Ultrafast electron dynamics after ionization by intense electromagnetic radiation

Michael Bonitz, Sebastian Bauch, Karsten Balzer and David Hochstuhl

Institut für Theoretische Physik und Astrophysik, Christian-Albrechts-Universität zu Kiel, D-24098 Kiel, Germany

Hamburg, December 14th, 2010

Bundesministerium für Bildung und Forschung

C A U Christian-Albrechts-Universität zu Kiel

Deutsche Forschungsgemeinschaft DFG

Introduction

Progress in experiments: new XUV/VUV (ω >10eV) sources

- radiation from higher-harmonics („table-top“)
- free-electron lasers, e.g. LCLS(Stanford), FLASH at DESY Hamburg
- planned: X-FEL at DESY and many more

New fields of physics

strong fields	intense UV/XUV radiation large photon energies in the non-perturbative regime
short time	ultrashort pulses (attosecond regime) time-resolved electronic processes in atoms, molecules, etc. examples: Auger decay, shake-up processes, ...

goal: better understanding of multi-electron atoms in (strong) laser fields
focus: ultrafast phenomena and correlation effects, chemical reactions, ...

Idea: Streak camera

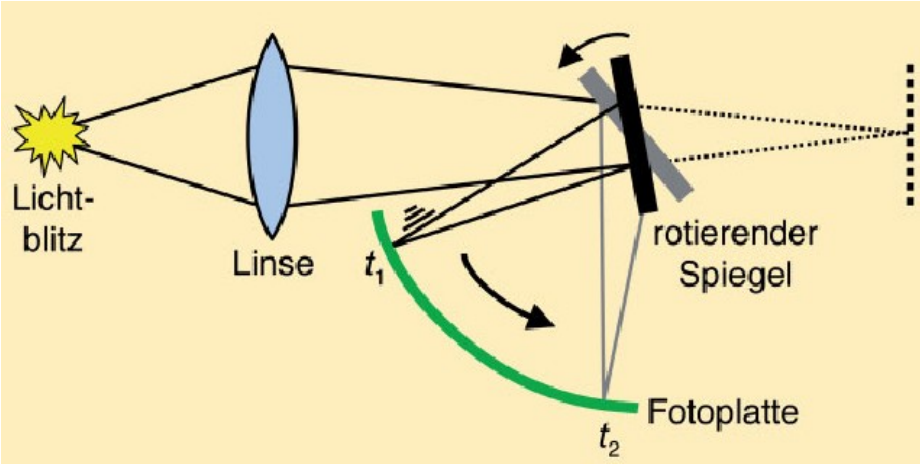


Fig.: Mechanical streak camera [1]

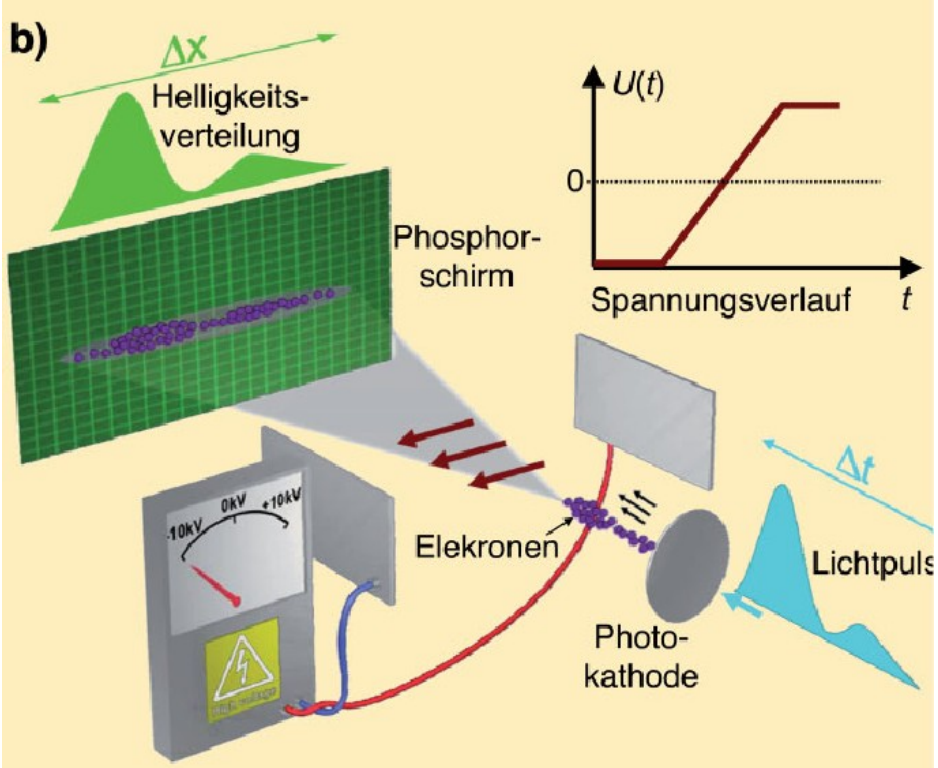
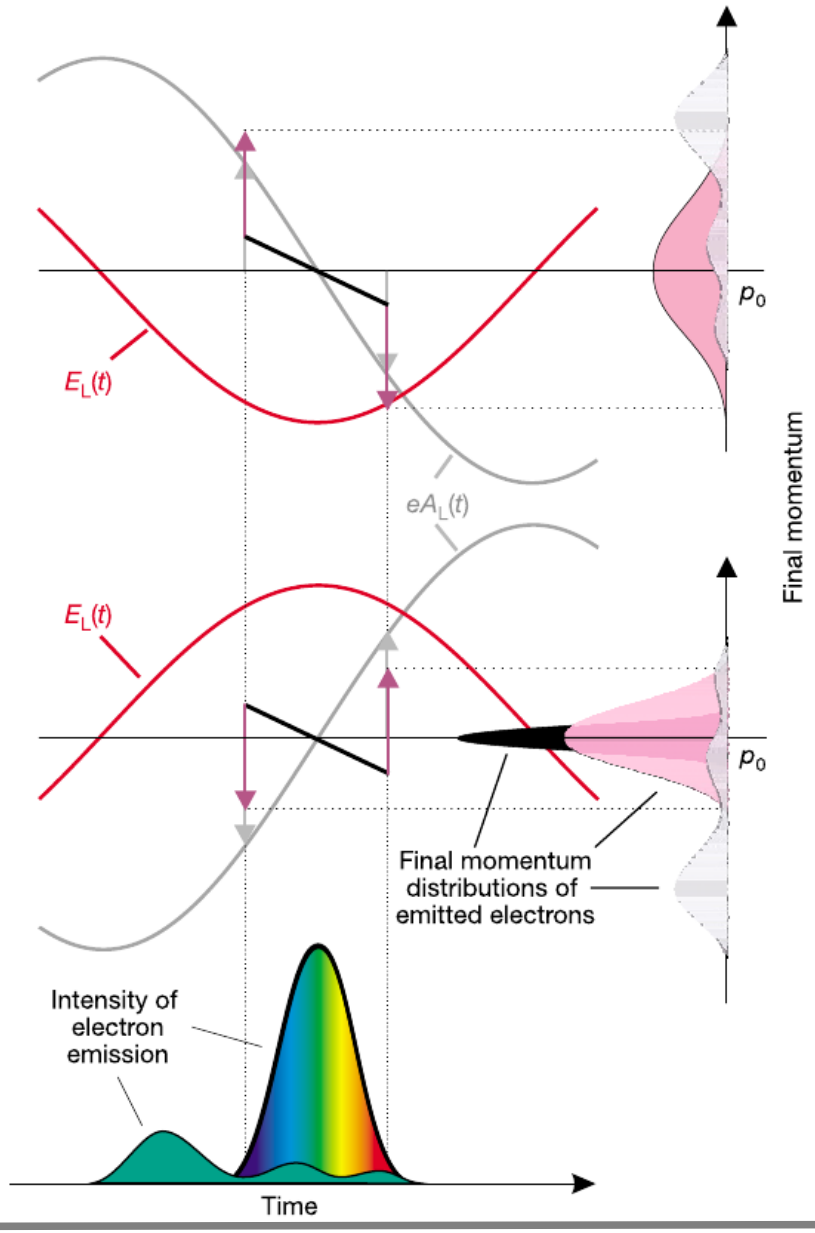


Fig.: Electrical streak camera [1]

- map temporal information onto spatial information
- method: time-dependent deflection „force“
- first realization: Sir Charles Wheatstone (1834) [2]
- limitation: mechanics (rotating mirror) or electric field change
- best time resolution reached: up to 200fs (!)

[1] M. Uiberacker et al., *Physik in unserer Zeit* **38,** 222 (2007) (Grafik)
[2] C. Wheatstone, *Phil. Trans. R. Soc. Lond.* **124,** 583 (1834)

Atomic transient recorder



Use laser field for deflection in momentum space via vector potential:

$$\mathbf{p}(t) = \mathbf{p}_0(t) - \frac{e}{c}\mathbf{A}(t)$$

Basic idea

Short XUV-pulse creates electron: $\mathbf{p}_0(t)$

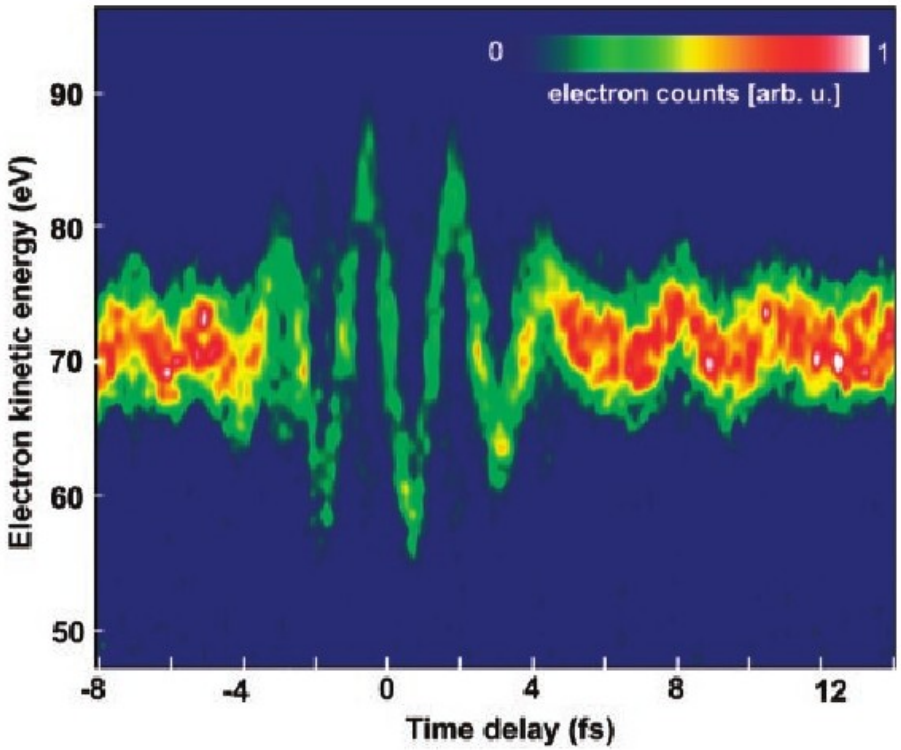
$\mathbf{p}_0(t)$ determined by emission process and XUV photon energy ω

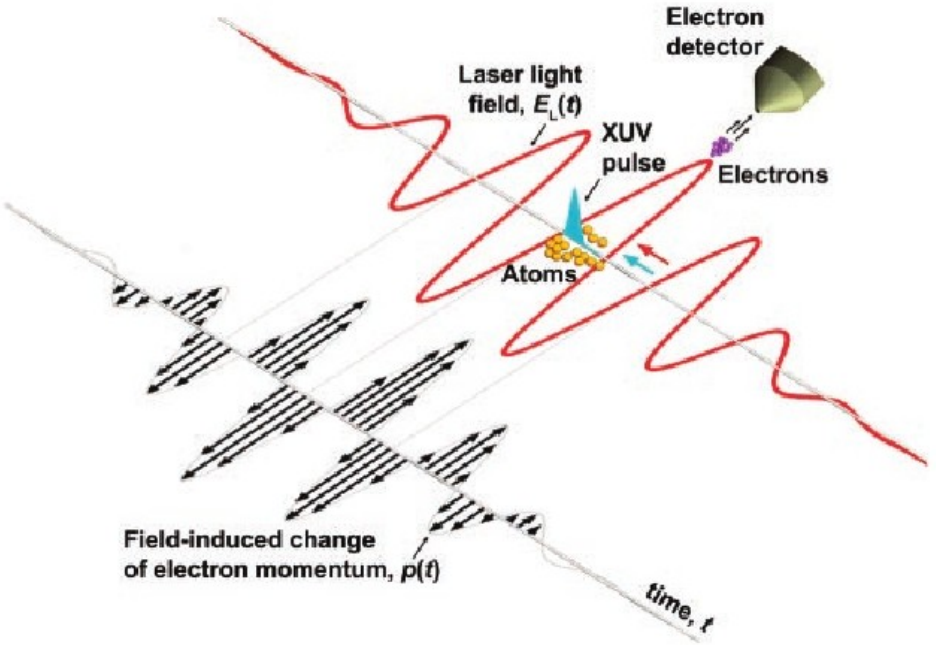
$\mathbf{A}(t)$ of a second (low frequency) pulse maps to momentum space

