(Sub)femtosecond control of electron dynamics in atoms, molecules and nanostructures

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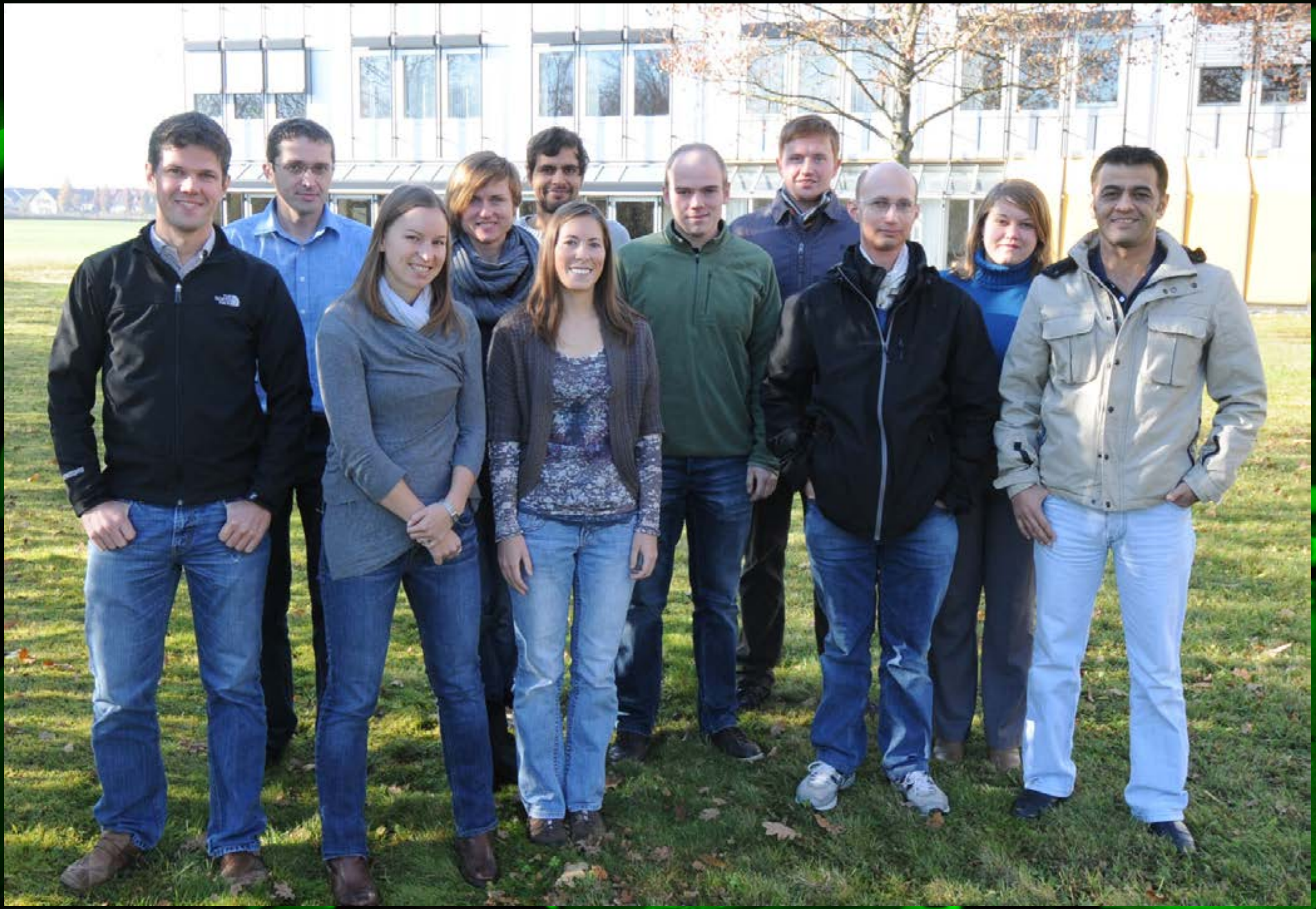

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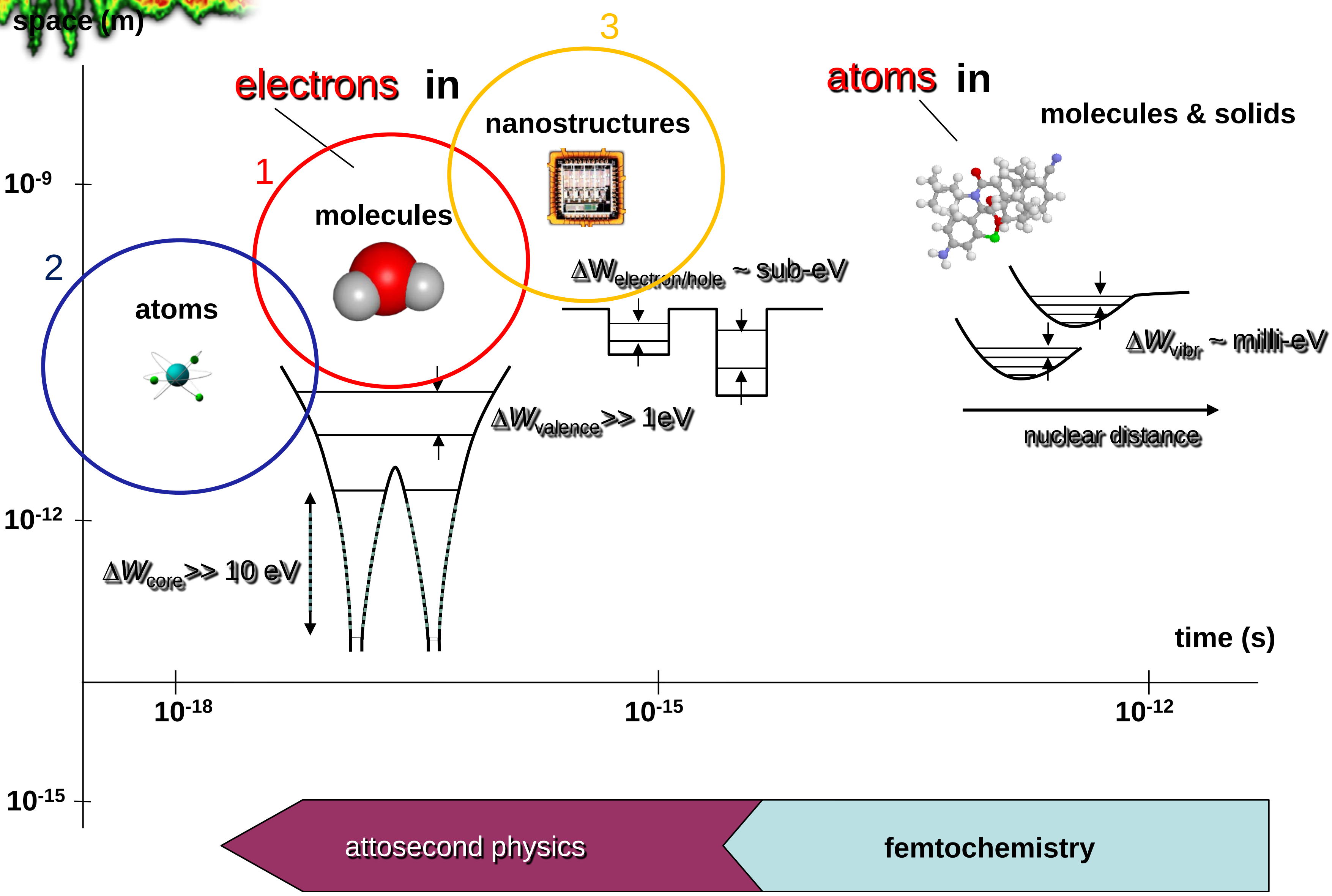
and others ...

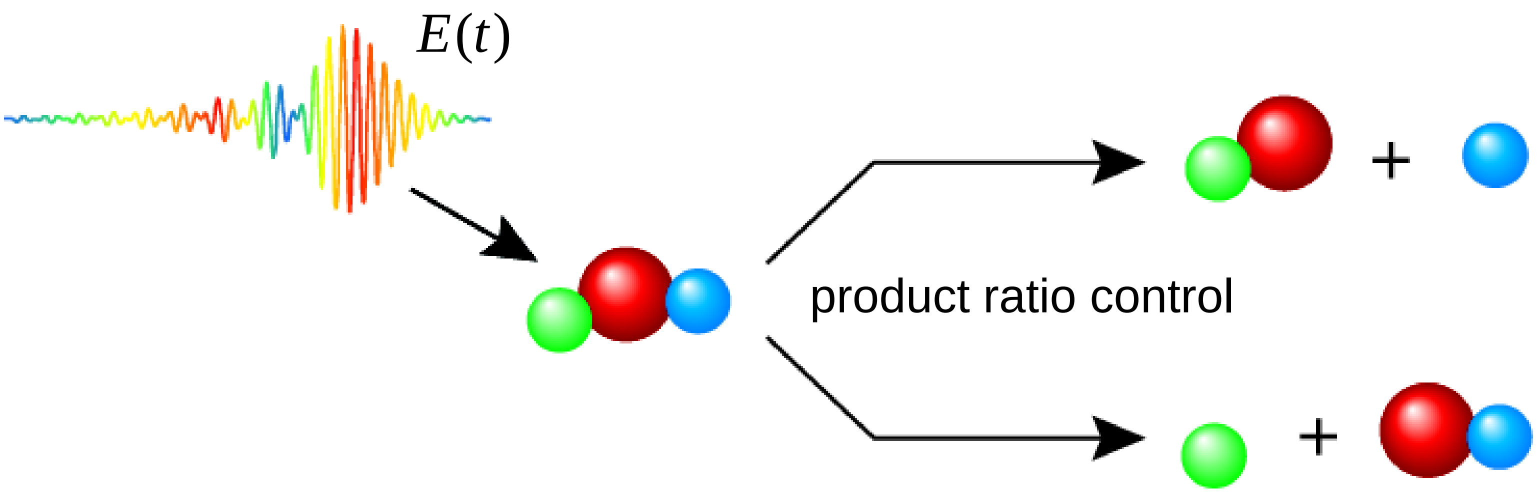
Motion on ultrashort timescales



Courtesy: F. Krausz

Controlling electrons in molecules

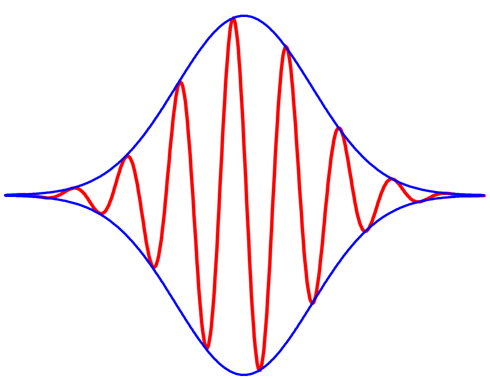
Coherent control via femtosecond pulse shaping



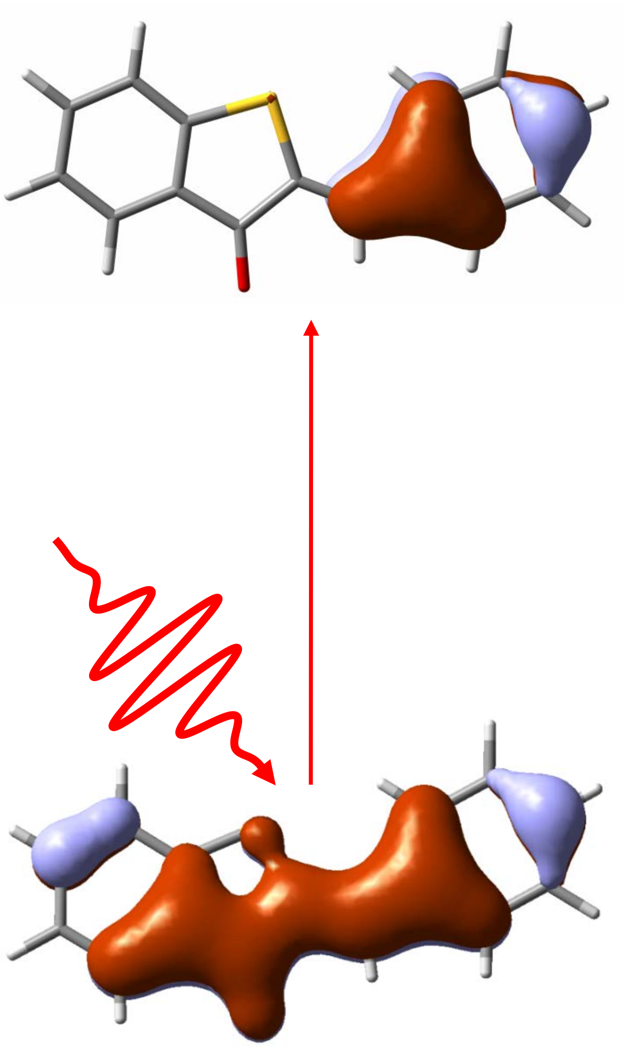
e.g. Wollenhaupt et al., *Ann. Rev. Phys. Chem*. **56**, 25 (2005)

