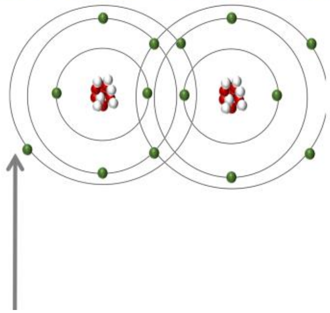
Markus Gühr, FLASH

Ultrafast x-ray summer school 2023

https://www-ssrl.slac.stanford.edu/newsletters/headlines/ultrafast_summerschool07_lg.jpg

The headline color is important

Electrons ‘bind’ nuclei to molecules.



Blue: Part of this lecture

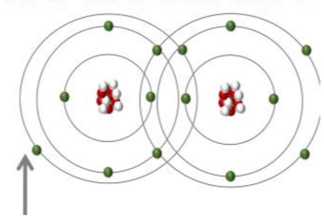
Black: More material for studying later

More formal BOA

For example - what is $<\psi_j(r|R)|\nabla_a|\psi_i(r|R)>$?

$H_e|\psi_i>=E_i|\psi_i>$

Quiz: Which particles are faster?



Red: Quiz for this lecture

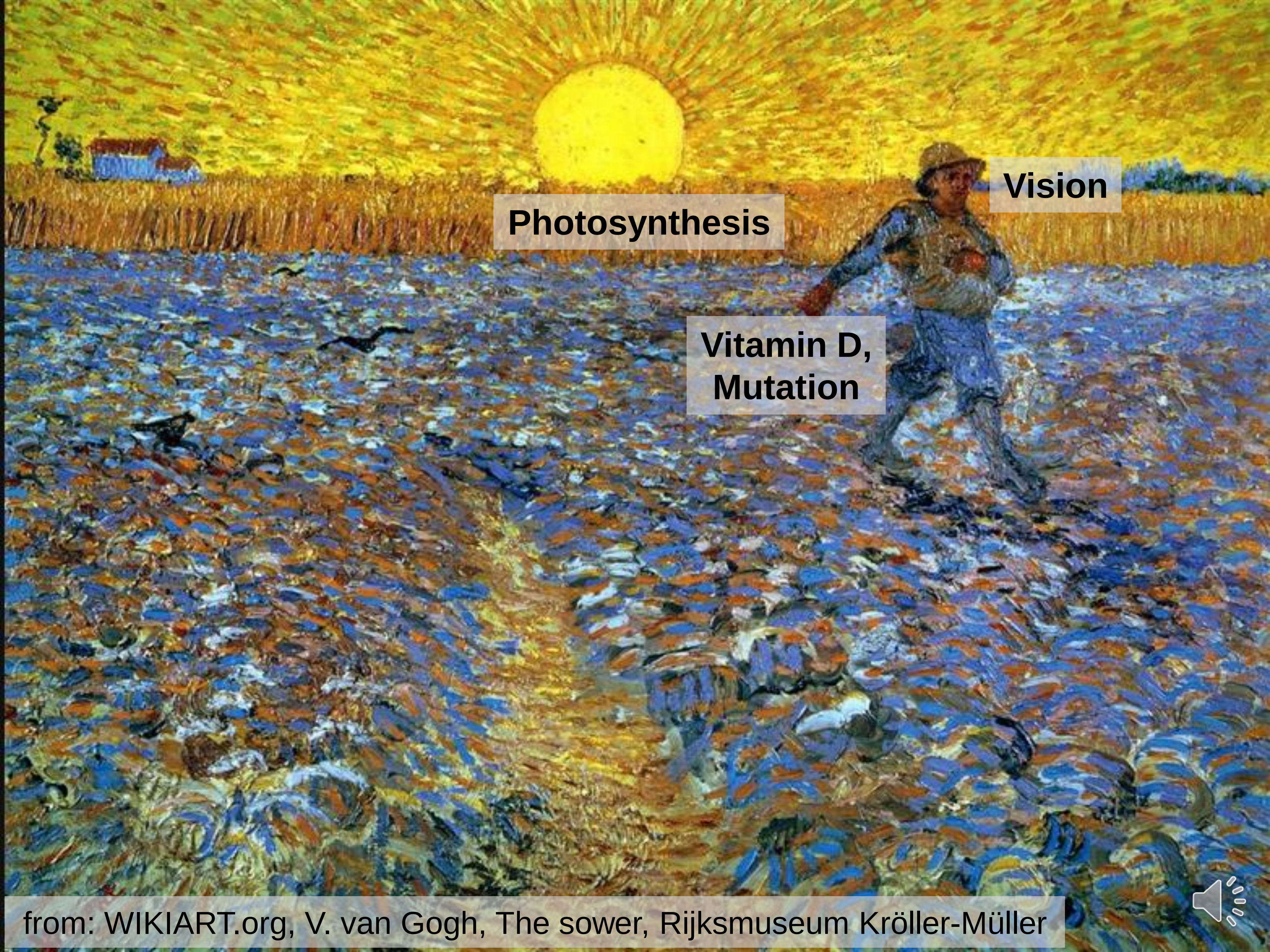
Quiz: Your educational background

What is the discipline of your highest degree?

a) Chemistry
b) Physics
c) Biology/Lifesciences
d) Engineering
e) Computer Science
f) Mathematics
g) other

https://pingo.coactum.de/events/966075

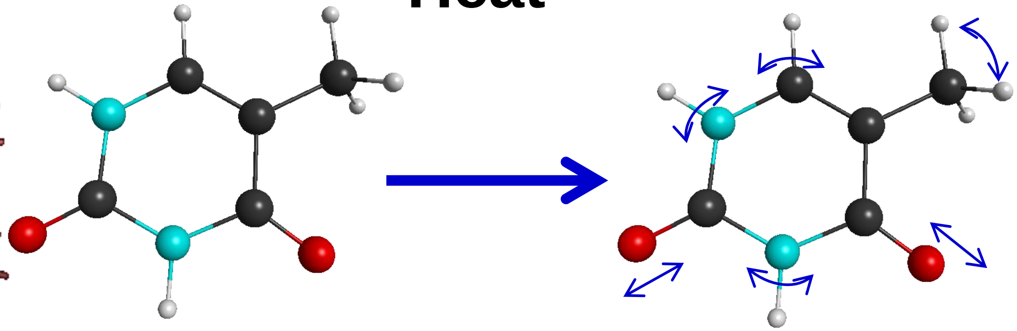

Chemical dynamics in the gas phase

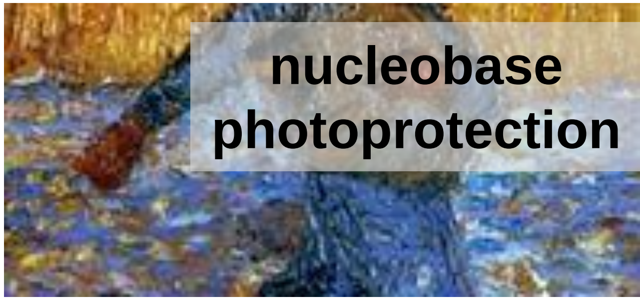


from: WIKIART.org, V. van Gogh, The sower, Rijksmuseum Kröller-Müller

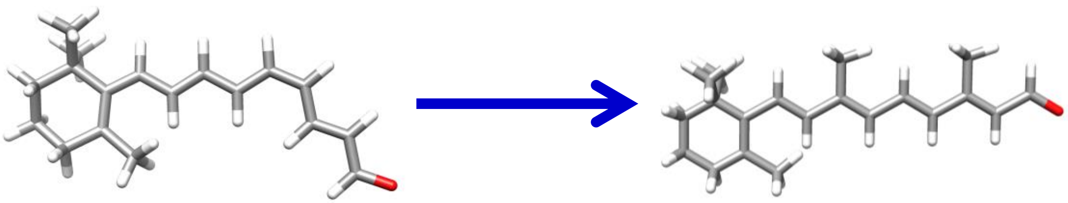
Transformation of light energy to other energies occur (ultra)fast.

Heat



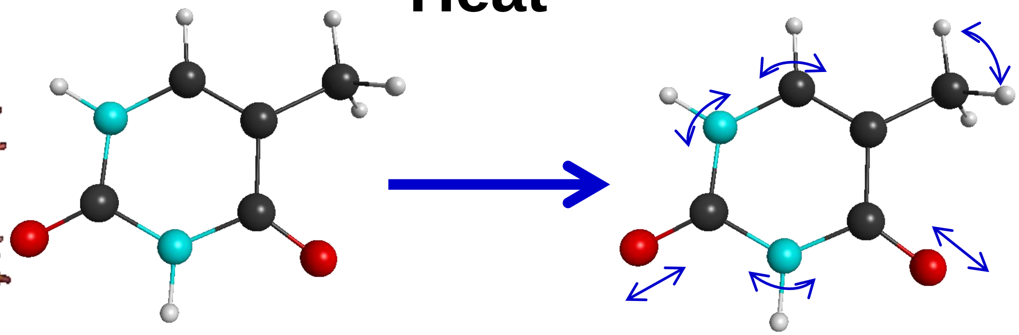


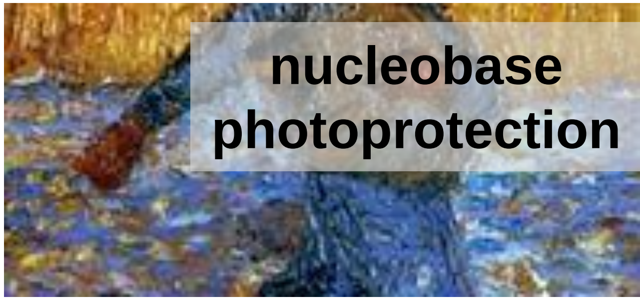
Chemical bond change



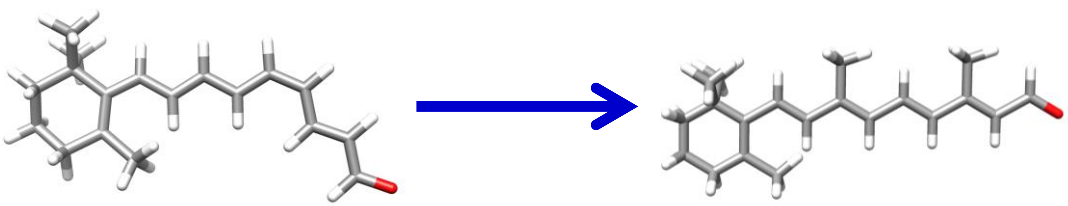
Quiz: Why are these processes happening on an ultrafast timescale?

Heat



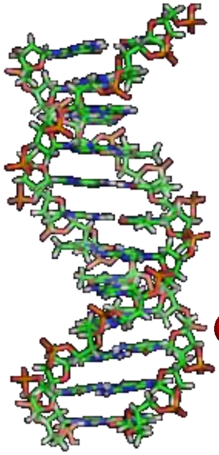


Chemical bond change

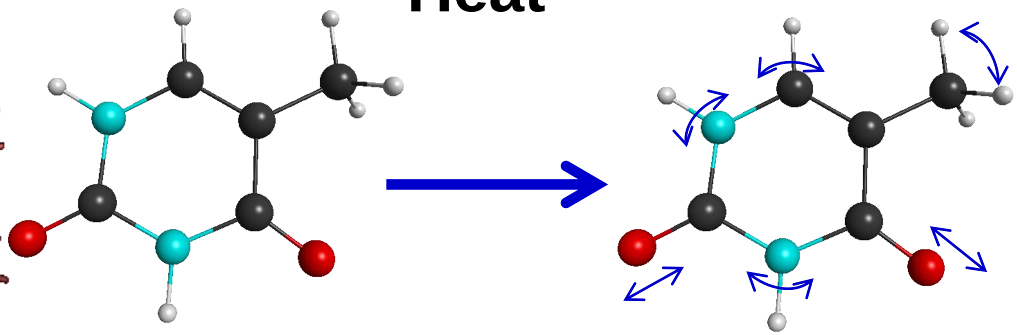


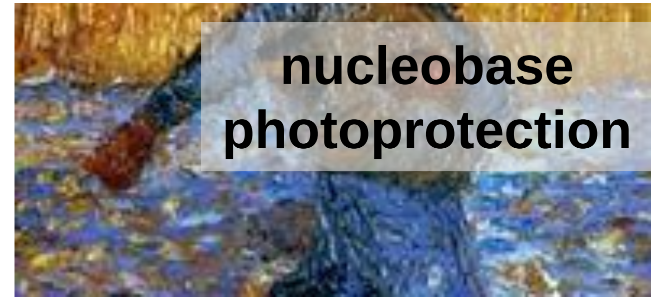
a) Because nuclei and electrons just move that fast
b) Because that might be a way to avoid competing processes
c) Because otherwise energy would not be stored by released again in form of light

Quiz: Why are these processes happening on an ultrafast timescale?

