Ultrafast Atomic and Molecular Dynamics – Capabilities at EuXFEL !?

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Ultrafast Atomic and Molecular Dynamics – Capabilities at EuXFEL !?

our expertise (FLASH):

- ion- and electron spectroscopy (time-resolved)
- coincidence measurements (REMI, COLTRIMS)

wish list (machine):

- sub-fs FEL pulses (10-100 uJ)
- XUV – soft X-ray regime (…100 eV …. 5 keV)
- Two-color pump-probe
- High rep-rate => CW operation

Ultrafast Atomic and Molecular Dynamics

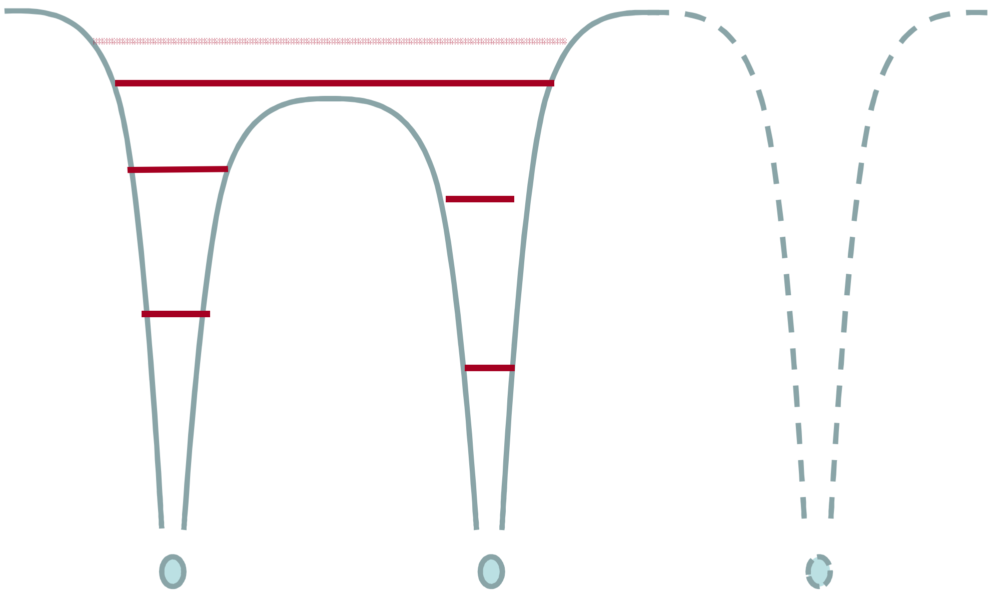
Where is the community ?

- 95% of all talks => electrons, nuclei, photons (Coulomb)
- Dynamics in molecules
  (chemistry, bio, catalysis, solids, magnetics(spin)…)
- Electron transfer and migration, transition-state dynamics,
  Isomerization, proton transfer, interatomic coulombic decay, …

What is the common basis and
what are the relevant time scales ?

Ultrafast Atomic and Molecular Dynamics

atom, molecule, cluster, ….solid



in ground-state:
mol. wave-function is static !!
no time dependence !!!

Ultrafast Atomic and Molecular Dynamics

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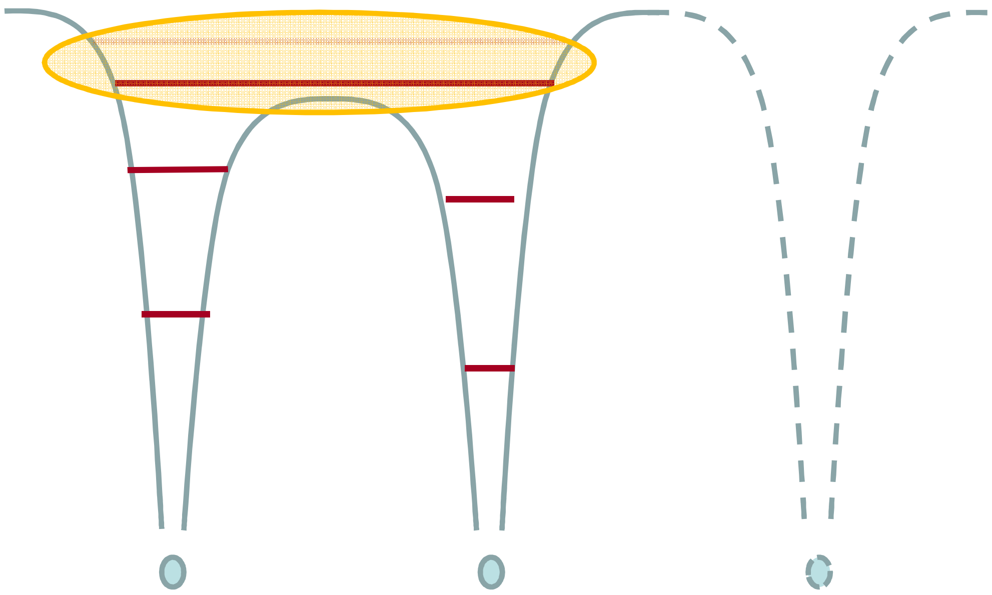
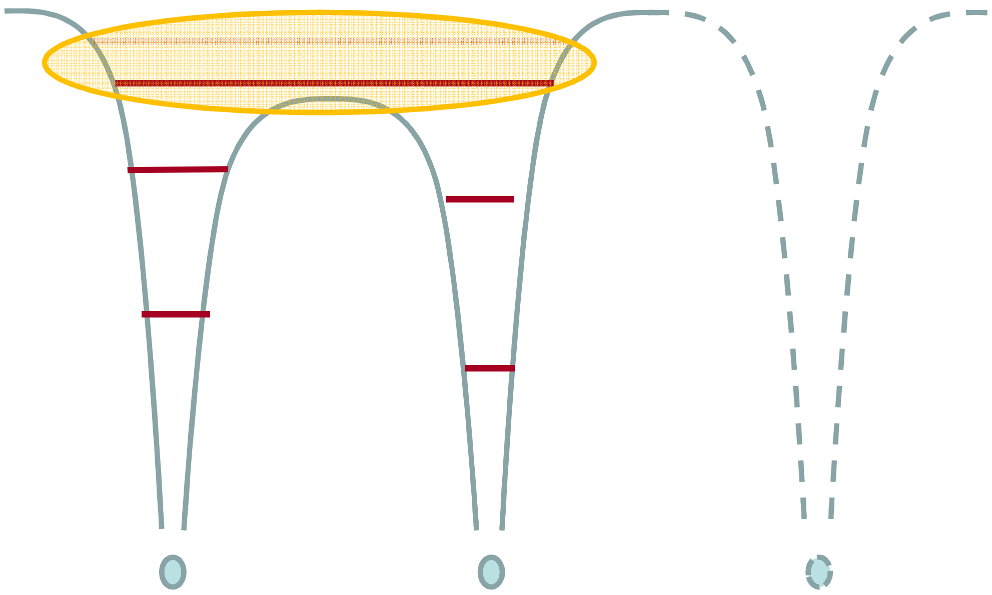


coherent super-position
of elec. states:

Ultrafast Atomic and Molecular Dynamics

atom, molecule, cluster, ….solid



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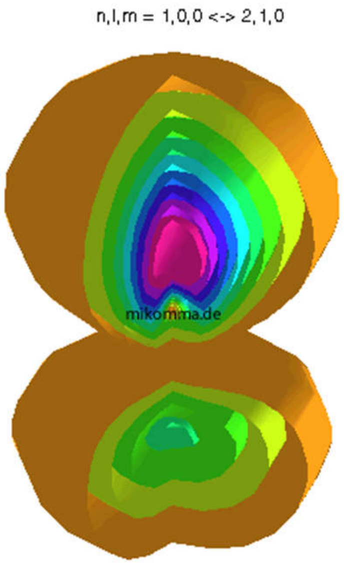
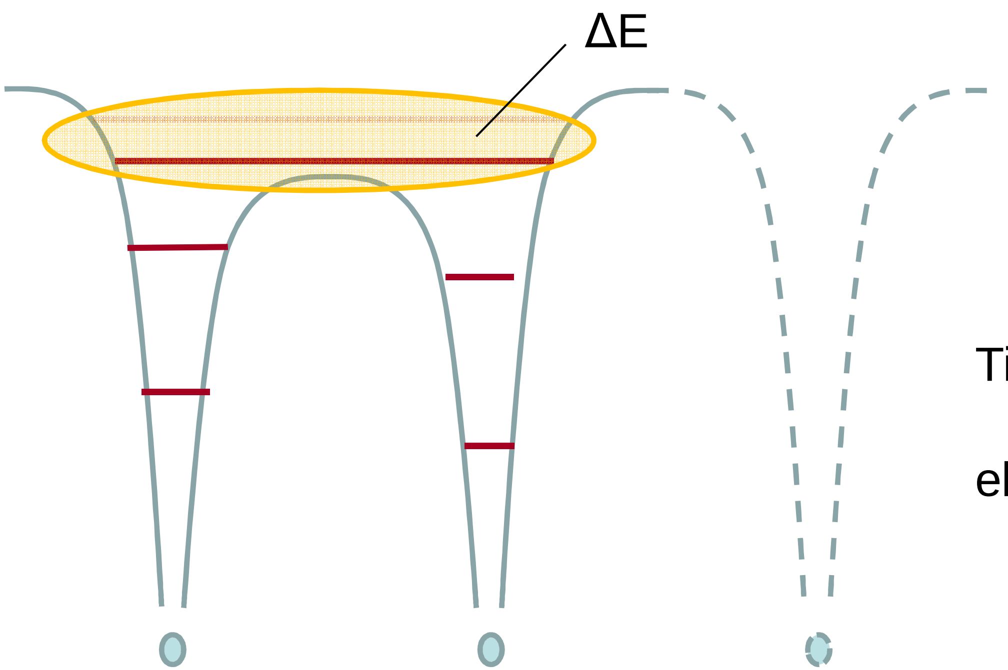