Knowledge of **emission** ⟷ Knowledge of **pulses**

[1] R. Kienberger et al., *Nature* **427,** 817 (2004) (Grafik)
[2] J. Itatani et al., *Phys. Rev. Lett.* **88,** 173903 (2002)

Breakthrough in experiments: attosecond streak camera





- enormous progress in experiments with XUV/IR pump-probe[1]
- ultrashort XUV pulses used for time-resolved measurement of EM field (vector potential) of an IR laser
- XUV pulse ionizes electrons at certain phase of IR field, serves as probe for vector potential of IR laser via momentum transfer
- kinetic energy of photoelectrons is measured

[1] e.g. F. Krausz, and M. Ivanov, *Rev. Mod. Phys.* **81,** 163 (2009)
[2] E. Goulielmakis et al., *Science* **305,** 1267 (2004) (Grafik)

Pulse characterization at FLASH: THz streak camera

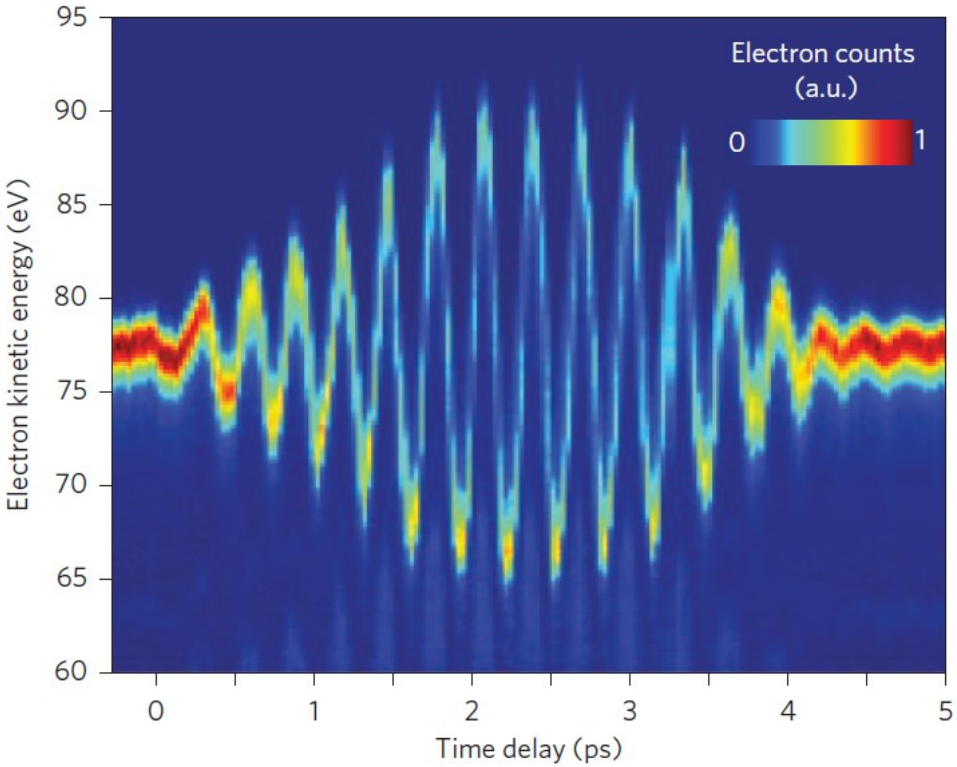


Fig.: sampled THz vector potential[1]

- typical XUV pulse durations: 5-60fs
- use THz (λ=90μm) pulse for deflection
- electron source: rare gas target atoms
- capability of single-shot characterization
- possibility for sorting of XUV pulses
- relies on response of atoms to *laser-assisted* XUV excitation

--> valuable information for all FLASH users

[1] U. Frühling et al., *Nat. Phot.* **3,** 523 (2009)

Challenges for theory/simulation

--> modelling of atomic processes and ionization including

- (sub-) femtosecond dynamics (e.g. XUV induced)
- long-time behavior (realistic detection process)

Theoretical approaches 1

One-electron dynamics (e.g. streak camera)

time-dependent Schrödinger equation (TDSE)
development of adequate (simple) models

Theoretical approaches 2

Multi-electron dynamics (correlation dynamics,..)

1. Few-particle TDSE
2. Multi-configuration time-dependent Hartree Fock (MCTDHF)
3. Nonequilibrium Green functions (NEGF)

Challenges for theory/simulation

--> modelling of atomic processes and ionization including

- (sub) femtosecond dynamics (e.g. XUV induced)
- long-time behavior (realistic detection process)

Theoretical approaches 1

One-electron dynamics (e.g. streak camera)

time-dependent Schrödinger equation (TDSE)
development of adequate (simple) models

Theoretical approaches 2

Multi-electron dynamics (e.g. correlation dynamics,..)

1. Few-particle TDSE
2. Multi-configuration time-dependent Hartree Fock (MCTDHF)
3. Non-equilibrium Greens functions (NEGF)

Time-dependent Schrödinger equation (TDSE)

laser assisted photoemission: concentrate on single-particle effects

consider lineraly polarized laser pulses in dipole approximation

$$i\frac{\partial}{\partial t}\Psi(\mathbf{r},t) = \left(-\frac{1}{2}\frac{\partial^2}{\partial \mathbf{r}^2} + V_b(\mathbf{r}) + \mathbf{r}\cdot\mathbf{E}(t)\right)\Psi(\mathbf{r},t)$$

Use “atomic units” (a.u.): $\hbar$=m_e=e=1
units: length 1au=a_B; time 1au=24as; energy: 1au=1Hartree=27.2eV;

V_b : binding (nucleus) potential, electrical field E(t) in length gauge

two-color pulse is modelled with (e.g. Gaussian) envelopes E(t)

$$E(t) = E_X(t,\tau)\cos\omega_X(t-\tau-t_0) + E_S(t,t_0)\cos\omega_S(t-t_0)$$

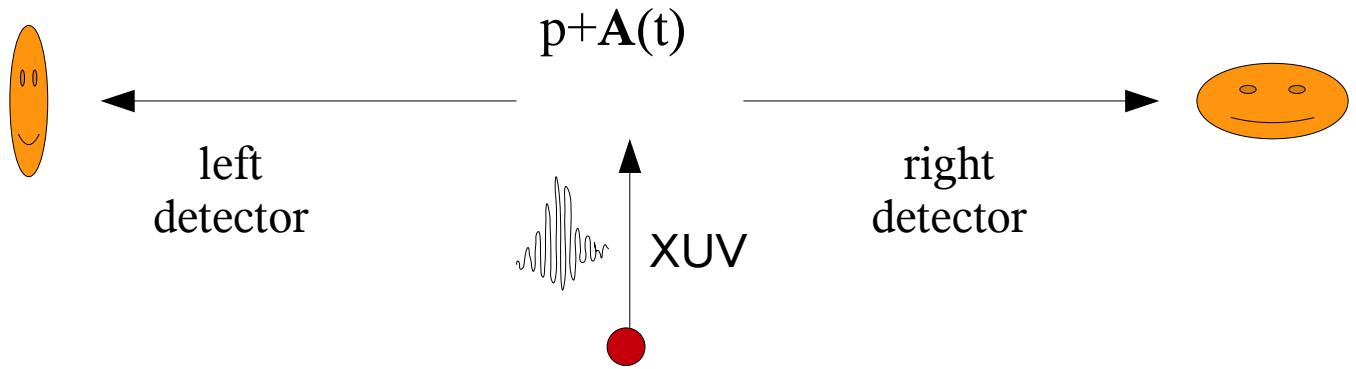
$$E_i(t,t_0) = E_i^0 \exp\left(-\frac{(t-t_0)^2}{2\tau_i^2}\right); i = \{X,S\}$$

X = XUV, S = streaking pulse (long wavelength)

for one-dimensional models, a regularized version of V_b is used:

$V_b(x) = -\frac{1}{\sqrt{(x^2+\kappa^2)}}$, κ: regularization parameter

Calculation of photoelectron spectra



final electron state after time propagation consists of three parts:

$$\begin{aligned} \psi_{\text{bound}} &= \psi(x,T)\ x = x_0 - r \ldots x_0 + r \\ \psi_{\text{left}} &= \psi(x,T)\ x < x_0 - r \\ \psi_{\text{right}} &= \psi(x,T)\ x > x_0 + r \end{aligned}$$

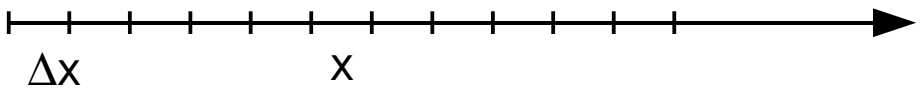
photoelectron spectrum obtained by Fourier transform:

$$\psi_{\text{bound,left,right}}(k) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \mathrm{d}x\, \exp(ikr)\psi_{\text{bound,left,right}}(x)\ .$$

Numerical approach to TDSE simulations

require: resolve dynamics of bound *and* free electrons

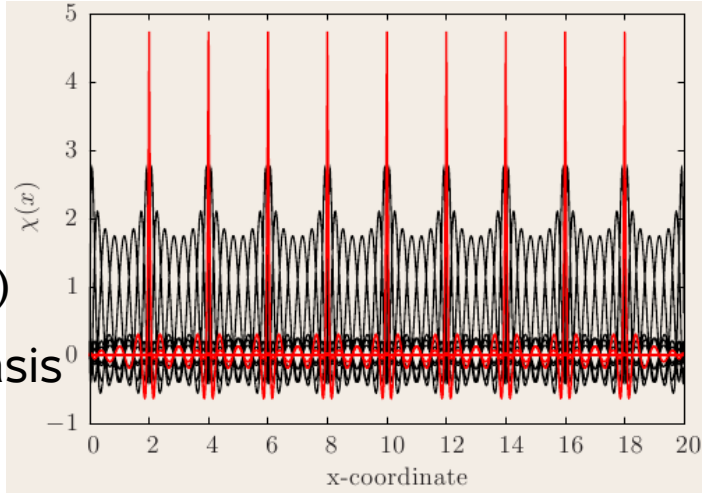
simplest method (intuitive): grid method



but: inefficient → need better approach

--> finite-element discrete variable approach (FE-DVR)

wave function represented in combined grid and basis like expansion[1,2]



Time-propagation: requirements

construct initial state by imaginary time propagation

evolve wave function in time with time step Δt = 0.001au...0.01au

large grids needed (up to 50000 au) due to high-energy electrons (~ 80eV)

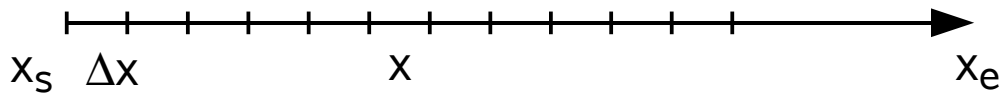
for e.g. THz streaking long evolution times (larger than 10000au)

[1] e.g. B.I.Schneider et al. *Phys. Rev. E* **73**, 036708 (2006)
[2] K. Balzer, S. Bauch and M.Bonitz, *Phys. Rev. A* **81,** 022510 (2010); *Phys. Rev. A* **82,** 033427 (2010)

Numerical approach to TDSE simulations

require: resolve dynamics of bound *and* free electrons

simplest method (intuitive): grid method

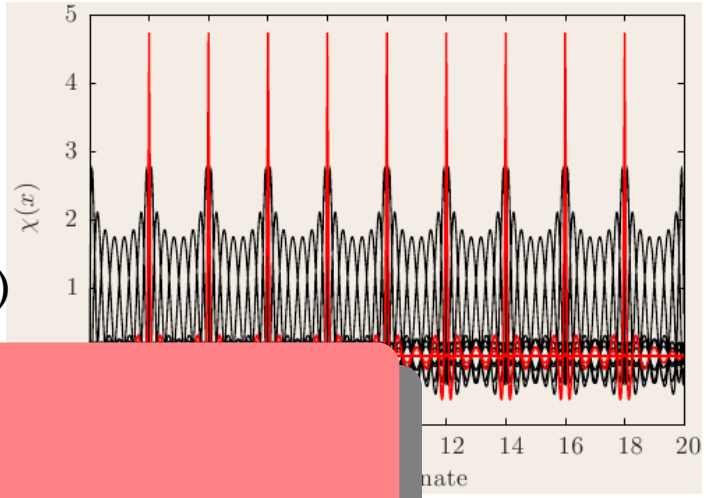


but: inefficient → need better approach

f--> inite-element discrete variable approach (FE-DVR)

wave function

expansion[1,2]



vary ω_S from IR to Thz:

longer wave length → longer times
longer times → larger grids (travelling wave packet)
→ smaller timestep (higher precision)