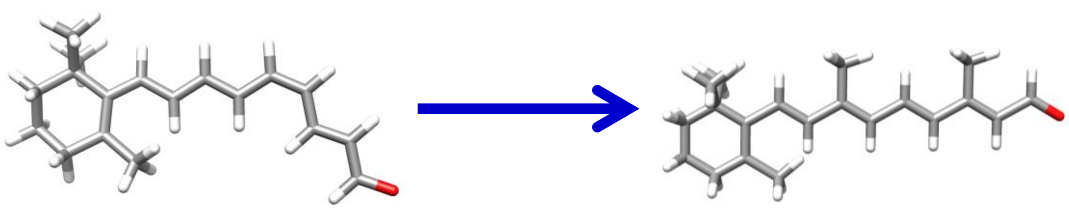


Heat





Chemical bond change



a) Because nuclei and electrons just move that fast
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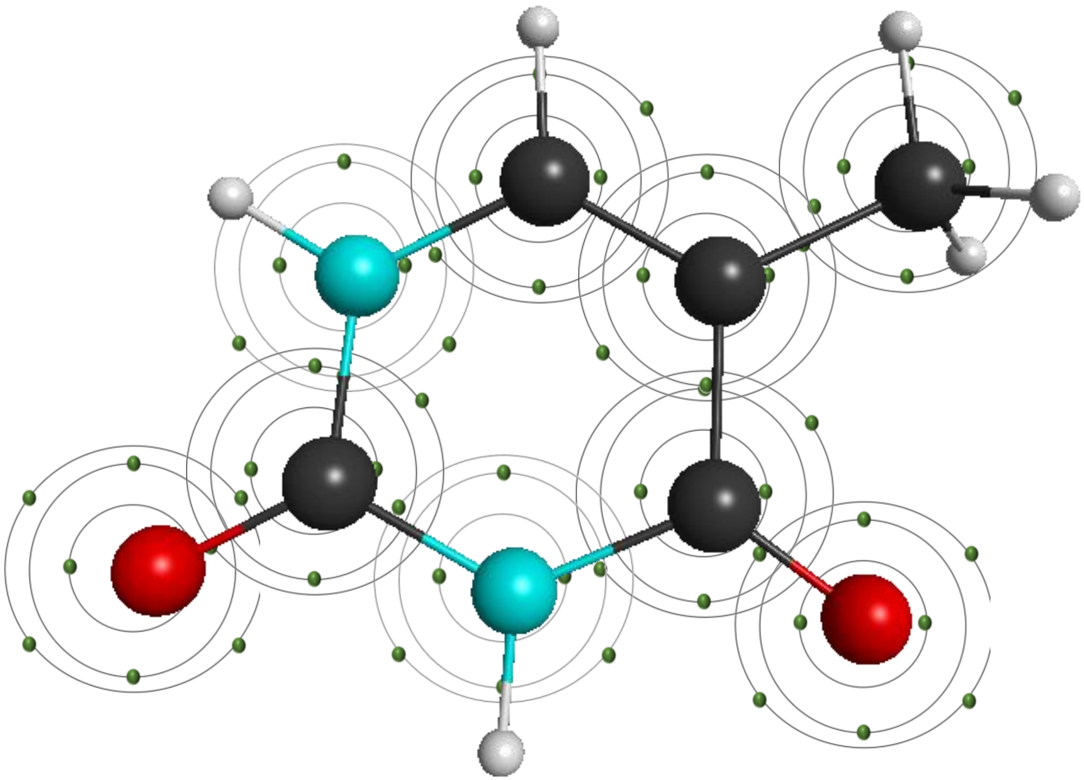
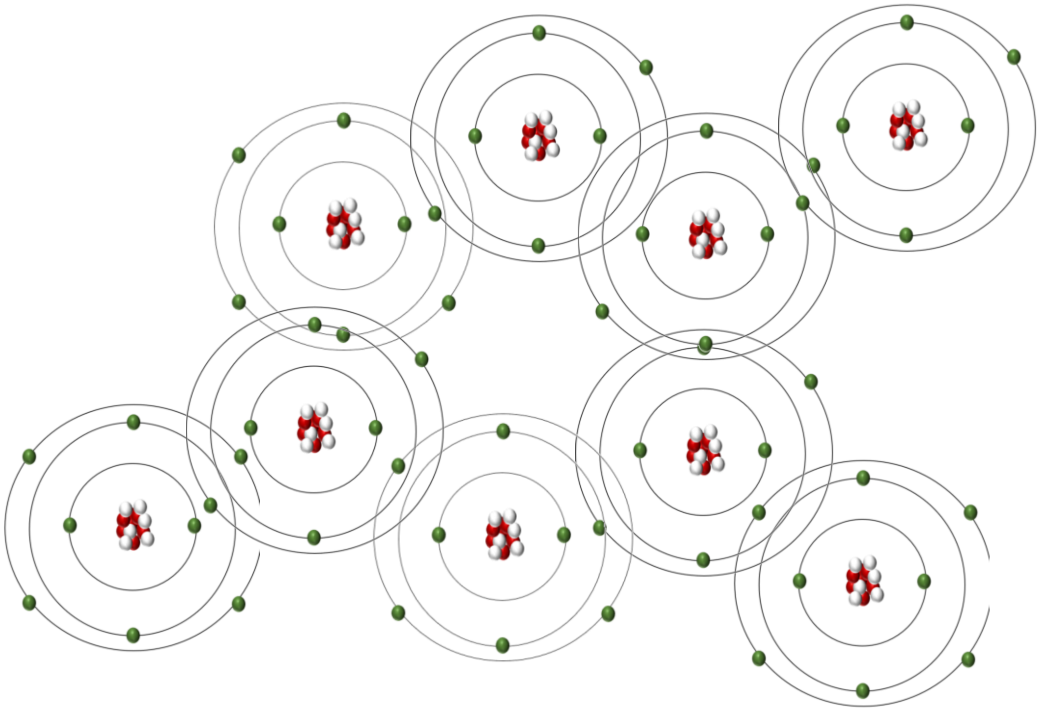
c) Because otherwise energy would not be stored by released again in form of light

Gas phase structural dynamics of molecules

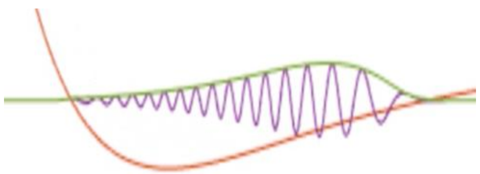


Gas phase structural dynamics of molecules

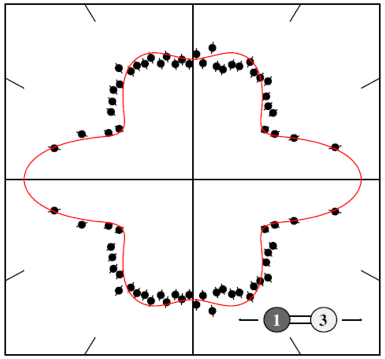


Why doing gas phase molecular studies?

Quantum manipulation possible



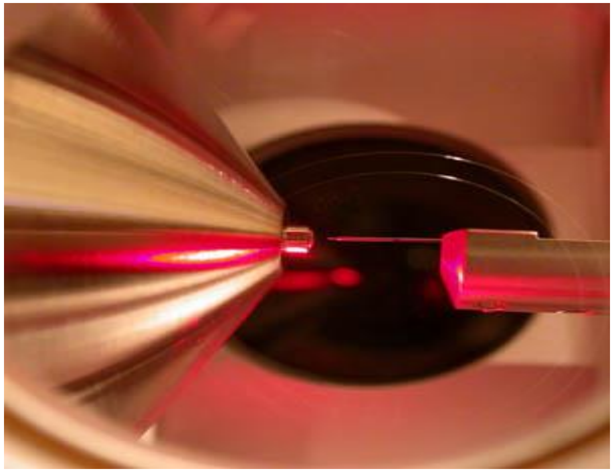
Charged particles available



Kastirke et al., PRX **10**, 021052 (2020)

1:1 comparison with high level calculations

Quite a lot of gas phase applications



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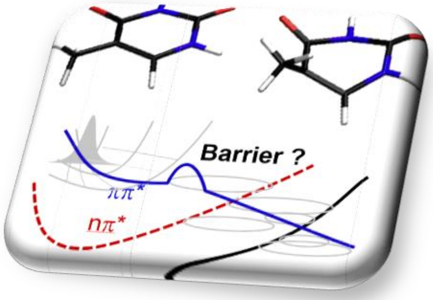
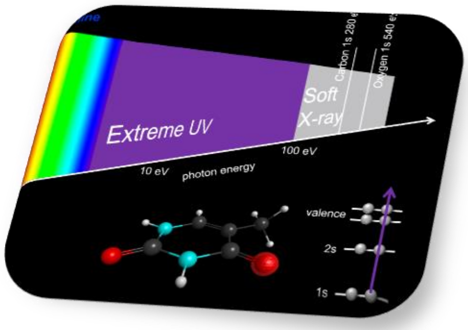
Outline

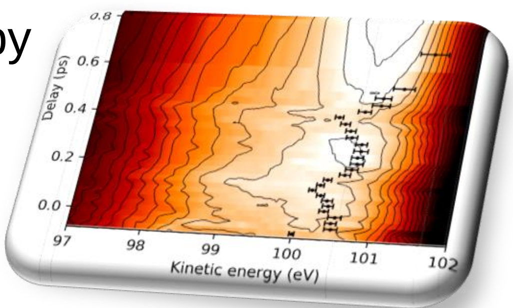
Basics: Coupled electronic and nuclear dynamics in molecules



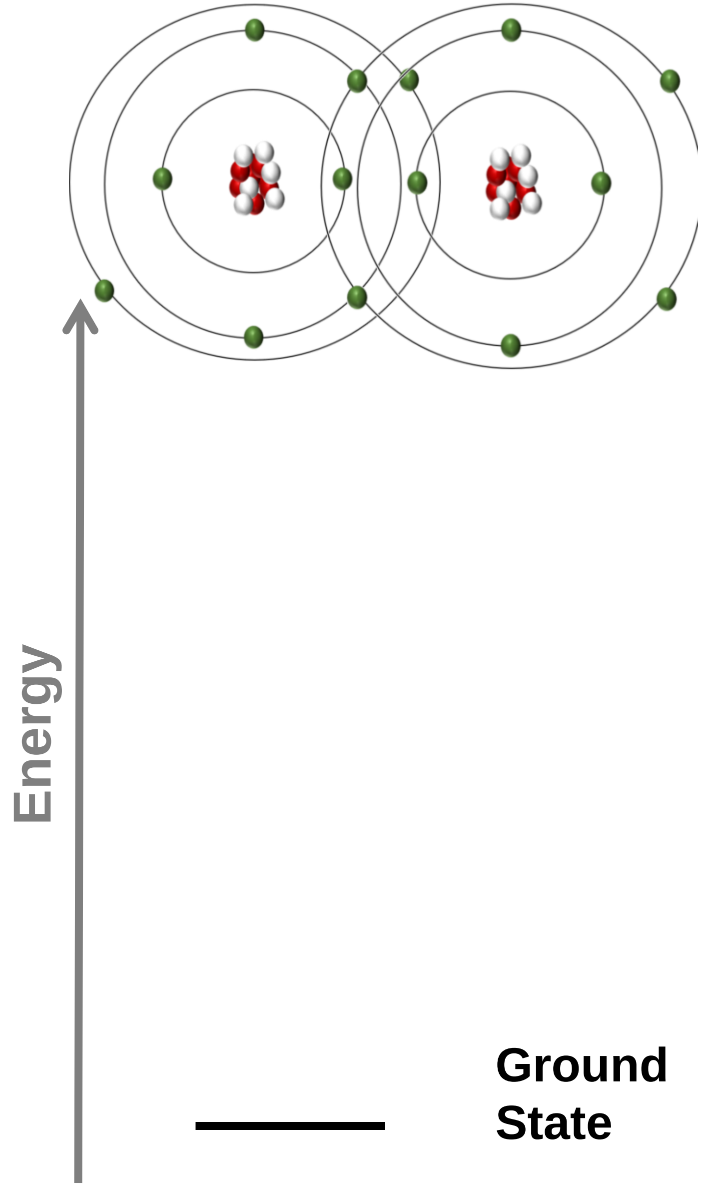
X-ray matter interaction



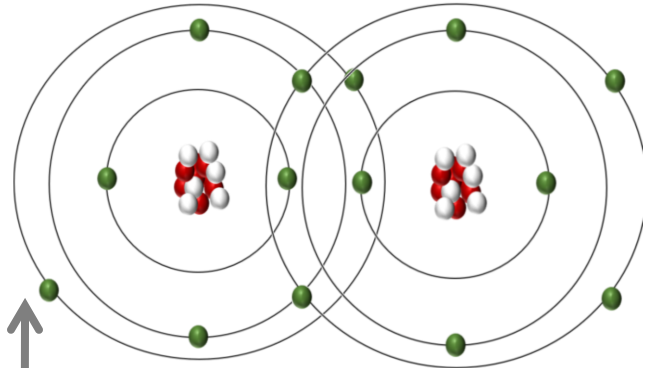
Two examples for ultrafast x-ray spectroscopy
resonant absorption
photoelectron spectroscopy

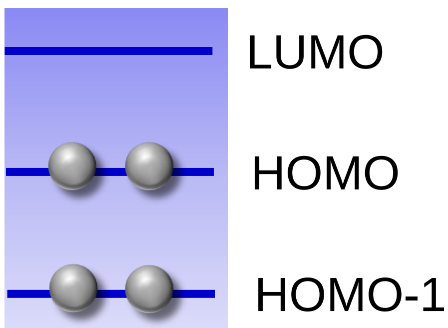


Electrons ‘bind’ nuclei to molecules.



Quiz: Which particles are faster?



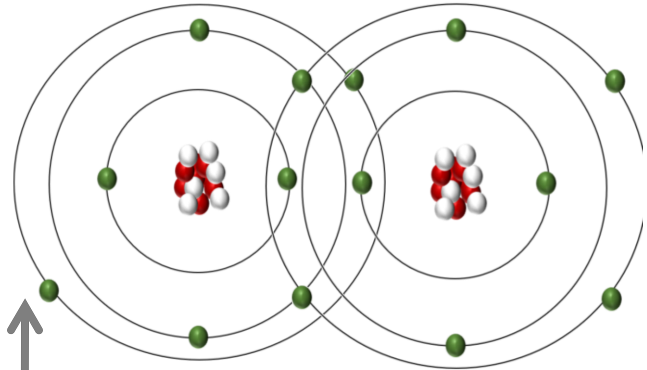


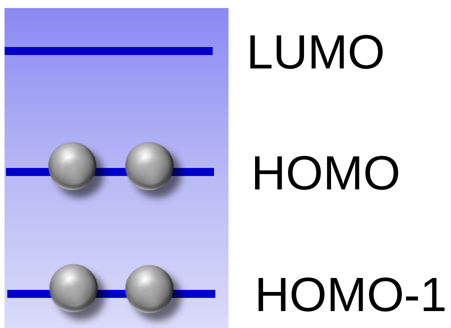
Electrons and nuclei are subject to the same Coulomb forces.
Which particles move faster under those forces?

