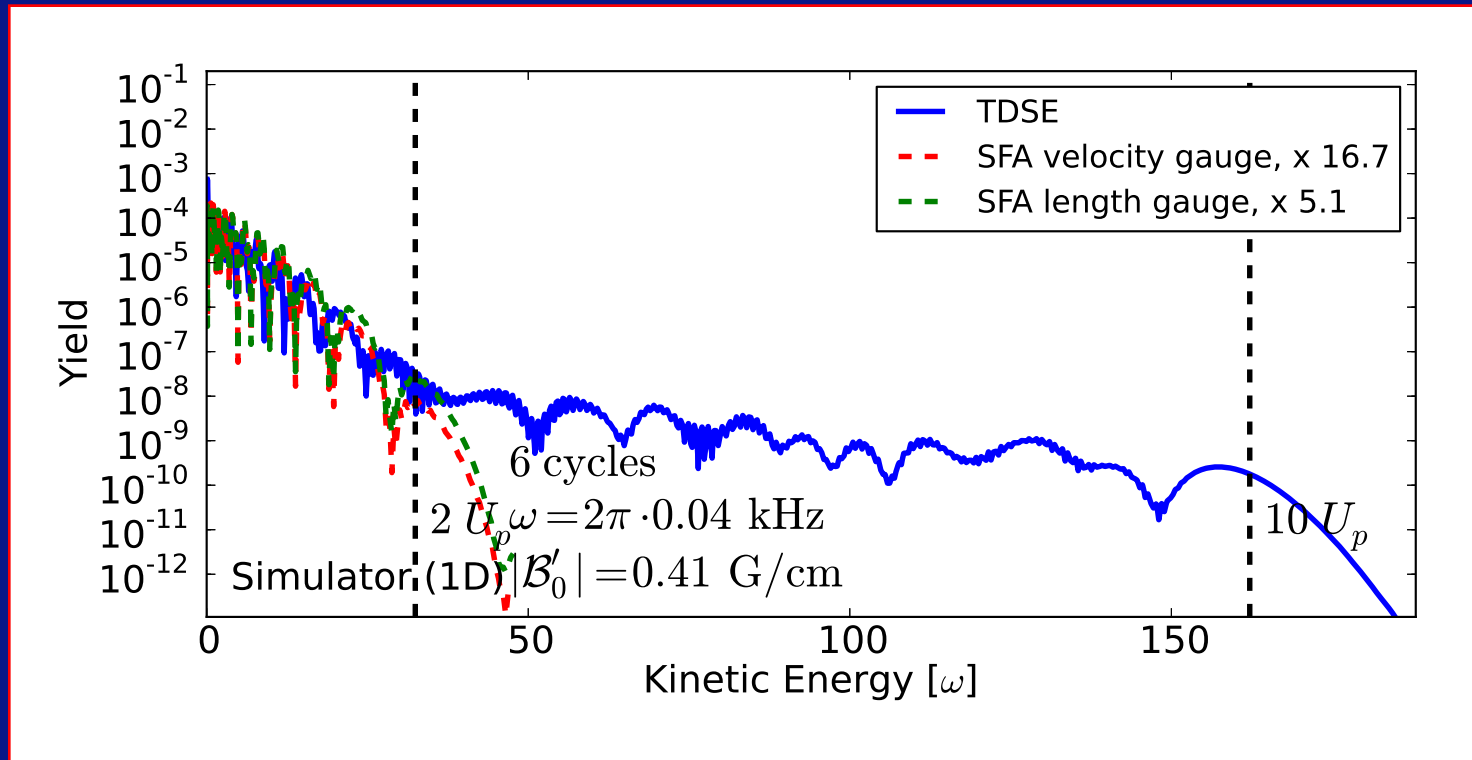


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other observables (e. g., excited states: “frustrated tunneling ionization”)
or using **many atoms** (e. g., one BEC) per simulated electron!

Towards experimental realization

The possibility of strong-field simulations with ultracold atoms has been discussed earlier: Arlinghaus and Holthaus [*Phys. Rev. A* **81**, 063612 (2010)].

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Note: Magnetic-field gradient would allow for **larger laser-parameter regime** (multiphoton/quasistatic).

Streaking ultrashort laser fields

A time-delayed (weak) attosecond pulse ionizes an atomic system dressed by an ultrashort intense femtosecond pulse (observed: kinetic energy of emitted electrons).