Increasing numerical challenge!

construct ir

evolve wave function in time with tem step Δt = 0.001...0.01

large grids needed (up to 50000 au) due to high-energy electrons (~ 80eV)

for e.g. THz streaking long evolution times (larger than 10000au)

[1] e.g. B.I.Schneider et al. *Phys. Rev. E* 73, 036708 (2006)
[2] K. Balzer, S. Bauch and M.Bonitz, *Phys. Rev. A* **81,** 022510 (2010); *Phys. Rev. A* **82,** 033427 (2010)

Semiclassical Superposition Approximation (SSA)[1,2]

Photoelectron distribution given by

$$J_{PES}(p) = \int_{-\infty}^{\infty} \mathrm{d}t \left(\frac{E_X^0(t)}{E_X^0} \right)^2 I_X[\Omega(p,t)] S[\Omega(p,t)] \ .$$

momentum transfer at moment *t*:

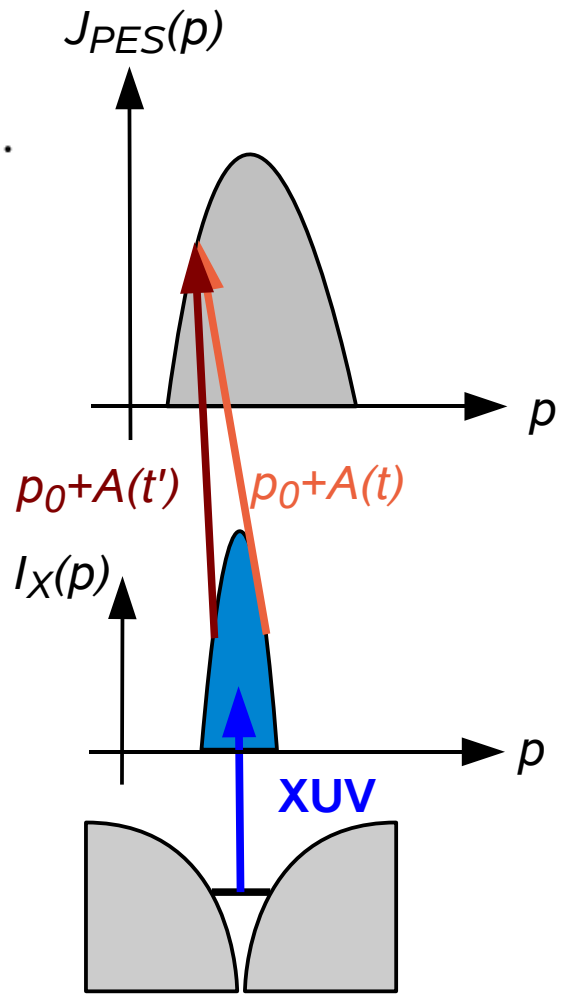
$$\Omega(p,t) = \frac{1}{2}\left(p - \frac{e}{c}A(t)\right)^2 + I_p$$

intensity distribution of the XUV pulse:

$$I_X(\omega) = |\mathcal{F}\left\{ E_X^0(t)\cos(\omega_X t)\right\}|^2 \ .$$

ionization cross section (of atom, solid etc.):

$$S(\omega) = |\langle\psi_f(\omega)|x|\psi_i\rangle|^2 \quad \text{with} \quad |\psi_f\rangle \propto \exp i\sqrt{2m\omega}r$$



Different parts of XUV spectrum contribute to same momentum p and are summed *incoherently*.

[1] E. Krasovskii, and M. Bonitz, *Phys. Rev. Lett.* **99**, 247601 (2007)

[2] E. Krasovskii, and M. Bonitz, *Phys. Rev. A* ***80***, 053421 (2009)

Semiclassical Superposition Approximation (SSA)

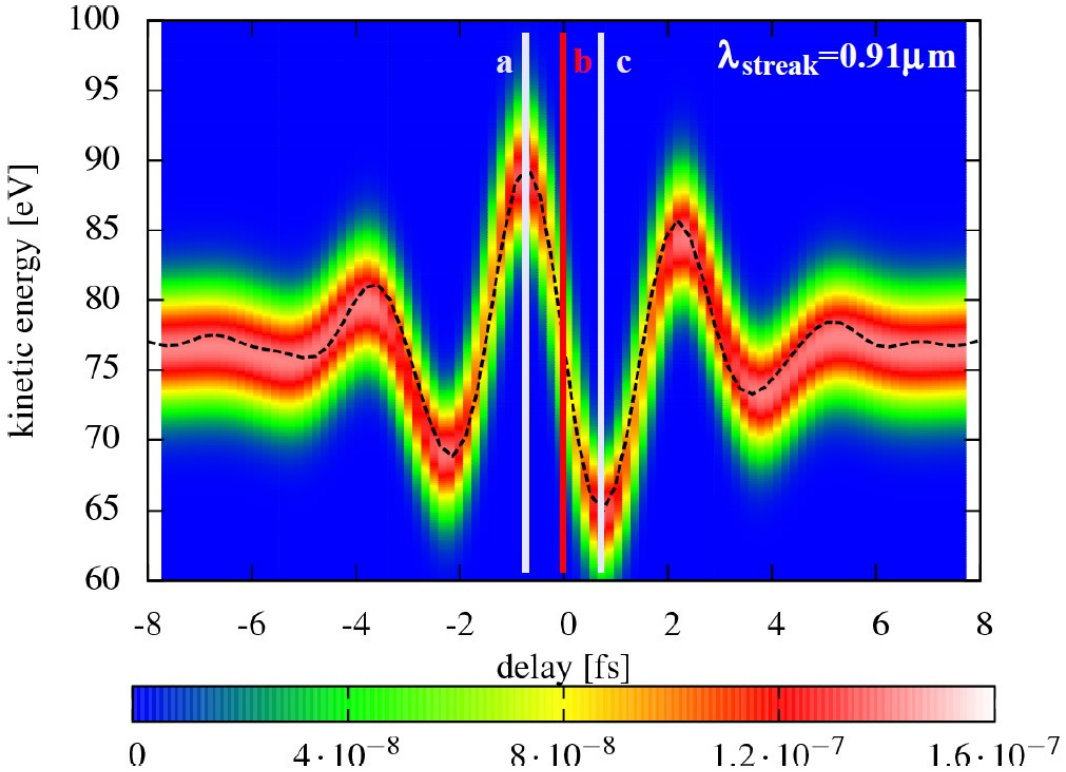


Fig.: TDSE

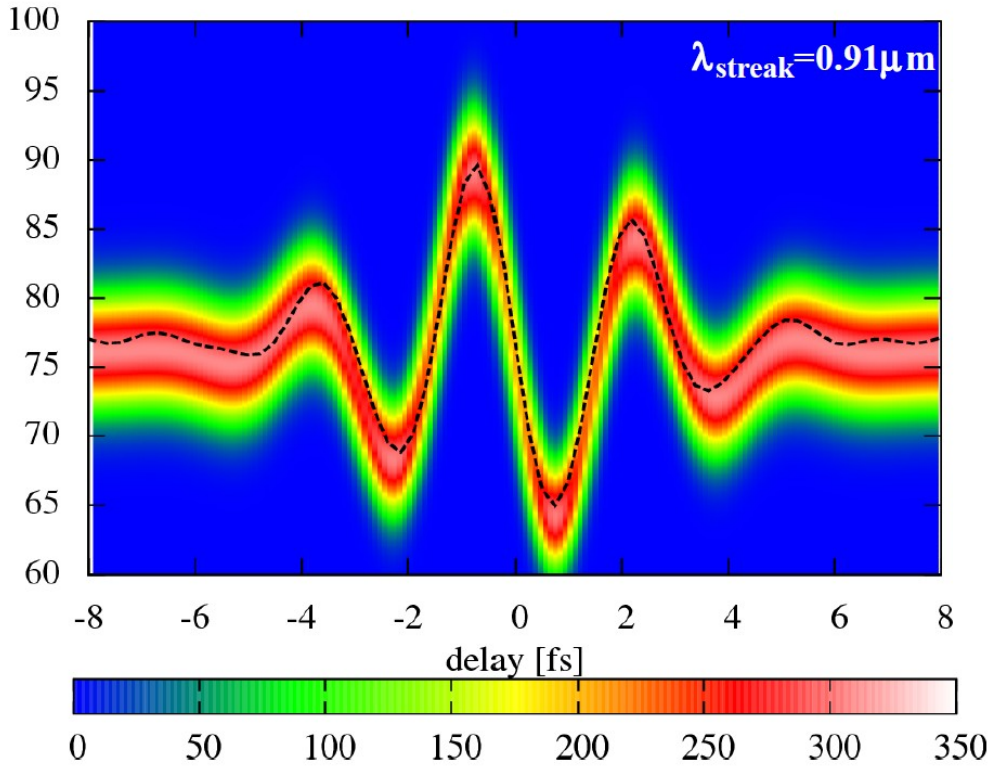


Fig.: SSA model

I_x^{peak} = 8.8 ·10^{13} W/cm^2 , ω_X = 91eV, ionization potential I_p=14.1eV and U_p=0.27eV

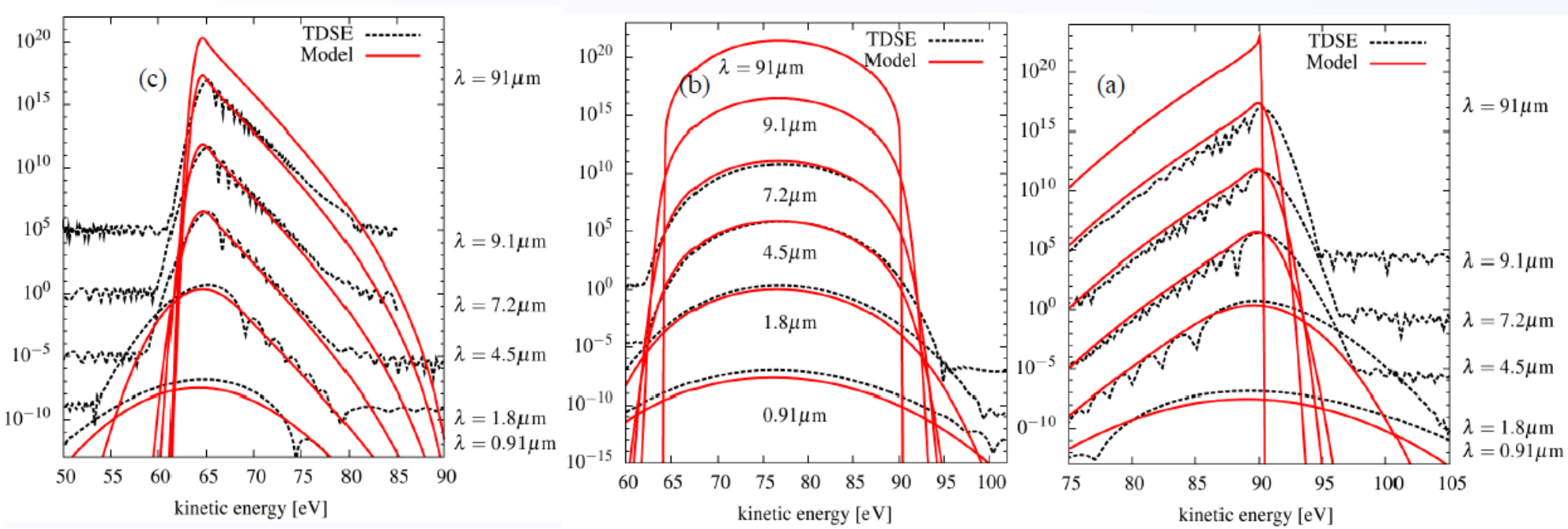
- dashed line: classically estimated kinetic energy of streaked photoelectrons ($W_0=\omega_X-I_p$)

$$E_{\mathrm{kin}} = W_0 \pm \sqrt{8W_0U_p}E(\tau - t_{0,\mathrm{streak}})\sin(\omega\tau - t_{0,\mathrm{streak}})$$

--> SSA model (right fig.) well reproduces the overall delay-dependence

Laser assisted ionization: PE line shapes from IR to THz streaking

A=min A=0 A=max



- vary ω_{streak} at fixed ponderomotive potential U_p=0.27eV

- use XUV pulse duration according to condition $\omega_{streak} \cdot \sigma_X$ = const

--> line shapes well reproduced by SSA model (logarithmic scale)

advantage: may avoid very large TDSE calculations (e.g. λ=91μm)

--> comparison with experiments under way

Challenges for theory & simulation

modelling of atomic processes and ionization, including

- (sub) femtosecond dynamics (e.g. XUV induced)
- long-time behavior (realistic detection process)

Theoretical approaches 1

One-electron dynamics (e.g. streak camera)

time-dependent Schrödinger equation (TDSE)
development of adequate (simple) models

Theoretical approaches 2

Multi-electron dynamics (correlation dynamics, correlated ionization etc.)