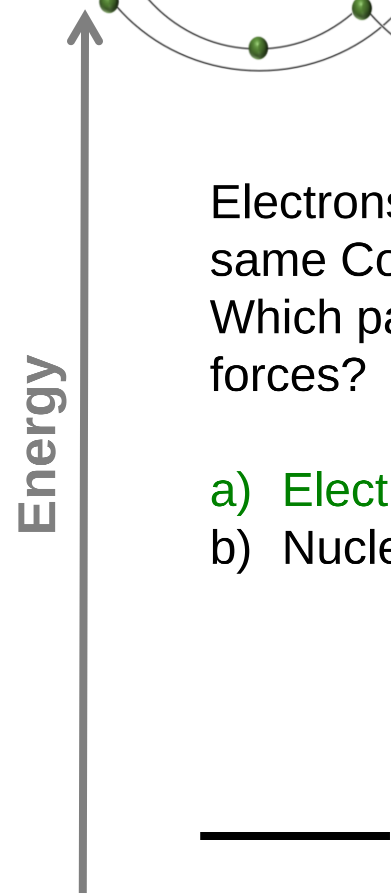
a) Electrons
b) Nuclei

Ground State

Quiz: Which particles are faster?







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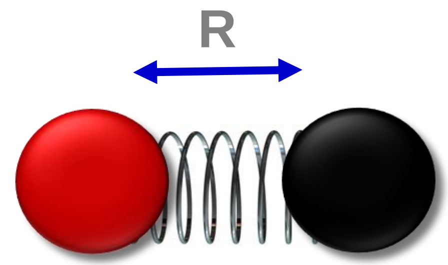
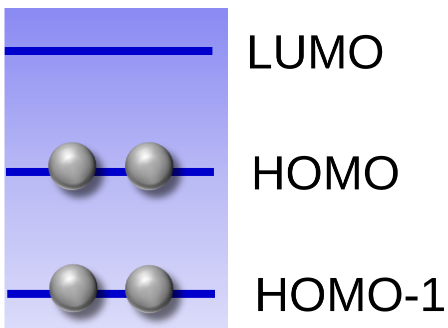
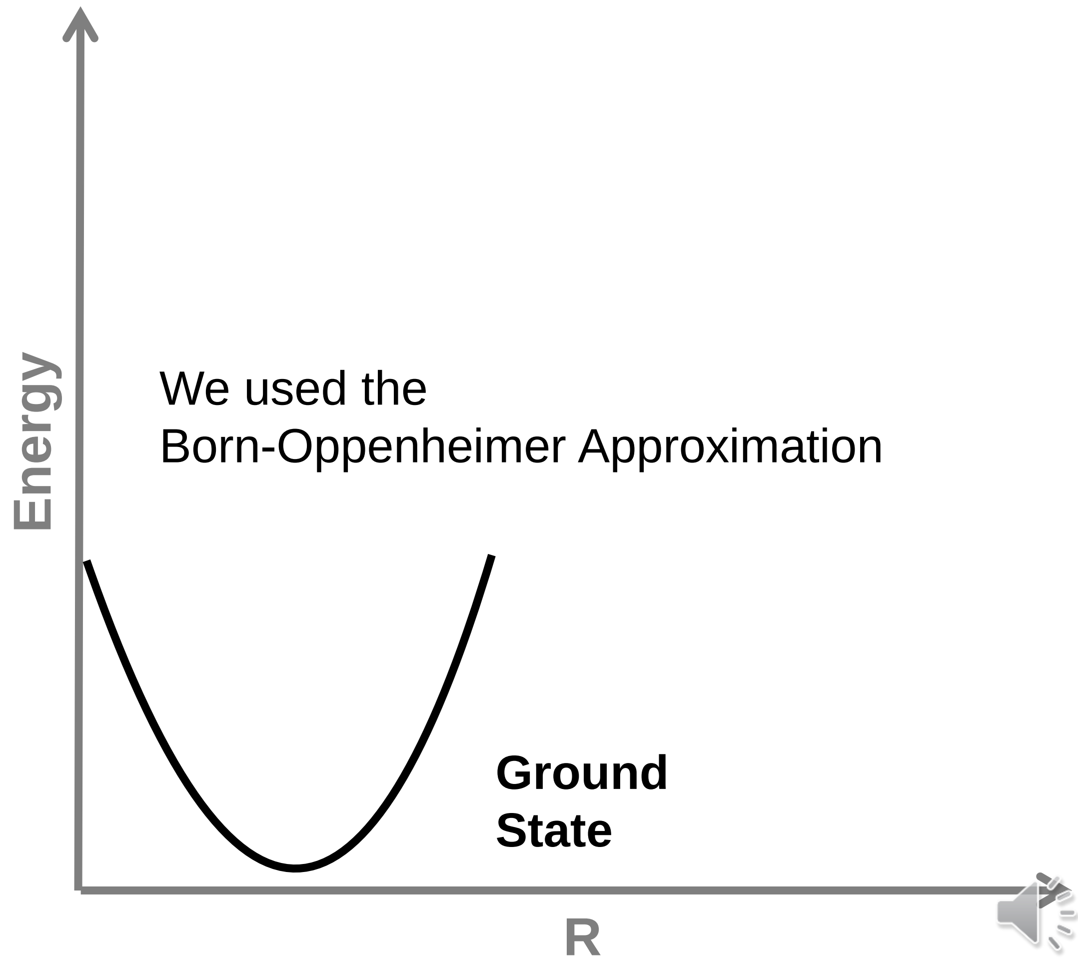
Ground State

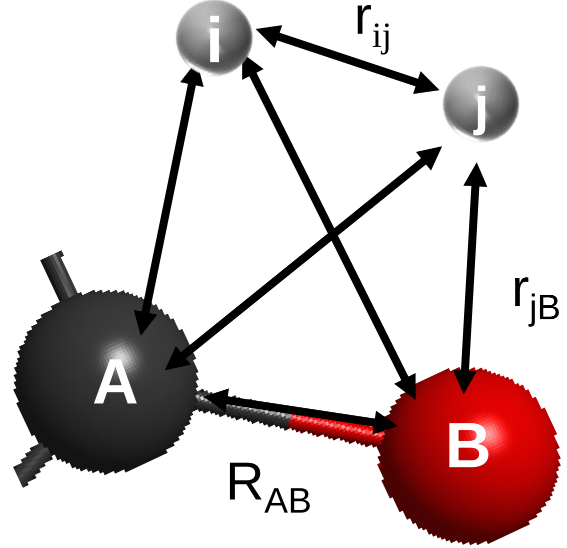
BOA allows for potentials for nuclei.







More formal BOA



$$H = H_e + T$$ Molecular Hamiltonian

$$T = \sum_{a=1}^{M} -\frac{\hbar^2}{2m_a}\nabla_a^2$$ Nuclear kinetic energy

$$H_e = \sum_{j=1}^{N} -\frac{\hbar^2}{2m_e}\nabla_j^2$$ Electronic Hamiltonian

$$-\sum_{j=1}^{N}\sum_{a=1}^{M}\frac{Z_a e^2}{r_{ja}}$$ Nuclear Electron attraction

$$+\sum_{j<k}\frac{e^2}{r_{jk}}$$ Electronic repulsion

$$+\sum_{a<b} Z_a Z_b \frac{e^2}{R_{ab}}$$ Nuclear repulsion

$$H = H_e + T$$
$$H\,|\psi(r,R)>= E\,|\psi(r,R)>$$
$$H_e\,|\psi_i(r\,|\,R)>= E_i\,|\psi_i(r\,|\,R)>$$
only parametric dependence on R
$$\psi(r,R) = \sum_i |\psi_i(r\,|\,R)\xi_i(R)>$$

David Reis‘ talk yesterday

More formal BOA

See f.i. Jensen
Introduction to Computational Chemistry
pp. 80

$$(H_e + T - E)\sum_i |\psi_i(r\,|\,R)\xi_i(R) > = 0$$

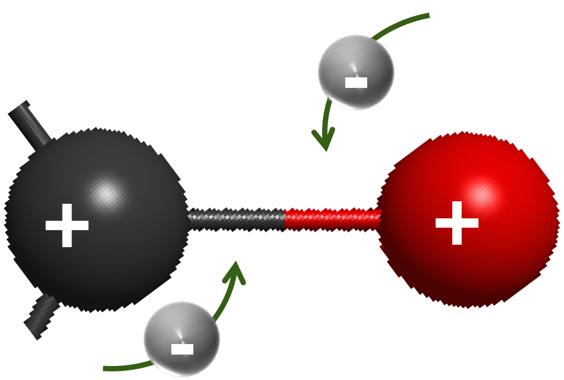
multiply by $<\psi_j(r\,|\,R)\,|$

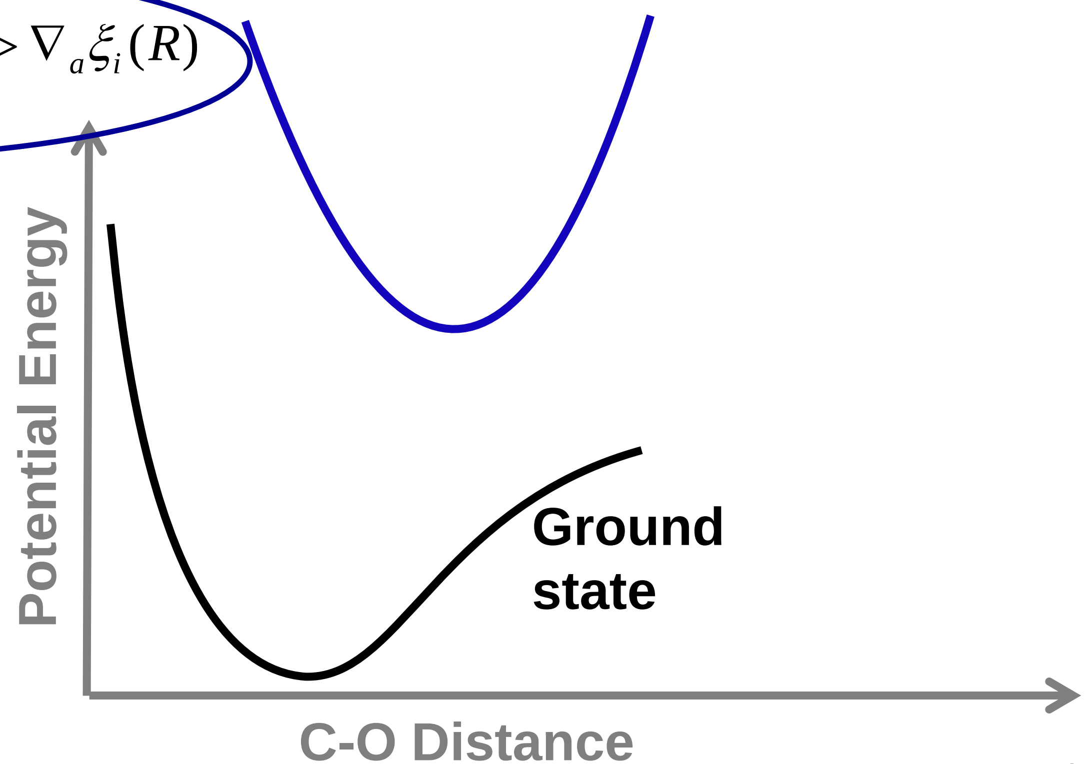
$$<\psi_j(r\,|\,R)\,|\,(H_e + T - E)\sum_i |\psi_i(r\,|\,R)\xi_i(R) > = 0$$

$$\Rightarrow (E_j(R) + T - E(R))\xi_j(R) = \sum_{i \neq j} <\psi_j(r\,|\,R)\,|\,T\,|\,\psi_i(r\,|\,R) > \xi_i(R)$$

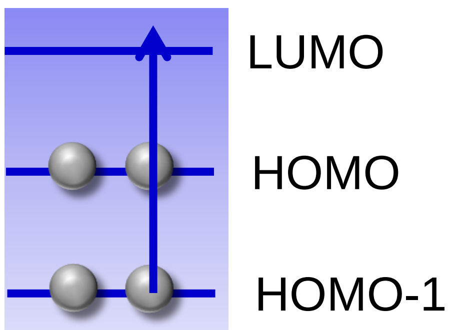
$$+ \sum_{i \neq j}\sum_a \frac{\hbar^2}{m_a} <\psi_j(r\,|\,R)\,|\,\nabla_a\,|\,\psi_i(r\,|\,R) > \nabla_a \xi_i(R)$$