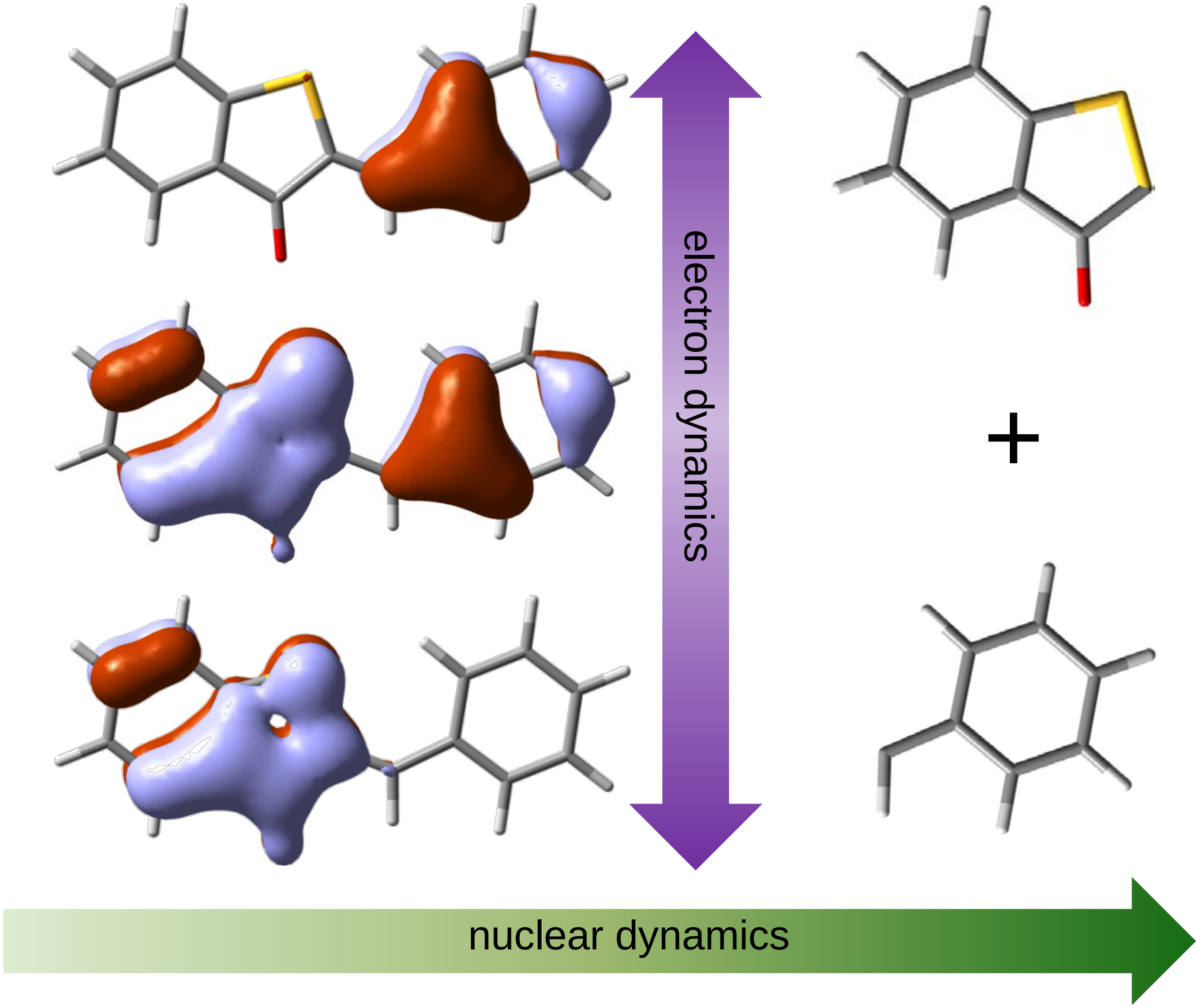
Control parameters via *E*(*t*):
intensity, frequency, polarization, pulse duration

Can the **electric field waveform** act as photonic reagent to control electronic motion?