1. Few-particle TDSE
2. Multi-configuration time-dependent Hartree Fock (MCTDHF)
3. Non-equilibrium Green's functions (NEGF)

Multi-electron effects in atomic ionization

two-electron TDSE (length-gauge):

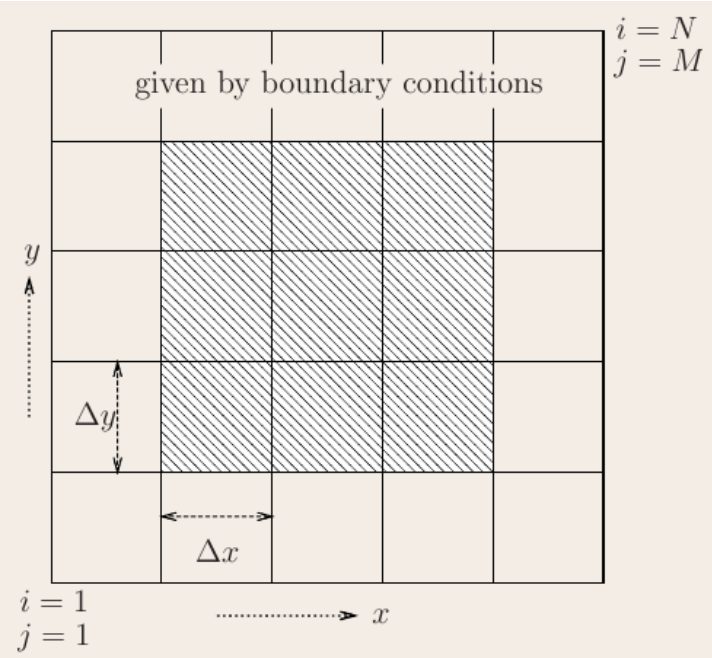
$$i\hbar\frac{\partial}{\partial t}\Psi(\boldsymbol{r}_1,\boldsymbol{r}_2,t)=\left\{-\frac{\hbar^2}{2m_e}\left(\frac{\partial^2}{\partial\boldsymbol{r}_1^2}+\frac{\partial^2}{\partial\boldsymbol{r}_2^2}\right)+\sum_i^2\left(V_{\text{bind}}(\boldsymbol{r}_i)+V_{\text{laser}}(\boldsymbol{r}_i,t)\right)+w(\boldsymbol{r}_1,\boldsymbol{r}_2)\right\}\Psi(\boldsymbol{r}_1,\boldsymbol{r}_2,t)$$

with two-color laser field in dipole approximation

$$V_{\text{laser}}(x_i,t) \quad = \quad -x_i\left[E^0_{\text{X}}e^{\left(-\frac{(t-\tau)^2}{2\sigma_{\text{X}}}\right)}\cos(\omega_{\text{X}}(t-\tau))E^0_{\text{IR}}e^{\left(-\frac{t^2}{2\sigma_{\text{IR}}}\right)}\cos(\omega_{IR}t)\right]$$

binding potential V_{bind} and binary interaction potential

$$w(x_1-x_2)=\frac{1}{\sqrt{(x_1-x_2)^2+1.0)}}$$



Multi-electron effects in ultrafast atomic ionization

two-electron TDSE (length-gauge):

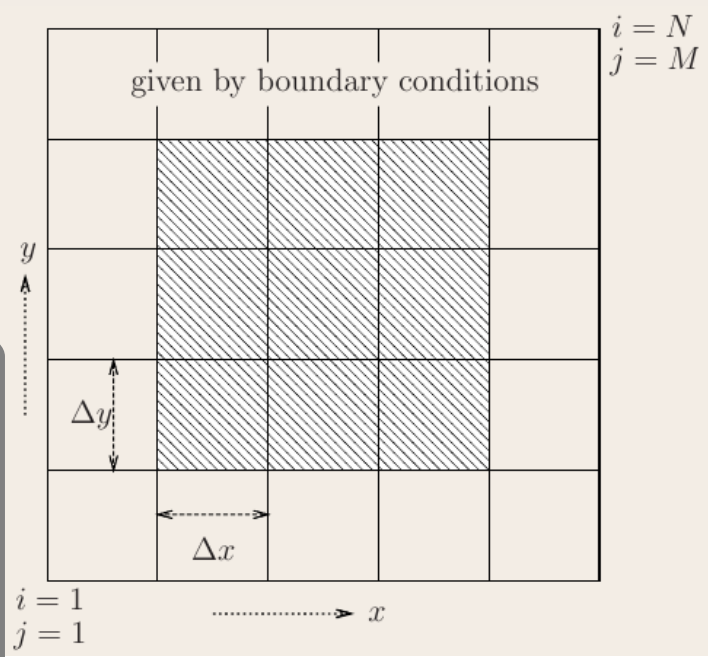
$$i\hbar\frac{\partial}{\partial t}\Psi(\boldsymbol{r}_1,\boldsymbol{r}_2,t) = \left\{-\frac{\hbar^2}{2m_e}\left(\frac{\partial^2}{\partial \boldsymbol{r}_1^2}+\frac{\partial^2}{\partial \boldsymbol{r}_2^2}\right)+\sum_i^2\left(V_{\text{bind}}(\boldsymbol{r}_i)+V_{\text{laser}}(\boldsymbol{r}_i,t)\right)+w(\boldsymbol{r}_1,\boldsymbol{r}_2)\right\}\Psi(\boldsymbol{r}_1,\boldsymbol{r}_2,t)$$

with two-color laser field in dipole approximation

$$V_{\text{laser}}(x_i,t) \quad = \quad -x_i\left[E^0_{\text{X}}e^{\left(-\frac{(t-\tau)^2}{2\sigma_{\text{X}}}\right)}\cos(\omega_{\text{X}}(t-\tau))E^0_{\text{IR}}e^{\left(-\frac{t^2}{2\sigma_{\text{IR}}}\right)}\cos(\omega_{IR}t)\right]$$

binding potential V_{bind} and binary interaction potential

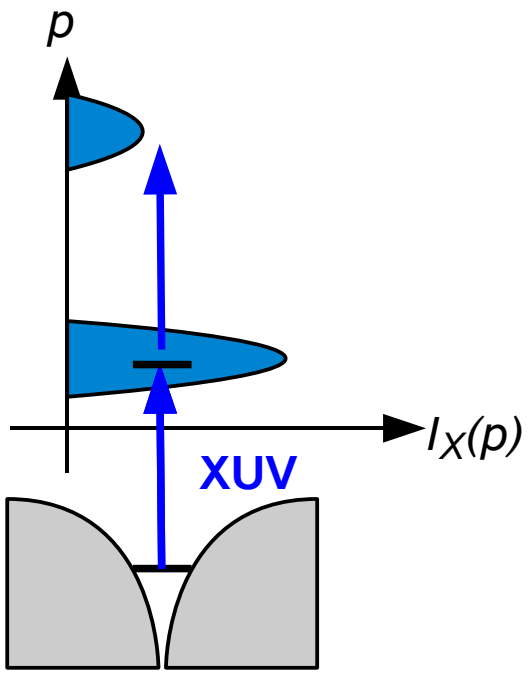
$$w(x_1-x_2) = \frac{1}{\sqrt{(x_1-x_2)^2+1.0)}}$$

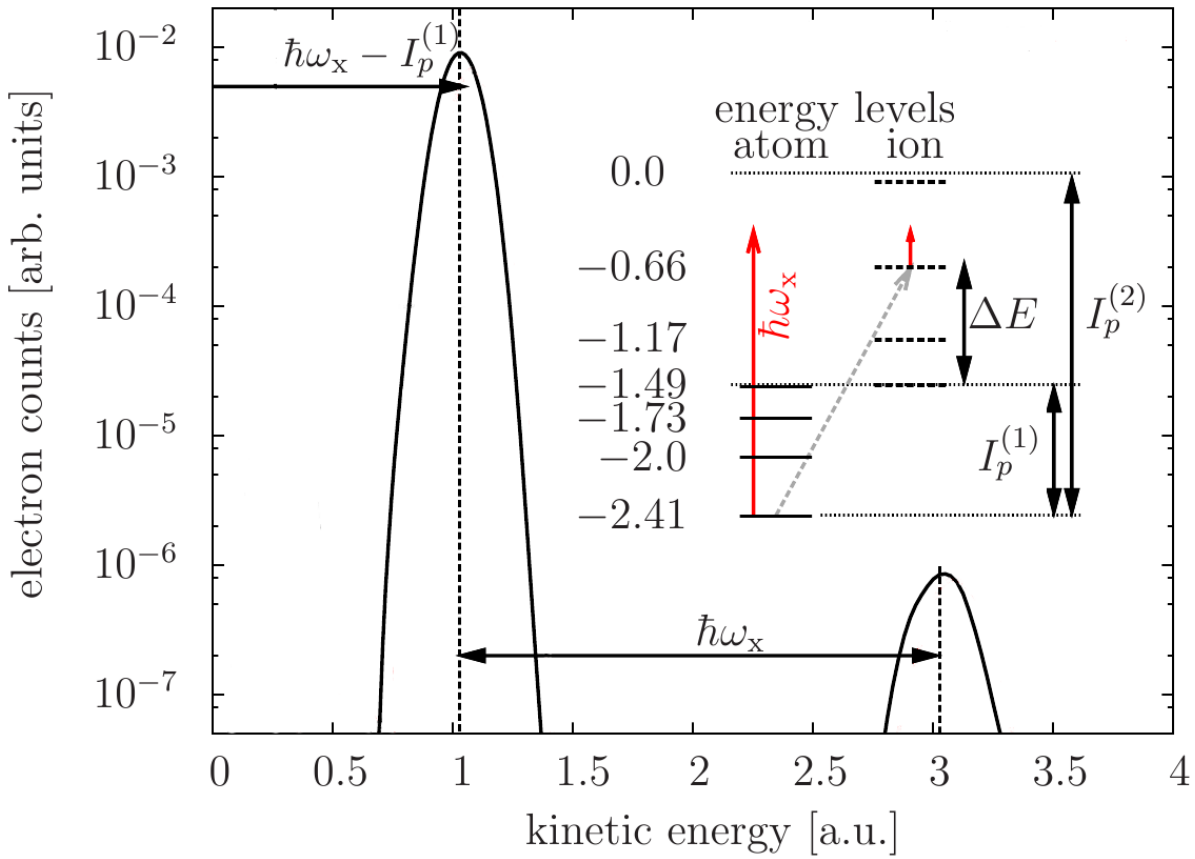


Extension from 1 to 2 particles

--> increase of system dimensionality
for 1D model system: solution of 2D TDSE
--> enormous increase in CPU time and grid requirements
- parallel code (up to several 100 CPUs) essential
- restriction to (near) IR probe pulses