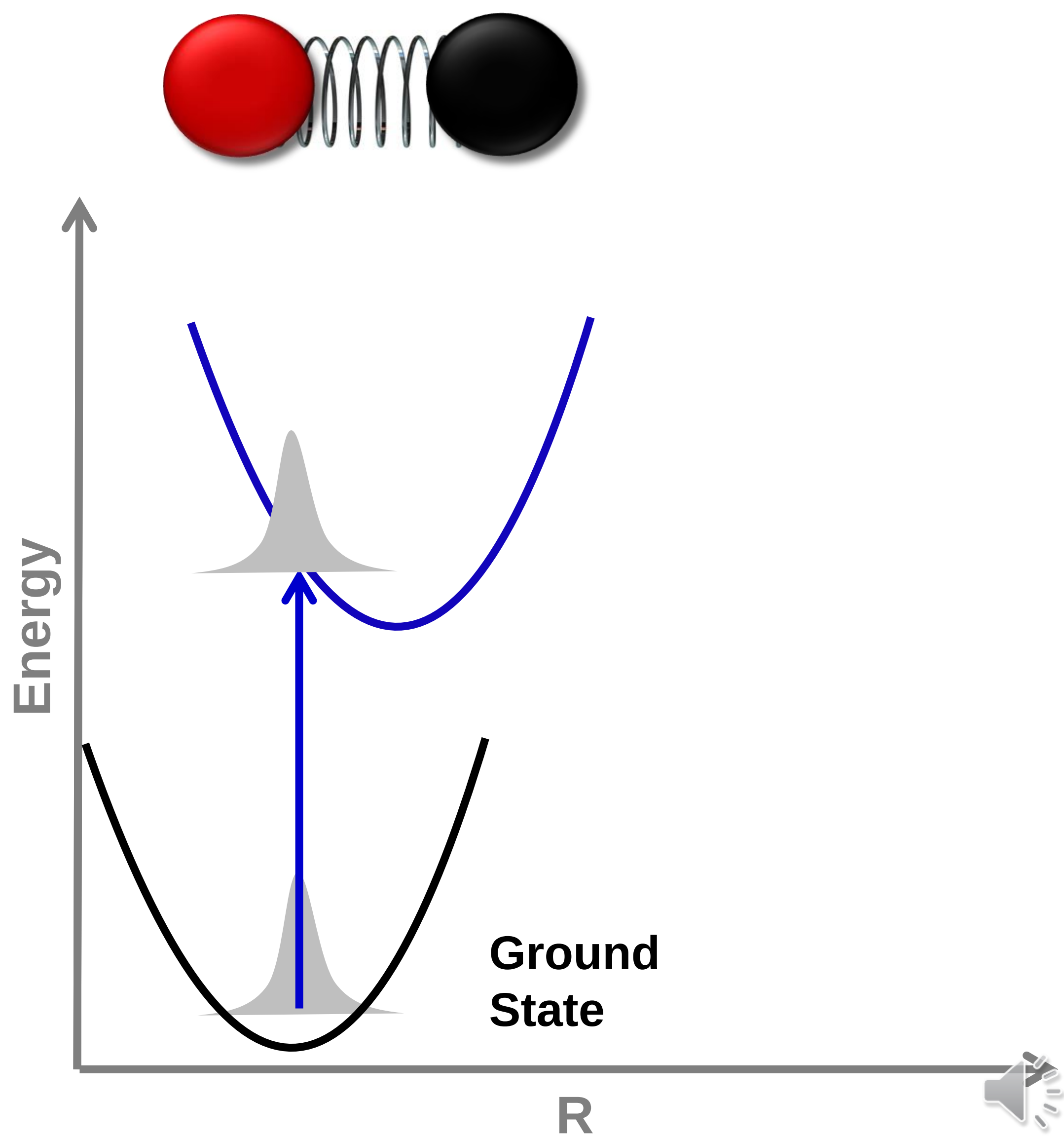
Neglected in BOA



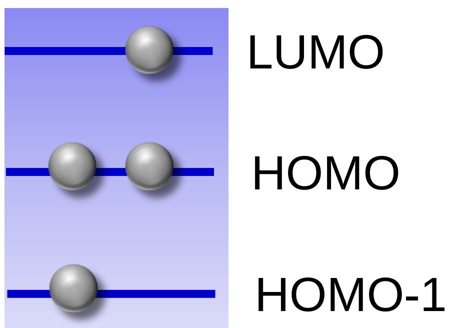


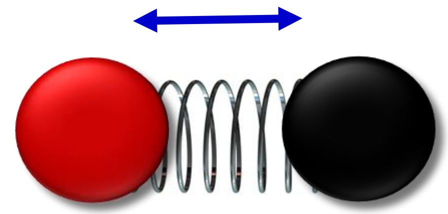
Light excitation couples to electrons.

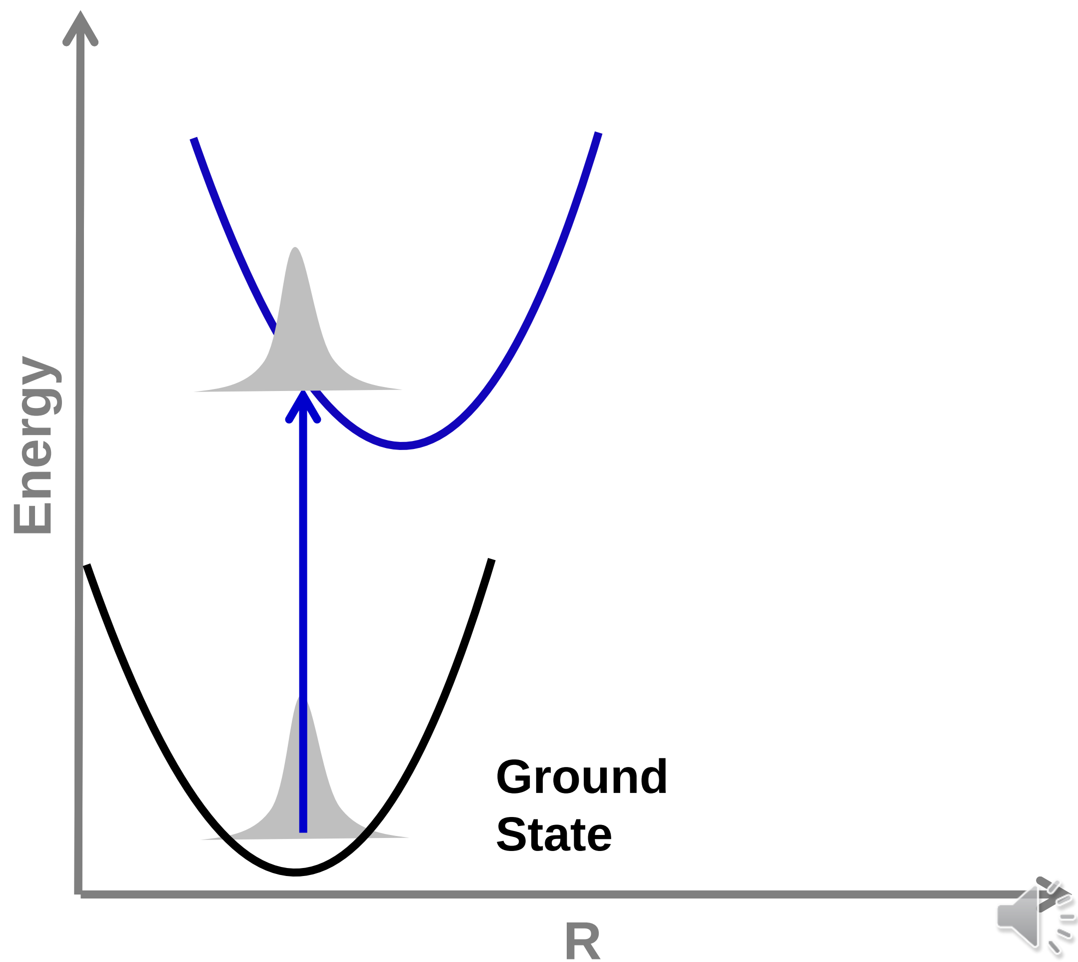




Electrons couple to nuclei.







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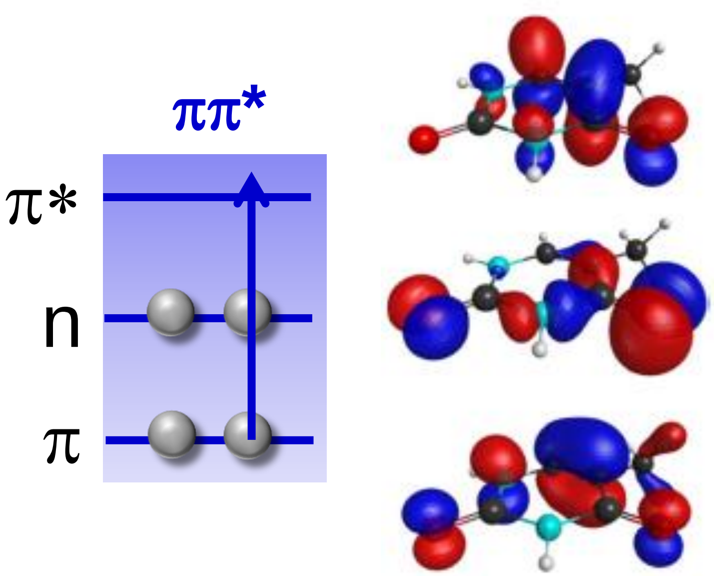

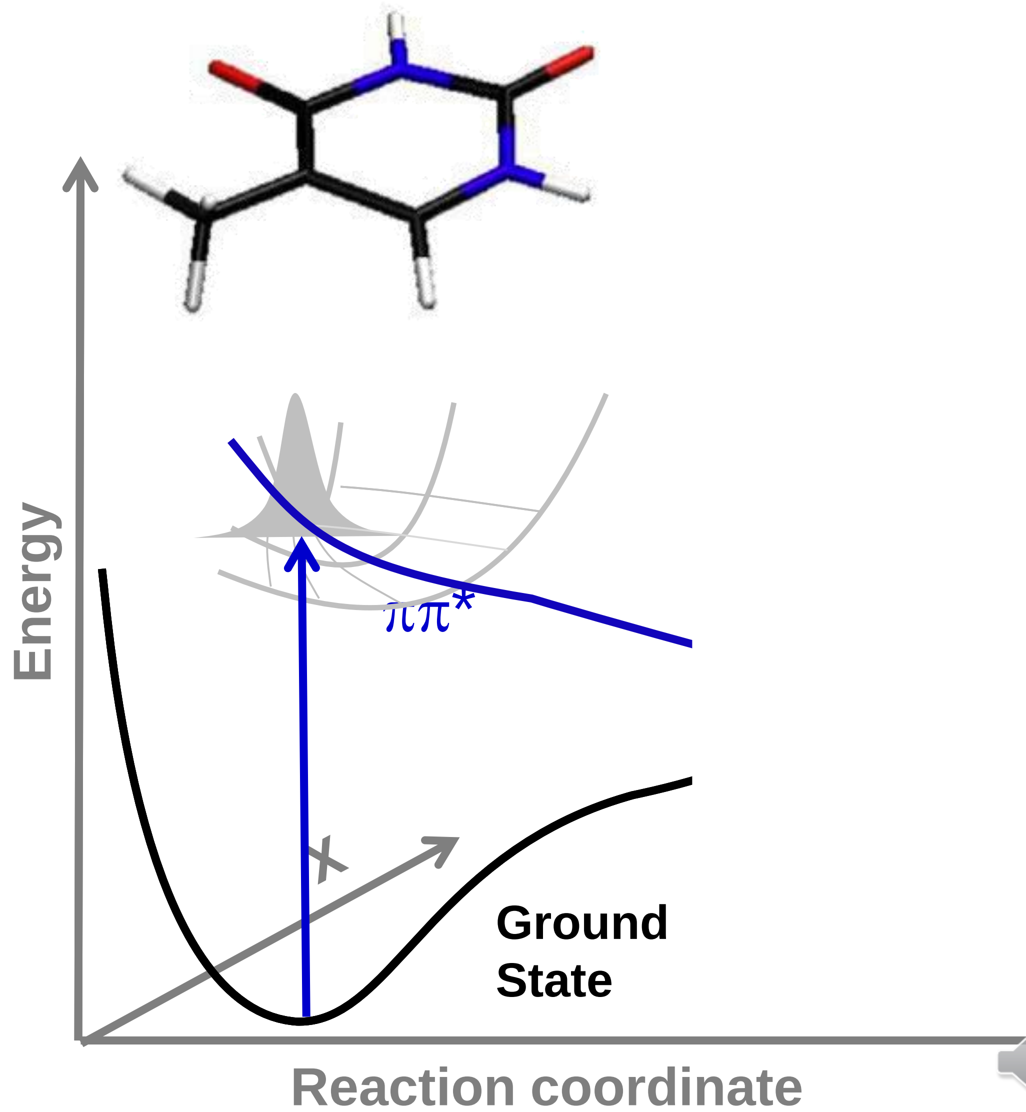

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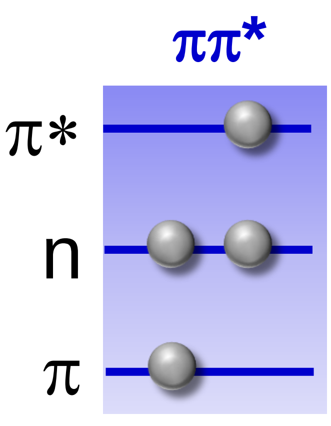
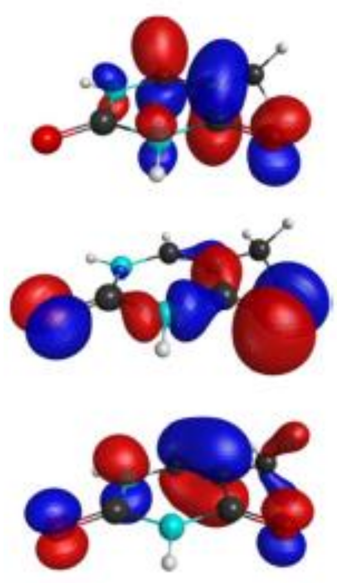
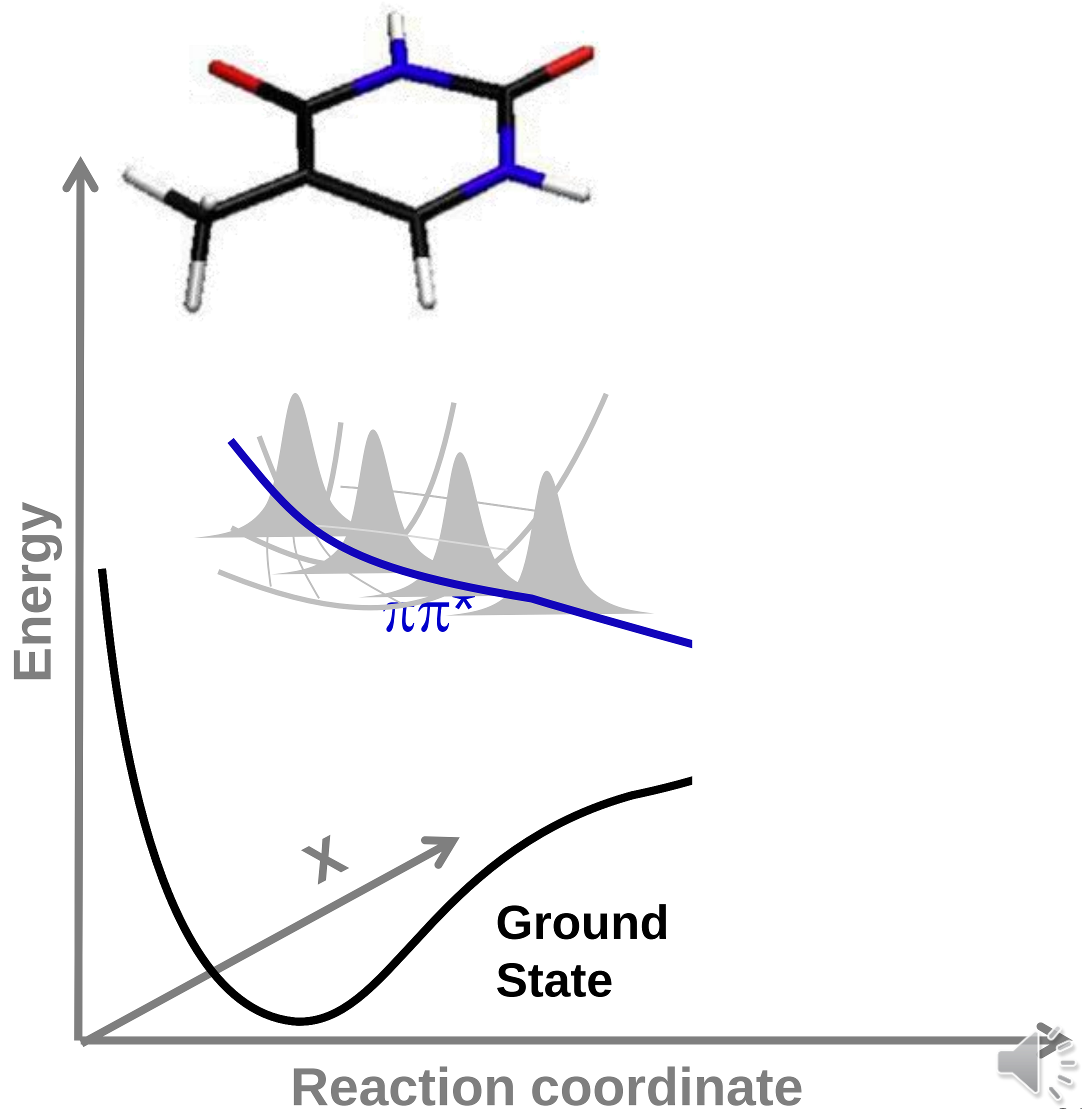
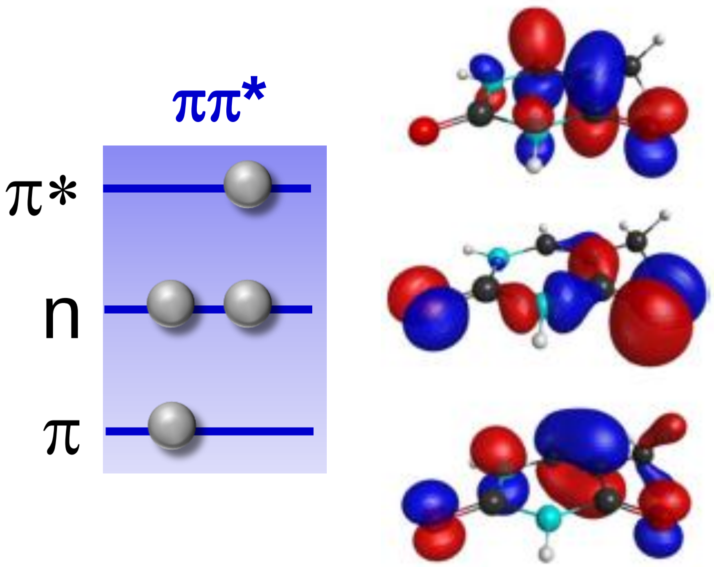