H-atom (1s and 2p)

picture from http://www.mikomma.de/fh/hydrod/hydoszi.htm

Ultrafast Atomic and Molecular Dynamics



coherent super-position
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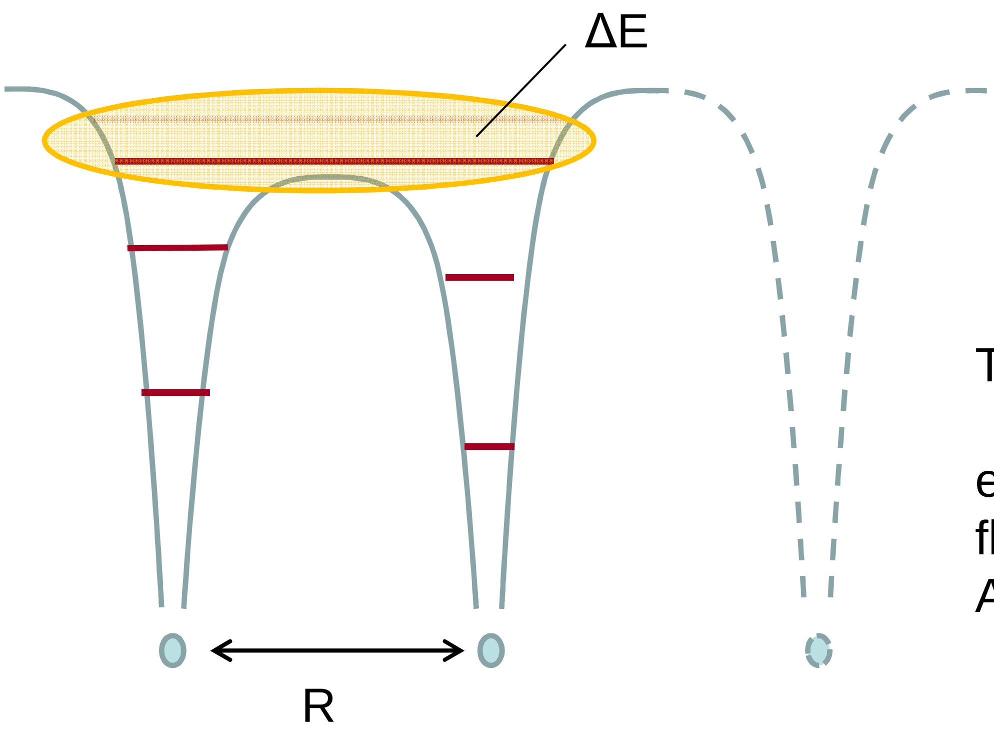
Time scales:

elec. motion (t=1/ΔE): ..100 as. … 10 fs

Ultrafast Atomic and Molecular Dynamics



coherent super-position
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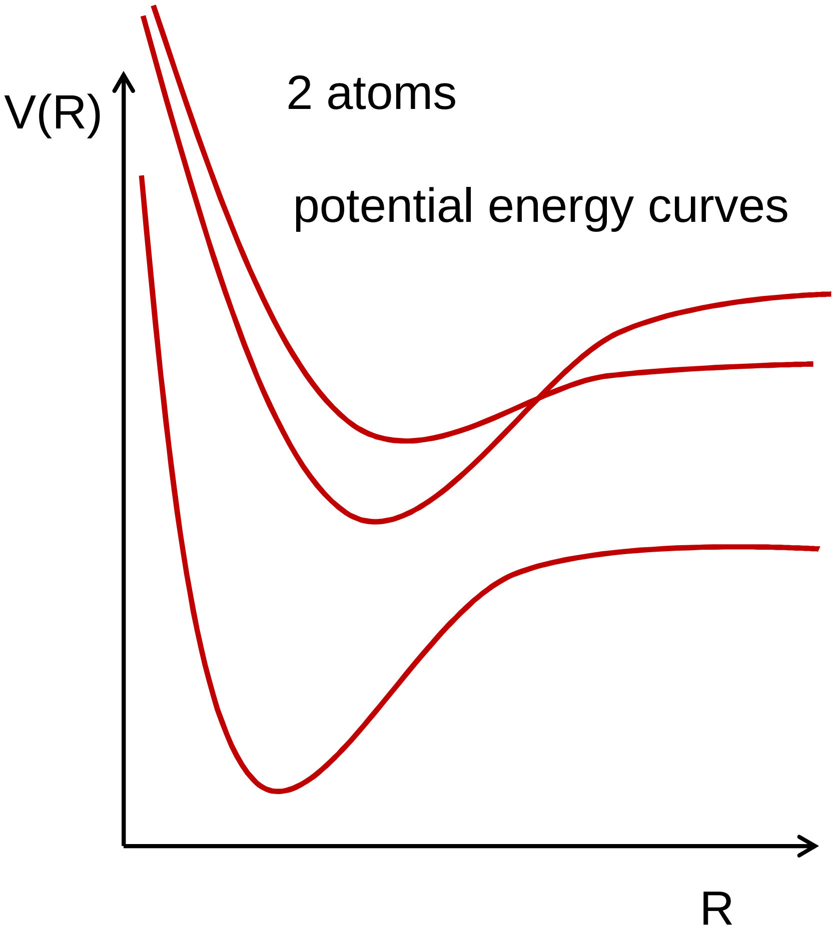
Auger-, auto-ionization: 5 fs … 100 fs…

Ultrafast Atomic and Molecular Dynamics



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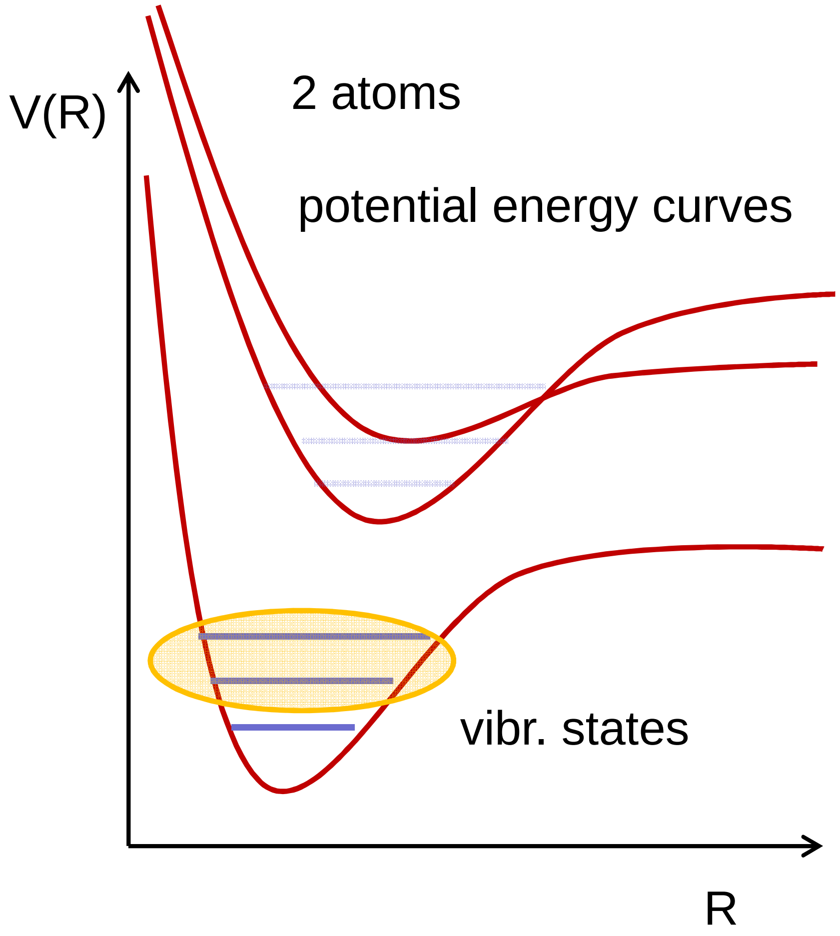
Born-Oppenheimer Approximation: $\Psi(r_e,R) = \Sigma\ \varphi(r_e;R) \cdot \Phi(R)$

Electrons adapt (adiabatically) to nuclear positions.
The nuclei move in the electronic potential V(R).

Ultrafast Atomic and Molecular Dynamics



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Ultrafast Atomic and Molecular Dynamics

3 atoms (or more)

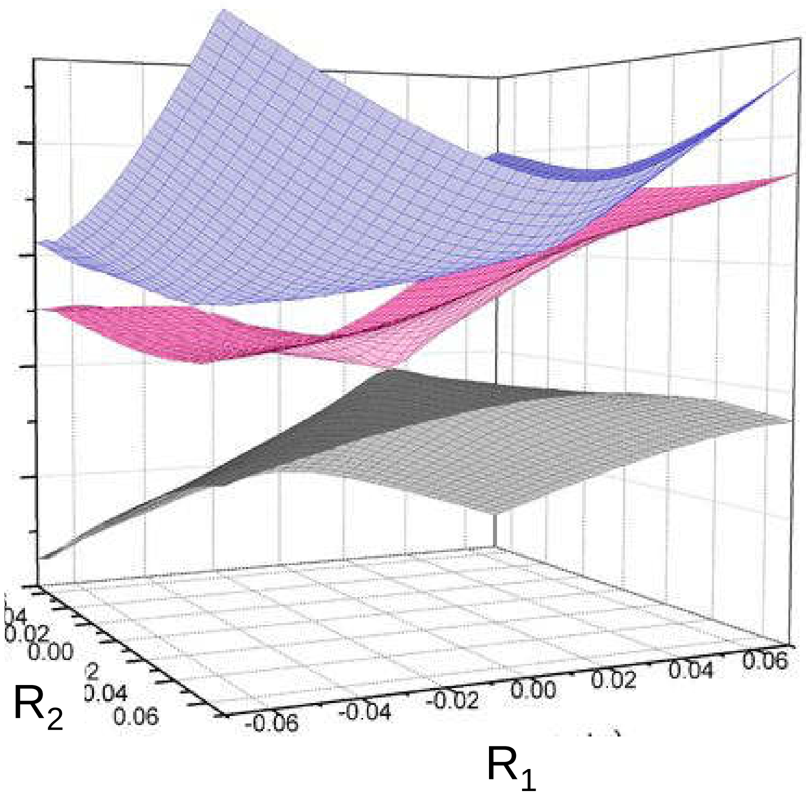

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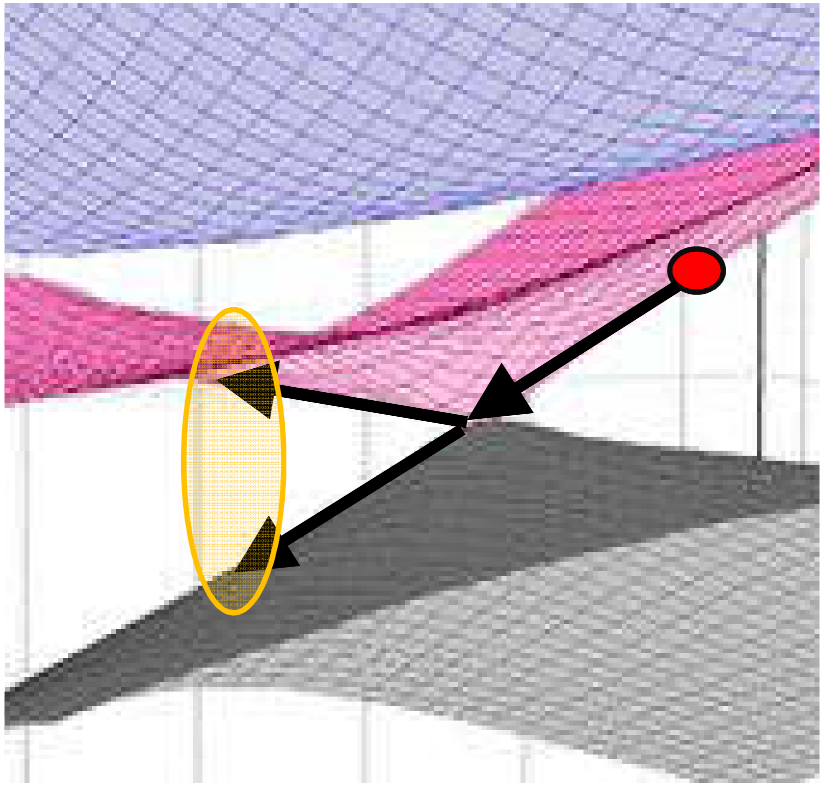
Figure from https://astro.temple.edu/~smatsika/Research.html