Streaking ultrashort laser fields

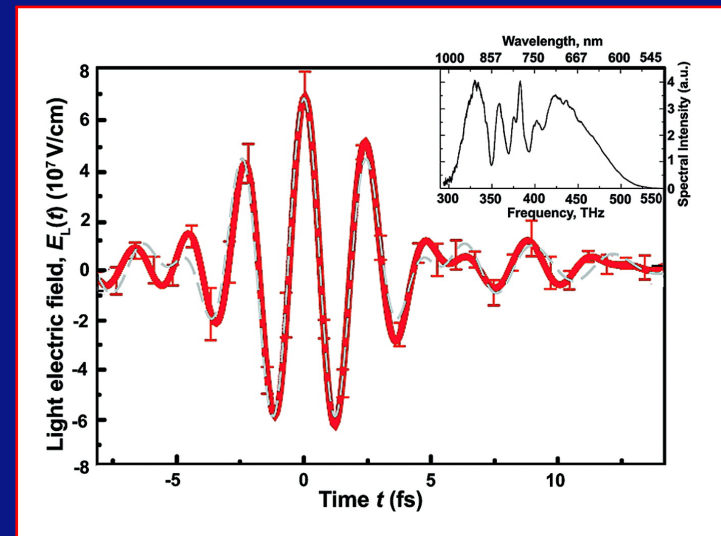
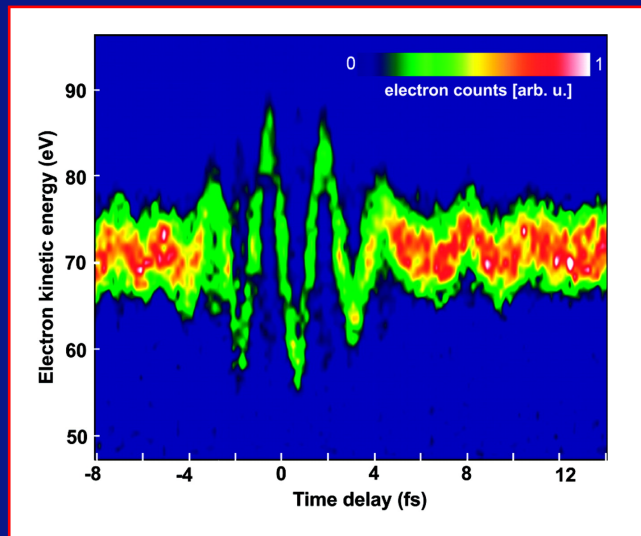
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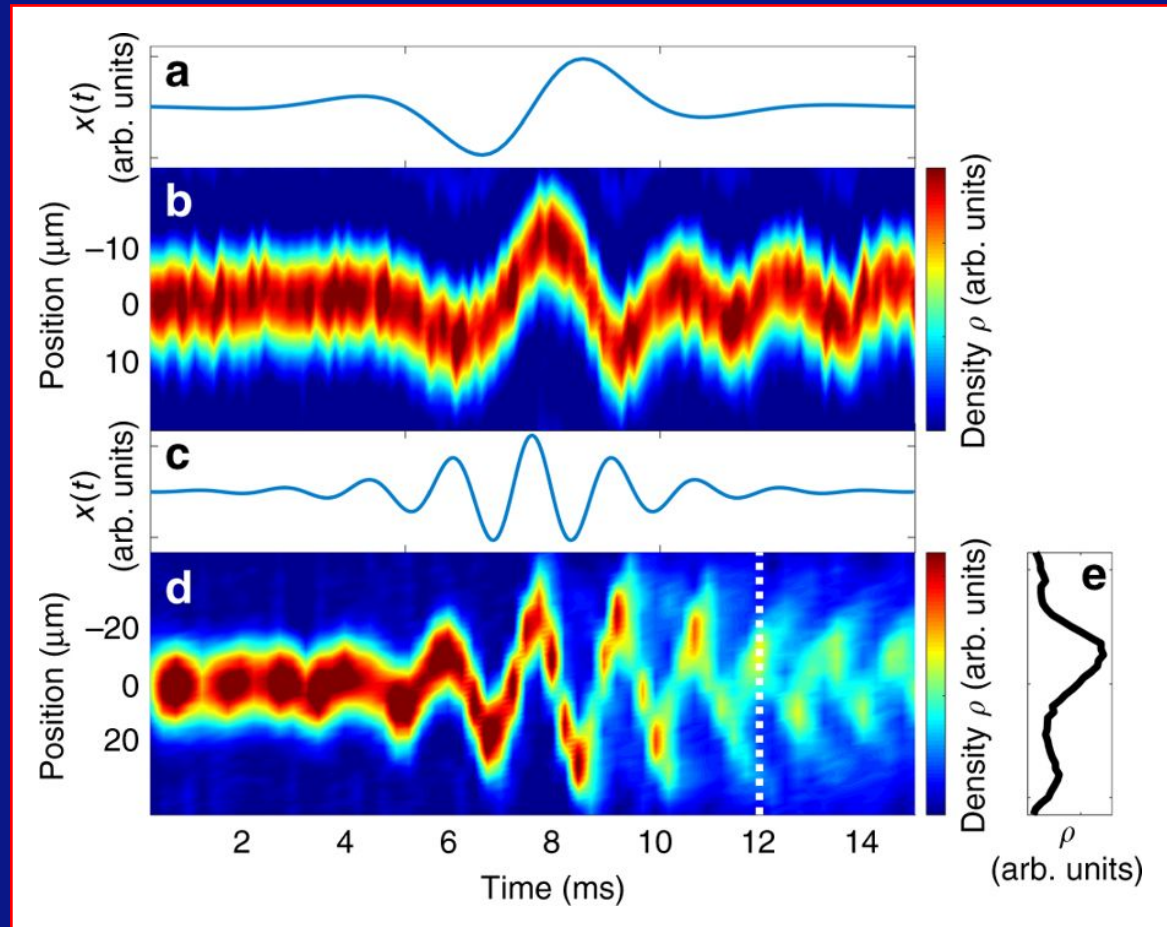
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[Goulielmakis *et al.*, *Science* **305**, 1267 (2004)]

Streaking using ultracold atoms



Experiment: periodically shaken single-trap strontium BEC.

[Senaratne *et al.*, *Nature Comm.* **9**, 2065 (2018)]

Summary

- *Lochfraß*: robust coherent wavepacket formation by means of time and spatially varying dissipation.
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New perspective: strongly (periodically) driven (few or many) ultracold atoms, possibly with (strong) interaction and structured dissipative environment.