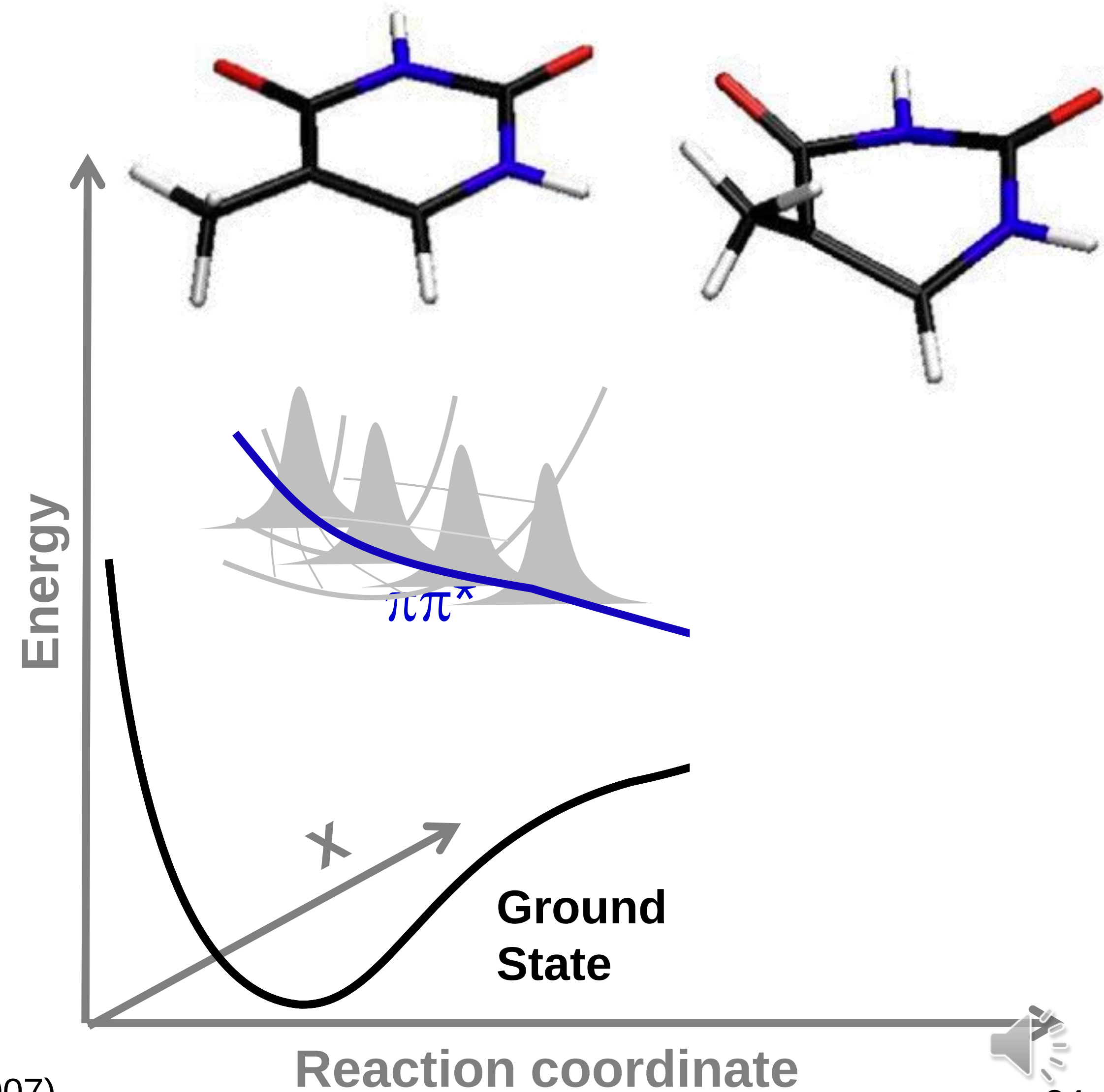




Electrons couple to nuclei.

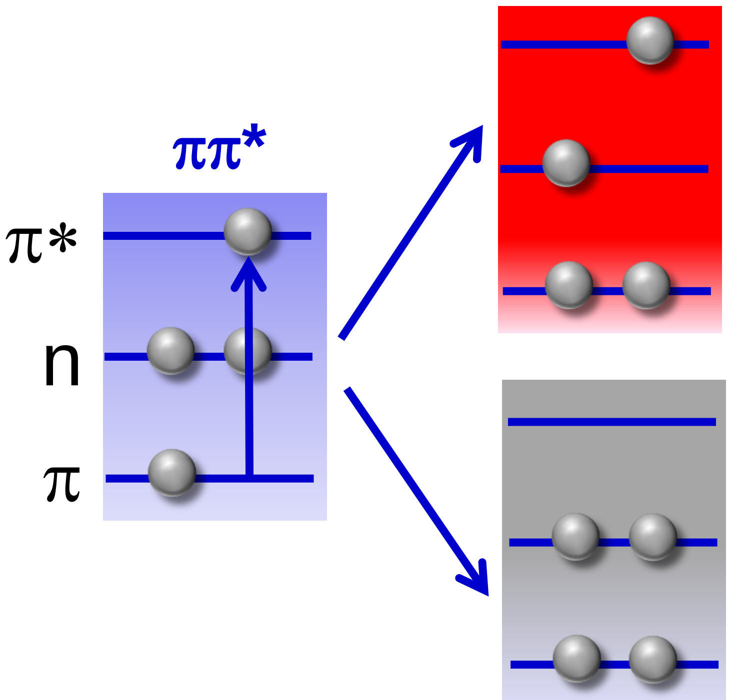
Interuclear separation changes
Angles in space change

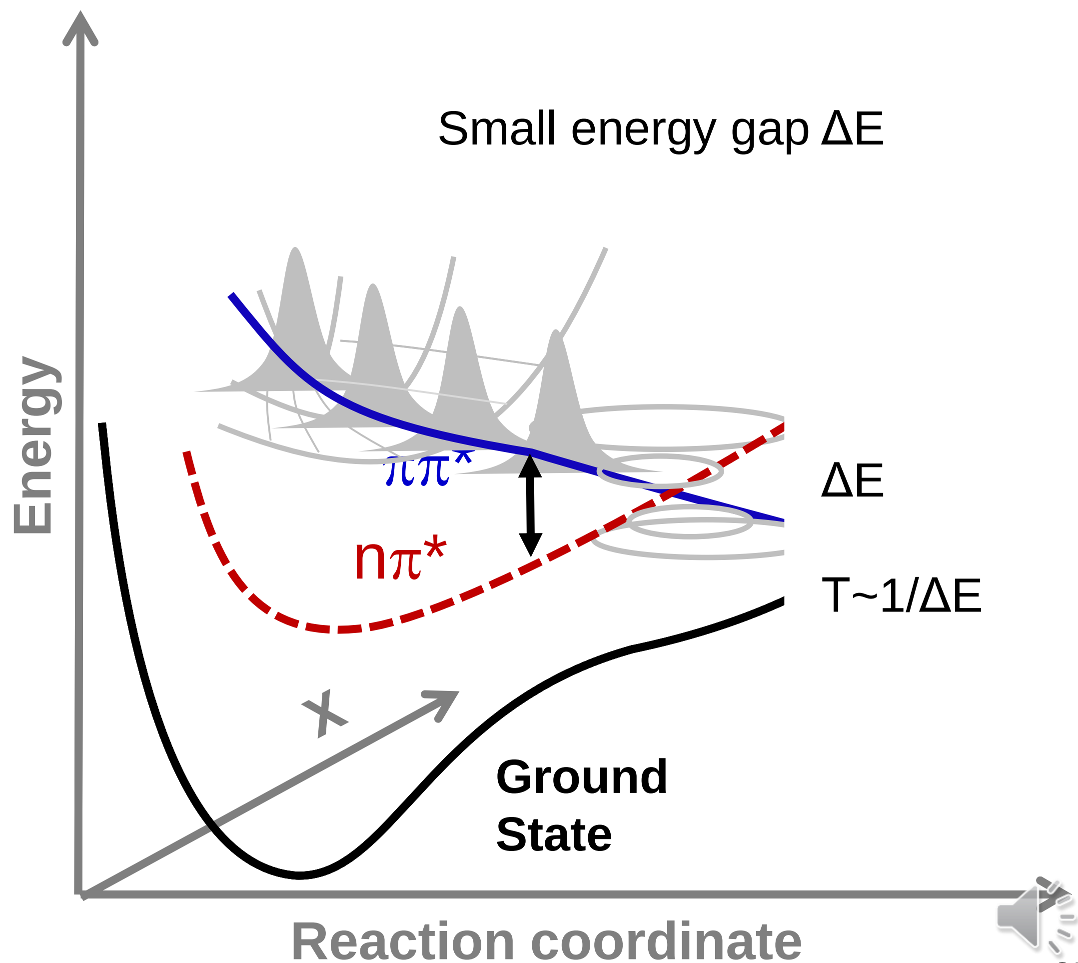




Geometries:
Hudock *et al.*,
J. Phys. Chem. A,**111**, 85 (2007)

Nuclei couple to electrons.





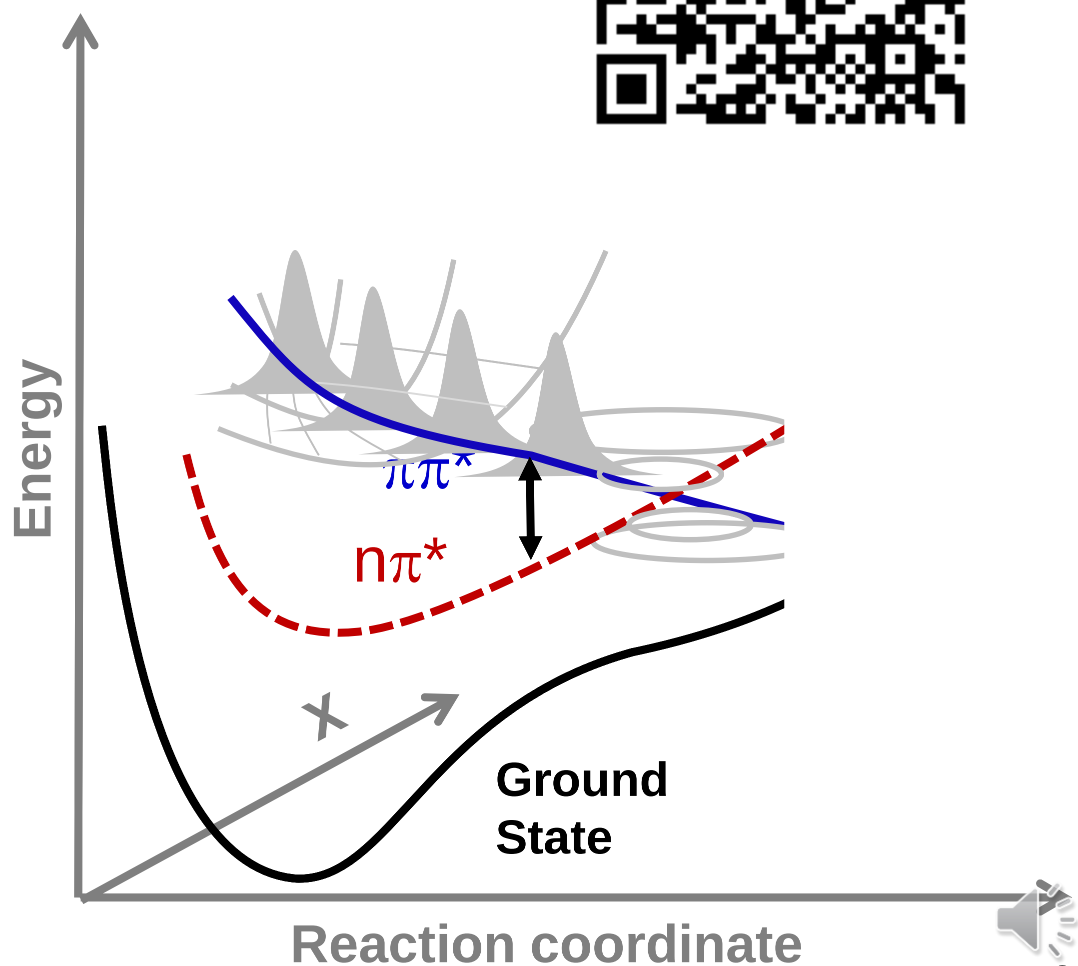
Wait a moment….