Ultrafast Atomic and Molecular Dynamics

Dynamics at conical intersection



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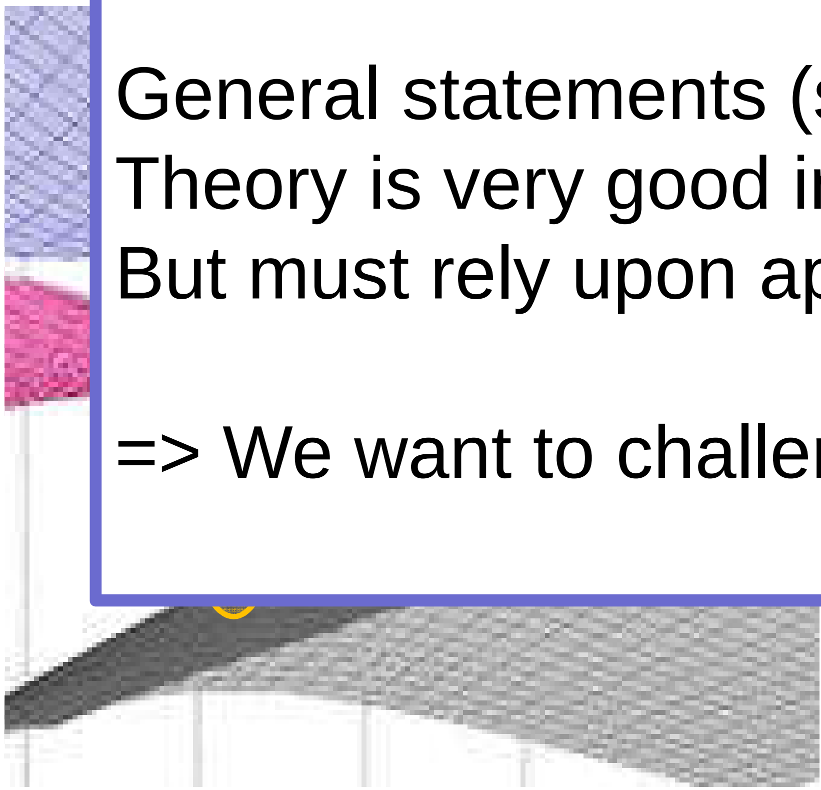


General statements (simplified):

Theory is very good in calculating structure !

But must rely upon approximations for dynamics !

=> We want to challenge (test) theory !!!

vibr. motion: 10 fs….100 fs

Born-Oppenheimer Approximation: ~~$\Psi(r_e,R) = \Sigma\, \varphi(r_e,R) \cdot \Phi(R)$~~

Figure from https://astro.temple.edu/~smatsika/Research.html

Experimental Approaches

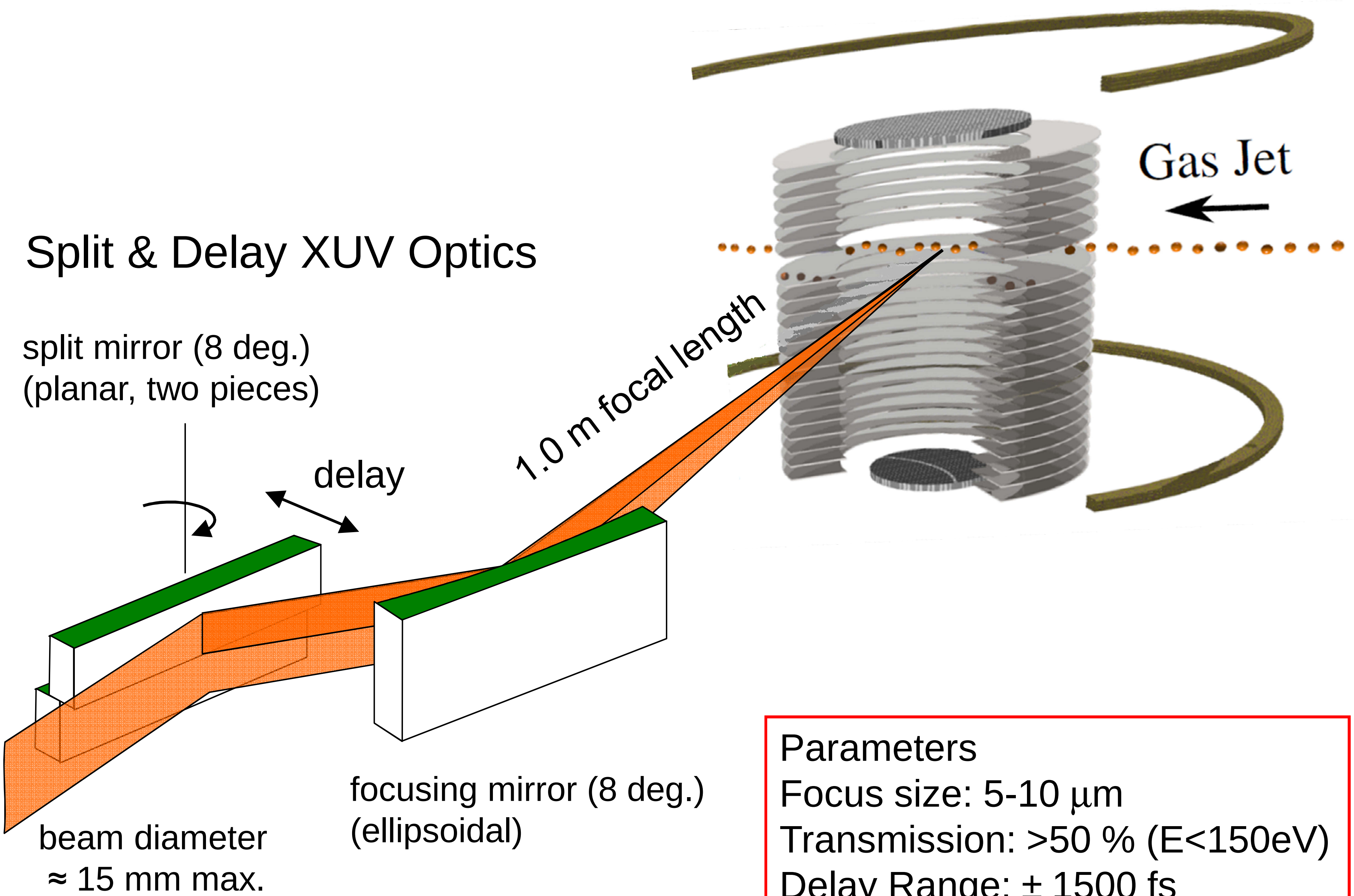
1. Laser based experiments (HHG)
2. FEL based experiments (FEL)

The ideal source
=> EuXFEL ??

	Pros	Cons
1. HHG	- High time-resolution - High rep. rate - Pulse to pulse stability	- Low XUV flux (no XUV pump-probe) - Low photon energies - (IR probe, i.e. strong field effects)
2. FEL	- High intensity (XUV pump-probe) - High photon energies (XUV to soft X-ray)	- Low time-resolution - Pulse to pulse fluctuations - Low rep. rate