Non Born-Oppenheimer dynamics in molecules





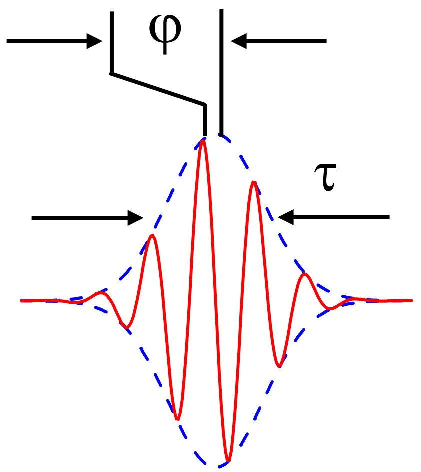
Courtesy Philipp von den Hoff

Spatial asymmetry in HD^+ (and H_2^+) dissociation – controlled via CEP

CEP-controlled dissociation of HD^+

V. Roudnev *et al.*, *PRL* **93** (2004) 163601

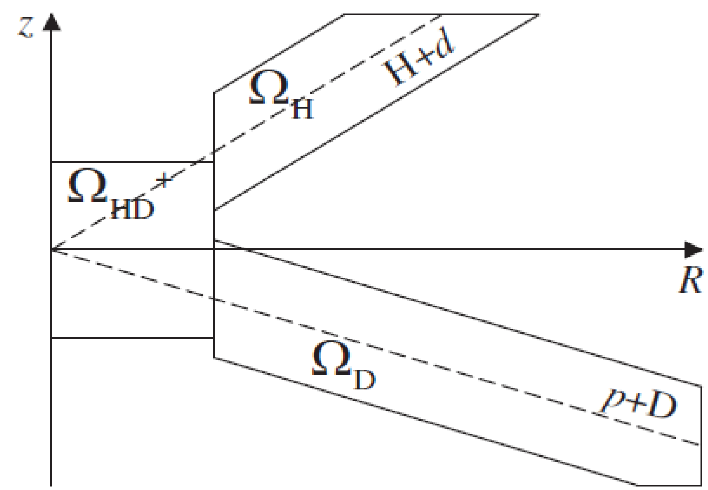
Few-cycle laser pulses



Envelope ***CEP***

$$E(t) = E_0(t)\cos(\omega t + \varphi)$$

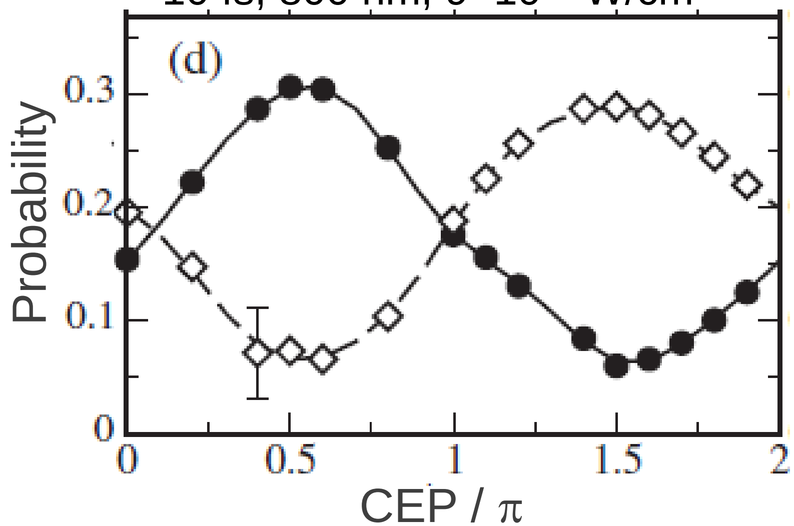
Frequency



$H + D^+$

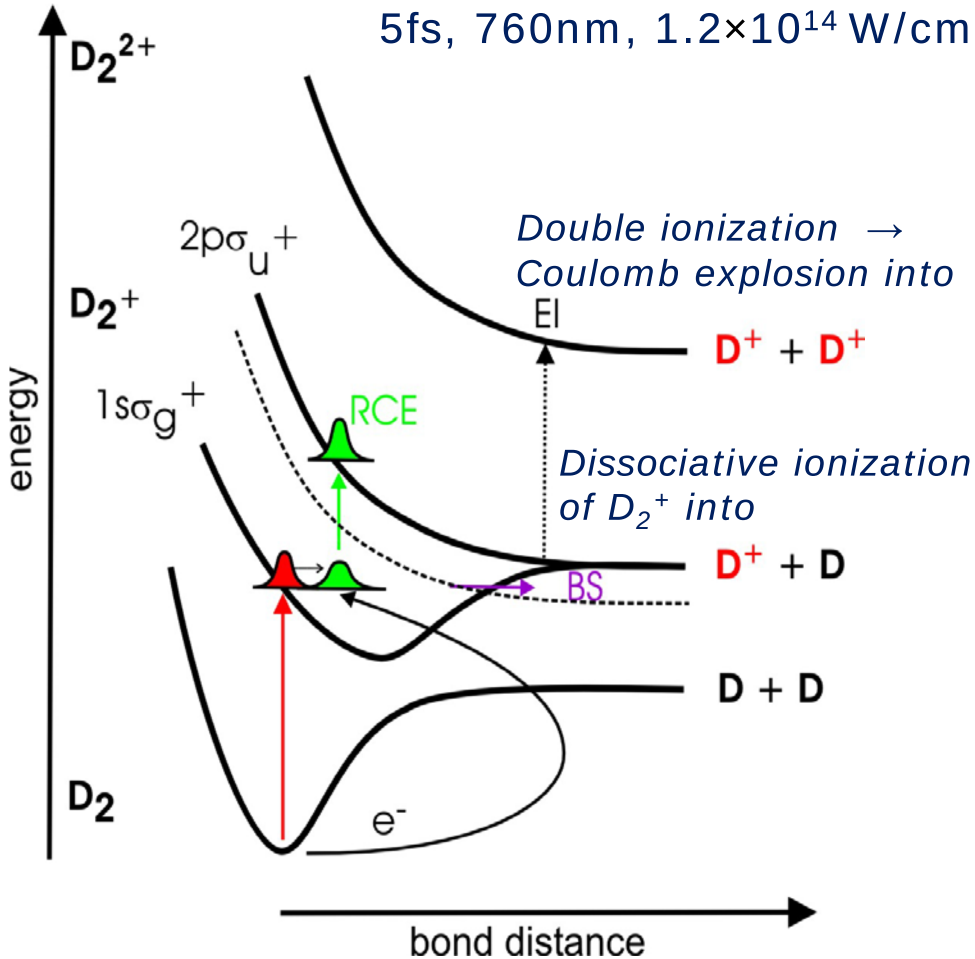
$H^+ + D$

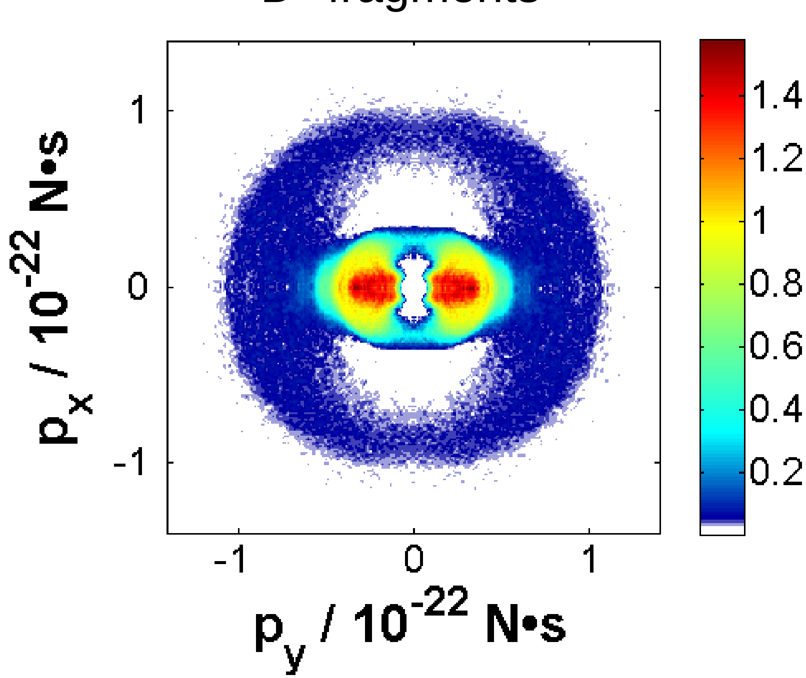
10 fs, 800 nm, 9×10^{14} W/cm^2



Experiment: strong-field dissociation of D_2

5fs, 760nm, 1.2×10^{14} W/cm^2



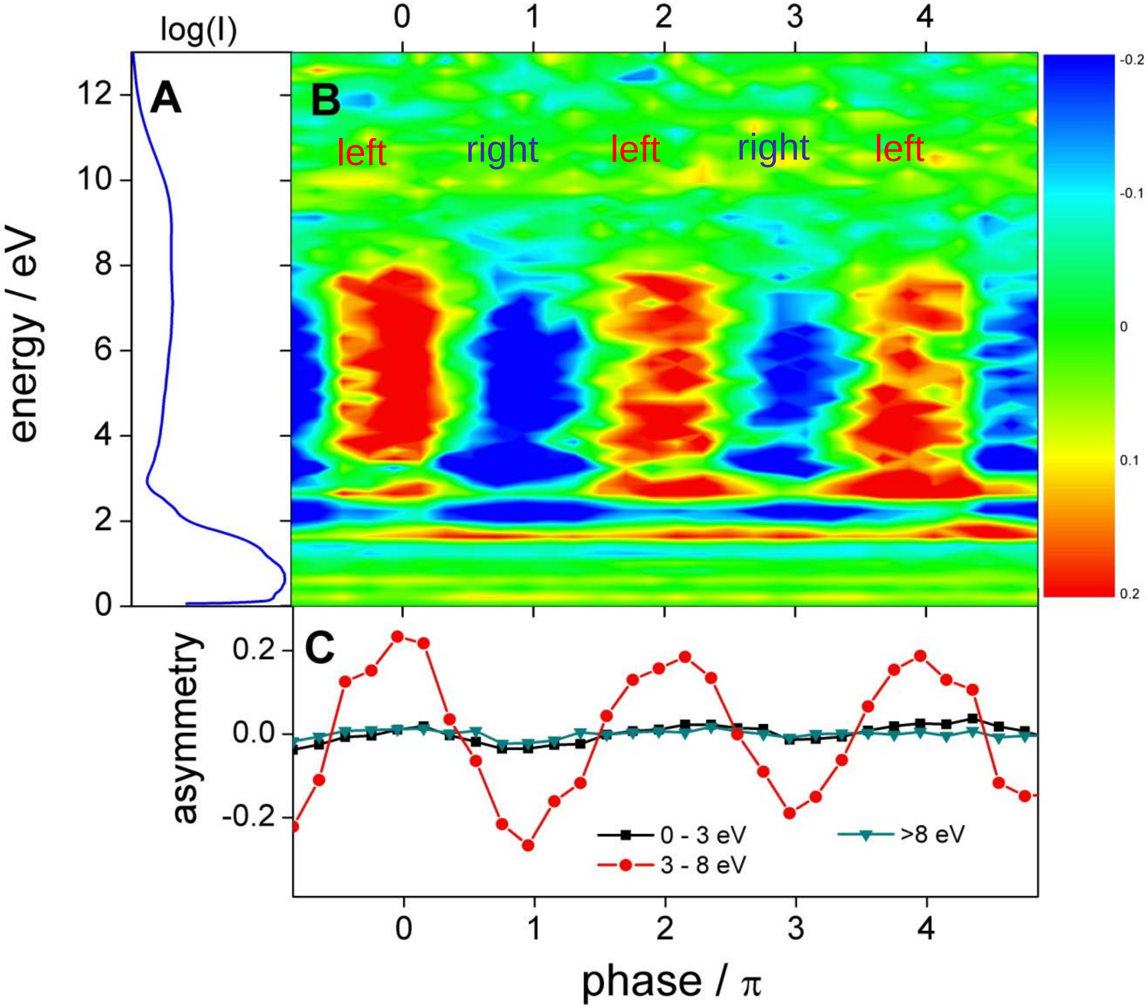


Kling *et al.*, *Science* **312** (2006) 246.

Hydrogen dissociation – emission of D^+

Asymmetry = (left – right) / (left + right)

Kling et al., Science 312 (2006) 246, Mol. Phys. 106 (2008) 455



=> Direction of D^+ emission is controlled by light-waveform

Control of the asymmetry - model

QM simulation of D_2^+
numerically solving the Schrödinger equation

$$i\frac{\partial\Psi(\vec{r},R;t)}{\partial t} = H\Psi(\vec{r},R;t)$$

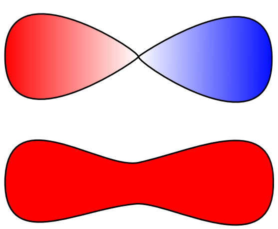
$$\Psi(\vec{r},R;t) \approx |g\rangle\psi_g(R;t) + |u\rangle\psi_u(R;t)$$

with the electronic states $|g\rangle$ ($1s\sigma_g$) and $|u\rangle$ ($2p\sigma_u$), nuclear wave packets $\psi_{g/u}(R;t)$

Change electronic basis to one with localization left / right

$|u\rangle$

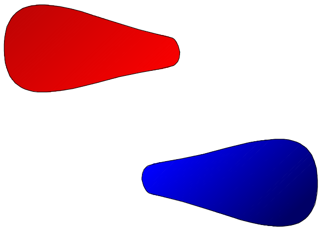
$|g\rangle$



⇨

$$|l\rangle = \frac{1}{\sqrt{2}}\left(|g\rangle + |u\rangle\right)$$

$$|r\rangle = \frac{1}{\sqrt{2}}\left(|g\rangle - |u\rangle\right)$$

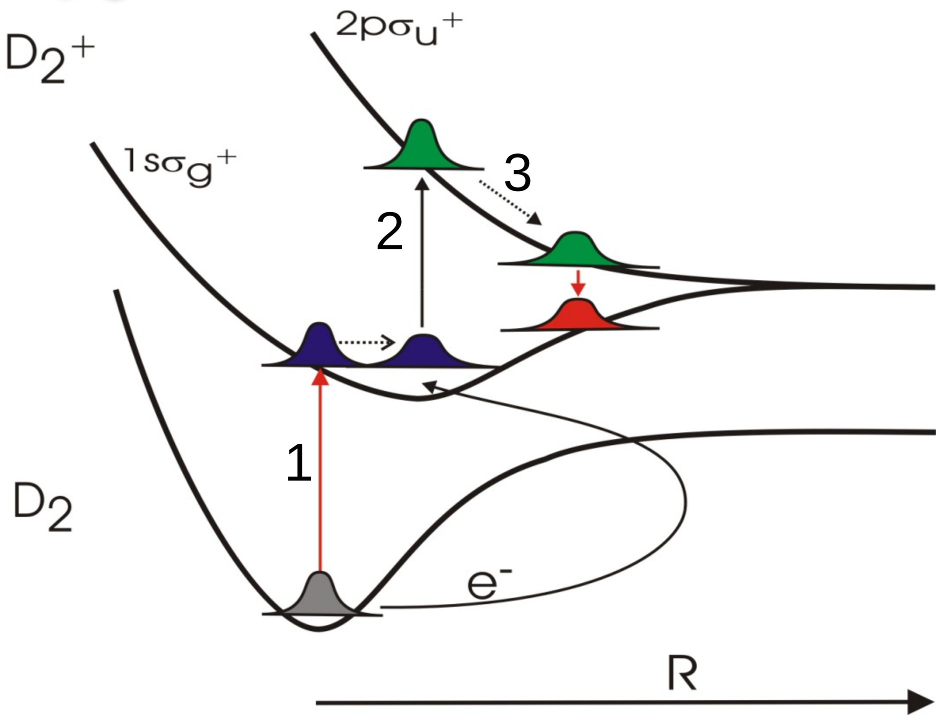


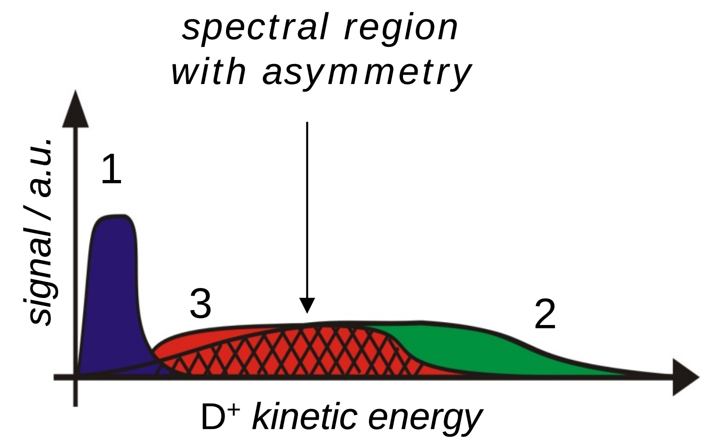
Asymmetry A(t):

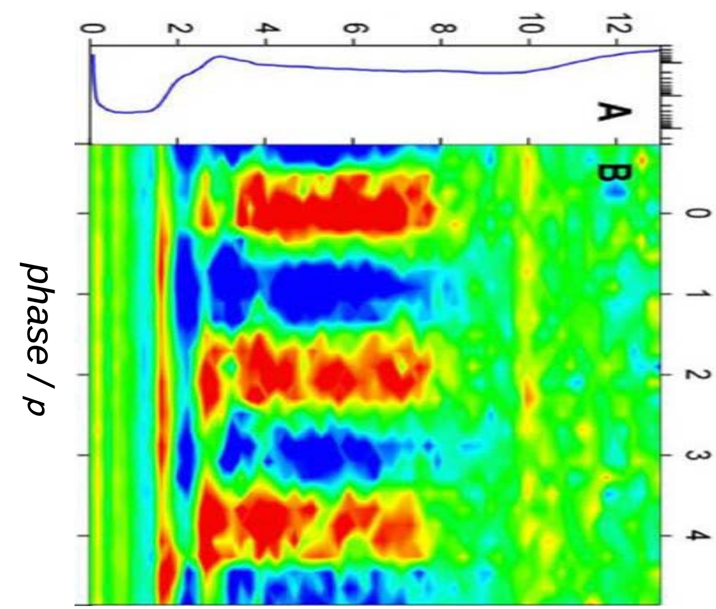
$$A(t) \equiv \frac{\int (P_l - P_r)dR}{\int (P_l + P_r)dR}$$

Kling et al., Science 312 (2006) 246.

Hydrogen dissociation – Mechanistic picture







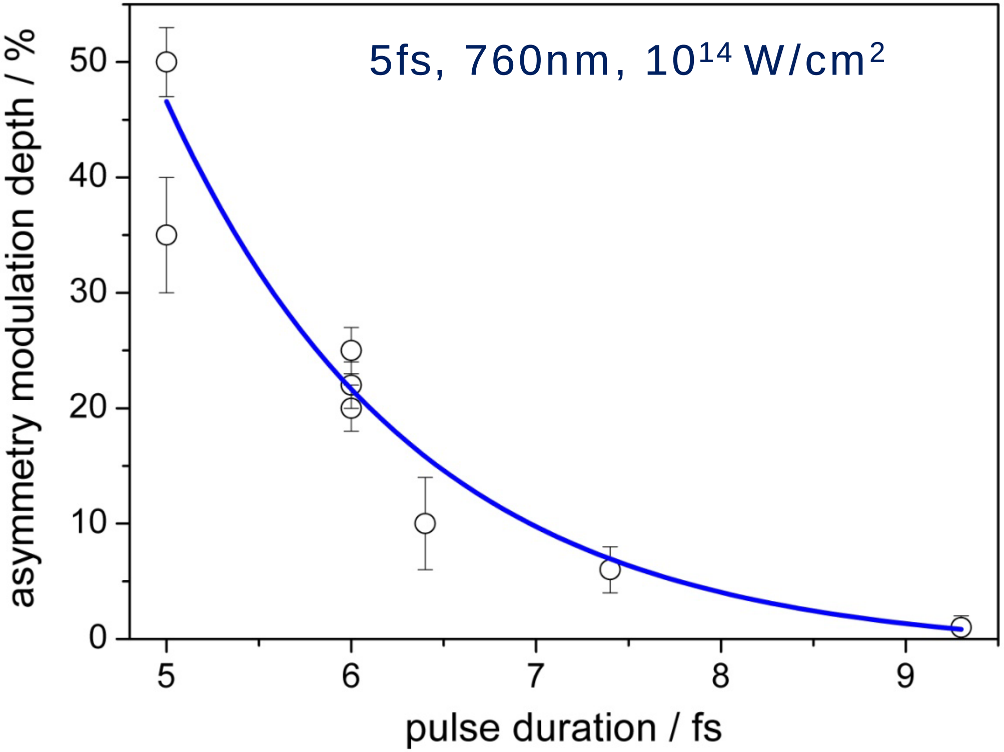
1 Ionization of D_2

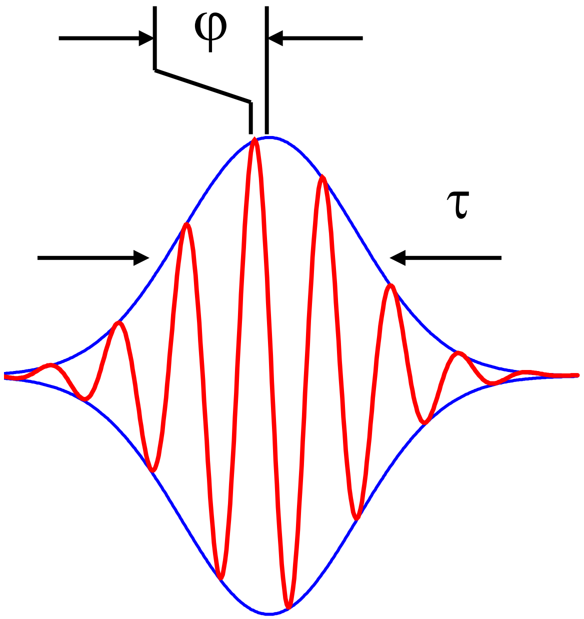
2 Recollisional excitation

3 Preparation of ***coherent superposition*** $(1s\sigma_g^+, 2p\sigma_u^+)$

The importance of pulse duration

*Kling et al., Mol. Phys. **106** (2008) 455*





Why are few-cycle pulses essential for (clear) observation of the phase dependence?