UV photo ionization: 1.) expected PE spectrum

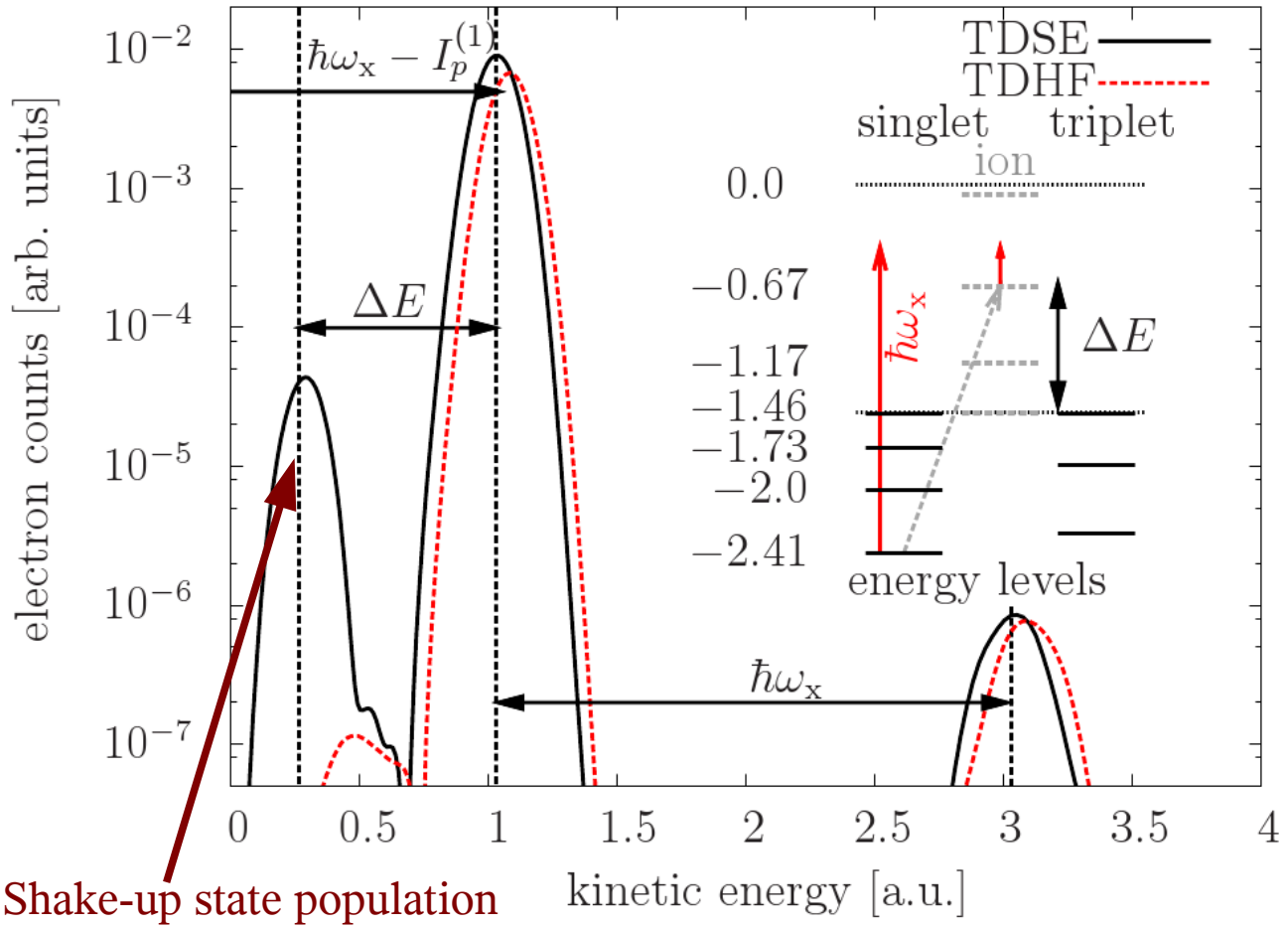




expect (single-active electron model): absorption of UV photon

in strong field: absorption of multiple photons

UV photo ionization: 2.) two-electron Computer experiment



XUV pump
Intensity 3.4 10^{14} W/cm²
Photon energy ω=**54 eV**
Duration τ=240 as

Model System
Two electrons in potential well

First ionization potential **25eV**
Second ionization pot. **65.5eV**

Helium-like system

--> significant deviation from simple one-electron picture

- reason: e-e correlations (electron shake up) produce additional slow electrons

interpretation confirmed by Hartree-Fock calculations (correlations neglected)

--> expect strong effect on laser-assisted double ionization

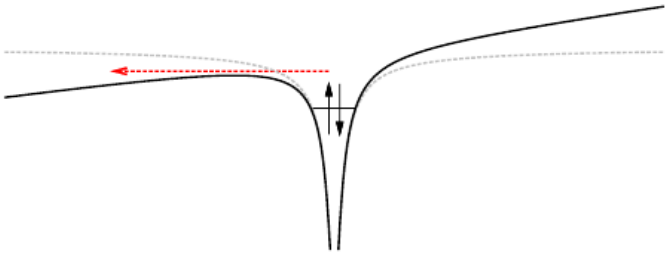
S. Bauch, K.Balzer and M. Bonitz, *Europhys. Lett.* **91,** 53001 (2010)

Correlation effects in strong field double ionization

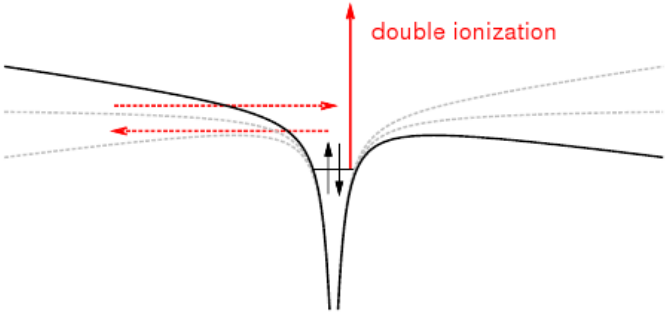
- sequential (uncorrelated) model (SAE) dramatically underestimates double ionization yield

--> correlated processes essential in double ionization

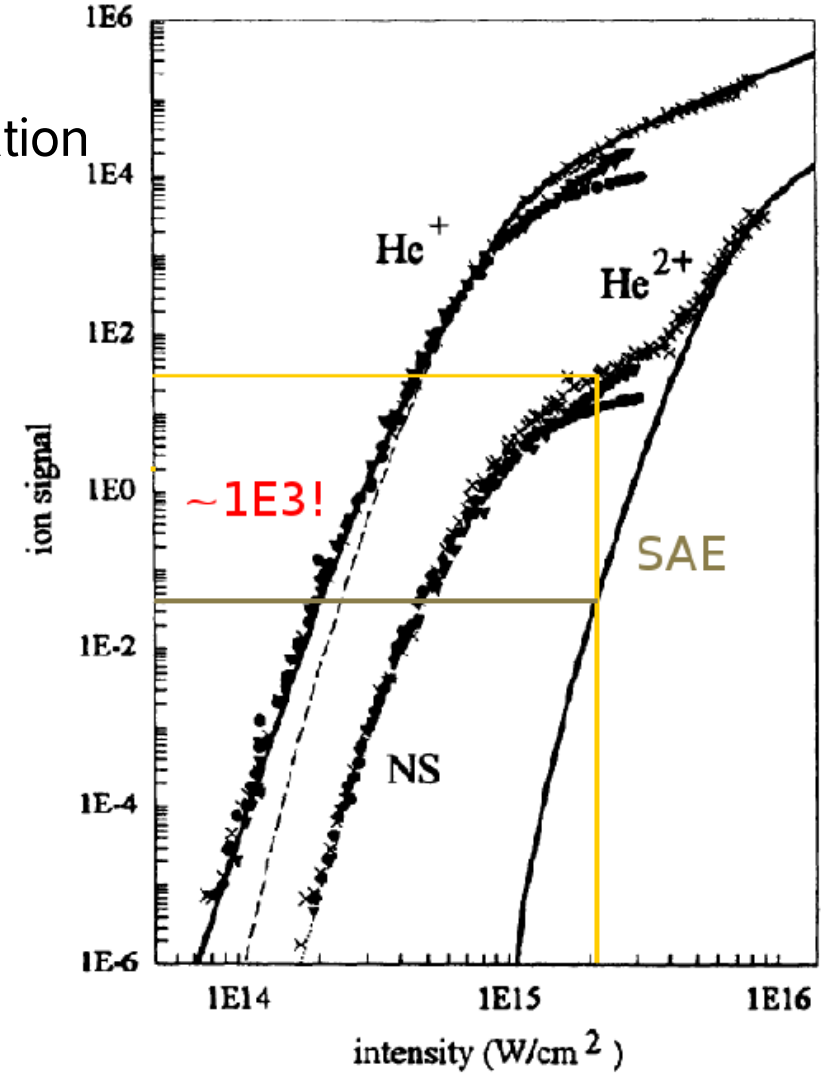
(1) one electron leaves binding potential



(2) field direction changes, electron accelerated towards the ion



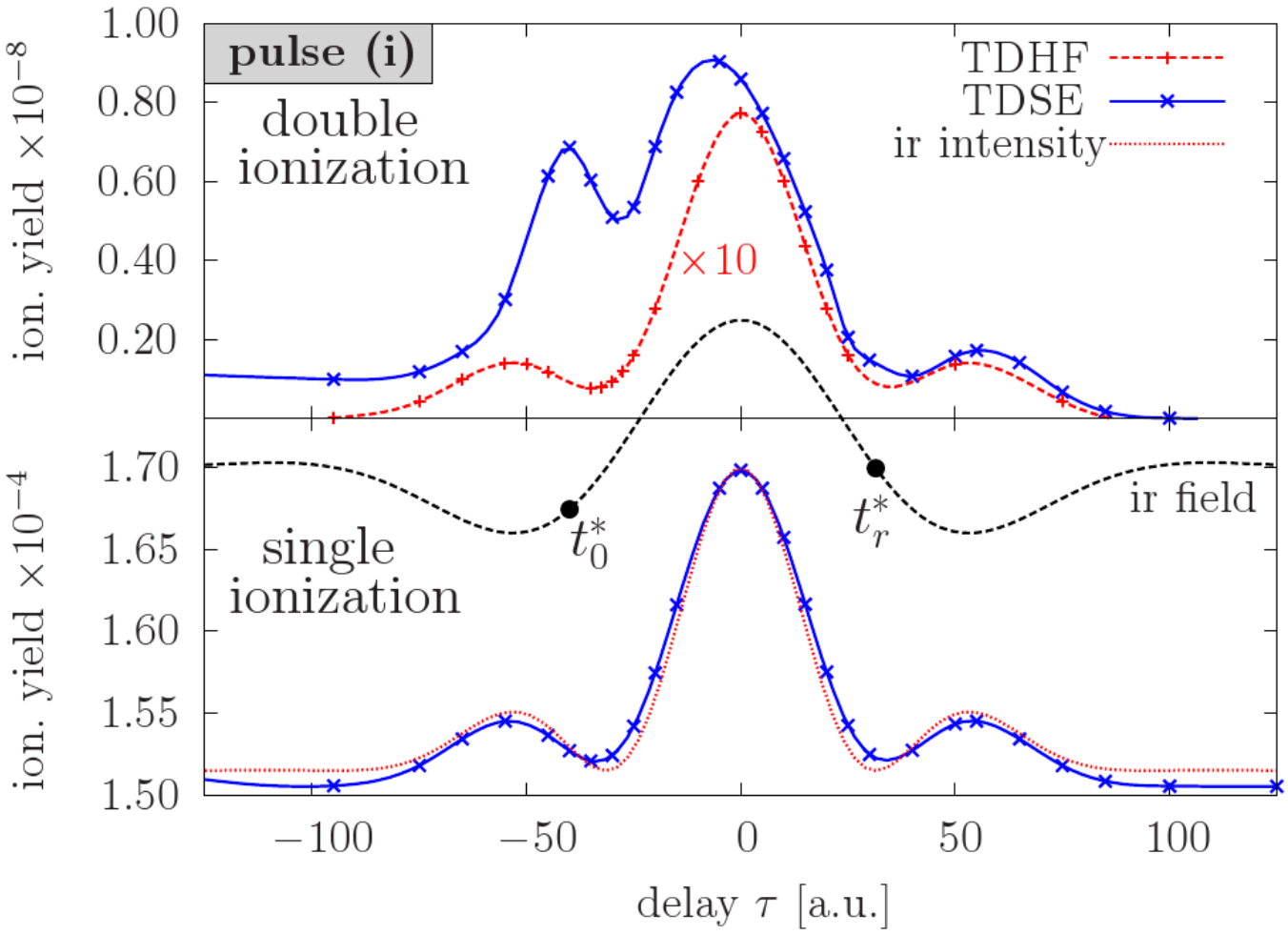
(3) "kicks off" second electron



Walker et al. PRL **73,** 1227 (1994)

Time dependence of double ionization?