Remember Jon‘s lecture.
Electronic motion can be thought of a superposition of at least two quantum states.

There is a small energy gap ΔE between the two quantum electronic states.

What does that mean for timescale (or period) of electron motion ?

a) It is long
b) It is short
c) I don‘t care, I need a coffee



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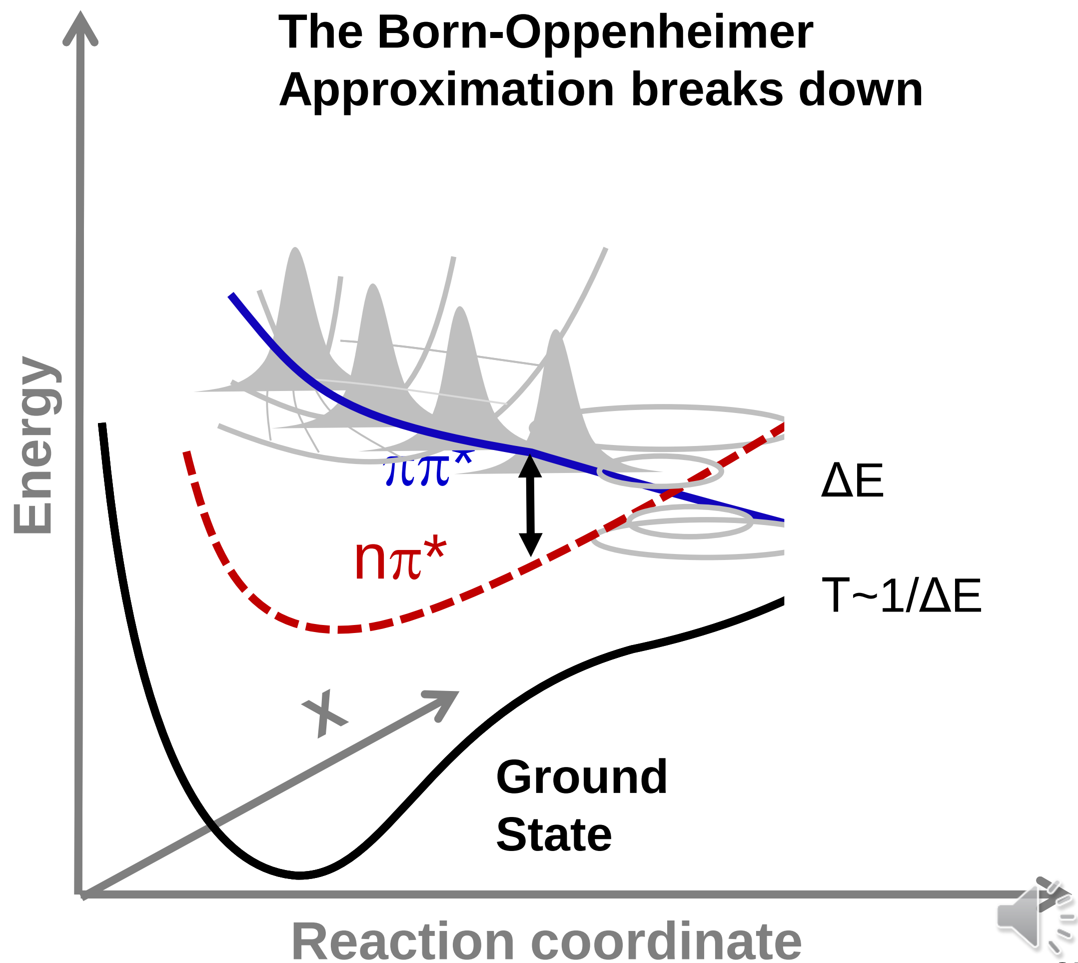
b) It is short
c) I don‘t care, I need a coffee

So, electrons move slower as the ΔE gets smaller!

The Born-Oppenheimer Approximation breaks down



More formal BOA

See f.i. Jensen
Introduction to Computational Chemistry
pp. 80

$$(H_e + T - E)\sum_i |\psi_i(r|R)\xi_i(R)> = 0$$

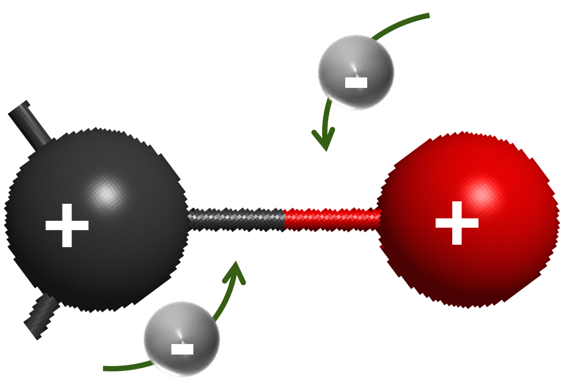
multiply by $<\psi_j(r|R)|$

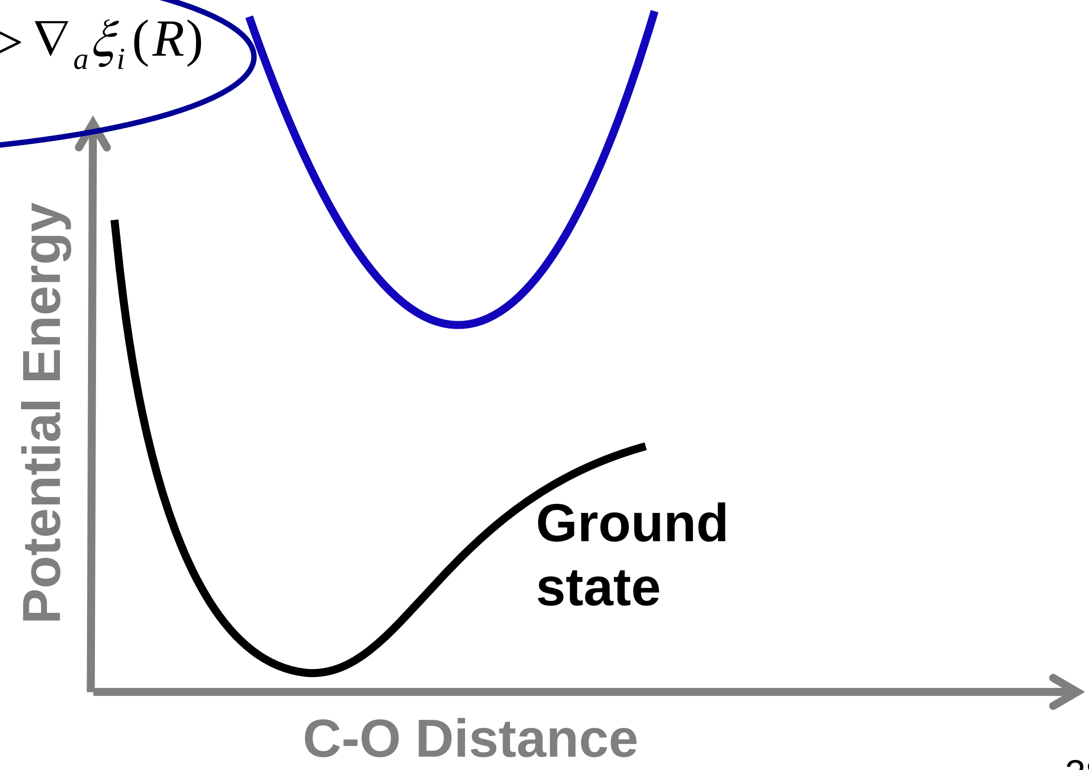
$$<\psi_j(r|R)|(H_e + T - E)\sum_i |\psi_i(r|R)\xi_i(R)> = 0$$

$$\Rightarrow (E_j(R) + T - E(R))\xi_j(R) = \sum_{i \neq j} <\psi_j(r|R)|T|\psi_i(r|R)>\xi_i(R)$$

$$+\sum_{i \neq j}\sum_a \frac{\hbar^2}{m_a}<\psi_j(r|R)|\nabla_a|\psi_i(r|R)>\nabla_a\xi_i(R)$$

Neglected in BOA





More formal BOA

For example - what is $<\psi_j(r|R)|\nabla_a|\psi_i(r|R)>$?

$$H_e|\psi_i>=E_i|\psi_i>$$

$$\Rightarrow \nabla_a(H_e|\psi_i>)=\nabla_a(E_i|\psi_i>)$$

$$\Rightarrow (\nabla_a H_e)|\psi_i>+H_e(\nabla_a|\psi_i>)=(\nabla_a E_i)|\psi_i>+E_i(\nabla_a|\psi_i>)$$

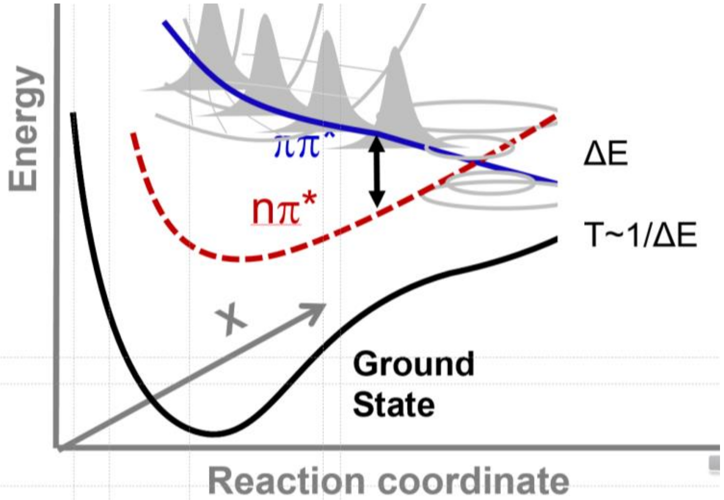
$$\Rightarrow <\psi_j|(\nabla_a H_e)|\psi_i>+<\psi_j|H_e(\nabla_a|\psi_i>)=(\nabla_a E_i)<\psi_j|\psi_i>+E_i<\psi_j|\nabla_a|\psi_i>$$

$$\Rightarrow <\psi_j|(\nabla_a H_e)|\psi_i>+E_j<\psi_j|\nabla_a|\psi_i>=E_i<\psi_j|\nabla_a|\psi_i>$$

$$\Rightarrow <\psi_j|\nabla_a|\psi_i>=\frac{<\psi_j|(\nabla_a H_e)|\psi_i>}{E_i-E_j}$$

The smaller splitting of adiabatic PES, the bigger non-BOA coupling!!!