Contributions from consecutive half-cycles with opposite sign (in laser-matter interaction)

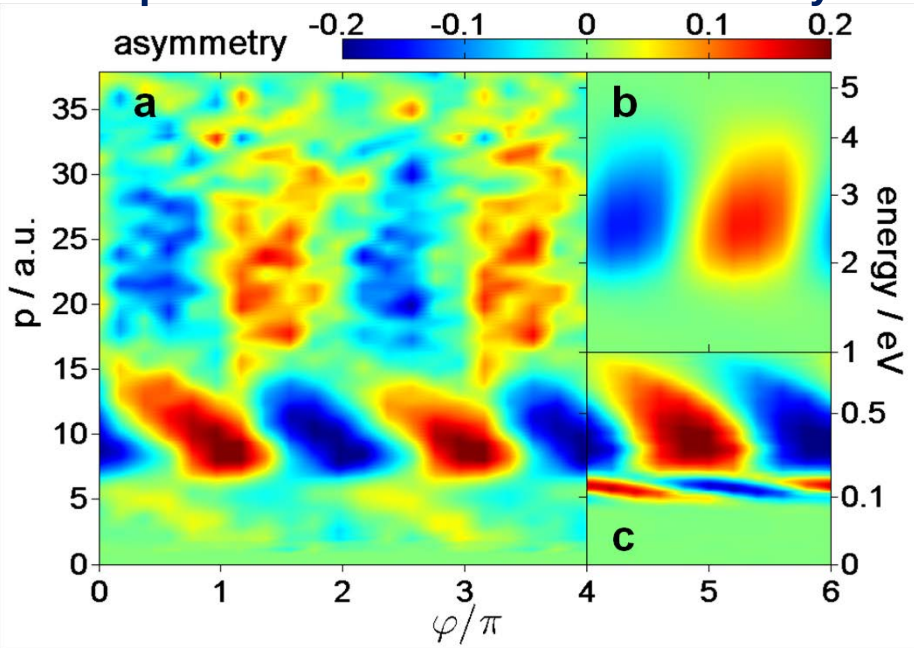
Exponential decay with pulse duration predicted by general theory:

V. Roudnev and B.D. Esry, *PRL* **99** (2007) 220406

Deuterium dissociation at 2.1 µm: D^+ ion emission asymmetry

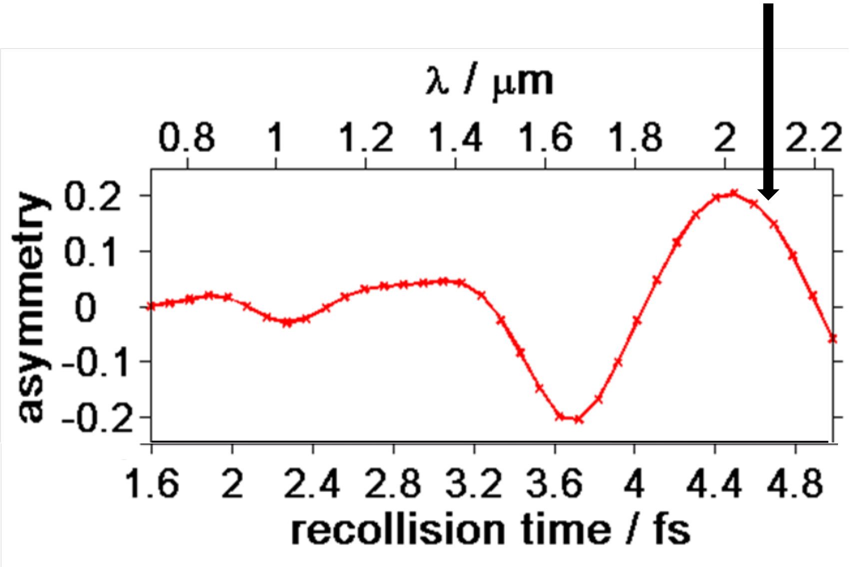
I. Znakovskaya *et al*., *PRL* **108** (2012) 063002

experiment **theory**



Recollisional excitation (RCE) channel

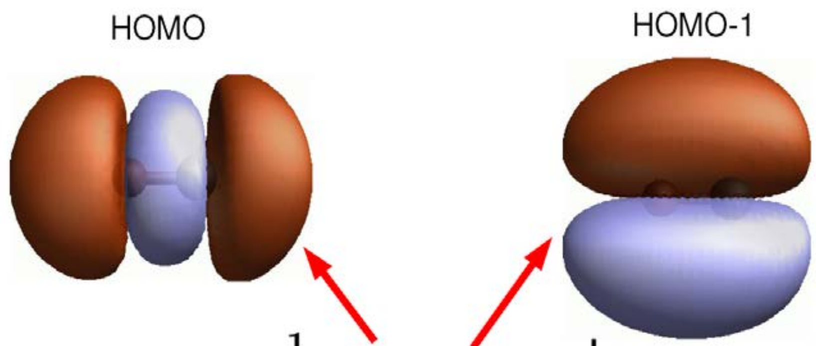
Bond softening (BS) channel



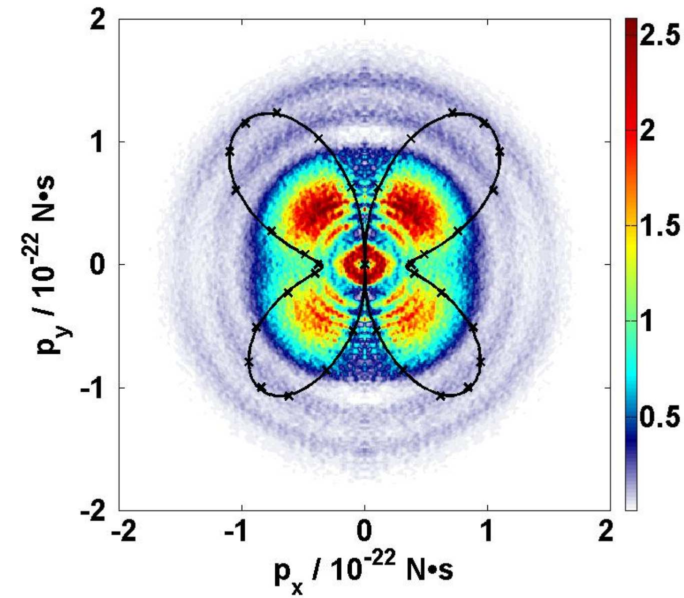
Wavelength dependence asymmetric dissociation (BS channel)

Light waveform driven molecular dynamics

Can we extend this idea to more complex systems?



$$\rho_{SP}(r;R) = \frac{1}{\sqrt{2}}(\varphi_1^* \pm \varphi_2^*) * \frac{1}{\sqrt{2}}(\varphi_1 \pm \varphi_2)$$
$$= \frac{1}{2}|\varphi_1|^2 + \frac{1}{2}|\varphi_2|^2 \pm \varphi_1^*\varphi_2$$



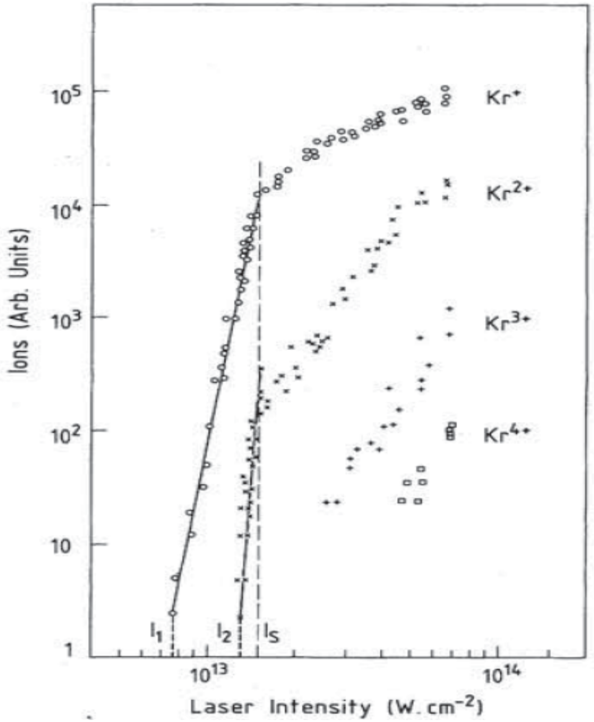
I. Znakovskaya *et al.*, **PRL** 103, 053002 (2009)

- orientation-dependent ionization selection [1]
- imaging of multiple (!) molecular orbitals from which ionization took place (here HOMO + HOMO-1) [2]
- strongly coupled electron-nuclear dynamics [3]

➞ *Unraveling such effects in the NBO dynamics of complex molecules is challenging !*

[1] I. Znakovskaya *et al.*, PCCP 13, 8653 (2011); [2] P. von den Hoff *et al.*, APB 98, 659 (2010); [3] I. Znakovskaya *et al.*, **PRL** 108, 063002 (2012)

Controlling correlated electron motion: non-sequential double ionization (NSDI)

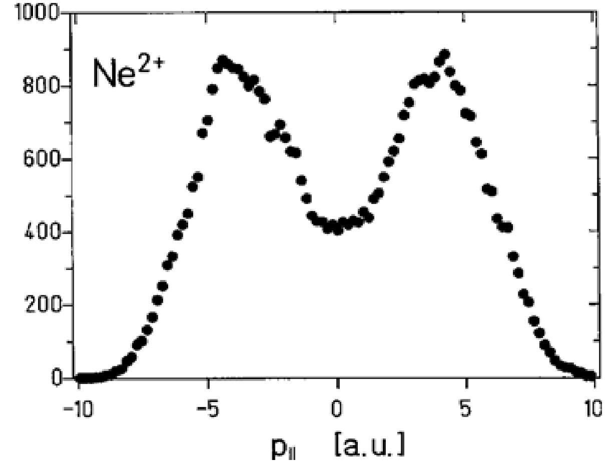


Manifestation of NSDI:

Knee structure

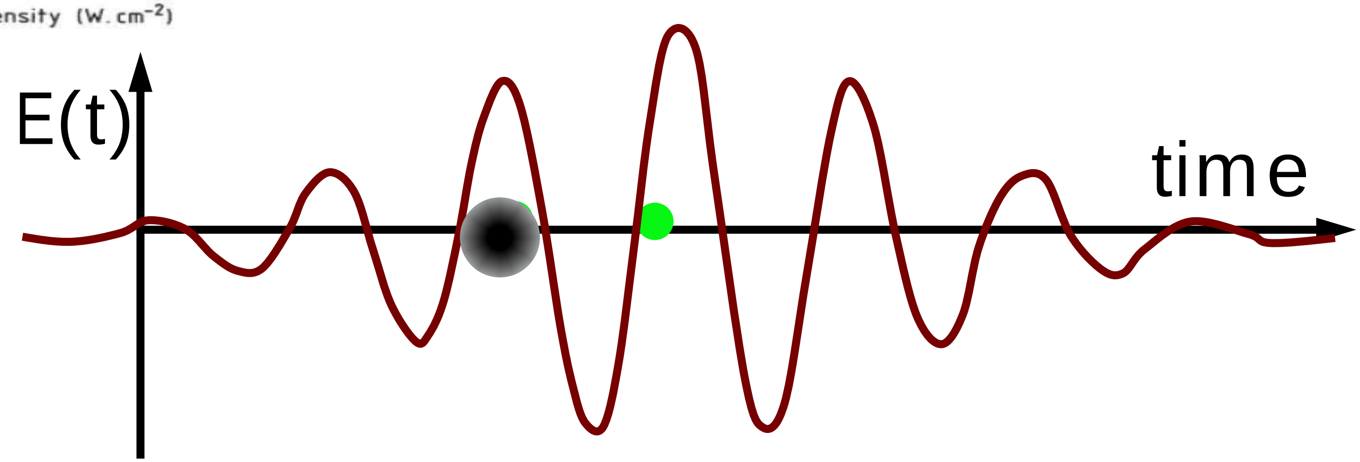
A. L`Huillier *et al*., *PRL* **48**, 1814 (1982)

Recollision mechanism apparent in ion recoil



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

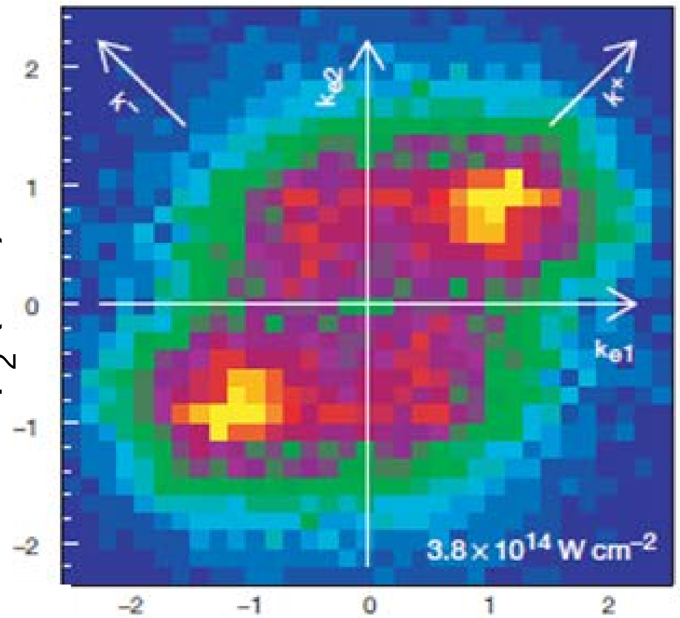
NSDI



Two-electron momentum distributions from coincidence measurements

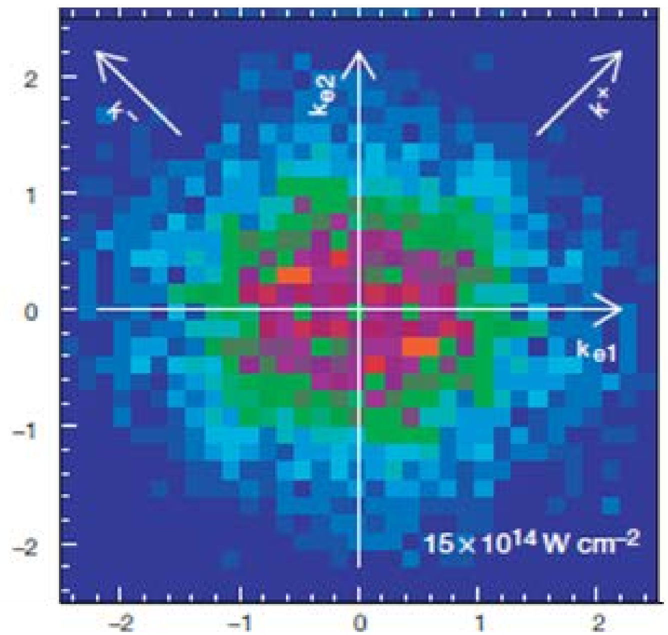
Th. Weber *et al*., *Nature* **405**, 658 (2000)