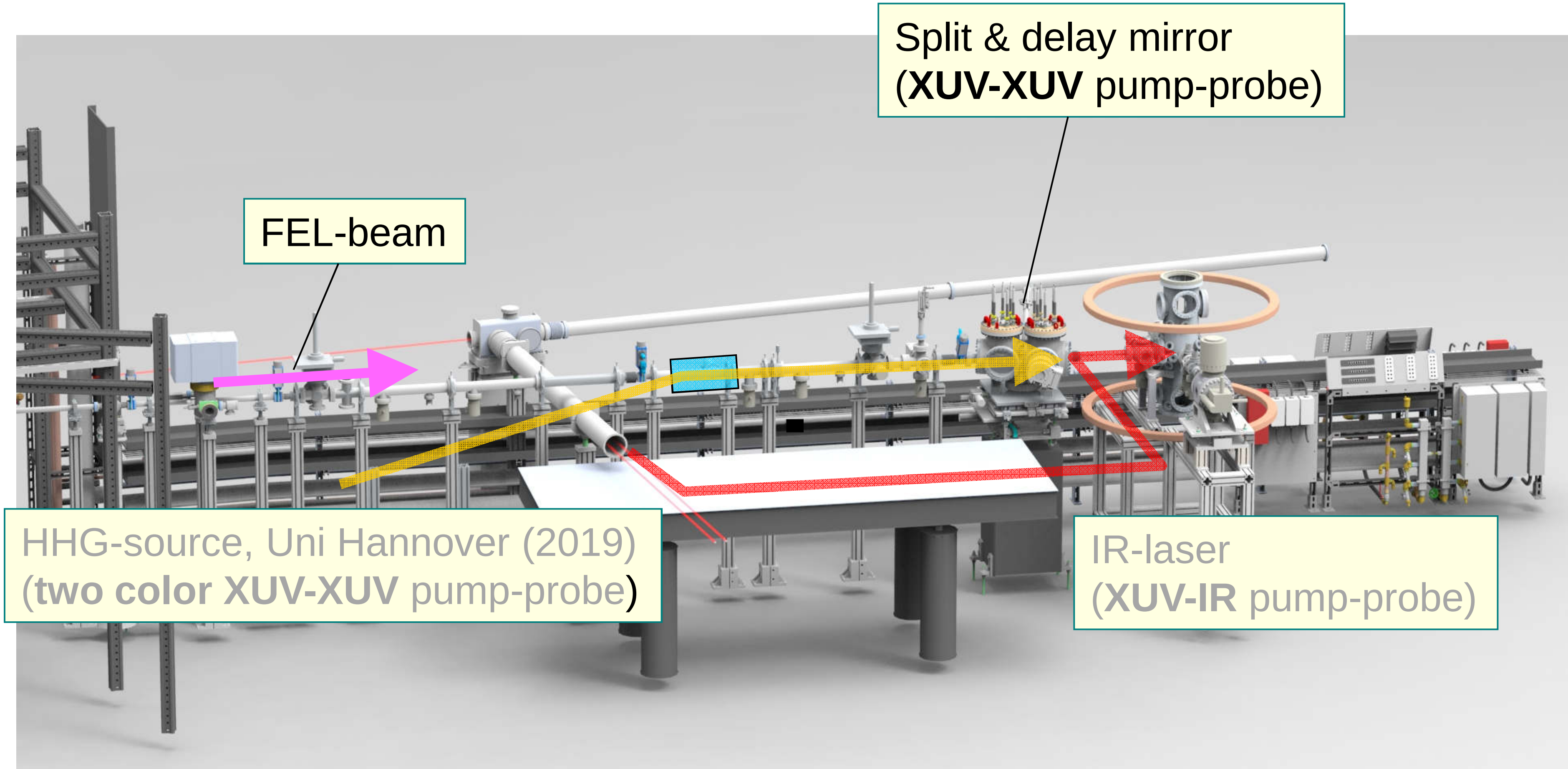
Reaction Microscope (REMI) at FLASH2

Split & Delay XUV Optics

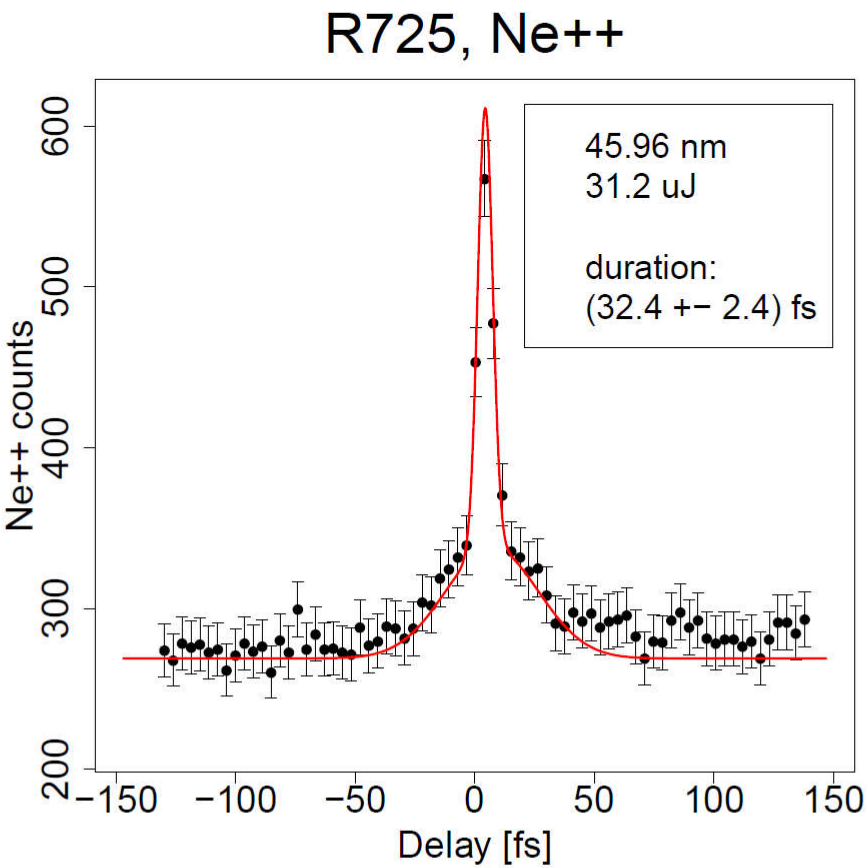


Parameters
Focus size: 5-10 μm
Transmission: >50 % (E<150eV)
Delay Range: ± 1500 fs

Reaction Microscope (REMI) at FLASH2

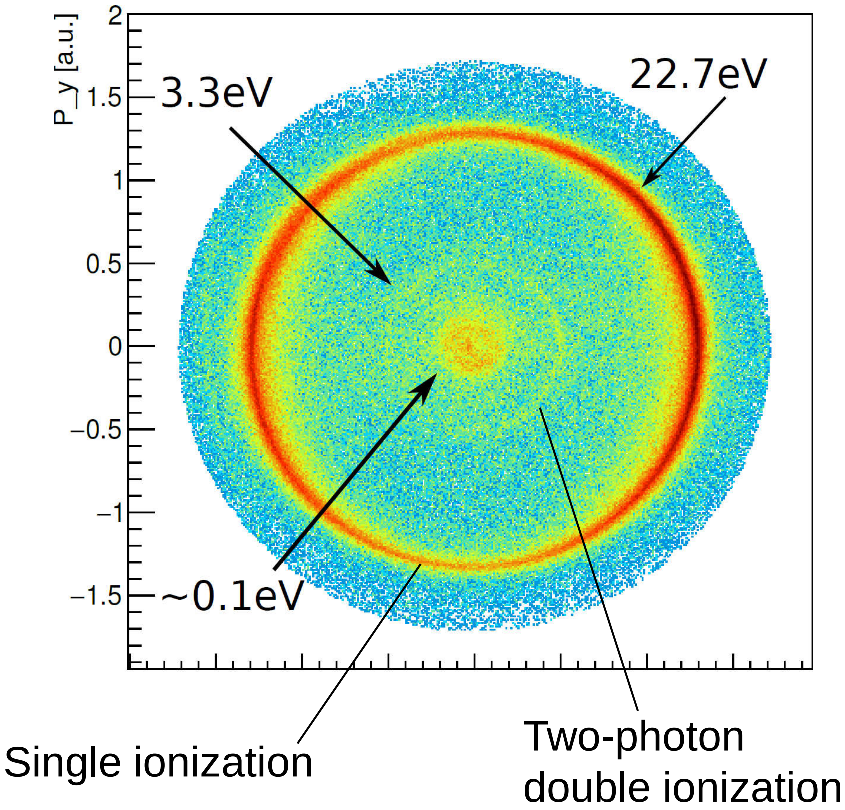


Autocorrelation Measurements

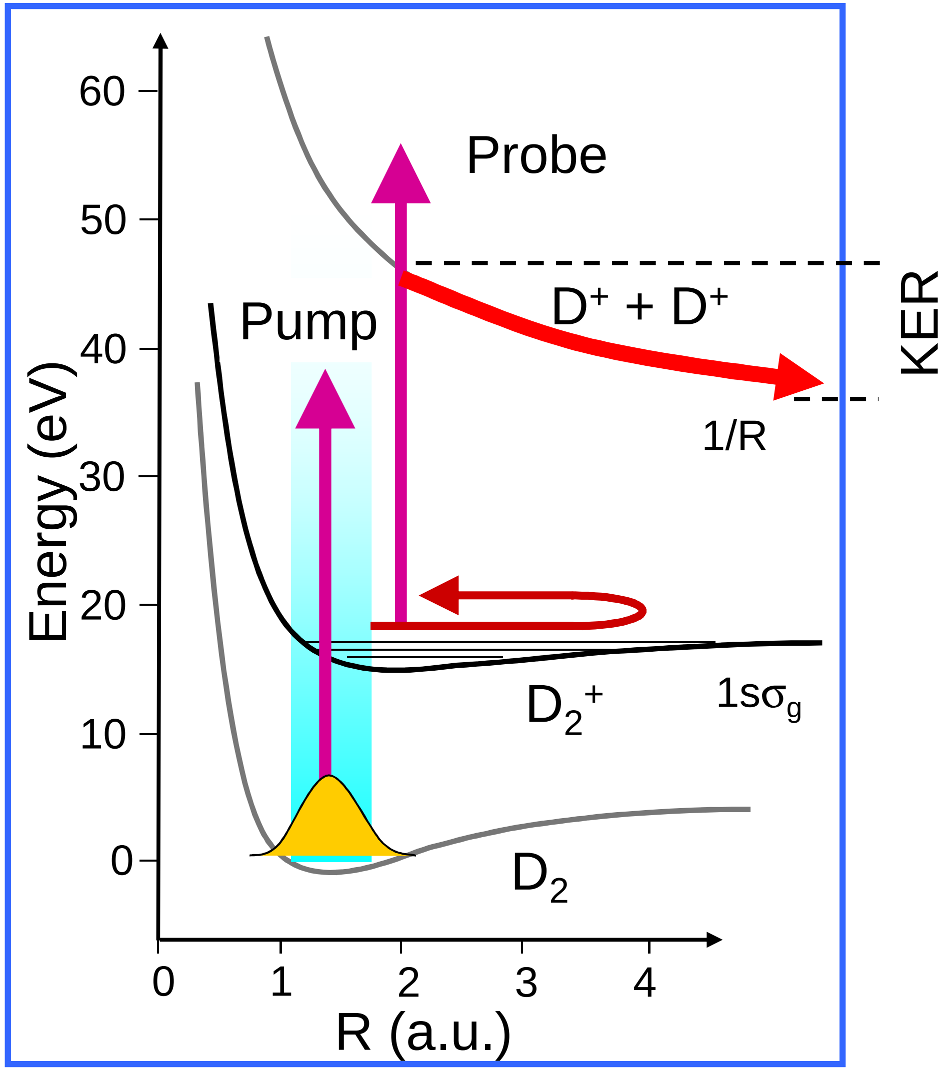


$E_\gamma = 27.0\,\mathrm{eV}$

Electron Spectrum:
Ionization of Ne (Eγ = 45 eV)



Pump – Probe with D_2 E_{ph} = 38 eV



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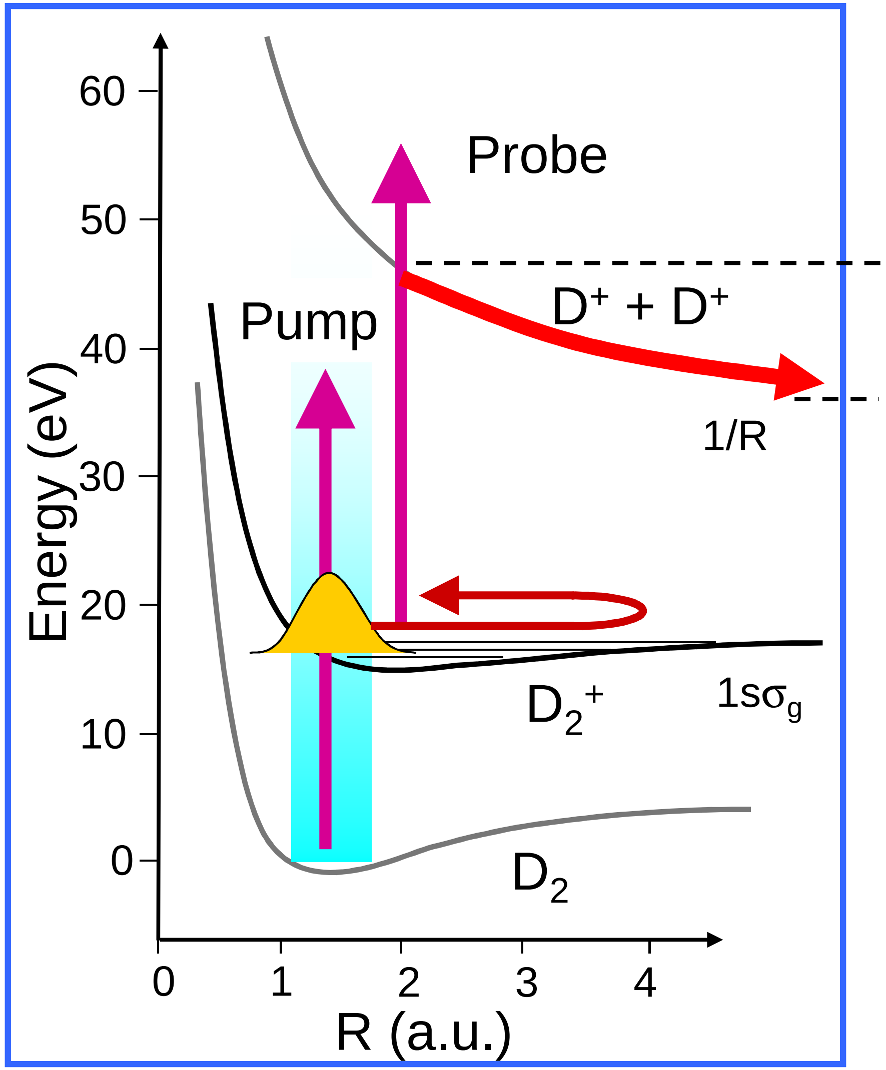