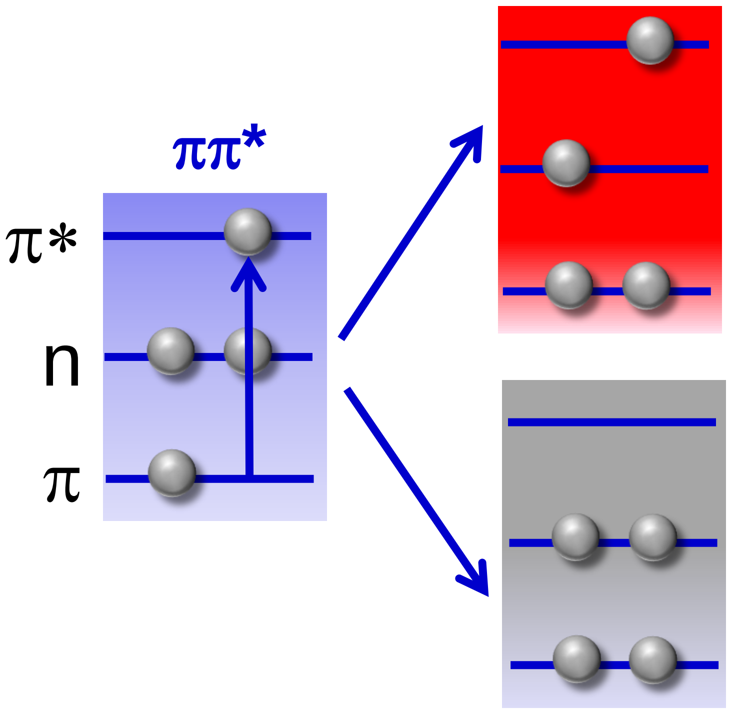
ΔE

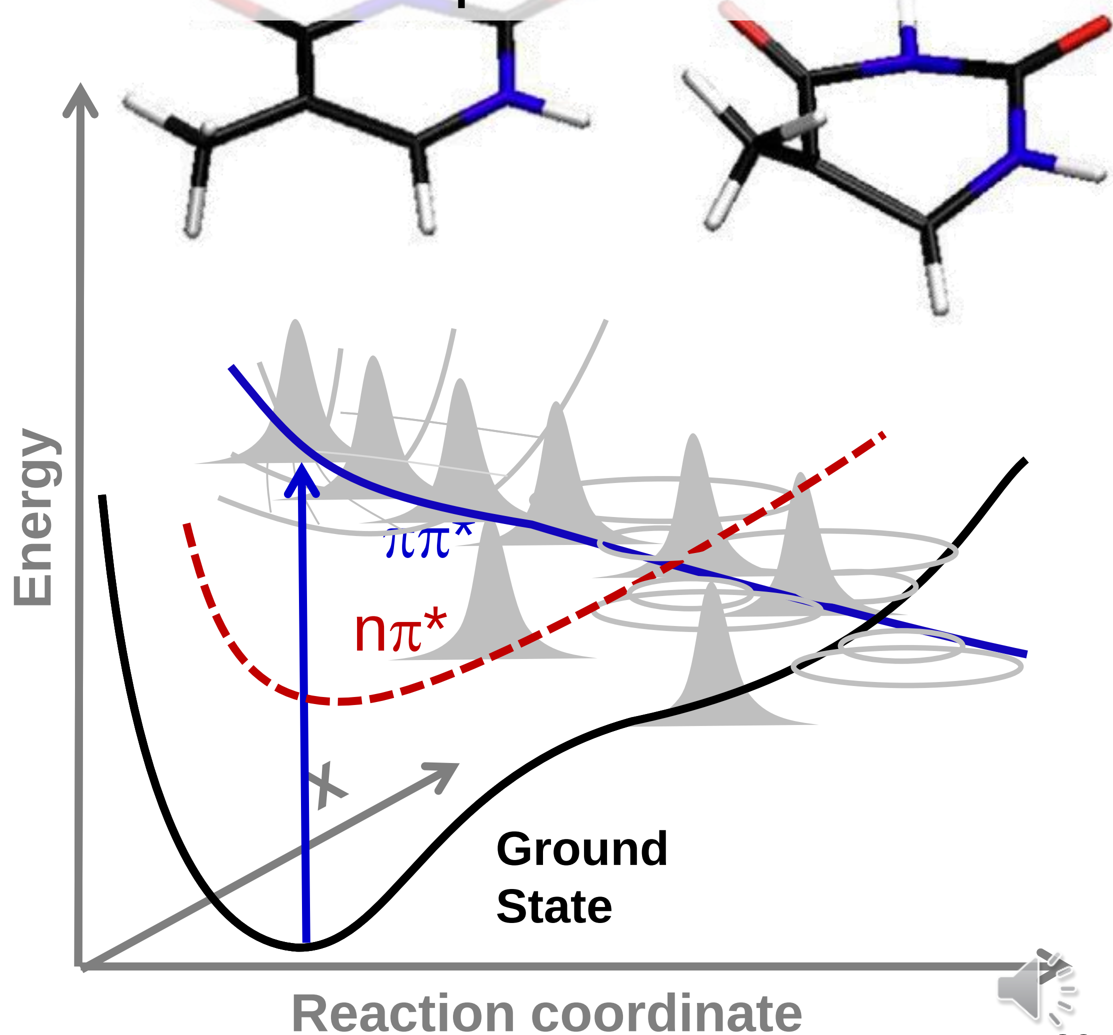


A real challenge for theory.

15 nuclei and 30 valence electons are able to move in 3d, and are coupled!

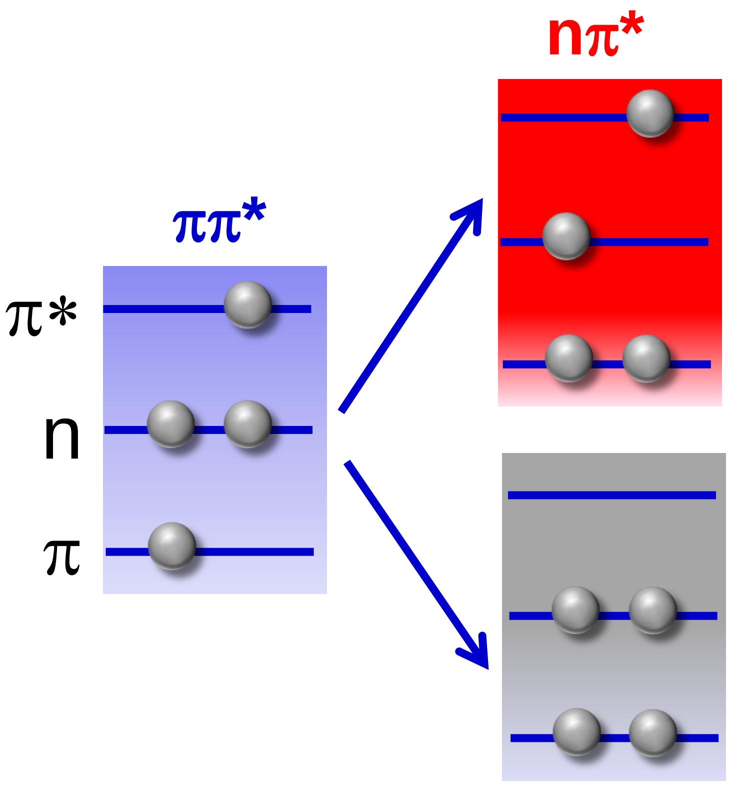
This needs serious approximations - which needs experimental tests!



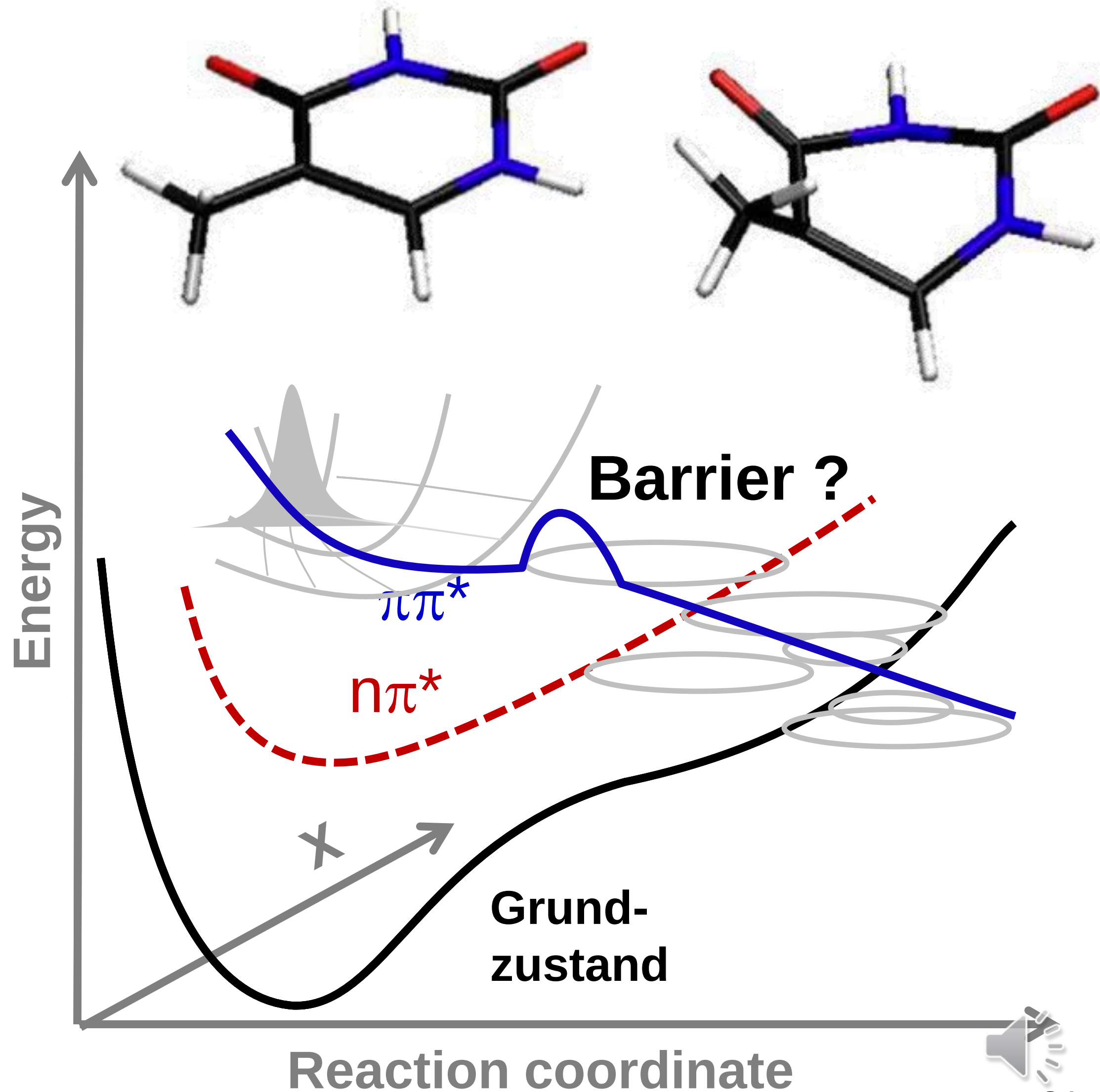


Two sides of the problem:

Electronic structure



Nuclear geometry



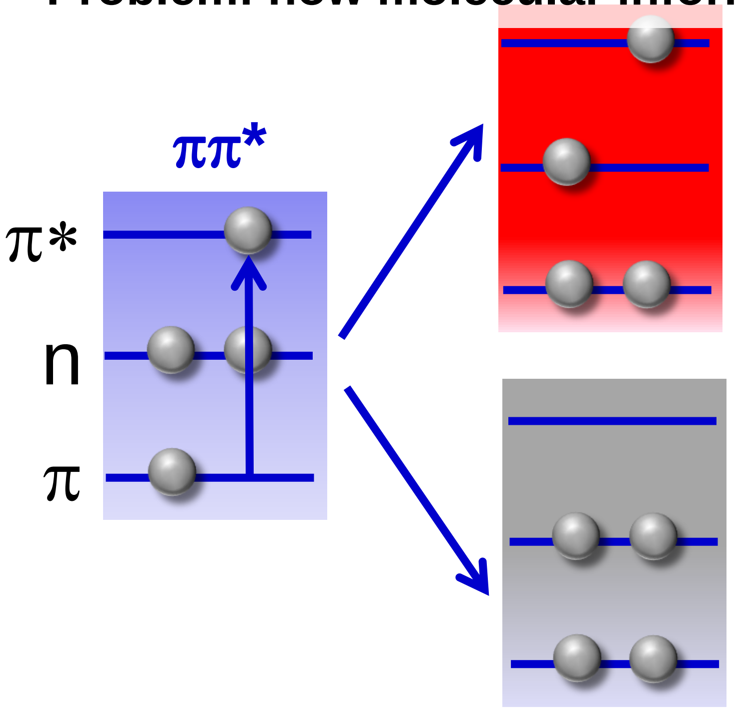
Asturiol et al.,
J. Phys. Chem. A,**113**, 10211 (2009)
Hudock et al.,
J. Phys. Chem. A,**111**, 85 (2007)

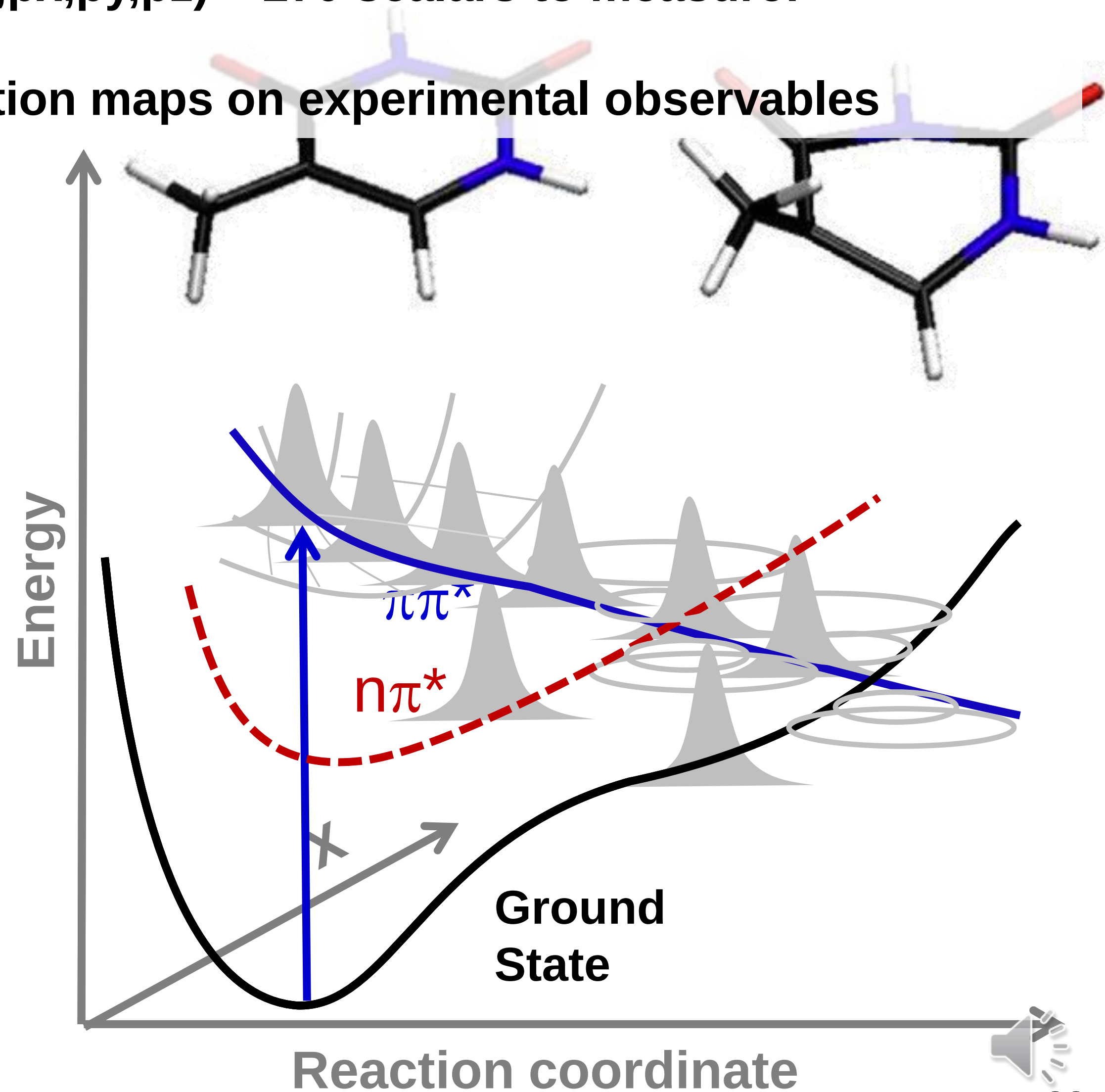
A real challenge for experiments too!