Ar^{2+} on the knee | **Ar^{2+} above the knee**



p_2 (a.u.)

p_1 (a.u.)



p_1 (a.u.)

strongly correlated electron momenta

uncorrelated electron momenta

=> NSDI is a good candidate to study the dynamics of correlated electrons

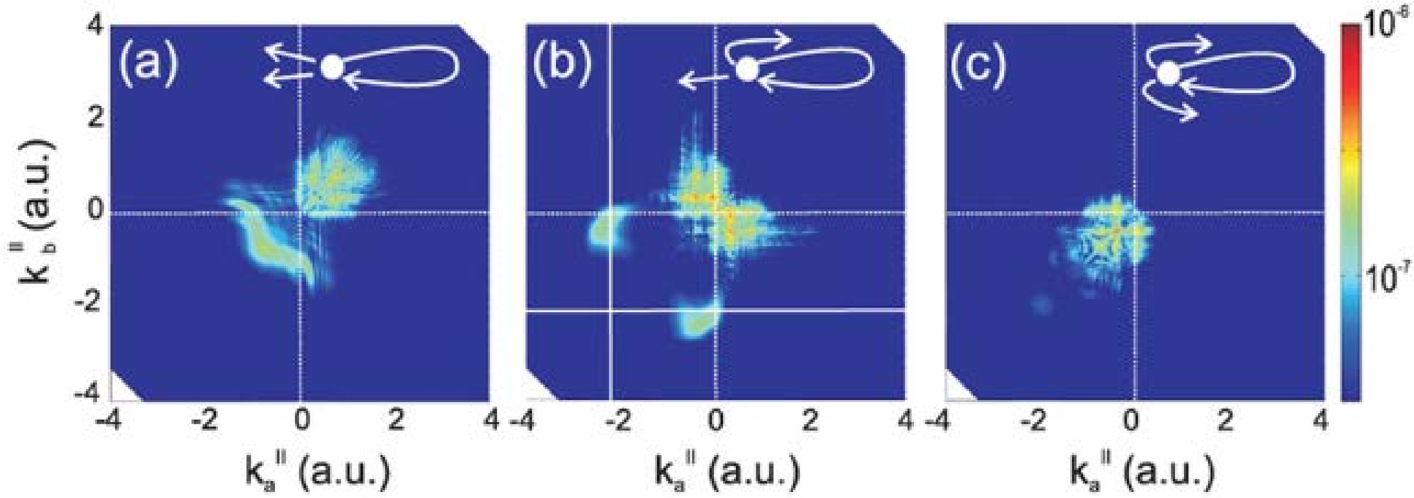
What happens for a single recollision?

Short pulses and CEP-control:

Multiple recollisions (for long pulses) complicate the dynamics !!!
→ Short CEP-controlled pulses can confine NSDI
to a single recollision event !

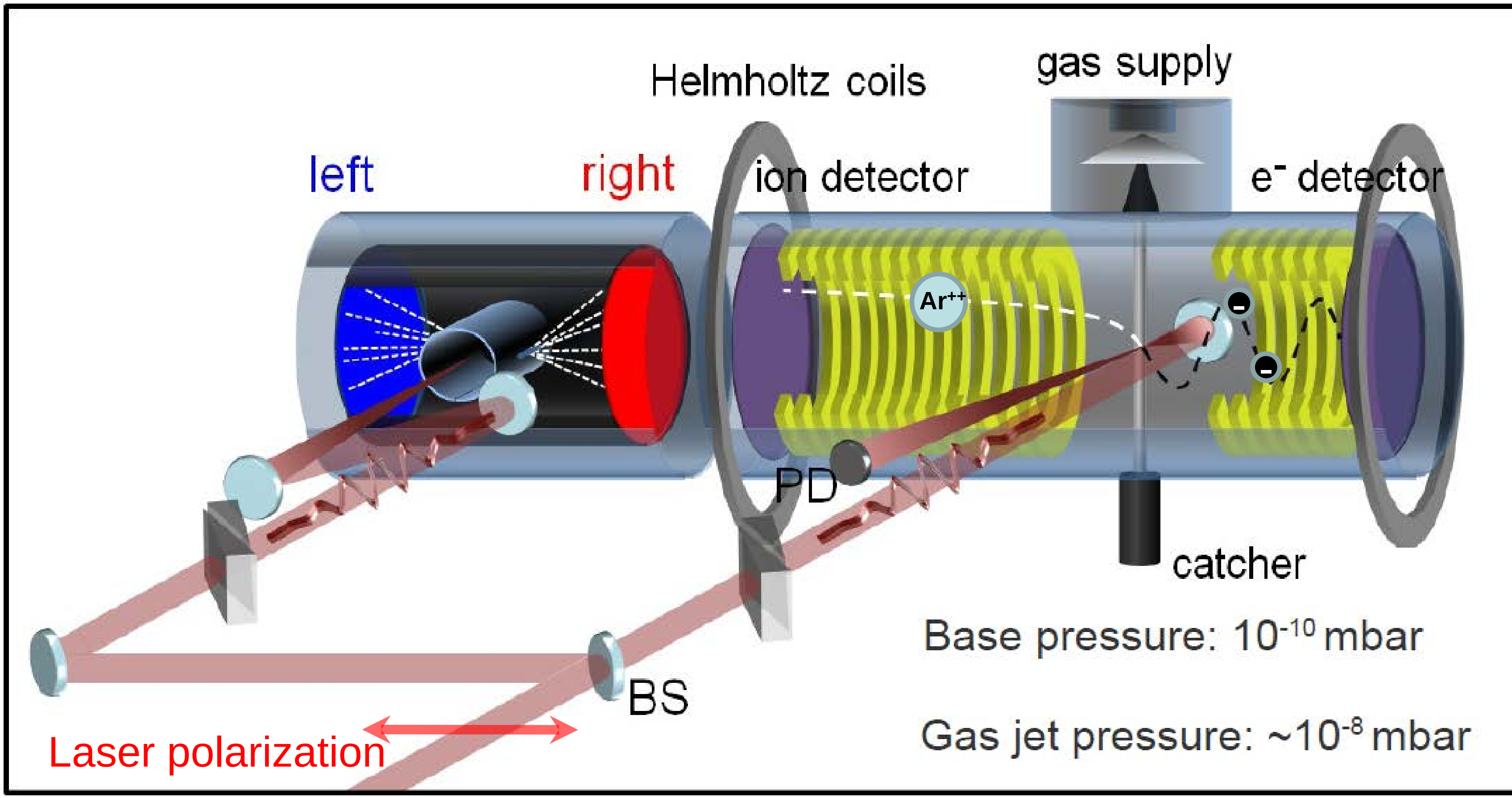
Energy and momentum sharing of the two electrons?
Time delay between the recollision and subsequent ionization?

Plenty of theory work: e.g. A. Staudte *et al.*, Phys. Rev. Lett. **99**, 263002 (2007)

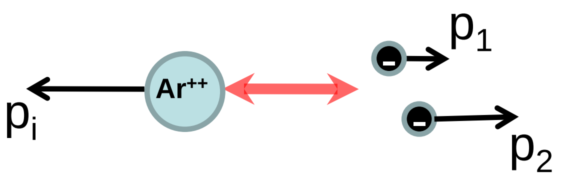


CEP-tagged coincidence experiments on correlated electron emission

Johnson et al., PRA 83, 013412 (2011); Rathje et al., JPB 45, 074003 (2012)



⇒ For each laser shot we record: ϕ and p_1,p_2 and p_i along the laser polarization direction.