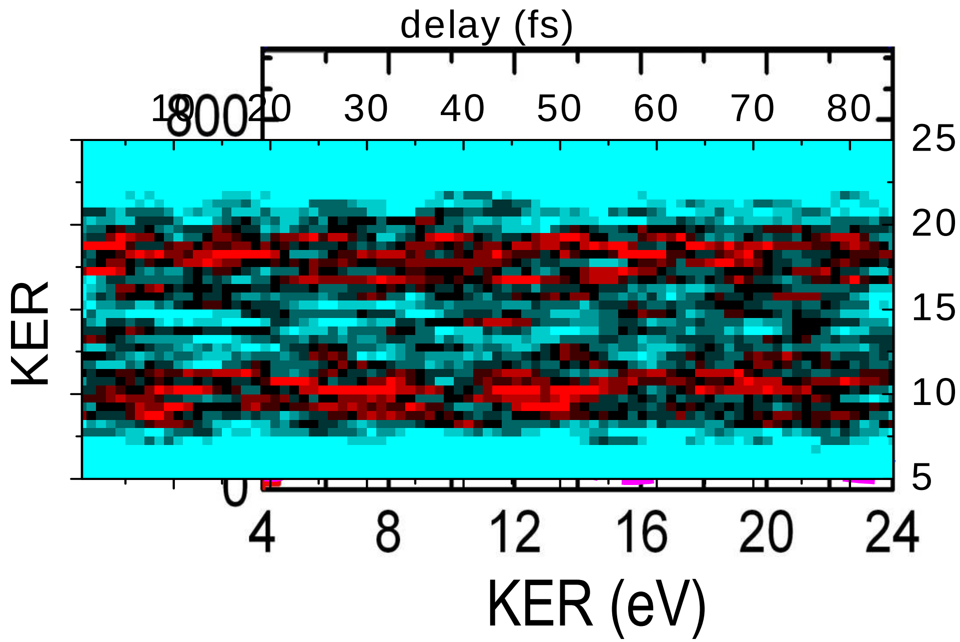

Y. Jiang et al., PRA 81 (2010)

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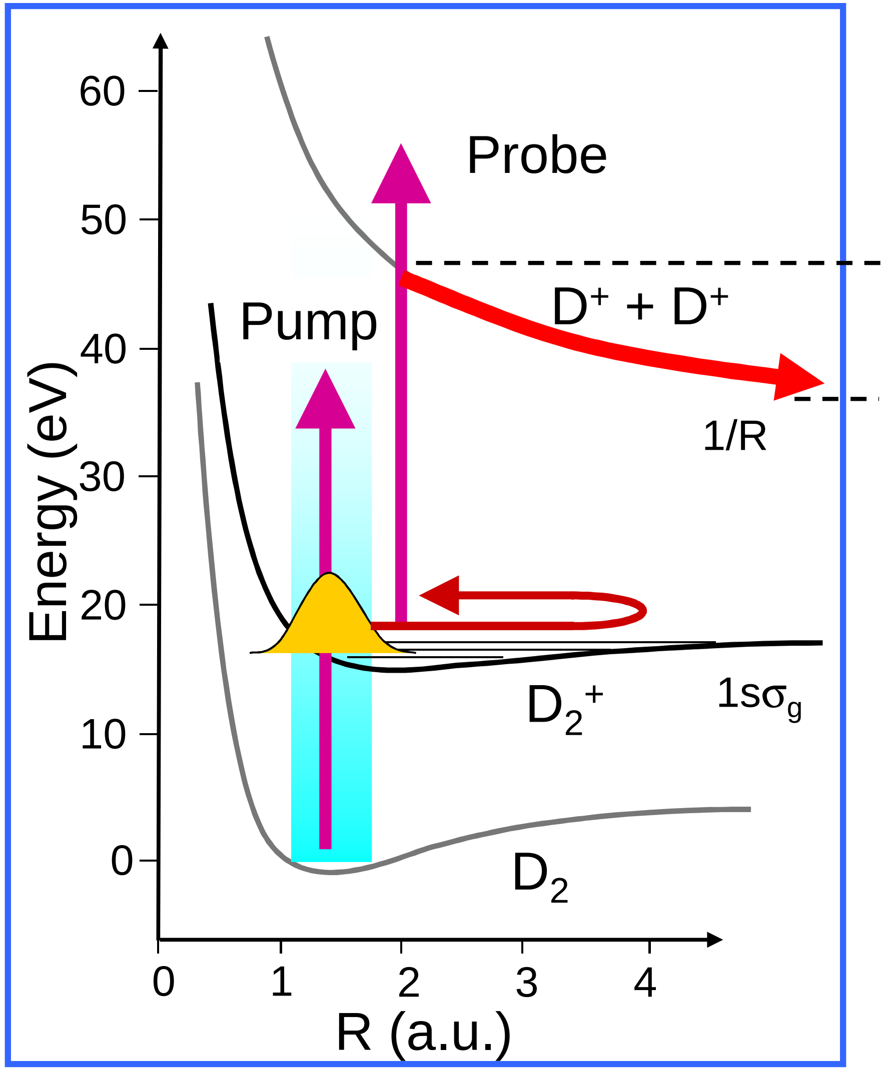

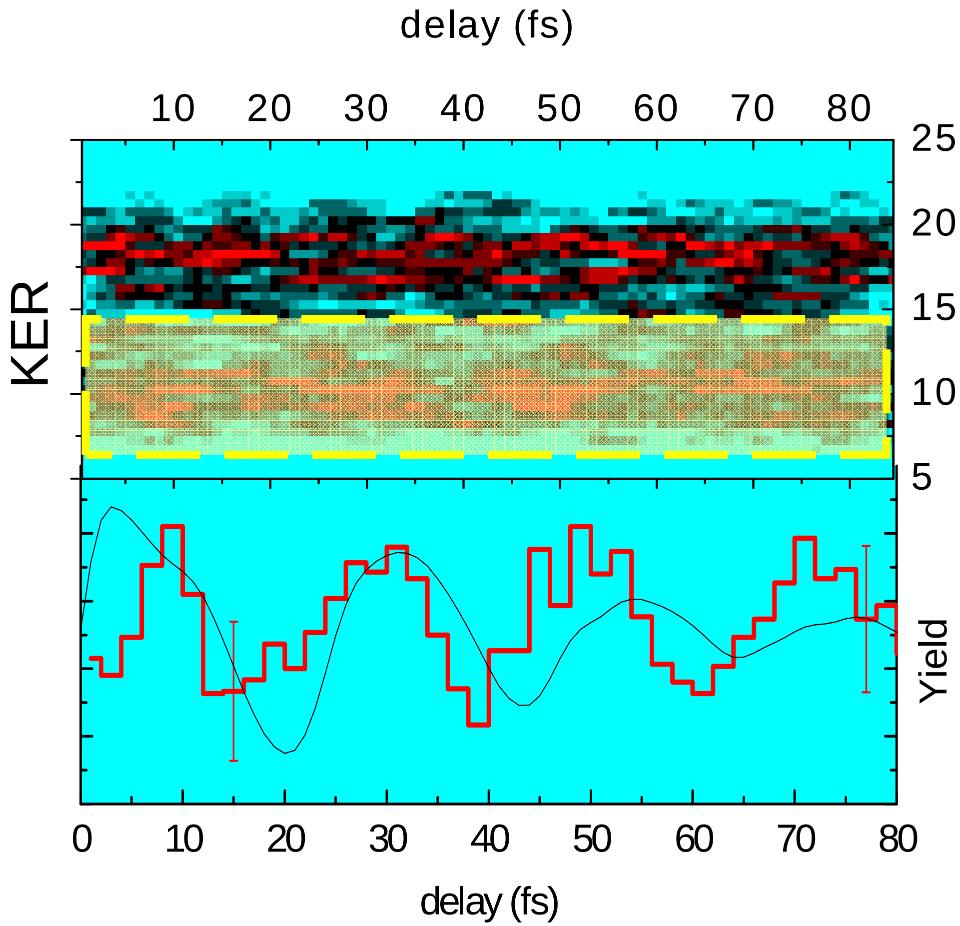

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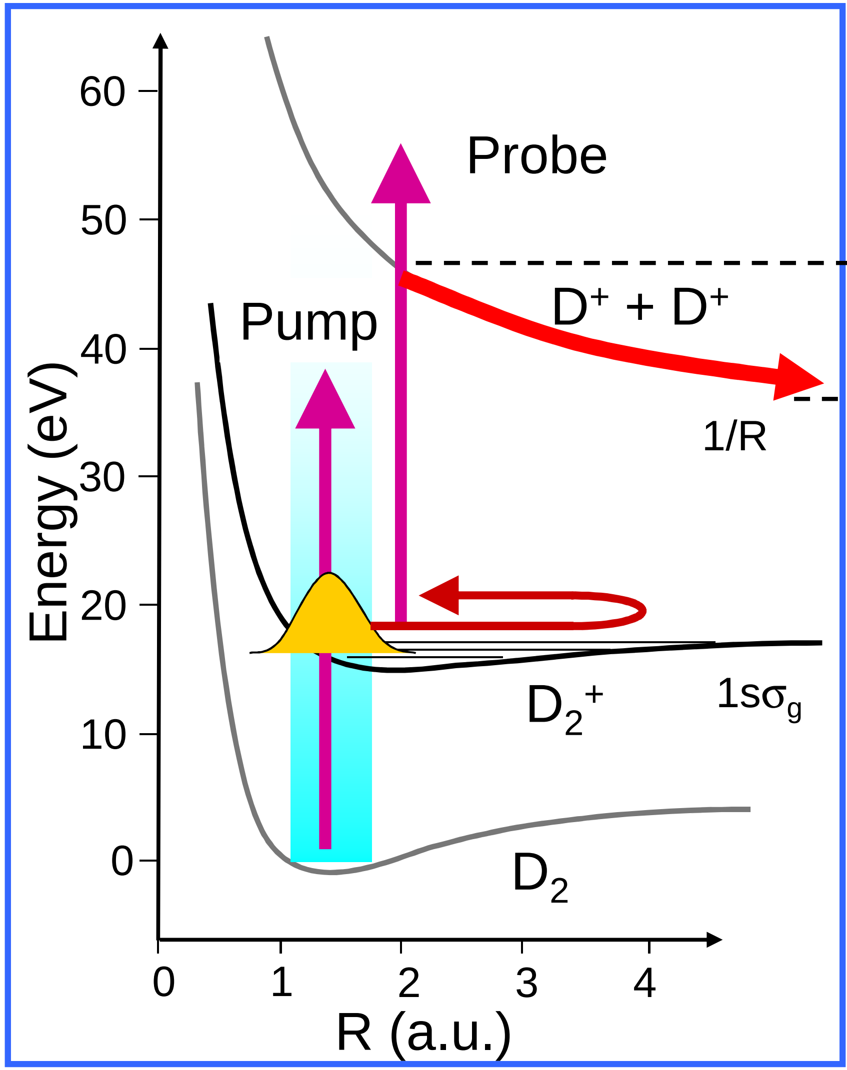