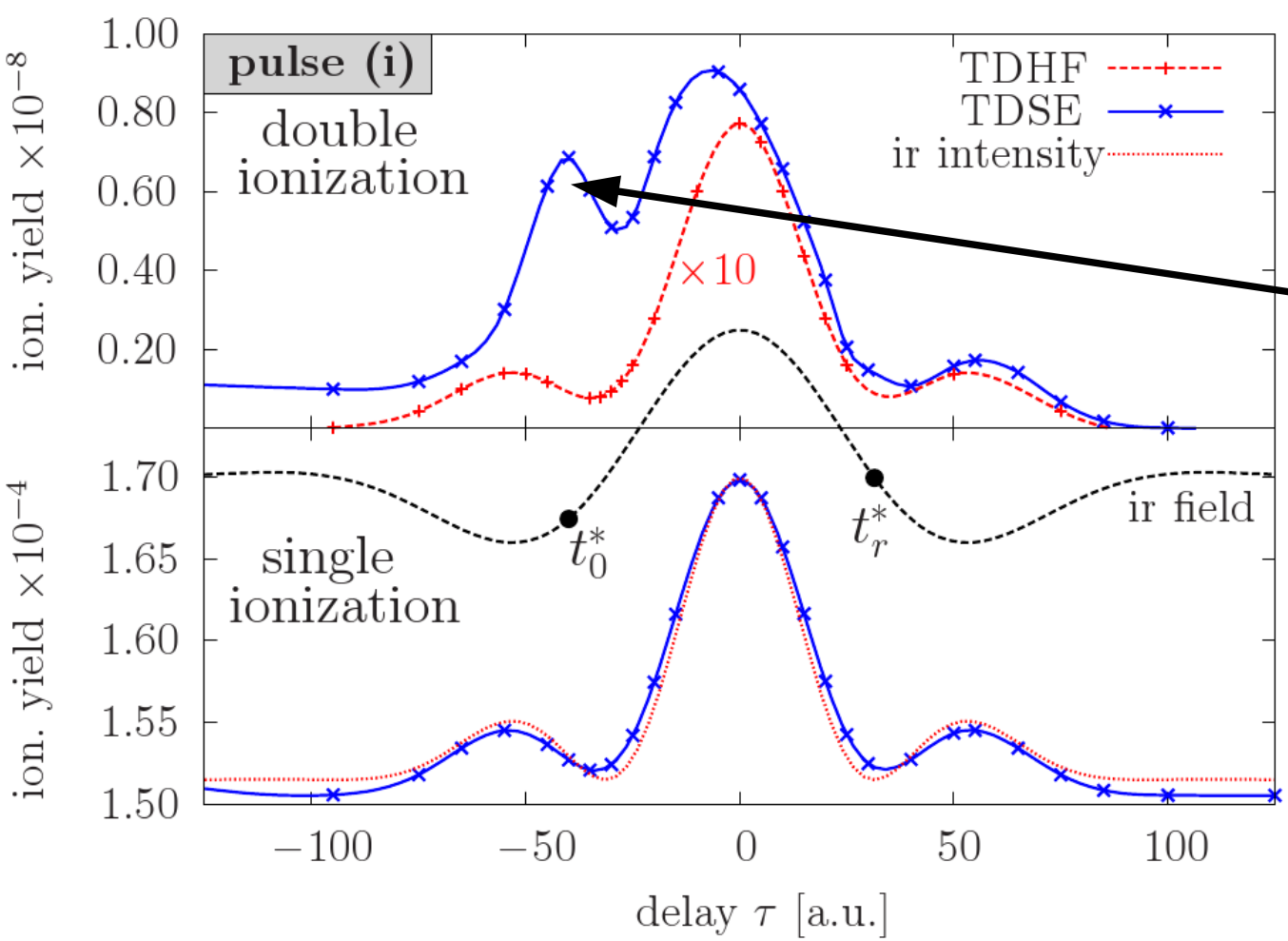
Time-resolved investigation of rescattering with XUV-IR pulses

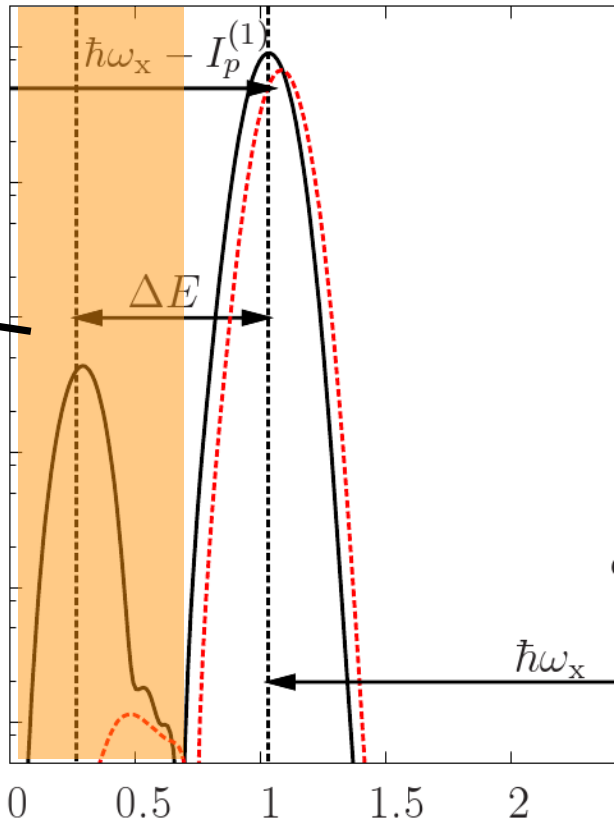


IR-Probe (i)
Intensity **8.8 10^{13}** W/cm^2
λ=900nm

S. Bauch, K.Balzer and M. Bonitz, *Europhys. Lett.* **91,** 53001 (2010)

Time-resolved investigation of rescattering with XUV-IR pulses





IR-Probe (i)
Intensity **8.8 10¹³** W/cm²
λ=900nm

- IR pulse accelerates UV-ionized electrons
- only slow electrons may re-scatter back on ion
- impact ionization of second e only in case of pre-excitaion
- shake-up mechanism is crucial

[1] S. Bauch, K.Balzer and M. Bonitz, *Europhys. Lett.* **91** 53001 (2010)

Classical model: “trajectories” of UV ionized electron

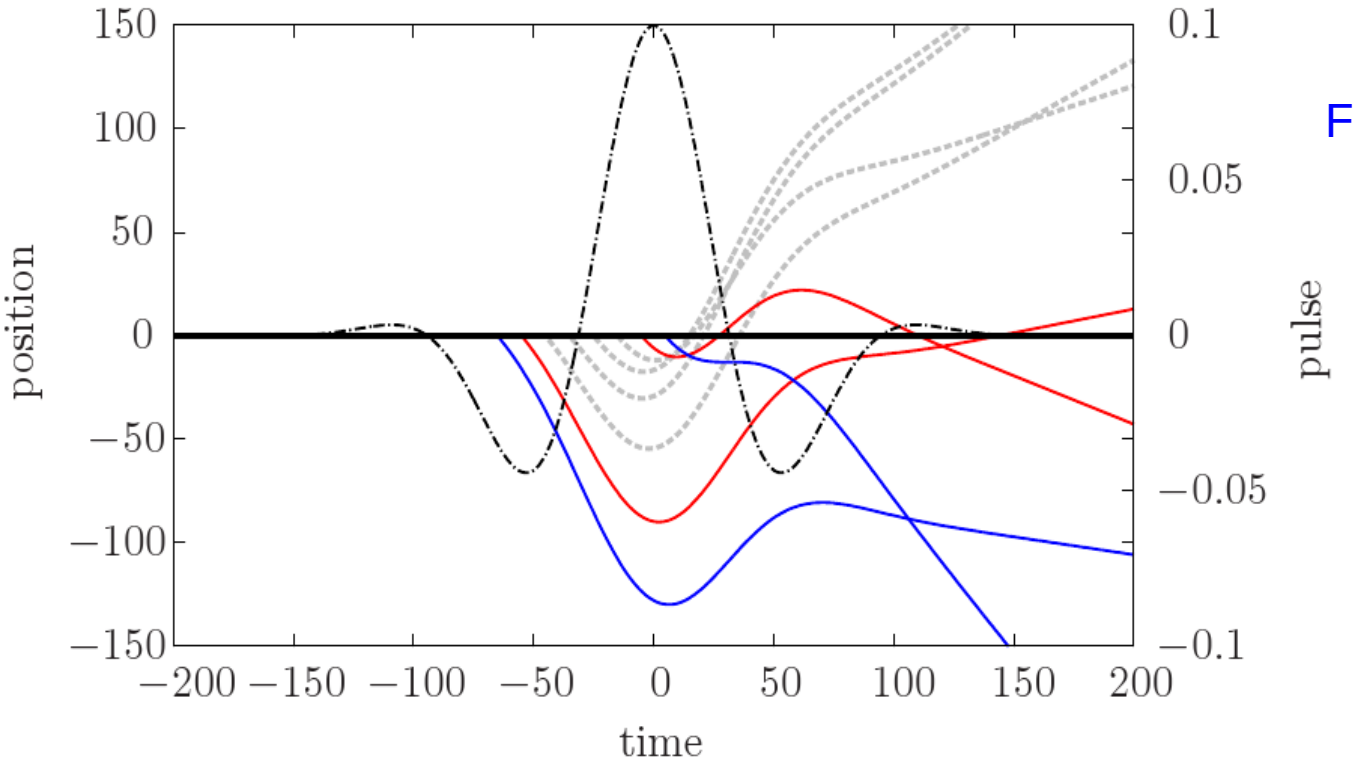


Fig.: Classical trajectories of UV ionized electron created at different delay times (IR field phases).

Blue trajectories do not rescatter

initial momentum of UV ionized electron: $p_0 = \pm[2(\omega_x - I_p^{(1)} - E_{\text{shakeup}})]^{1/2}$

Time-dependent momentum: $p(t,t_0) = p_0 + \frac{1}{c}A(t) = p_0 - \int_{t_0}^{t} \mathrm{d}\bar{t}\; E(\bar{t})$

classical trajectories: $x(t,t_0) = p_0(t-t_0) - \int_{t_0}^{t} \mathrm{d}\bar{t} \int_{t_0}^{\bar{t}} \mathrm{d}\bar{\bar{t}}\, E(\bar{\bar{t}})$

Classical model: “trajectories” of UV ionized electron

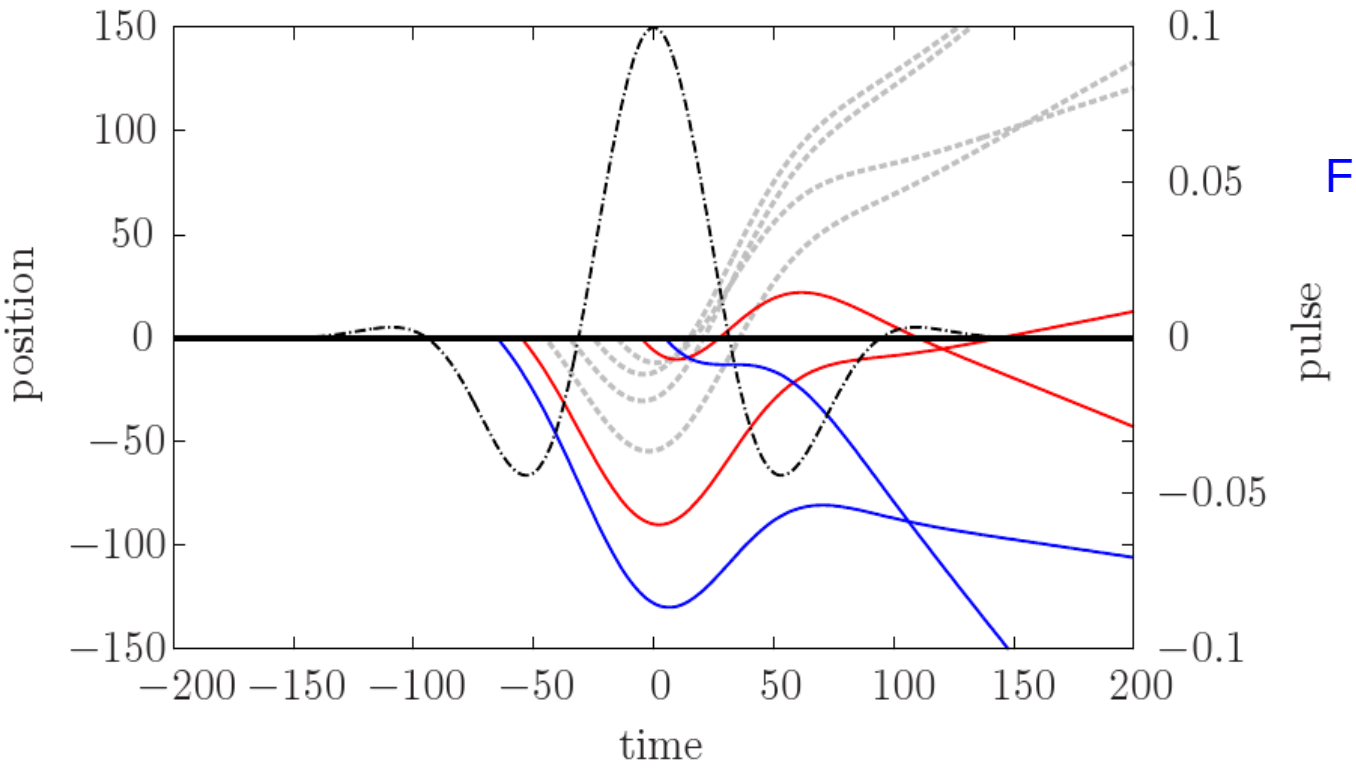
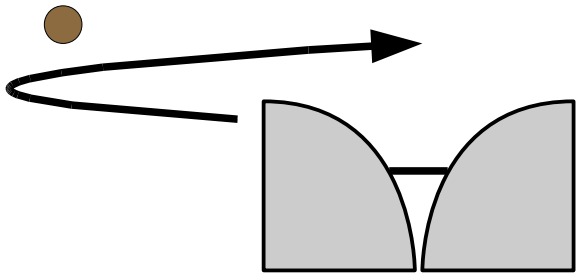


Fig.: Classical trajectories of UV ionized electron created at different delay times (IR field phases).