Correlated two-electron momentum distribution

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

Target gas:
Argon

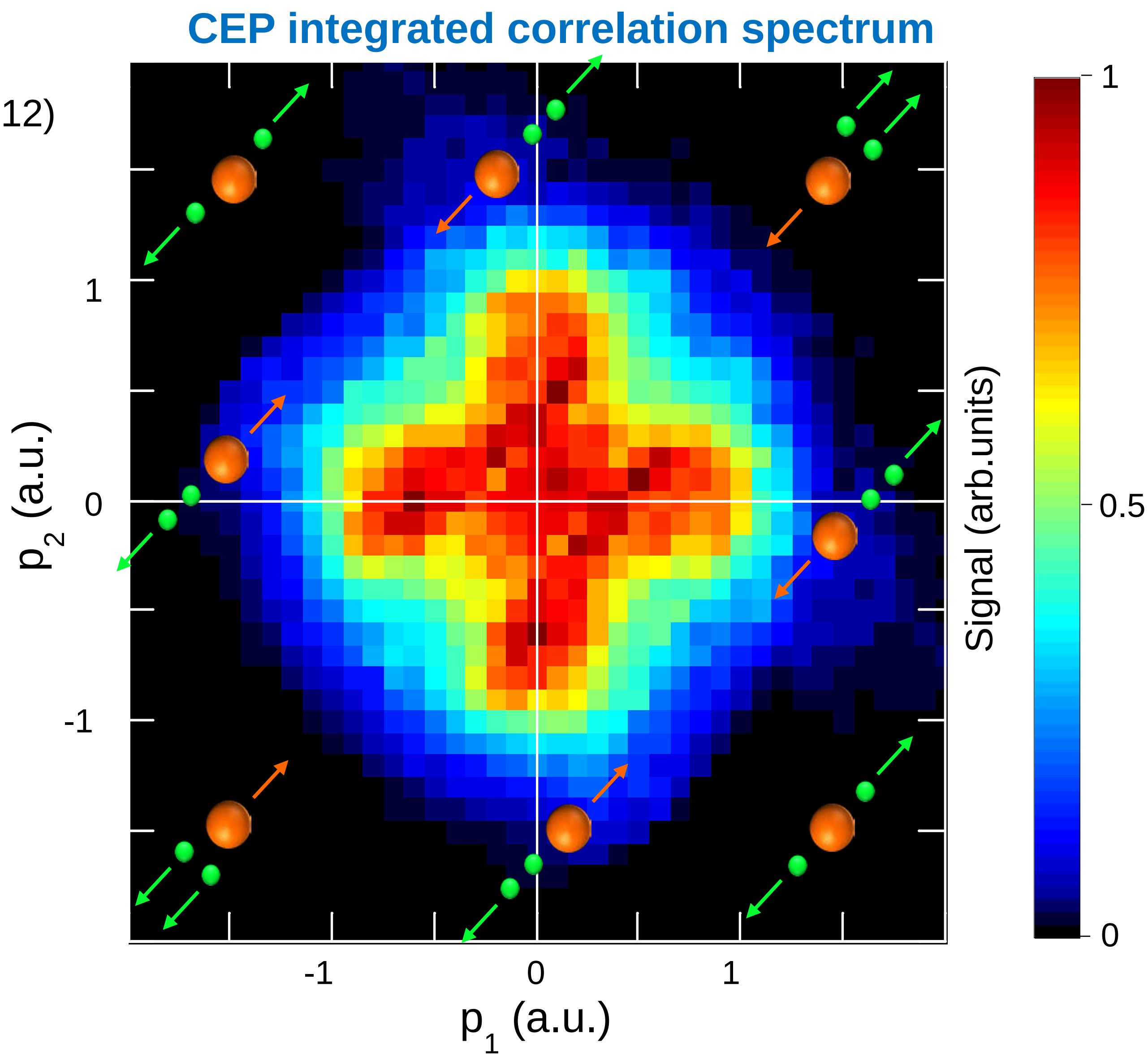
Pulse duration:
~4 fs

Wavelength:
750 nm

Peak intensity:
~3 x 10^{14} W/cm^2

Acquisition time:
~30 hours

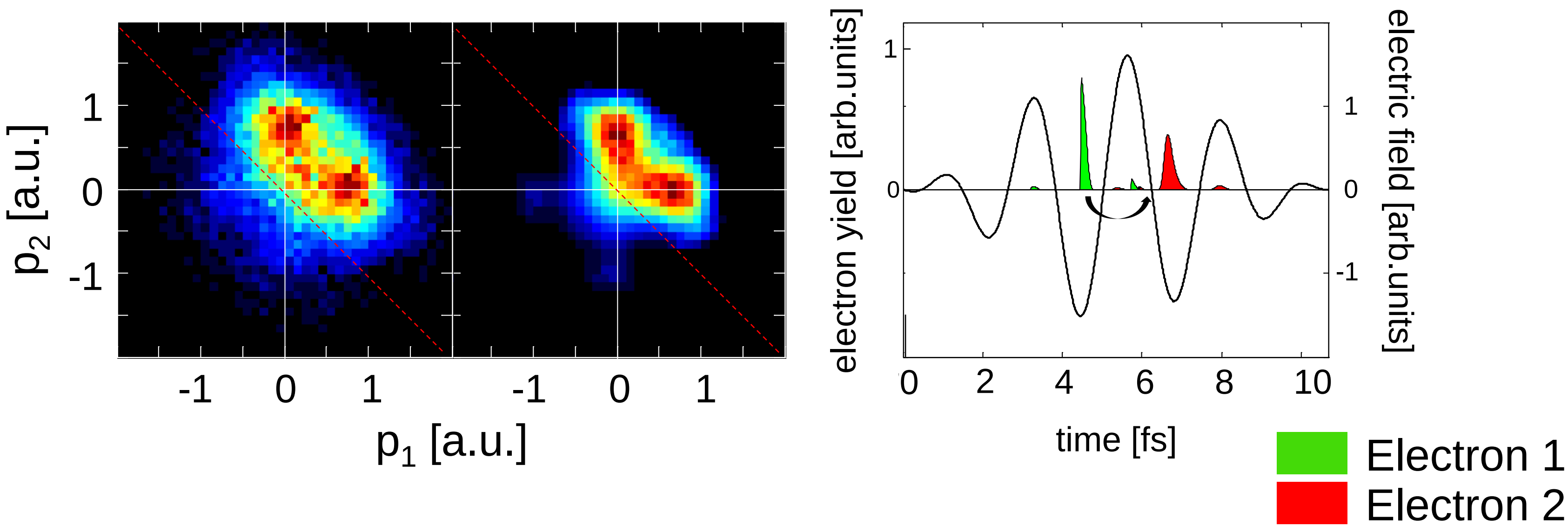
# of recorded Ar^{2+} ions:
~50 000



Sub-cycle dynamics

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

CEP with maximum asymmetry in the left right Ar^{2+} ions yield



- **The second electron** carries **high momentum**, while the first electron stays close to zero

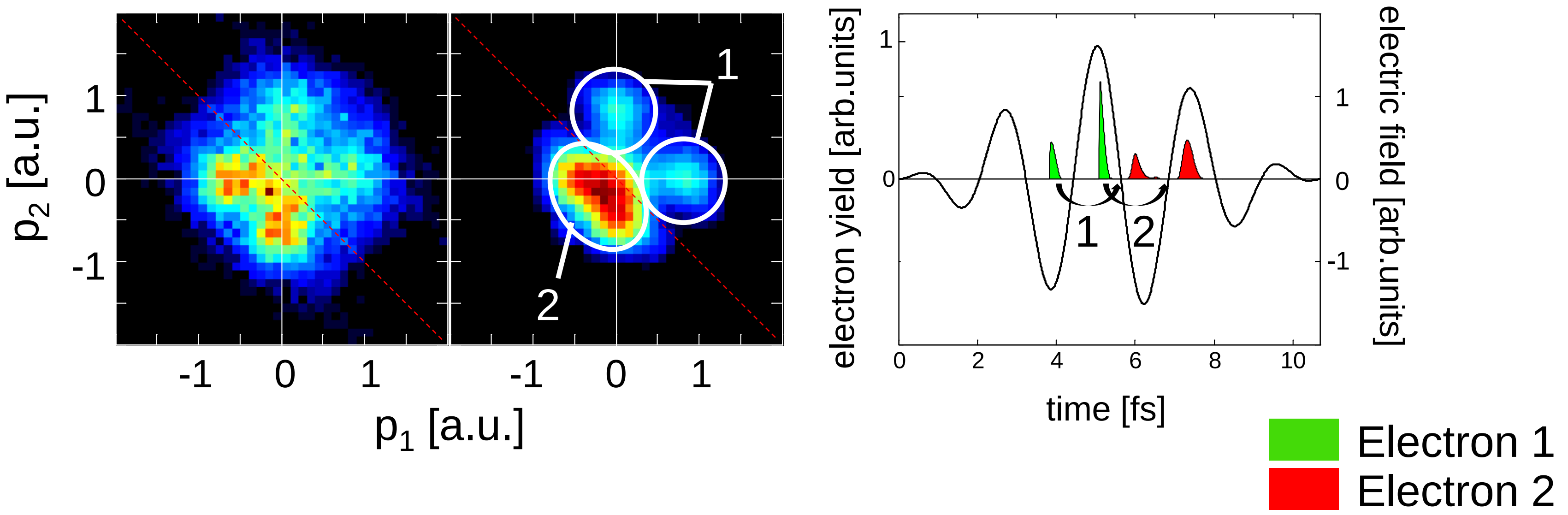
- The highest ionization probability of the second electron is reached (**210 +/- 40) attoseconds before** the field maximum

The calculation predicts **230 as**

Sub-cycle dynamics

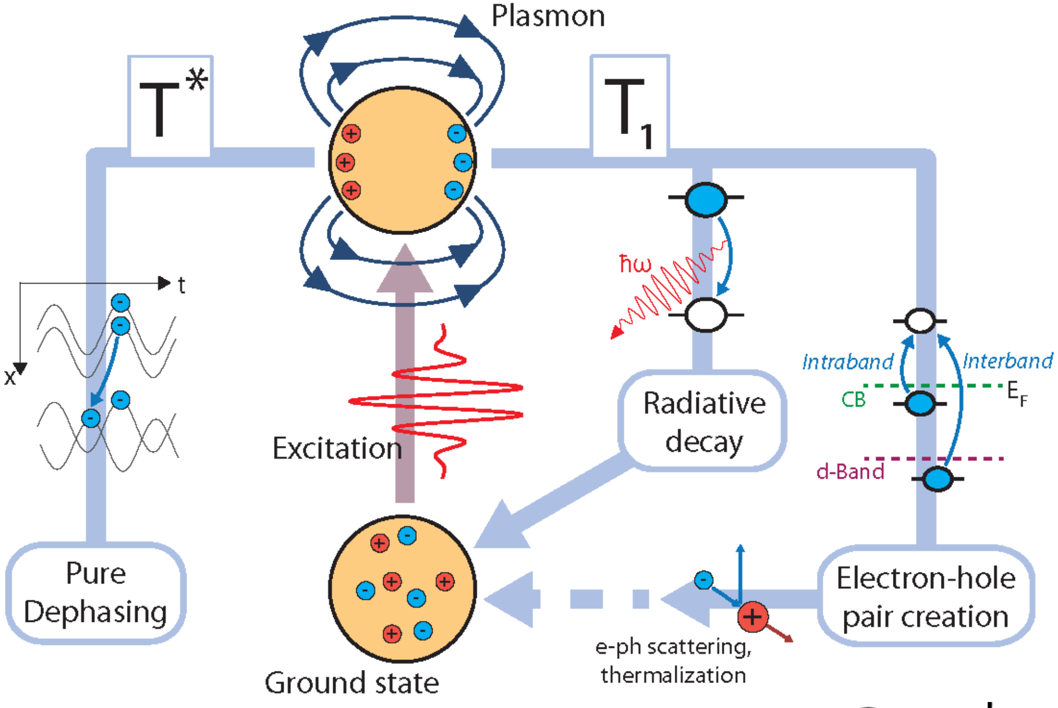
B. Bergues *et al*., *Nature Commun*. 3, 813 (2012)

CEP with zero asymmetry in the left right Ar^{2+} ions yield



- If the CEP is shifted by 90°, **2 consecutive recollision** events contribute
- Both recollision events can be **distinguished** in the experiment

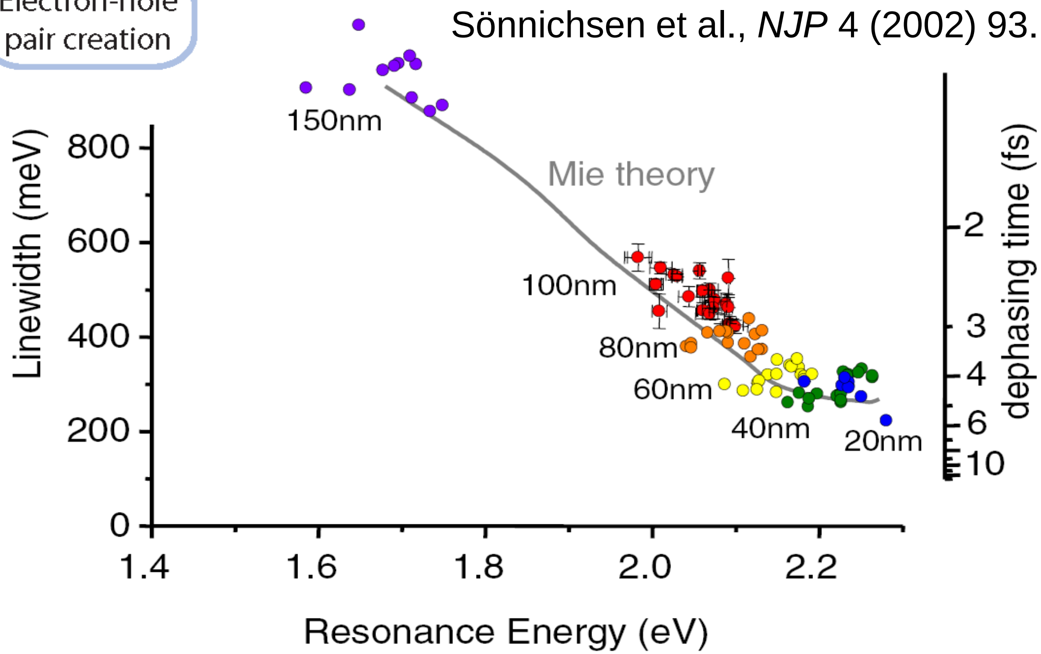
Controlling collective electron motion in nanostructures



Süßmann et al., *in „Attosecond Physics“, Wiley*

Ultrashort (few-fs) dephasing times

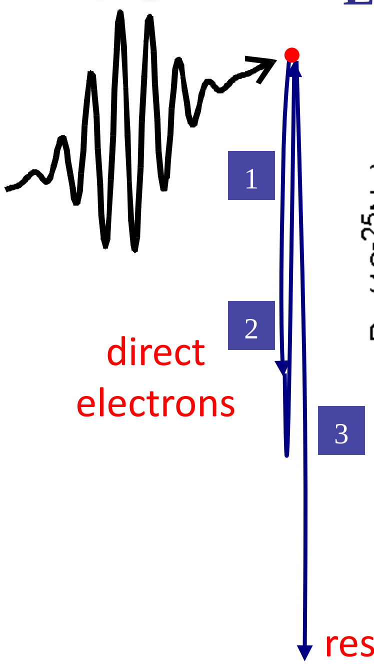
Sönnichsen et al., *NJP* 4 (2002) 93.