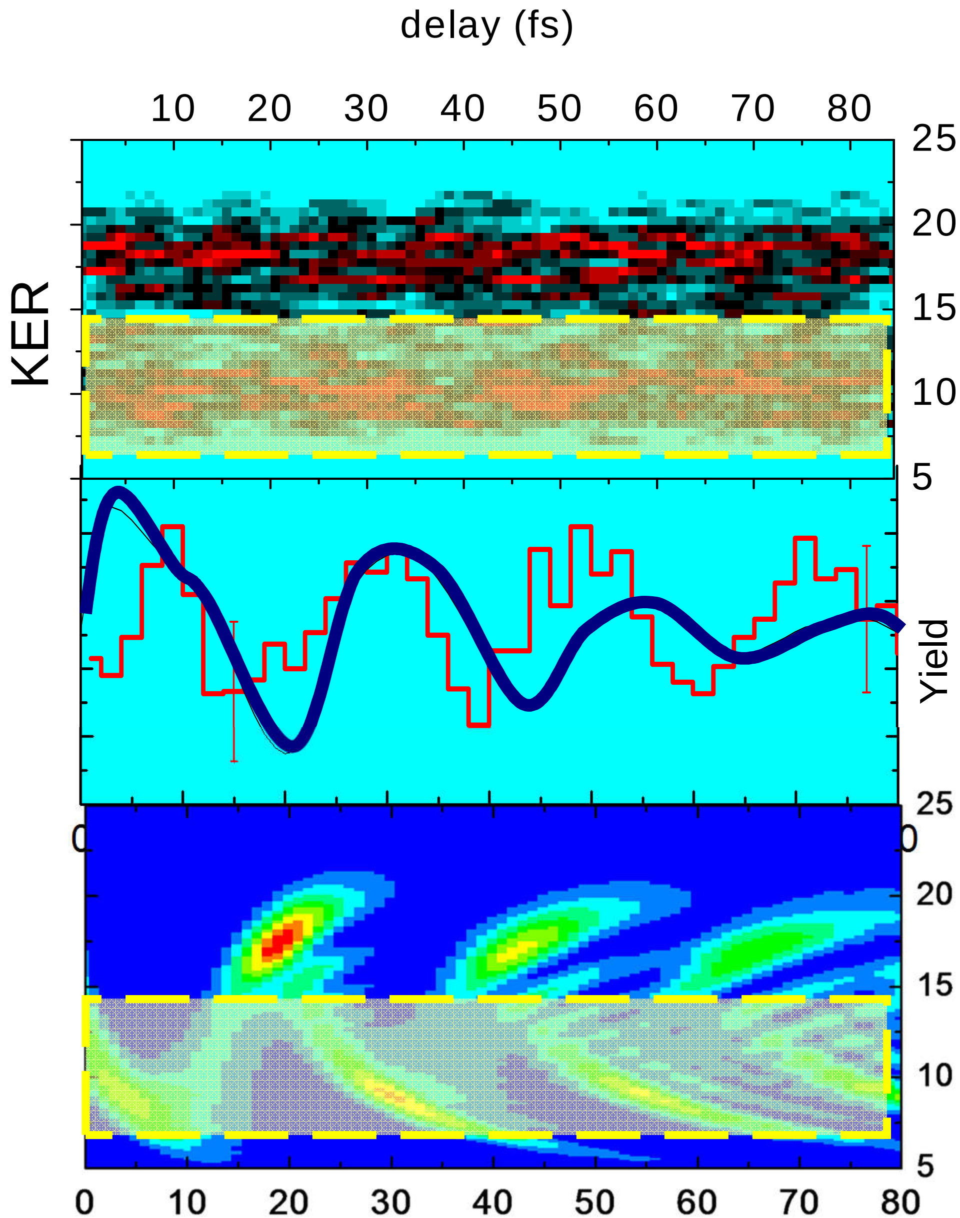

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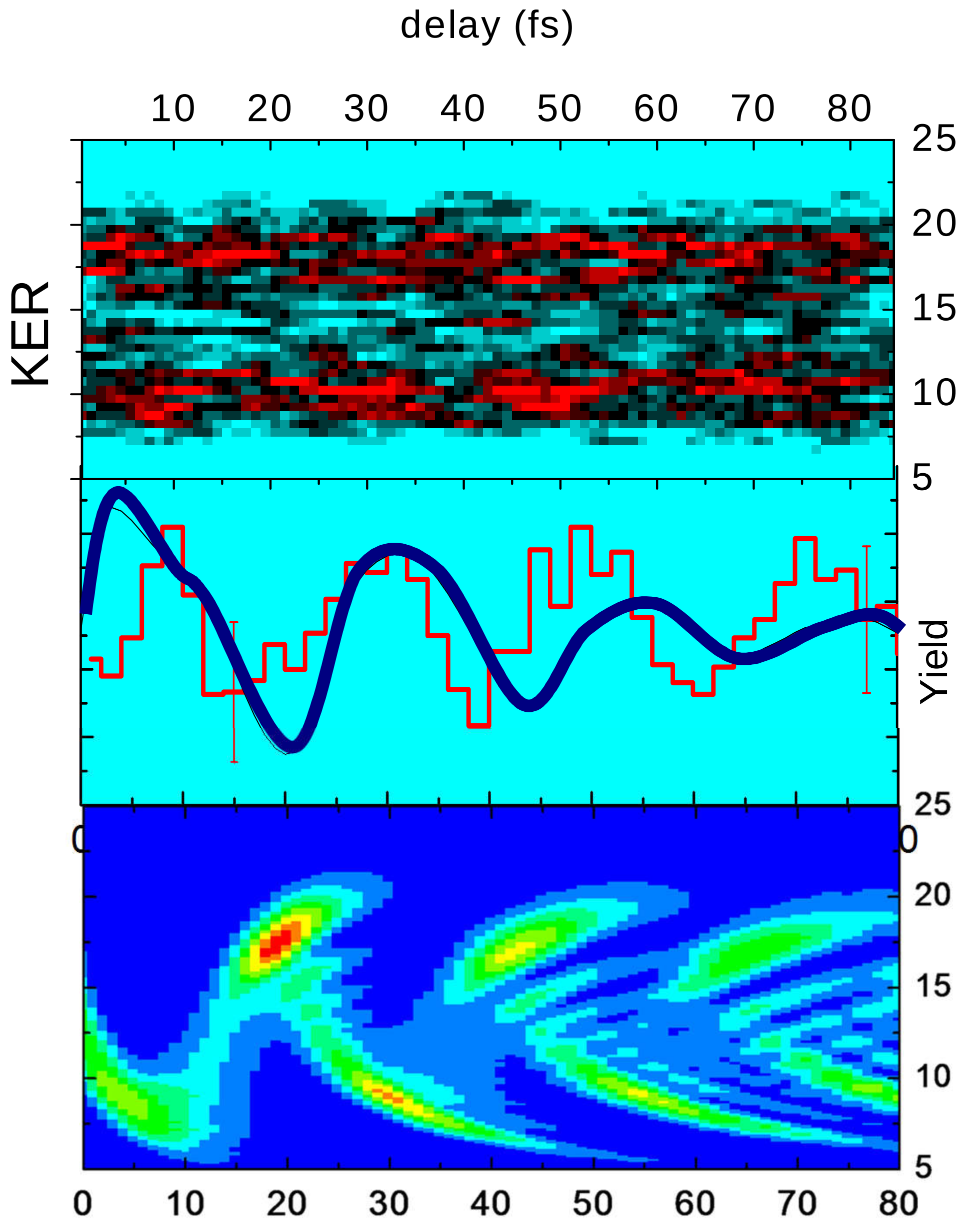
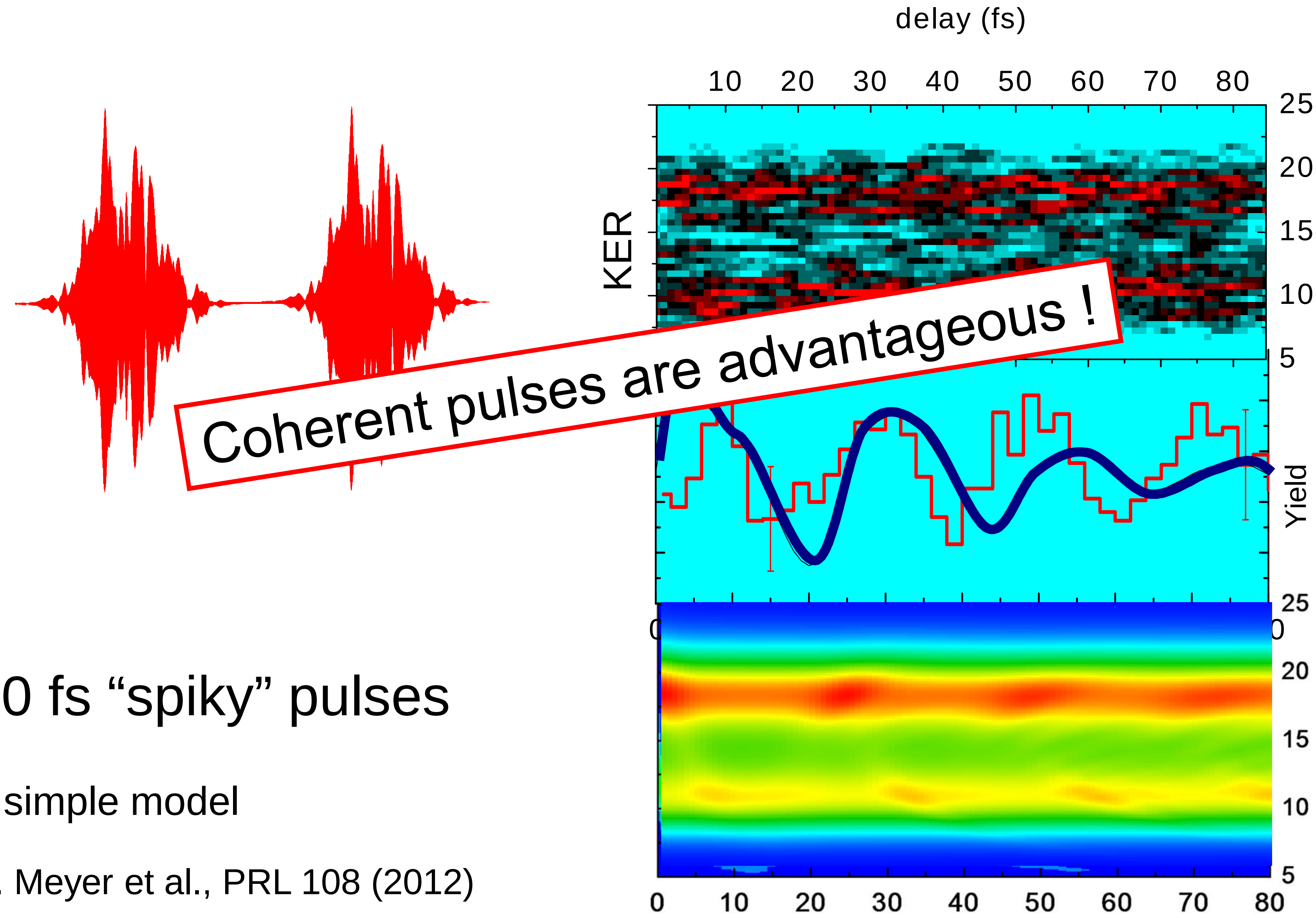


Calculation with δ pulses
(F. Martin)