Blue trajectories do not rescatter



Classical model: “trajectories” of UV ionized electron

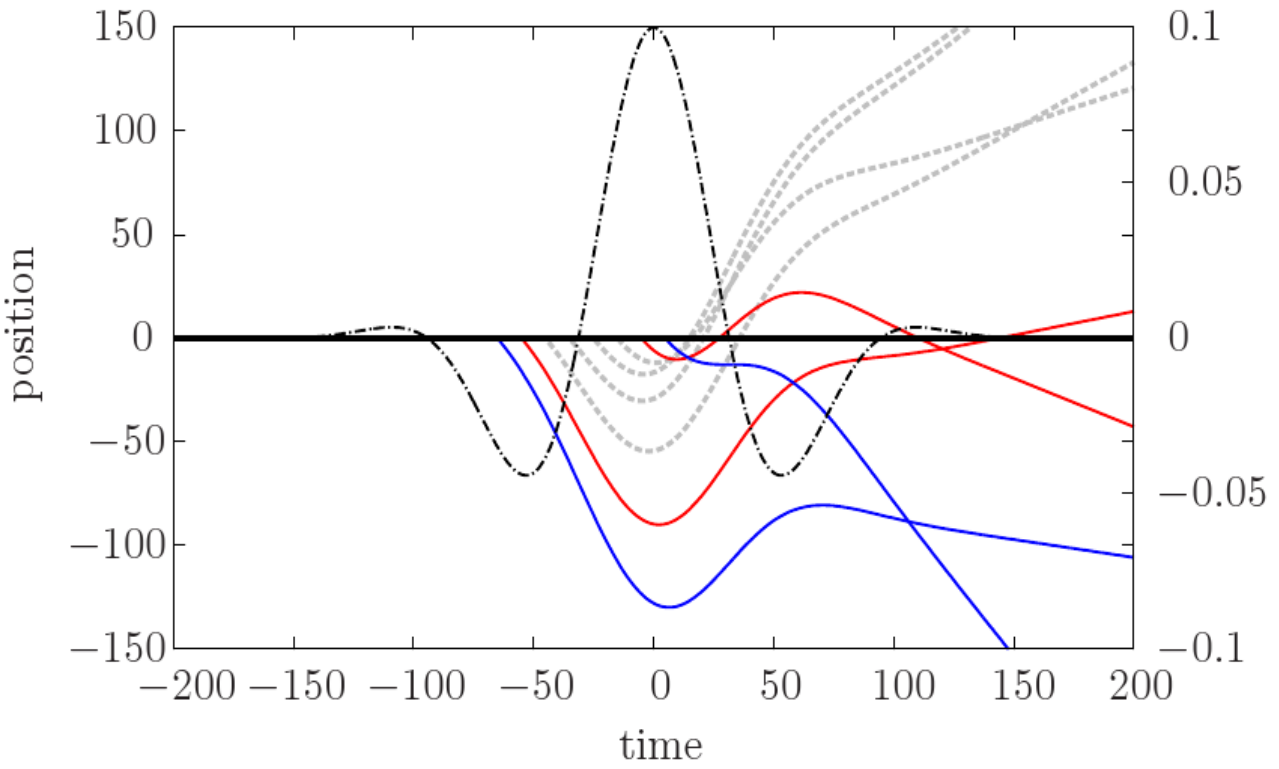
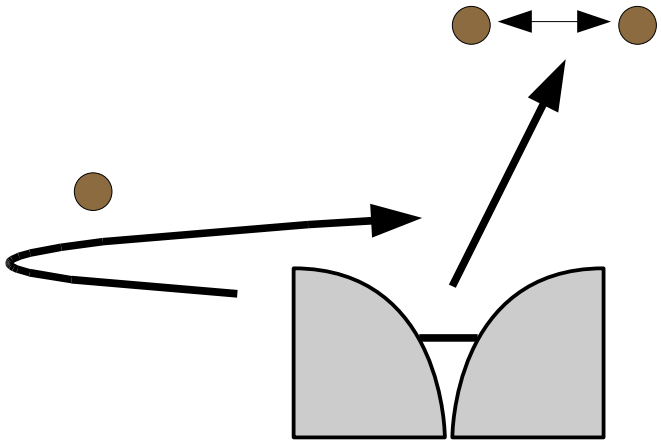
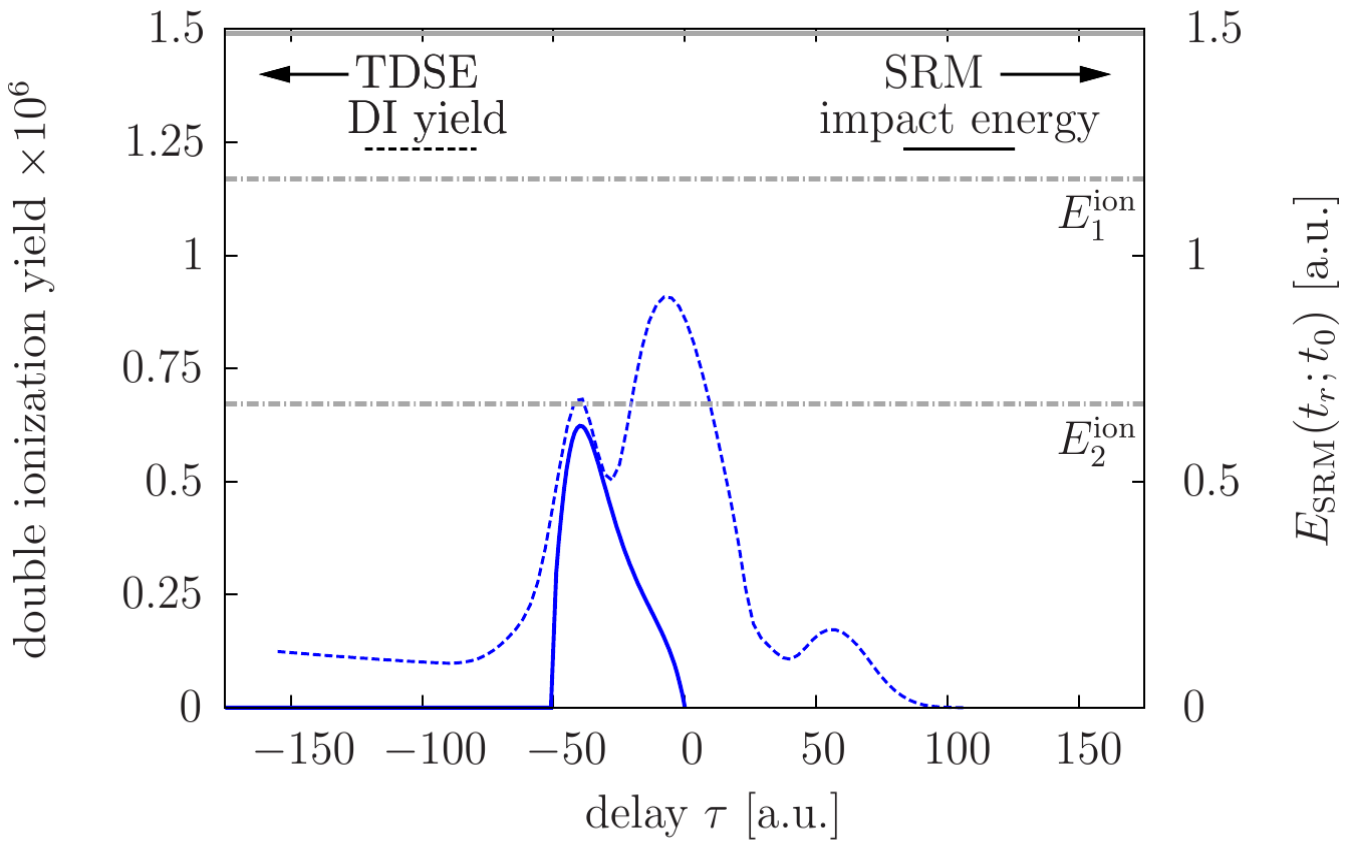


Fig.: Classical trajectories of UV ionized electron created at different delay times (IR field phases).

in case of sufficiently large kinetic energy: impact ionization



Classical model: impact energy of recolliding electron

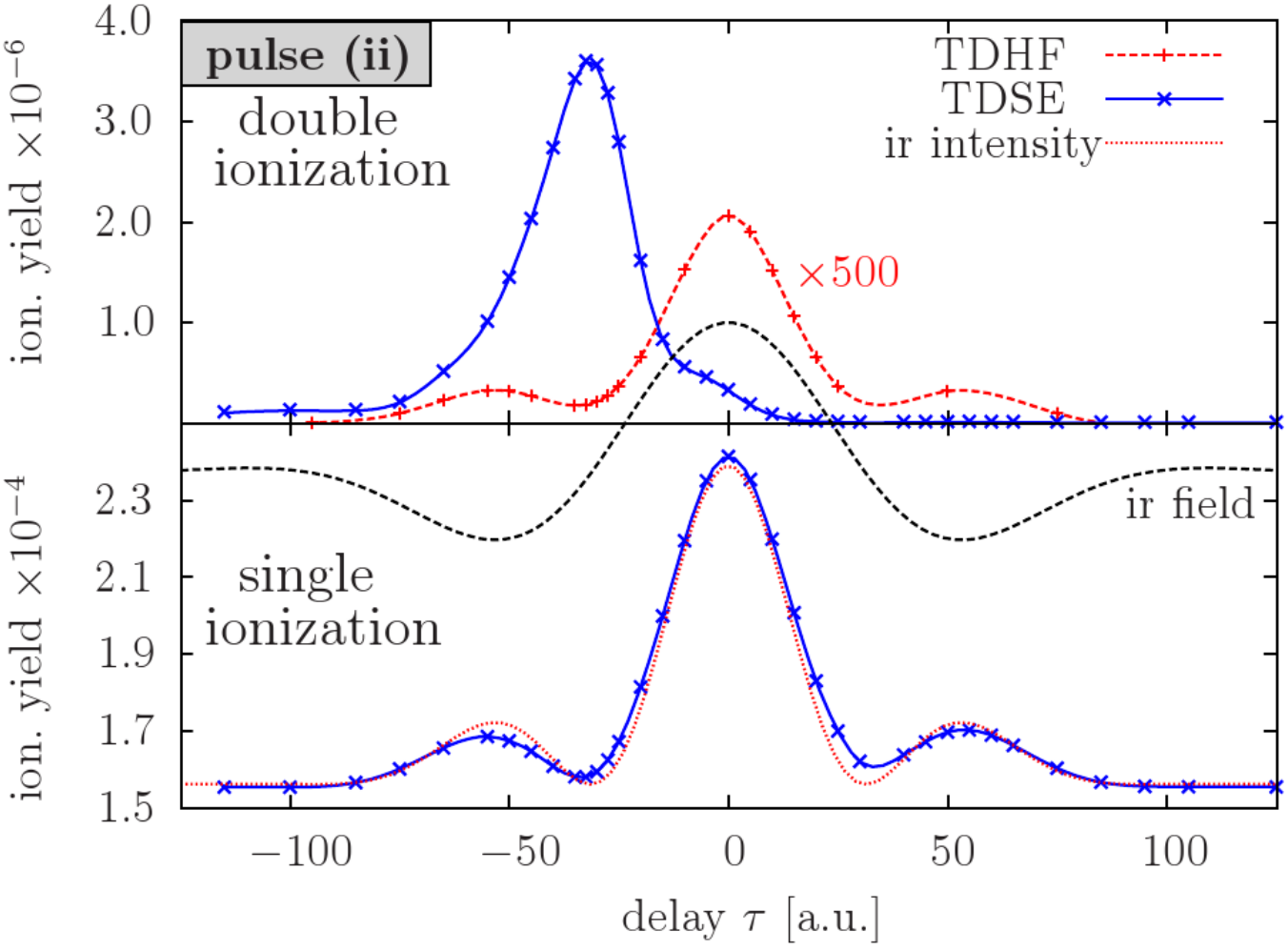


Full line: energy of electron returning to ion at time t_r : $E_{\mathrm{return}} = \frac{p^2(t_r)}{2m}$