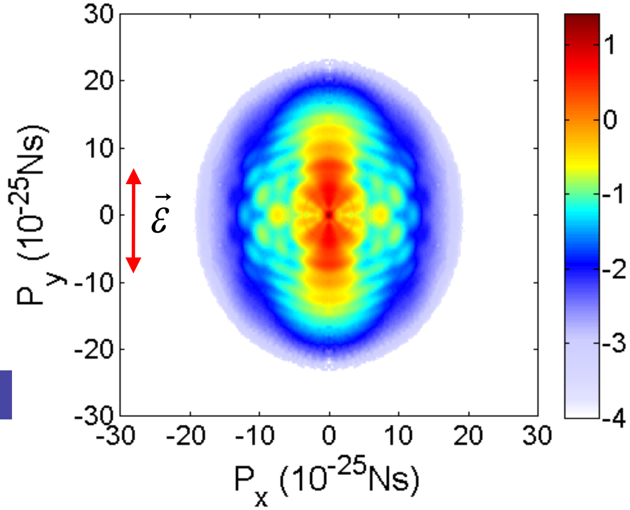


Fastest dynamics on the 100 as timescale

Controlling electron acceleration in nanolocalized near-fields

Electron emission from atoms

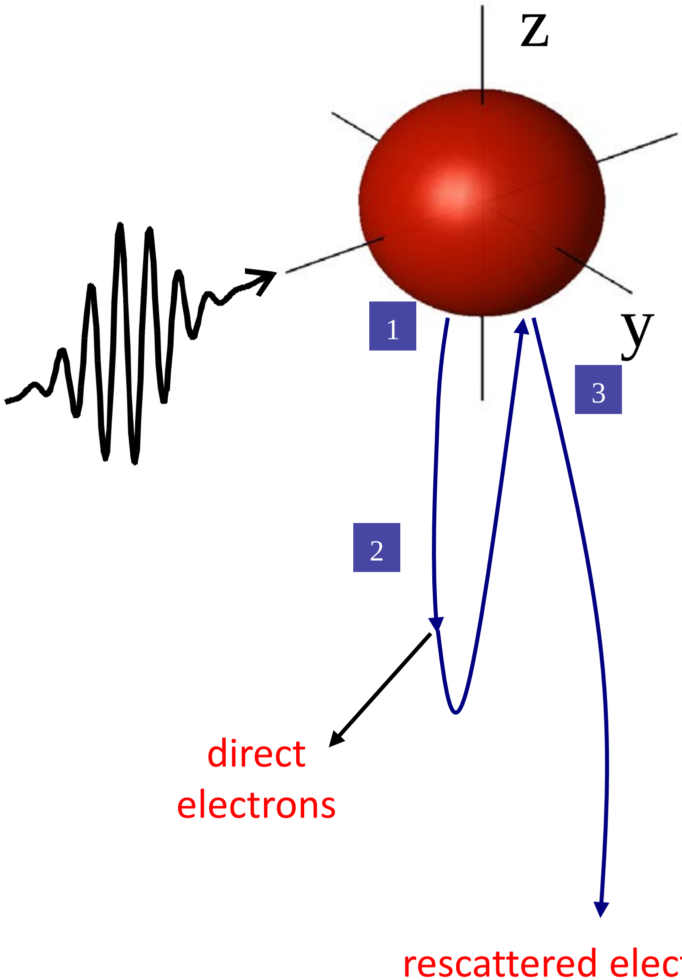




rescattered electrons

classical cutoff energy: **10 U_p**

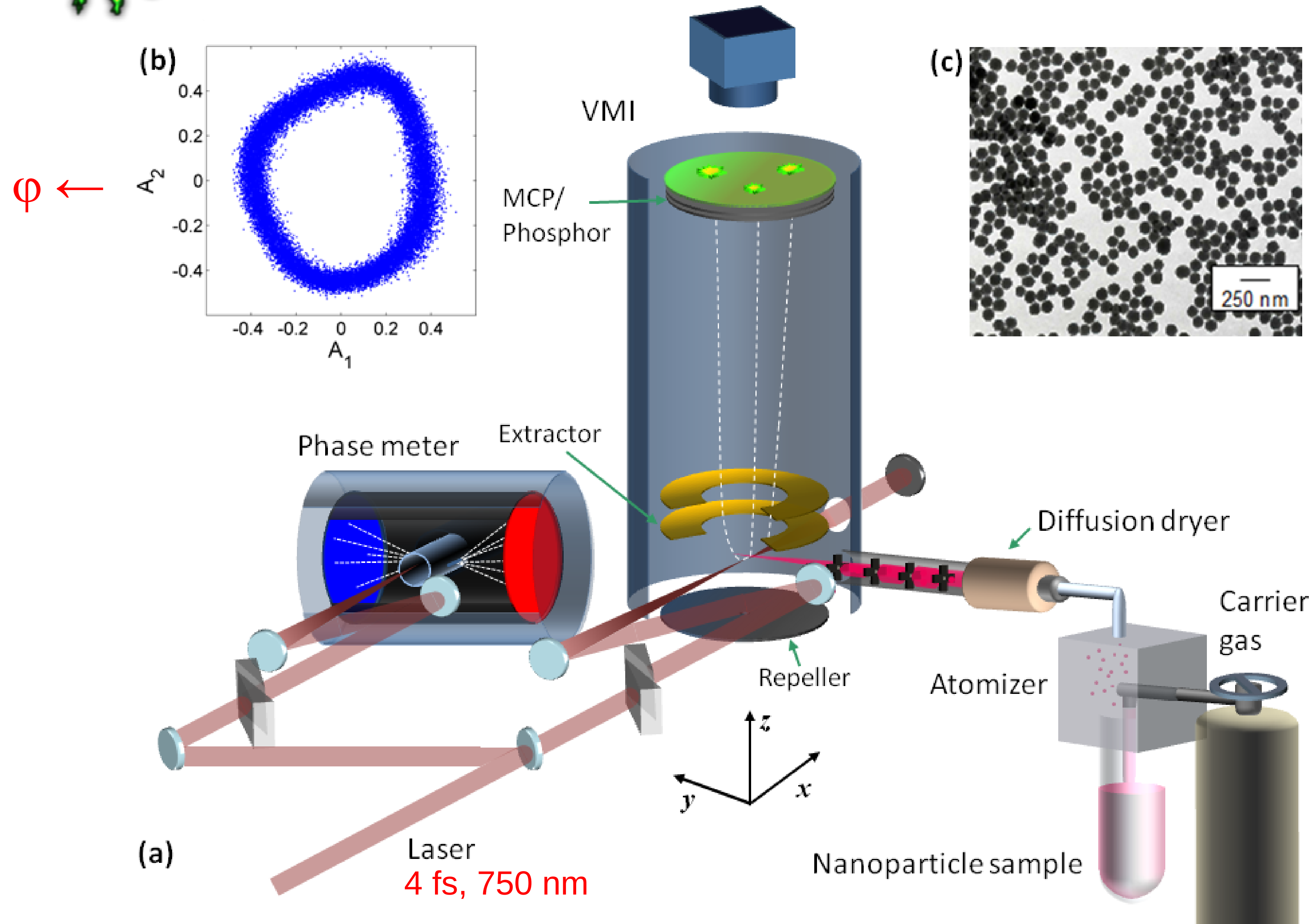
Electron emission from nanoparticles



Phase-tagged imaging of the electron emission from nanoparticles

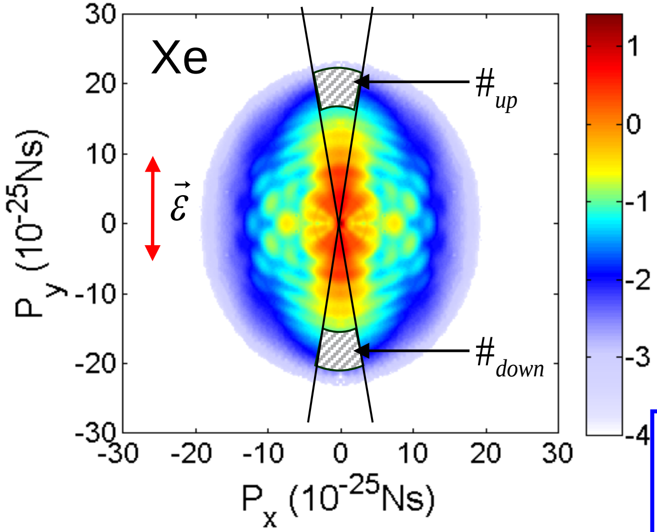
www.attoworld.de

F. Süßmann *et al*., *Rev. Sci. Instr*. 82, 093109 (2011); S. Zherebtsov et al., *NJP* (2012)



Measured electron distribution & light waveform control

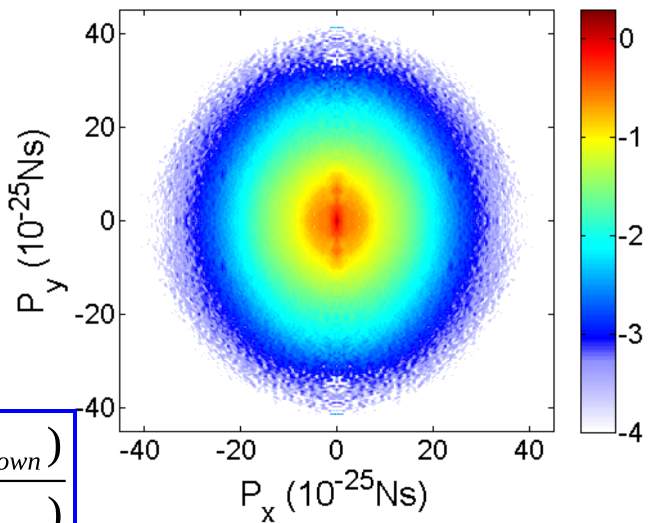
S. Zherebtsov *et al.*, *Nature Phys.* 7, 656 (2011)

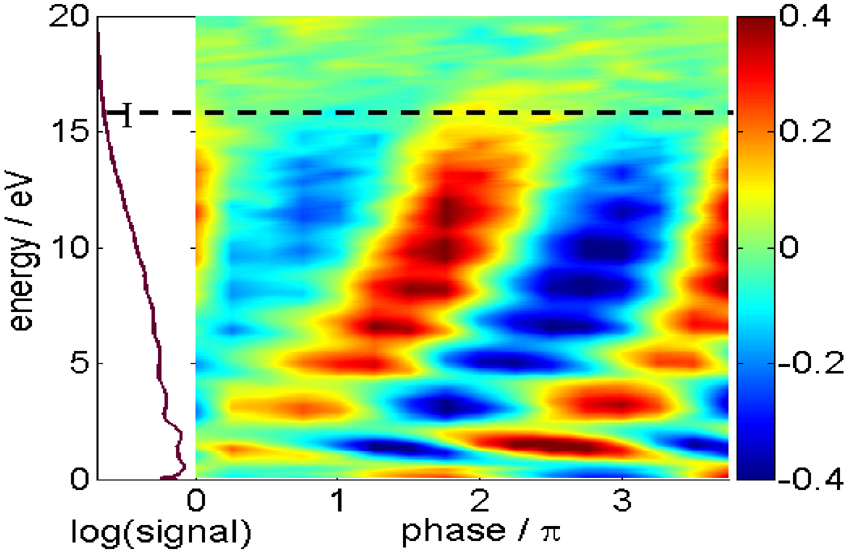


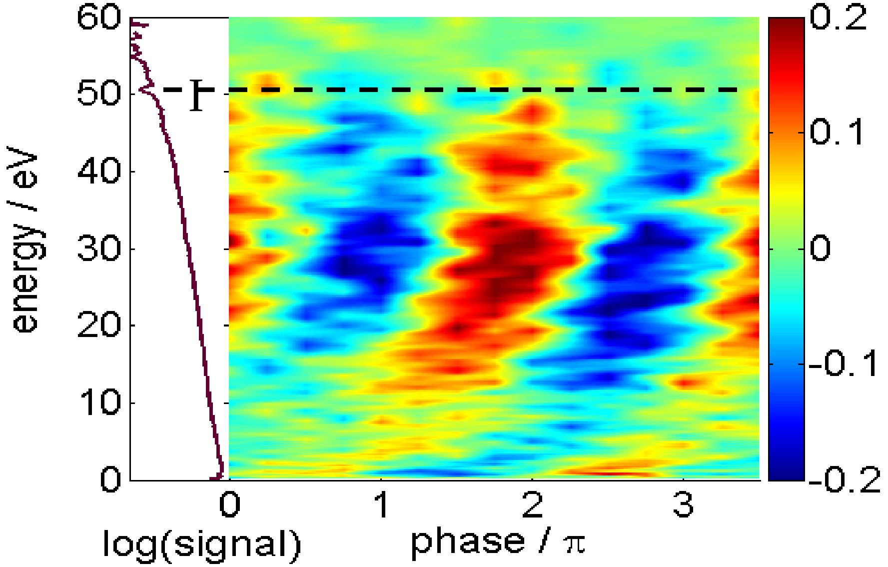
Electron emission from Xe via ATI was used as reference