Pump – Probe with D_2 E_{ph} = 38 eV



40 fs "spiky" pulses

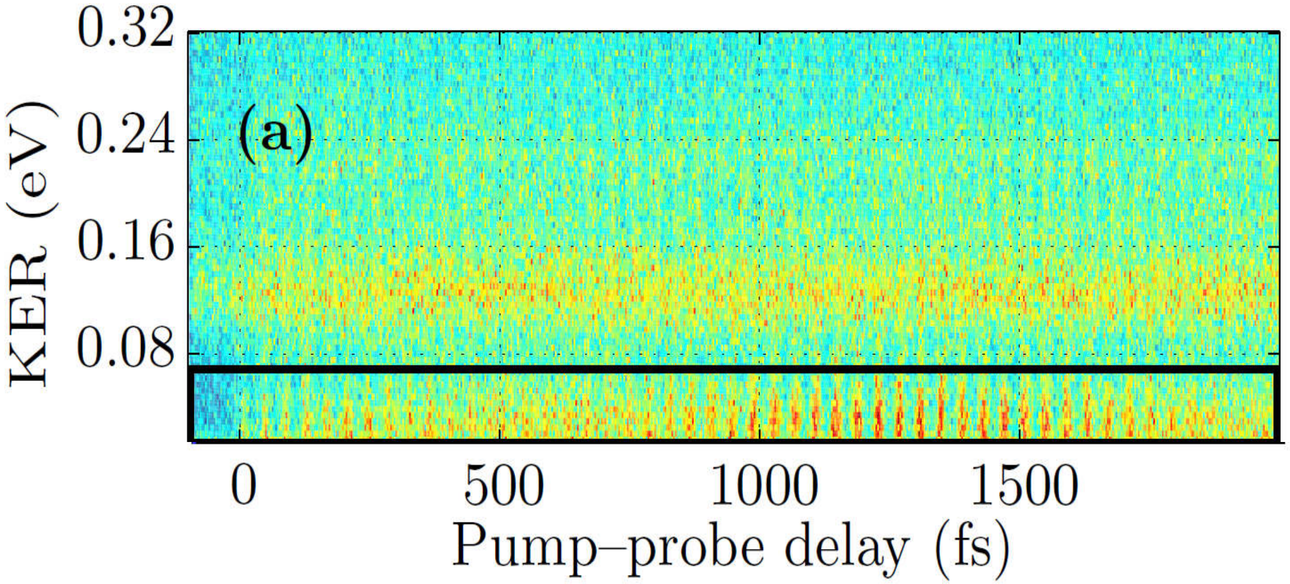
a simple model

K. Meyer et al., PRL 108 (2012)

Ionization and Dissoziation of O_2

HHG-IR experiment

1. step: ionization (XUV) $O_2 \Rightarrow O_2^+ + e^-$
2. step: dissoziation (IR) $O_2^+ \Rightarrow O^+ + O^o$



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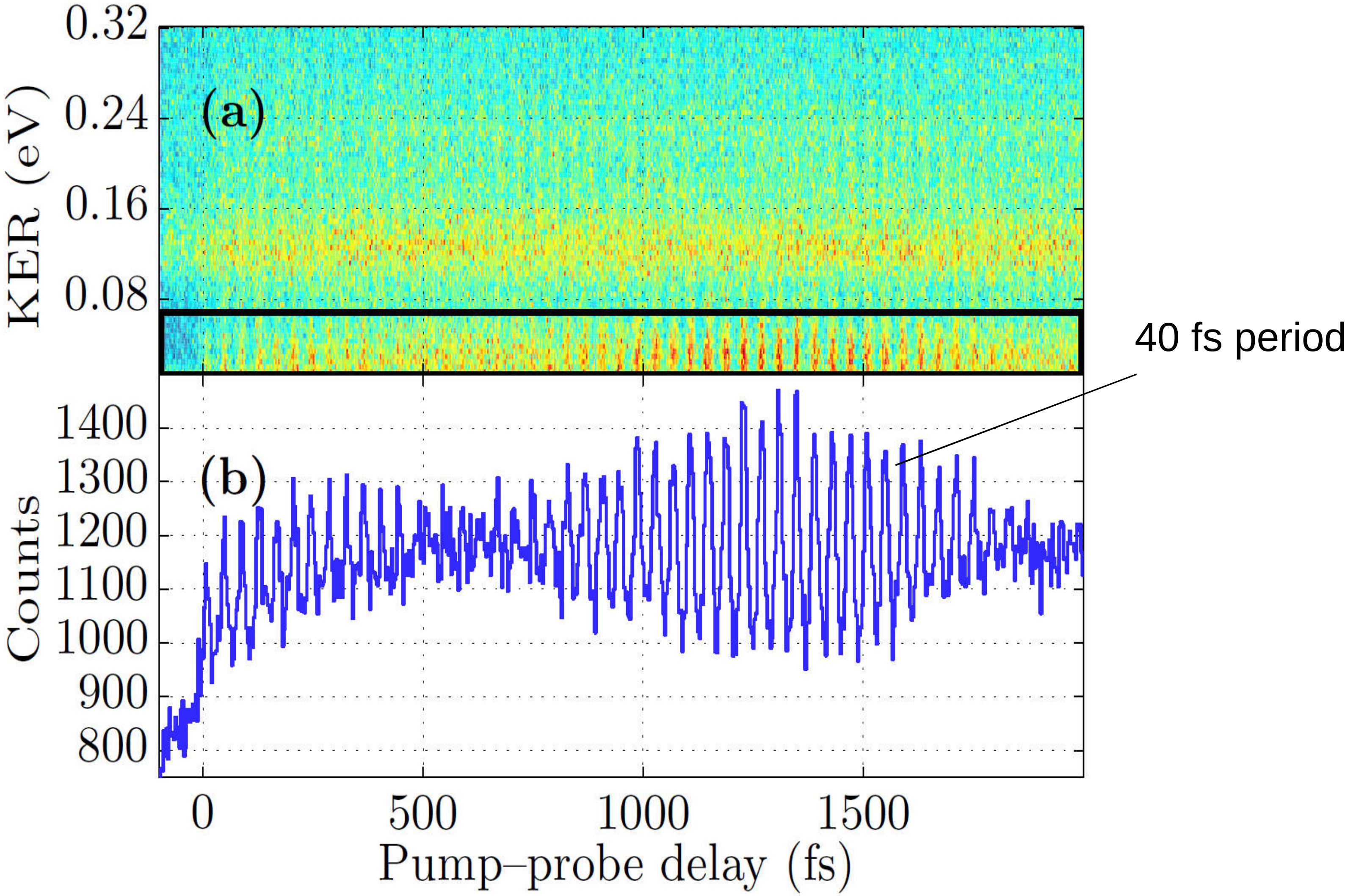

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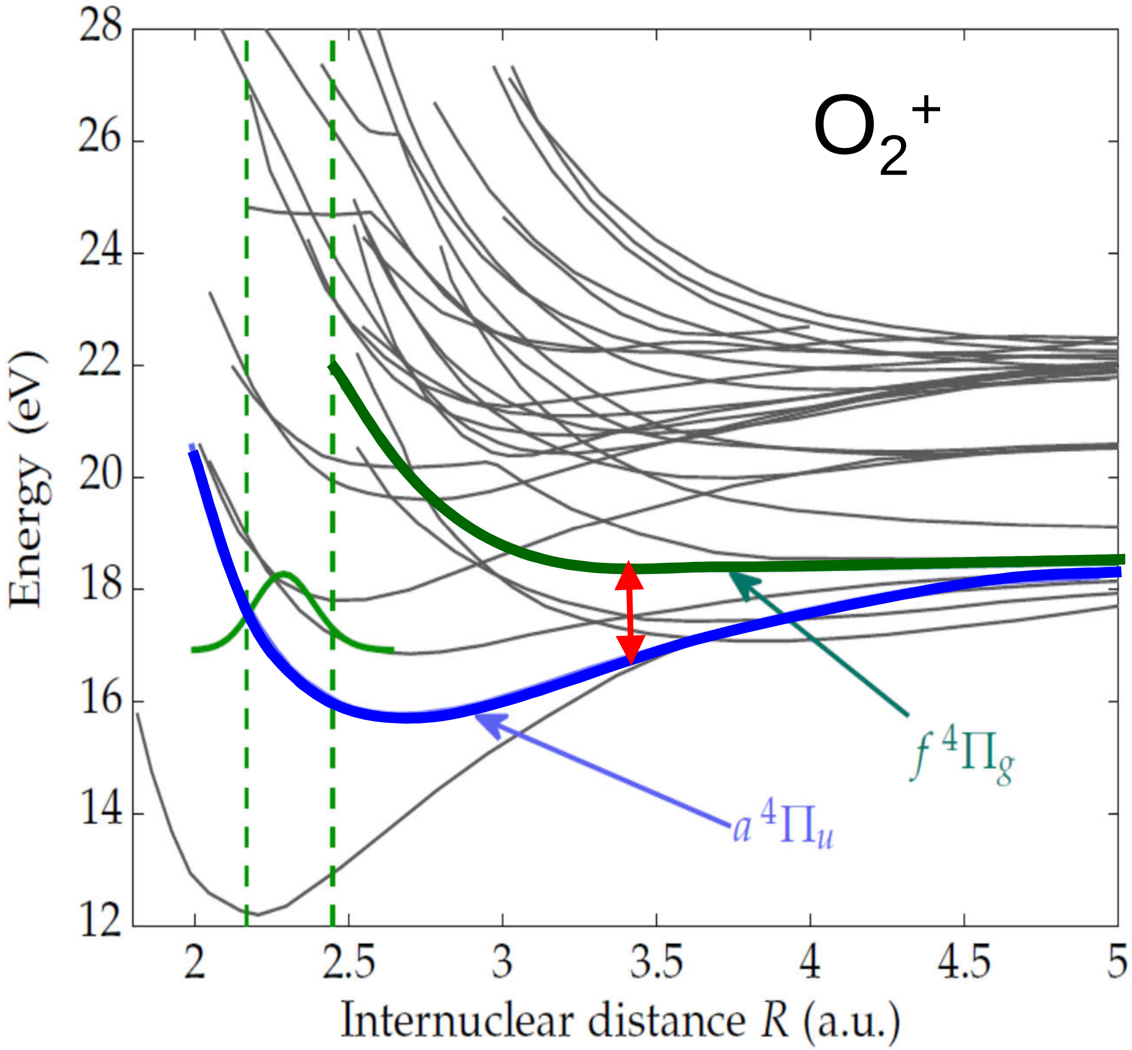