--> delay dependence coincides with maximum of delay-dependent double ionization

--> both electrons are (directionally) *correlated* due to Coulomb interaction

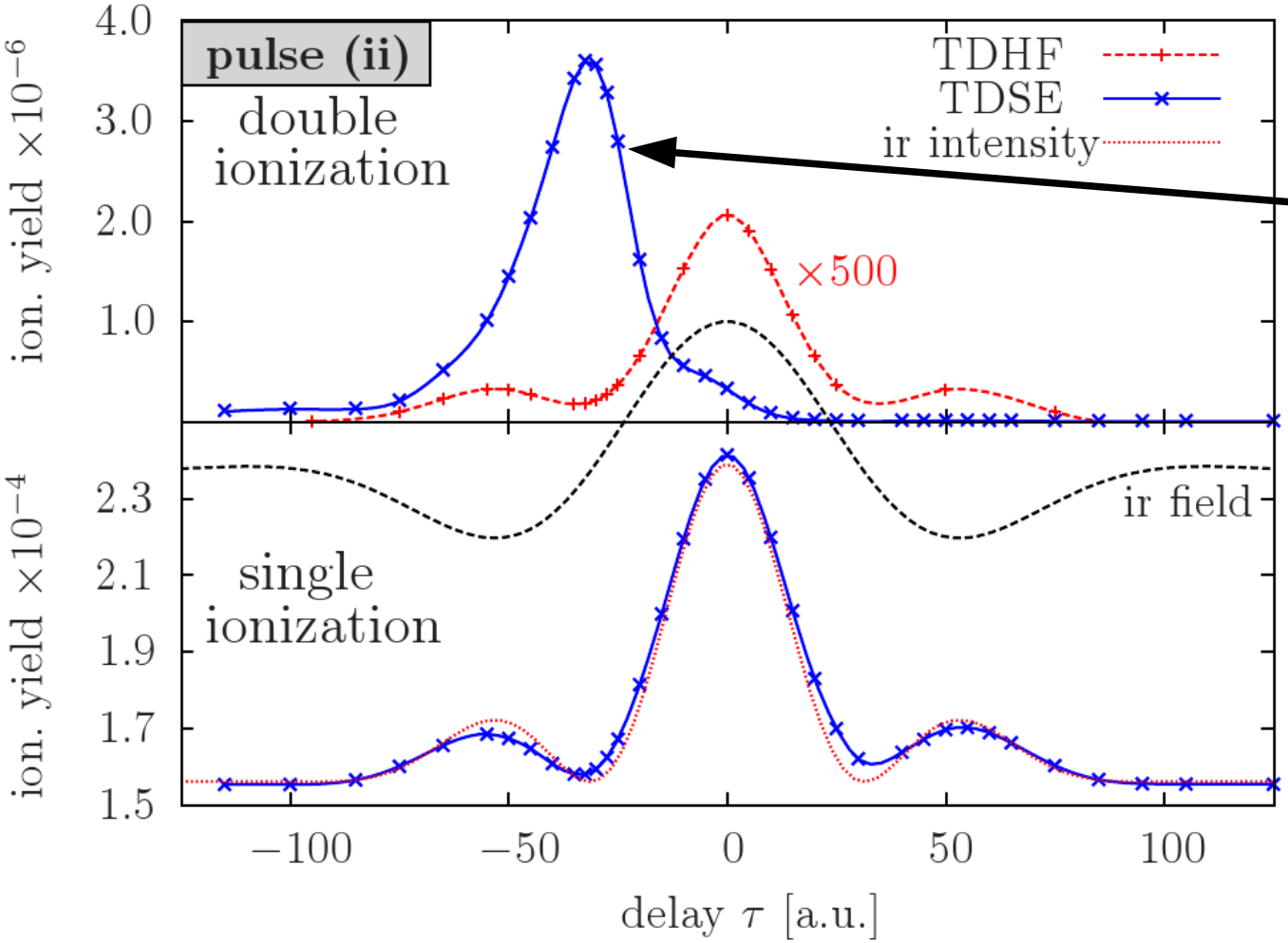
Optimize double ionization yield: increase IR intensity (x4)

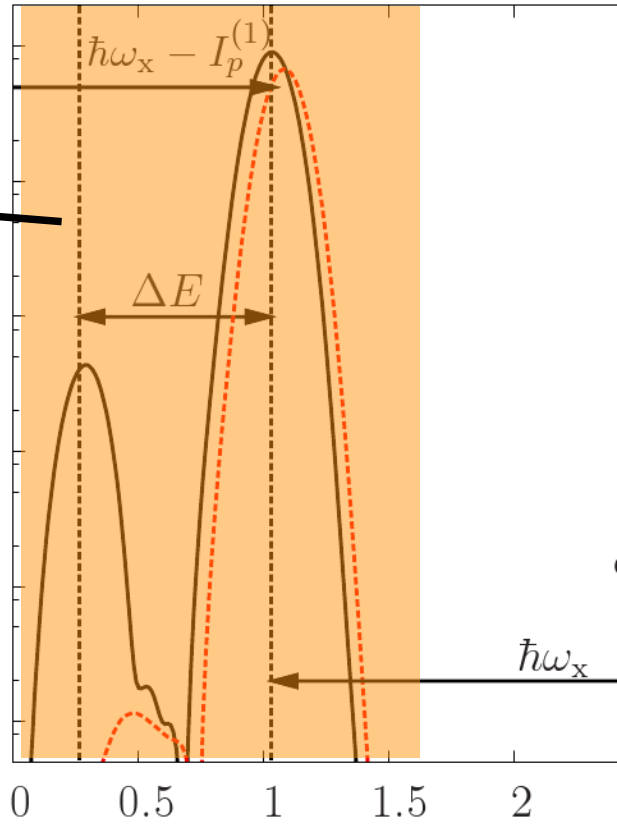


IR-Probe (ii)
Intensity **3.4 10^{14}** W/cm²
λ=900nm

S. Bauch, K.Balzer and M. Bonitz, *Europhys. Lett.* **91,** 53001 (2010)

Optimize double ionization yield: increase IR intensity (x4)





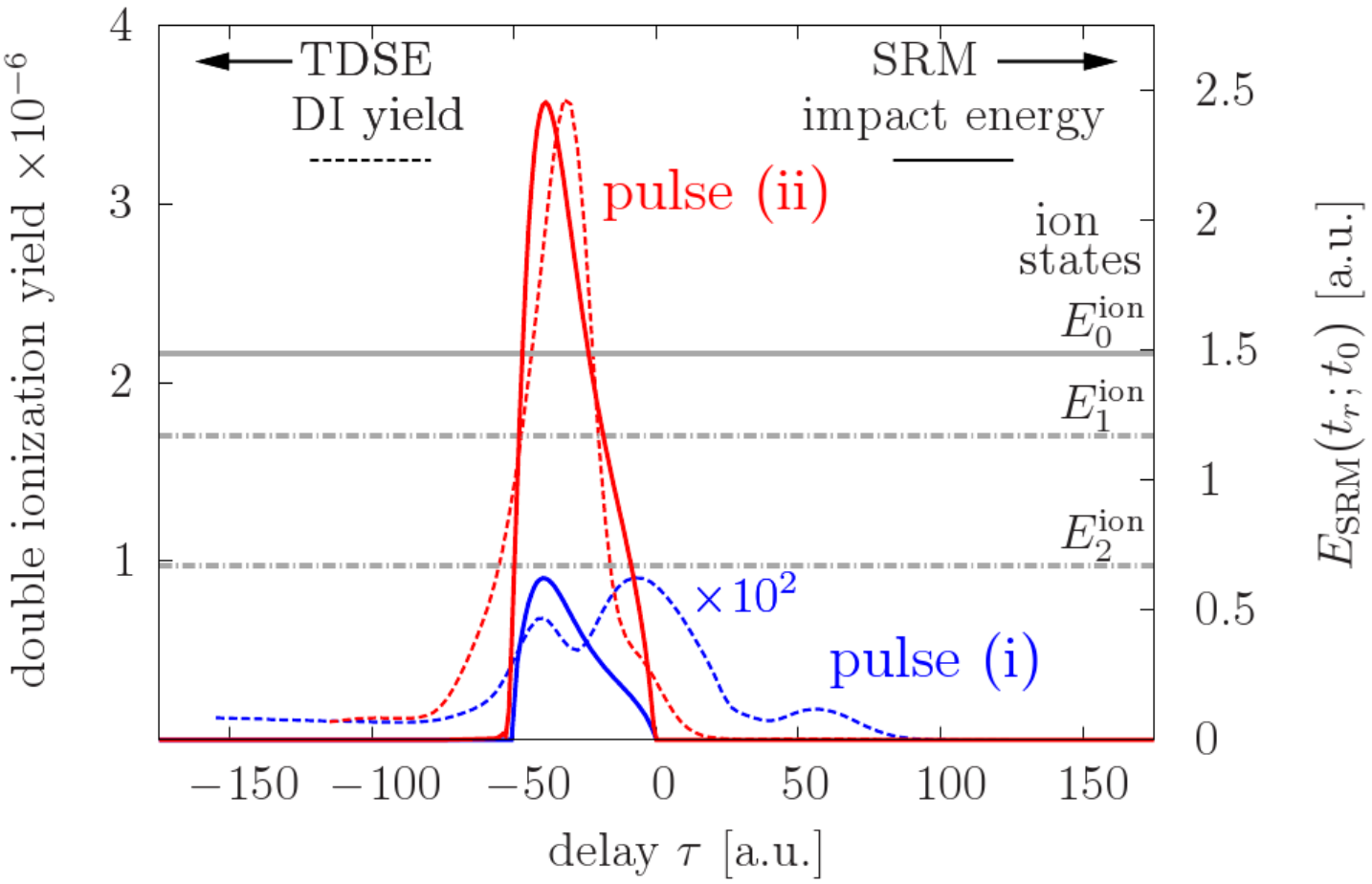
IR-Probe (ii)
Intensity **3.4 10¹⁴** W/cm²
λ=900nm

- all ionized electrons can re-scatter
- returning energy sufficient for impact ionization

--> enormous increase of double ionization

S. Bauch, K.Balzer and M. Bonitz, *Europhys. Lett.* **91,** 53001 (2010)

Double ionization yield vs. classical return energy



IR-Probe
Pulse (i):
Intensity **8.8 10^{13}** W/cm²
Pulse (ii):
Intensity **3.4 10^{14}** W/cm²

DI yields increases by factor 300!

quantum-mechanical and semi-classical results for DI agree

--> dominant mechanism: rescattering with impact ionization (plus shake-up)

S. Bauch, K.Balzer and M. Bonitz, *Europhys. Lett.* **91,** 53001 (2010)

Limitations of TDSE. Alternative approaches

- Straightforward TDSE solutions limited to approximately N=2..4 particles
- Exponential increase of memory/CPU time
- Approximate methods (wave function or density matrix) based necessaryl

Theoretical approaches 1

One-electron dynamics (e.g. streak camera)

time-dependent Schrödinger equation (TDSE)
development of adequate (simple) models

Theoretical approaches 2

Multi-electron dynamics (correlation dynamics, correlated ionization etc.)

1. Few-particle TDSE
2. Multi-configuration time-dependent Hartree Fock (MCTDHF)
3. Non-equilibrium Greens functions (NEGF)

Summary

1. **Theoretical modelling of t-resolved photoionization experiments**

   Streaking: one-electron simulations feasible but challenging

   useful: (semiclassical) models

2. **Electronic correlation effects:**

   relevant for ionization processes, rich physics

3. **Theoretical approaches to fs-dynamics of correlated systems:**

   i) time-dependent Schrödinger equation

   ii) multi-configuration time-dependent Hartree Fock

   iii) nonequilibrium Green's functions

Capabilities of theoretical approaches

	TDSE Time-dependent Schrödinger equation	MCTDHF Multi-configuration TD Hartree-Fock	NEGF Non-equilibrium Green's functions
Quantity	$\|\Psi(\boldsymbol{r}_1,\ldots,\boldsymbol{r}_N)\rangle$	$\|\Psi(\boldsymbol{r}_1,\ldots,\boldsymbol{r}_N)\rangle$	$G(\boldsymbol{r},t;\boldsymbol{r}',t')$
Level of correlation	complete	no correlation (HF) … complete	HF: no correlation Born: ~2/3 correlation energy
Particle number	N=1,2	N≤20; Effort ~ $\binom{2M}{N}$	N independent (limited by basis)
System size	>10000 a.u. (N=1) 1000 a.u (N=2)	depends on M and N	~100 a.u.
time pro-pagation	~t	~t	~t^2 (~t ?)
Pump-probe	one- and two-particle systems	depends on M and N generally possible	Not feasible
Many-particle systems	Not possible	Not possible	e.g. solids, plasmas through self energies

Summary and Outlook

3. **Theoretical approaches to fs-dynamics of correlated systems:**
   i) time-dependent Schrödinger equation
   ii) multi-configuration time-dependent Hartree Fock
   iii) nonequilibrium Green's functions

There is no „holy grail“ solving all problems!

All methods have very different strengths and limitations,
they complement each other.

Further developments and smart combinations are required.