$$Asymmetry(E) = \frac{(\#_{up} - \#_{down})}{(\#_{up} + \#_{down})}$$

SiO$_2$ nanoparticles

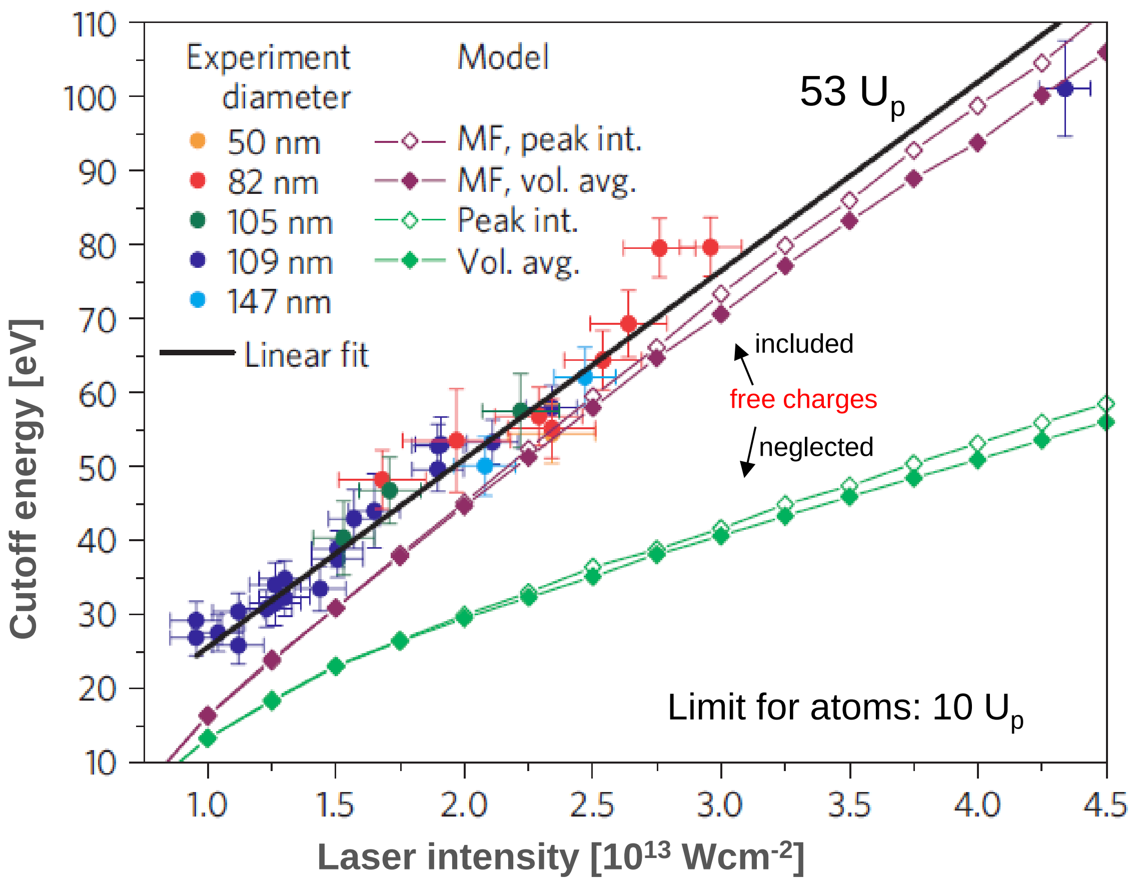




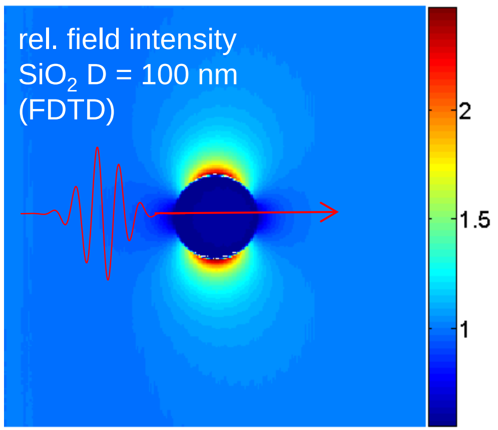


Acceleration of electrons from nanoparticles

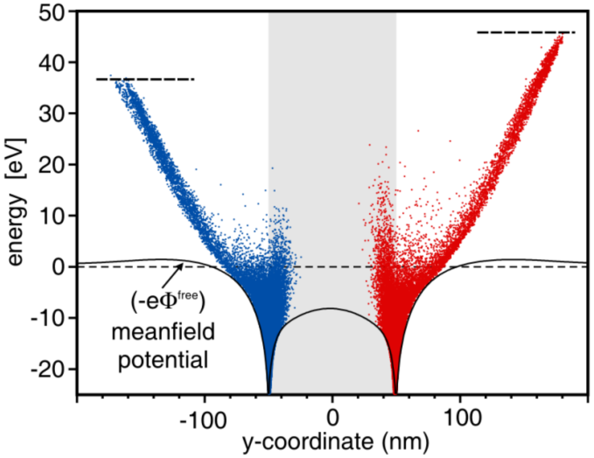
Electron cutoff energy vs. intensity (SiO_2)



dielectric enhancement

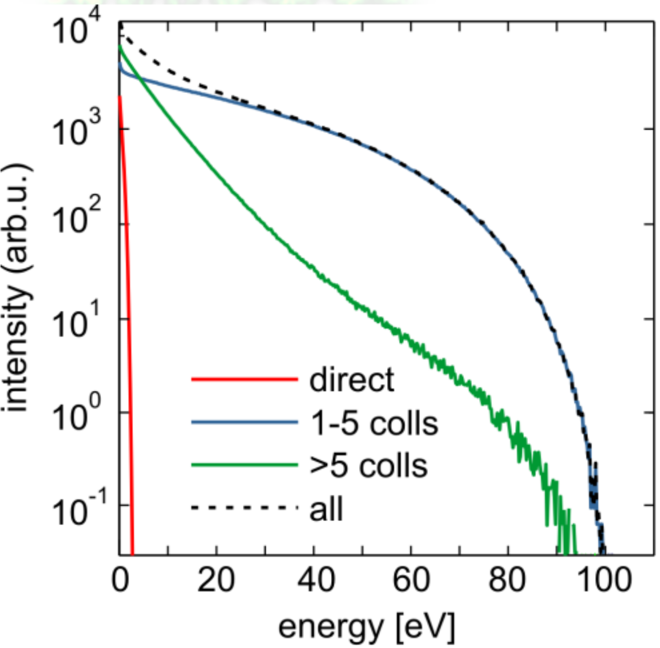


local trapping fields



S. Zherebtsov *et al., Nature Physics* **7**, 656 (2011); S. Zherebtsov *et al., New J. Phys*., NJP **14**, 075010 (2012)

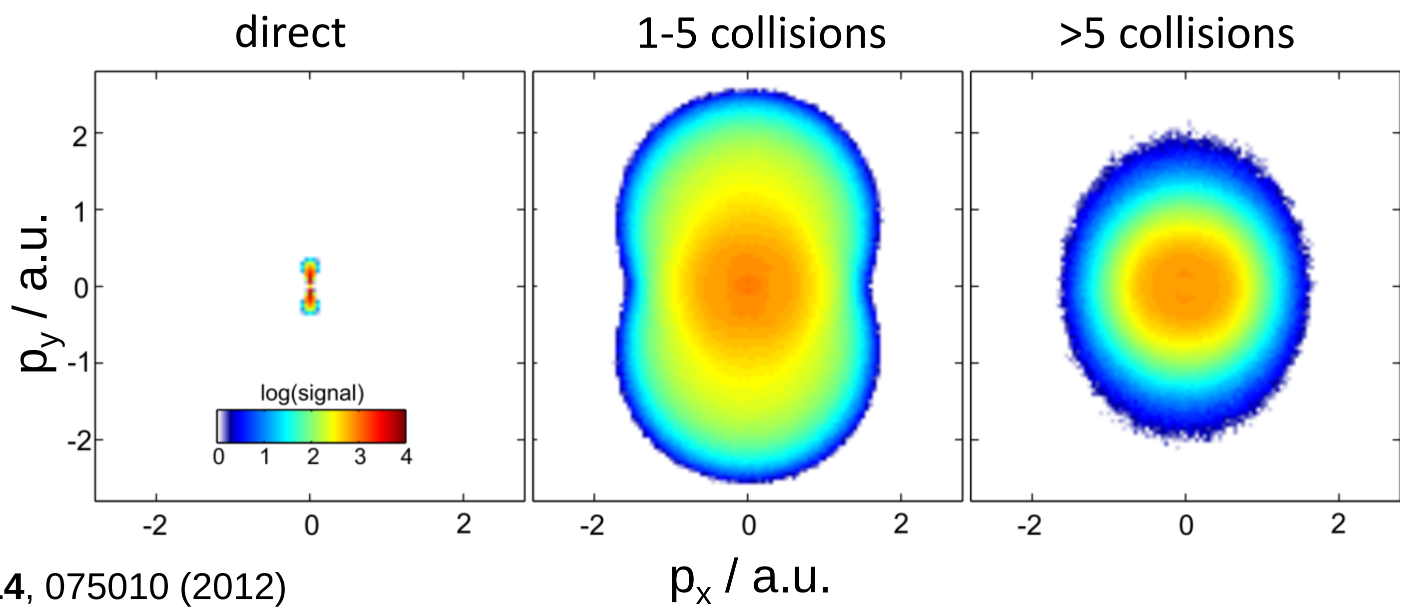
Disentangle few from many-collision dynamics



Theoretical simulations (C. Peltz, T. Fennel)

How many collisions do electrons undergo?

Low number of collisions for high-energy electrons !



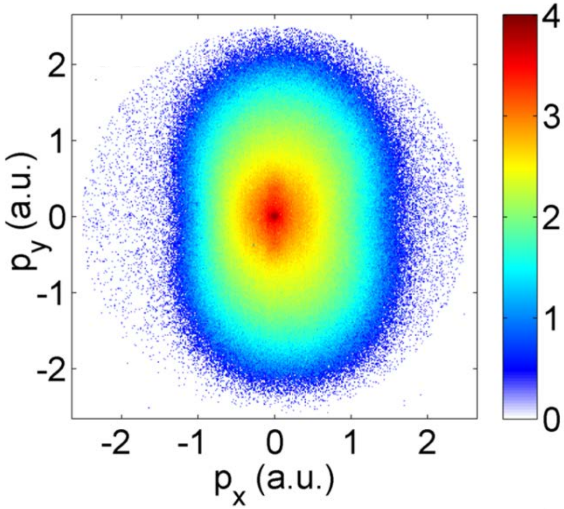
S. Zherebtsov *et al*., NJP **14**, 075010 (2012)

Disentangle few from many-collision dynamics

S. Zherebtsov *et al*., NJP **14**, 075010 (2012)

Analysis of data containing „thermal“ contributions

Total electron yield



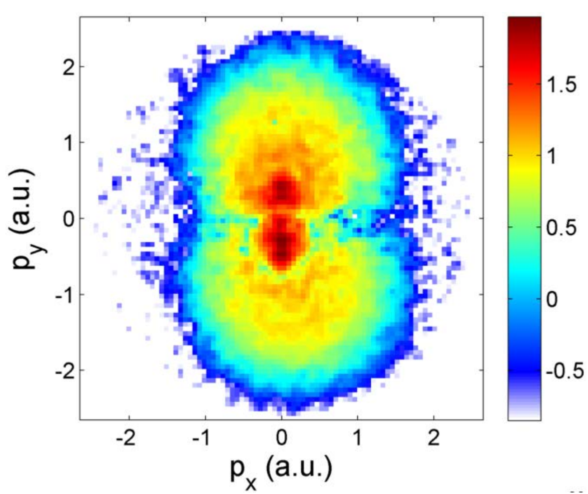
Electron yield oscillates with CEP

Determine *amplitude* and *phase offset* of oscillation

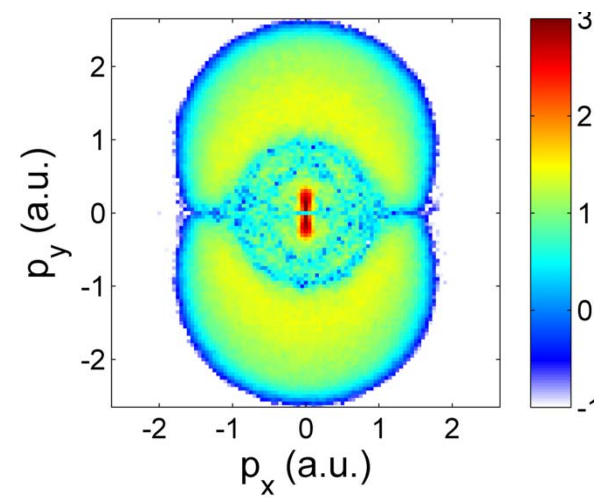
$$Y(p_x, p_y) = Y_0(p_x, p_y) \times \cos(\omega t + \Delta\varphi)$$

CEP-dependence allows to obtain insight into few-cycle dynamics

Amplitude CEP-dep. Signal Y_0 (p_x,p_y)

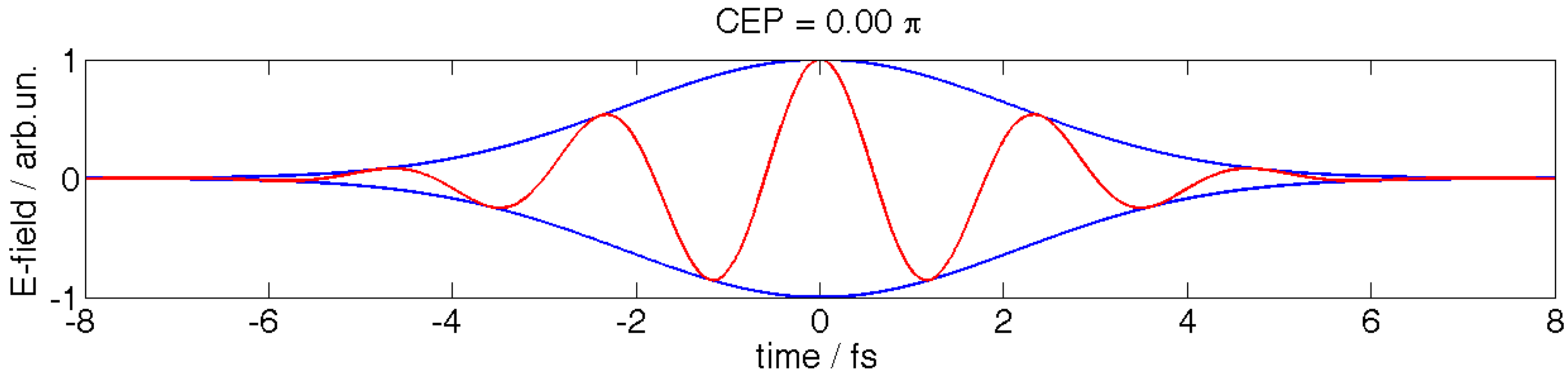


Theory



Conclusions

Light-waveform control of electron dynamics



Examples for the control of strong-field processes with the CEP:

- *Correlated electron emission from atoms (NSDI)*
- *Strongly coupled electron-nuclear dynamics (beyond BO) in molecules*
- *Electron emission, rescattering and acceleration in nanostructures*

Outlook:

- *Control with arbitrary shaped waveforms, see e.g.*
  Wirth et al., Science **334**, 195 (2011)
- *Optimal Control (using feedback for optimization)*

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Thank you for your attention!

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