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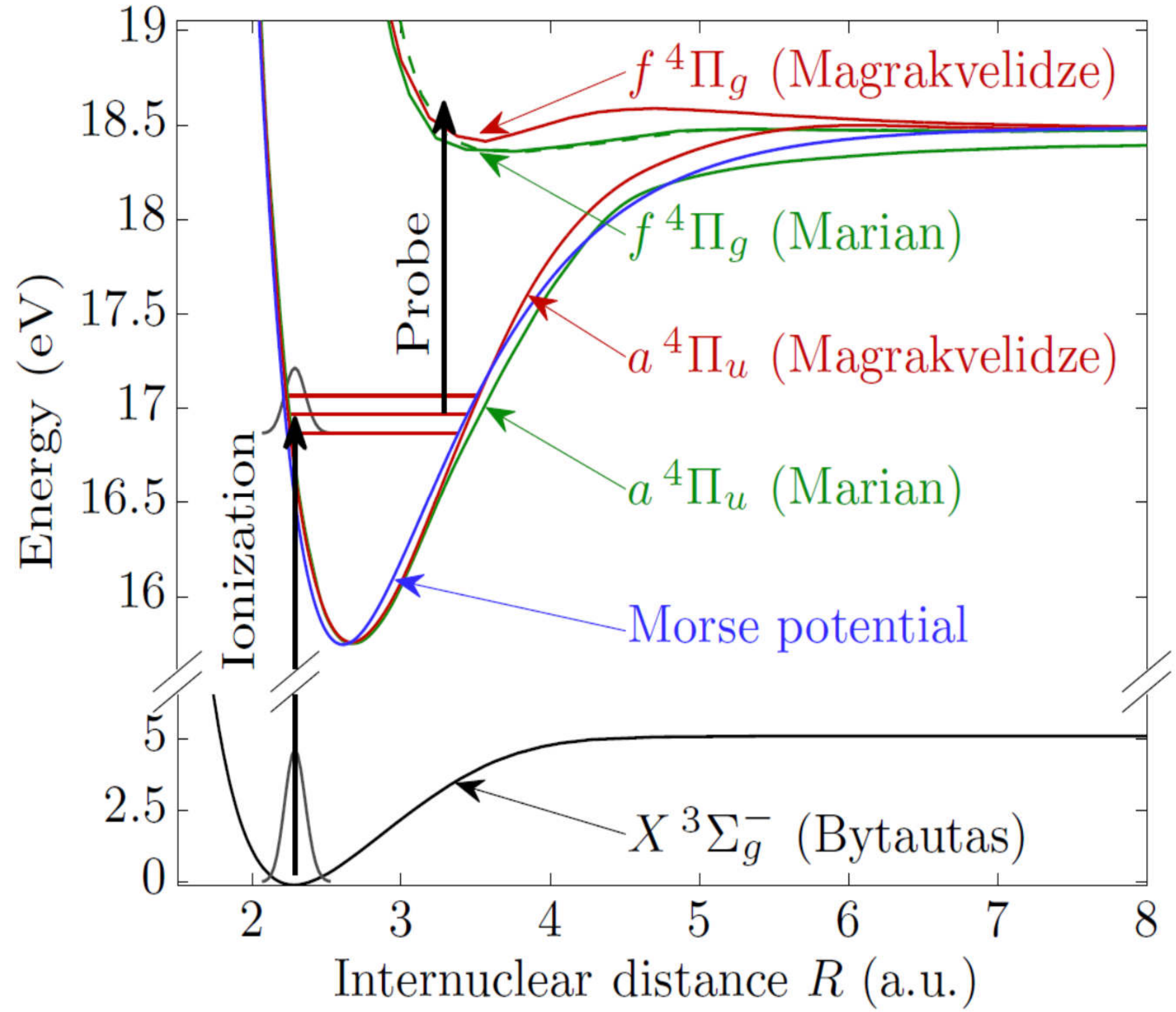

Ionization and Dissoziation of O_2

HHG-IR experiment

1. step: ionization (XUV) $O_2 => O_2^+ + e^-$
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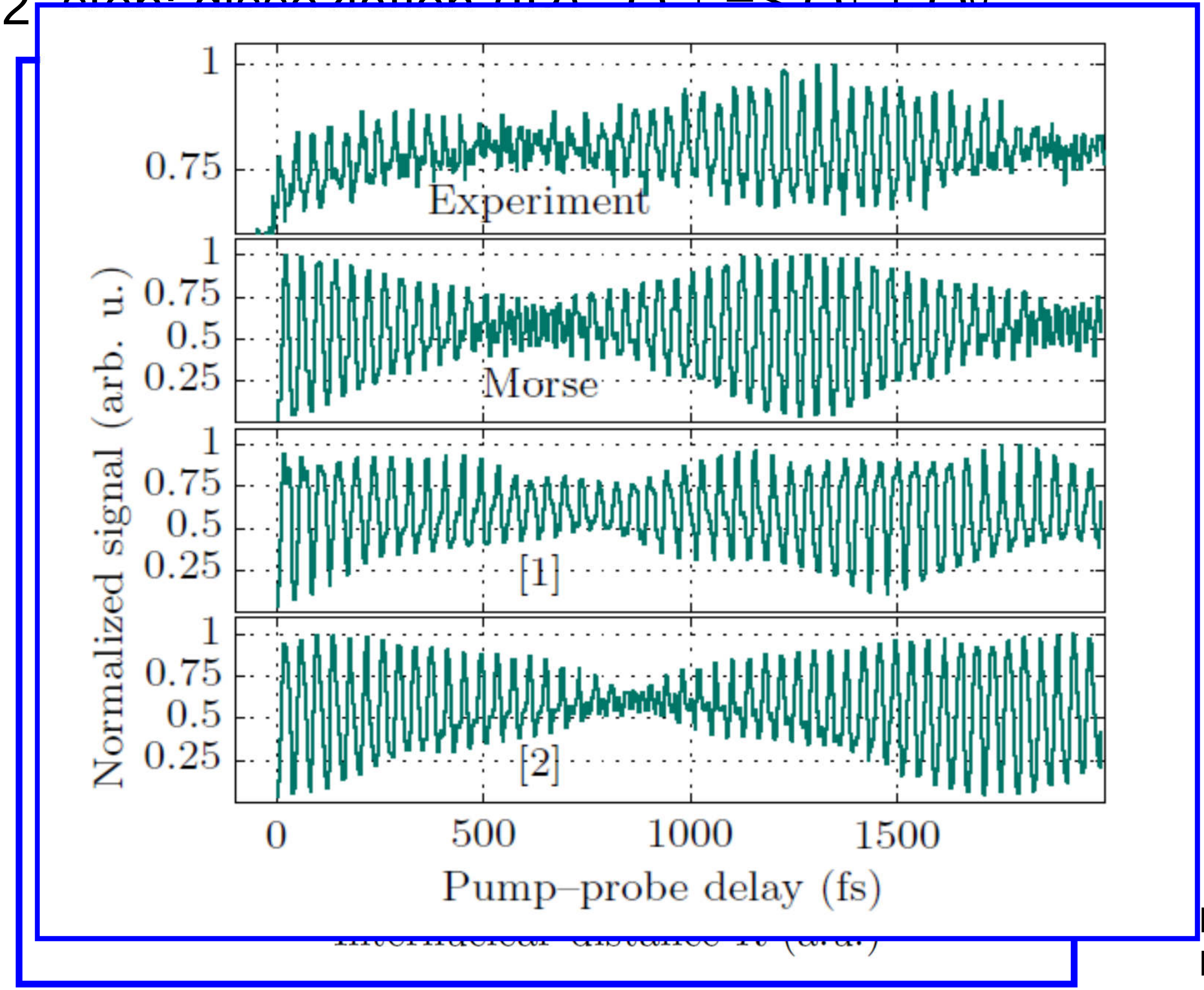
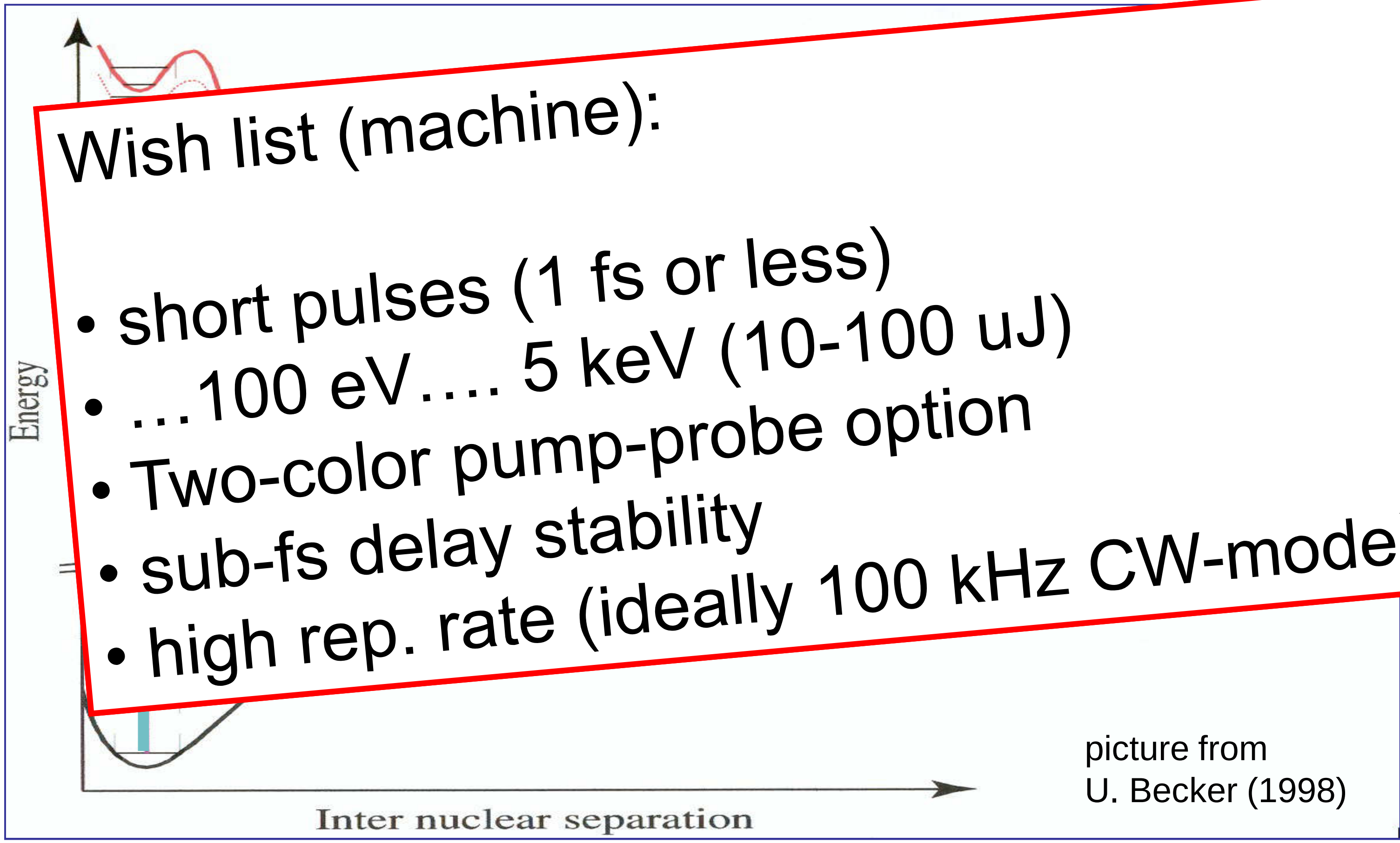


P. Cörlin et al.,
Phys. Rev. A 91 (2015)

Conclusion

FEL pump ($h\nu_1$) – FEL probe ($h\nu_2$)



Wish list (machine):

- short pulses (1 fs or less)
- …100 eV…. 5 keV (10-100 uJ)
- Two-color pump-probe option
- sub-fs delay stability
- high rep. rate (ideally 100 kHz CW-mode)

picture from
U. Becker (1998)

End