15 nuclei and 30 valence electons are able to move in 3d, and are coupled!

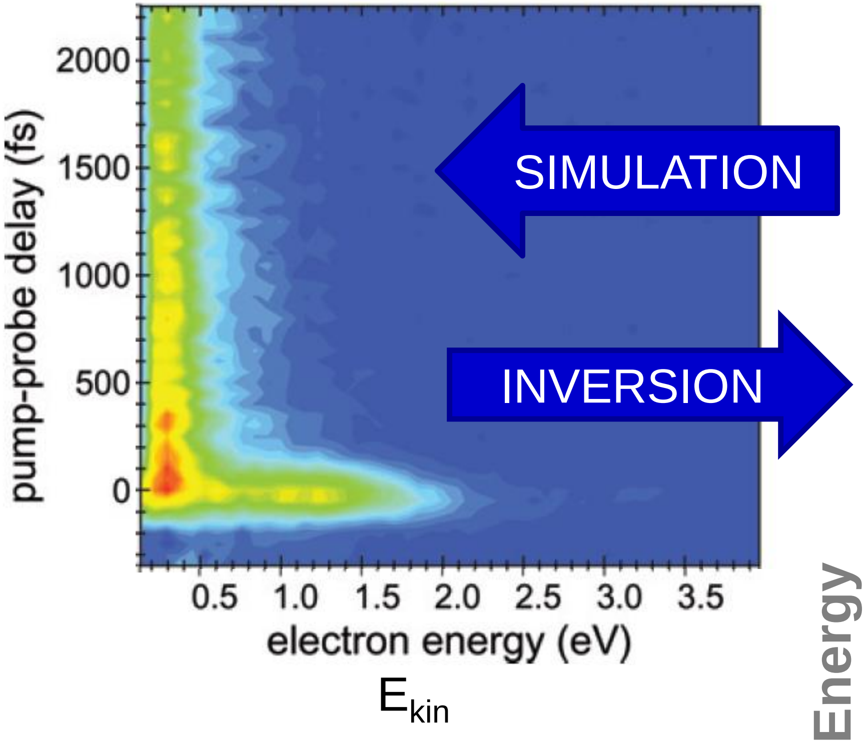
each particle has 6 scalars (x,y,z,px,py,pz) – 270 scalars to measure!

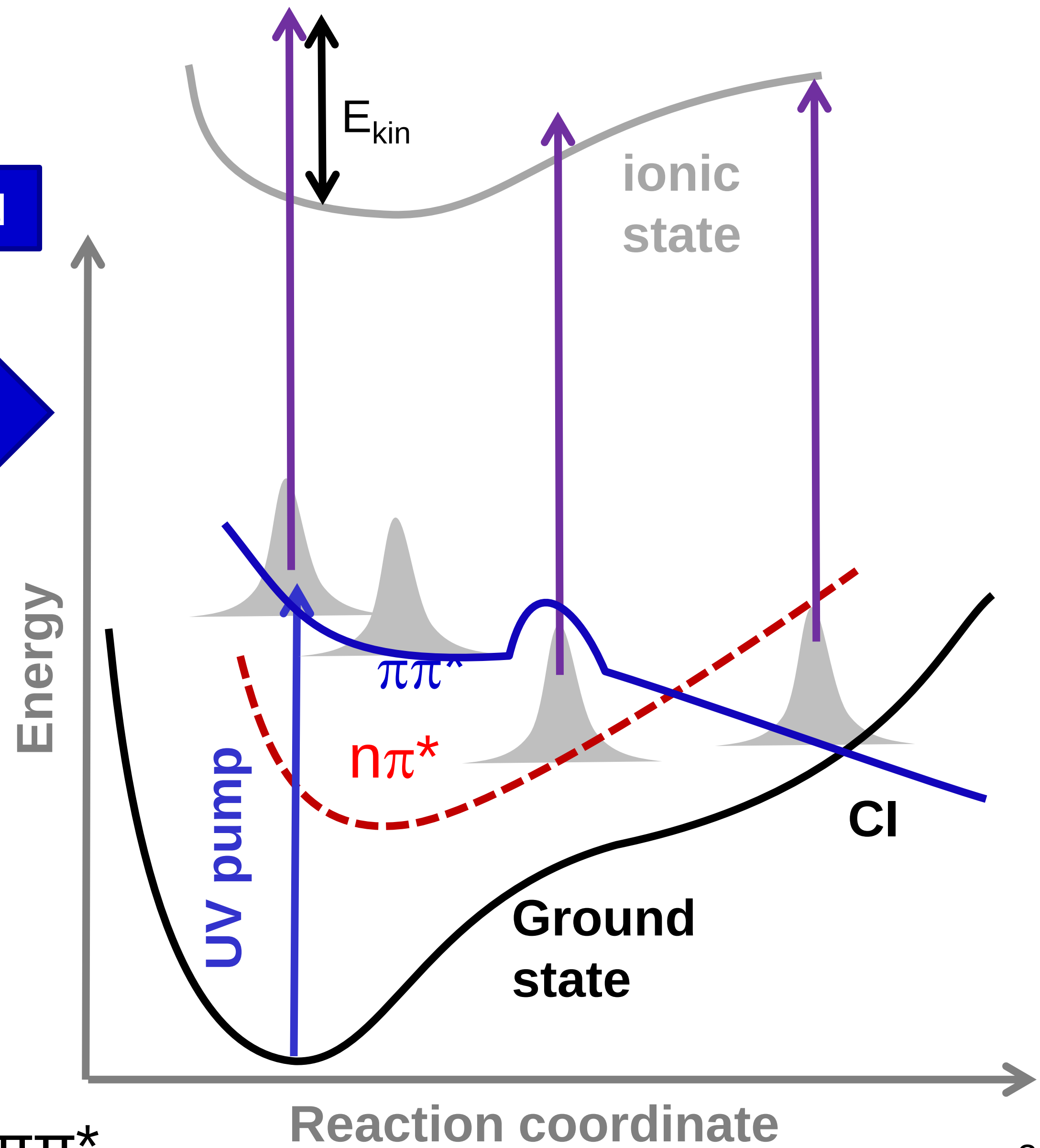
Problem: how molecular information maps on experimental observables





Observable inversion is impossible!





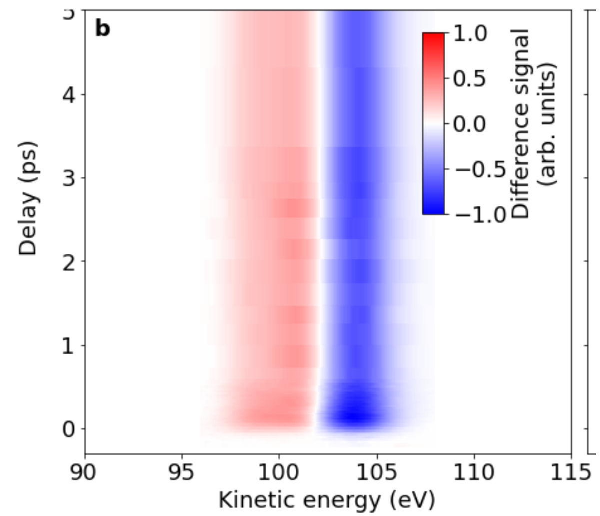
Ullrich et al. PCCP **6**, 2796 (2004)
Asturiol et al.,
J. Phys. Chem. A,**113**, 10211 (2009)
Hudock et al.,
J. Phys. Chem. A,**111**, 85 (2007)

Few picosecond lifetime of ππ*

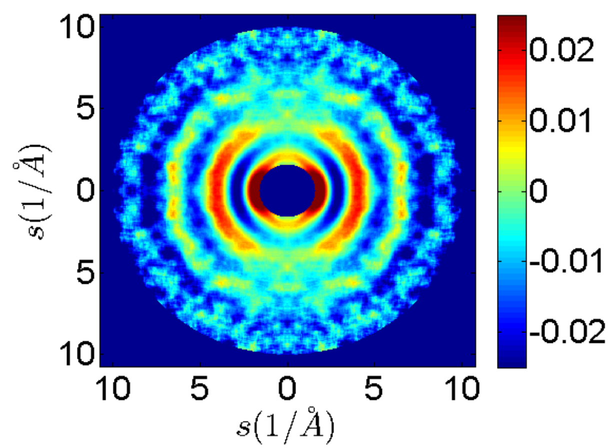
The more *different* observables, the easier it is to find something out about a molecule.

From the book cover of:
‚Gödel, Escher, Bach‘ by R. Hofstadter
20th anniversary edition, Perseus Books 1999

Ultrafast X-ray spectroscopy



Ultafast Hard x-ray/electron diffraction

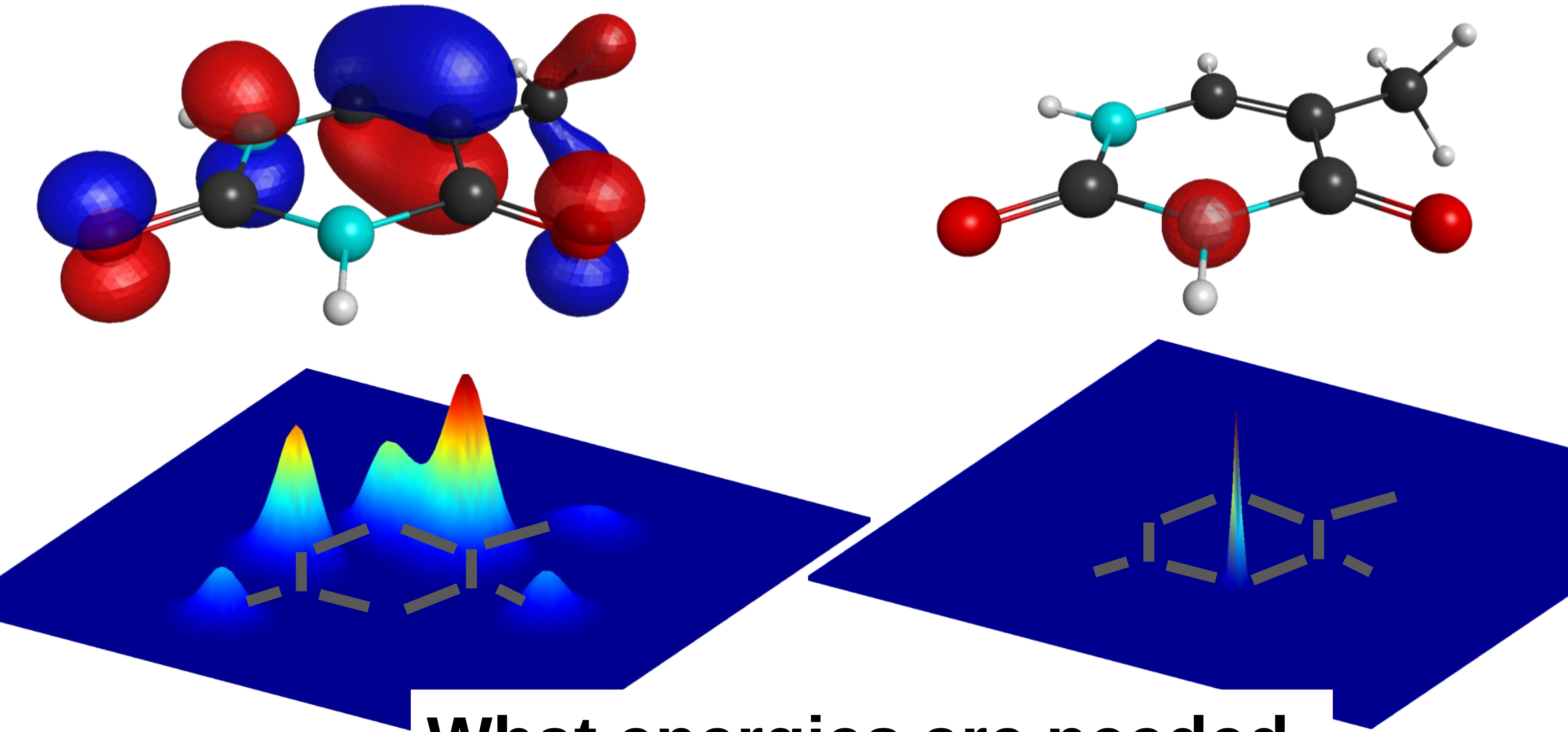


Why x-rays?

a) Local electronic structure and nuclear geometry

Delocalized valence orbital

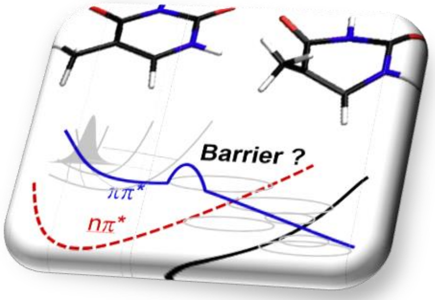
Localized core orbital

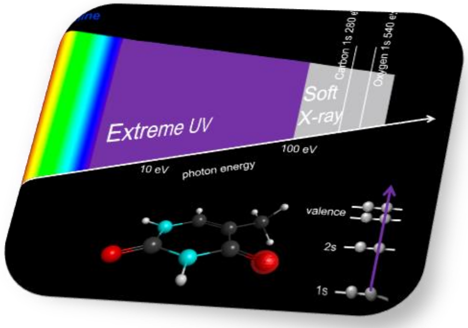


What energies are needed to talk to core electrons?

Outline

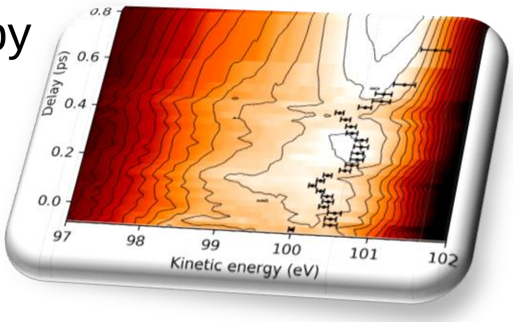
Basics: Coupled electronic and nuclear dynamics in molecules





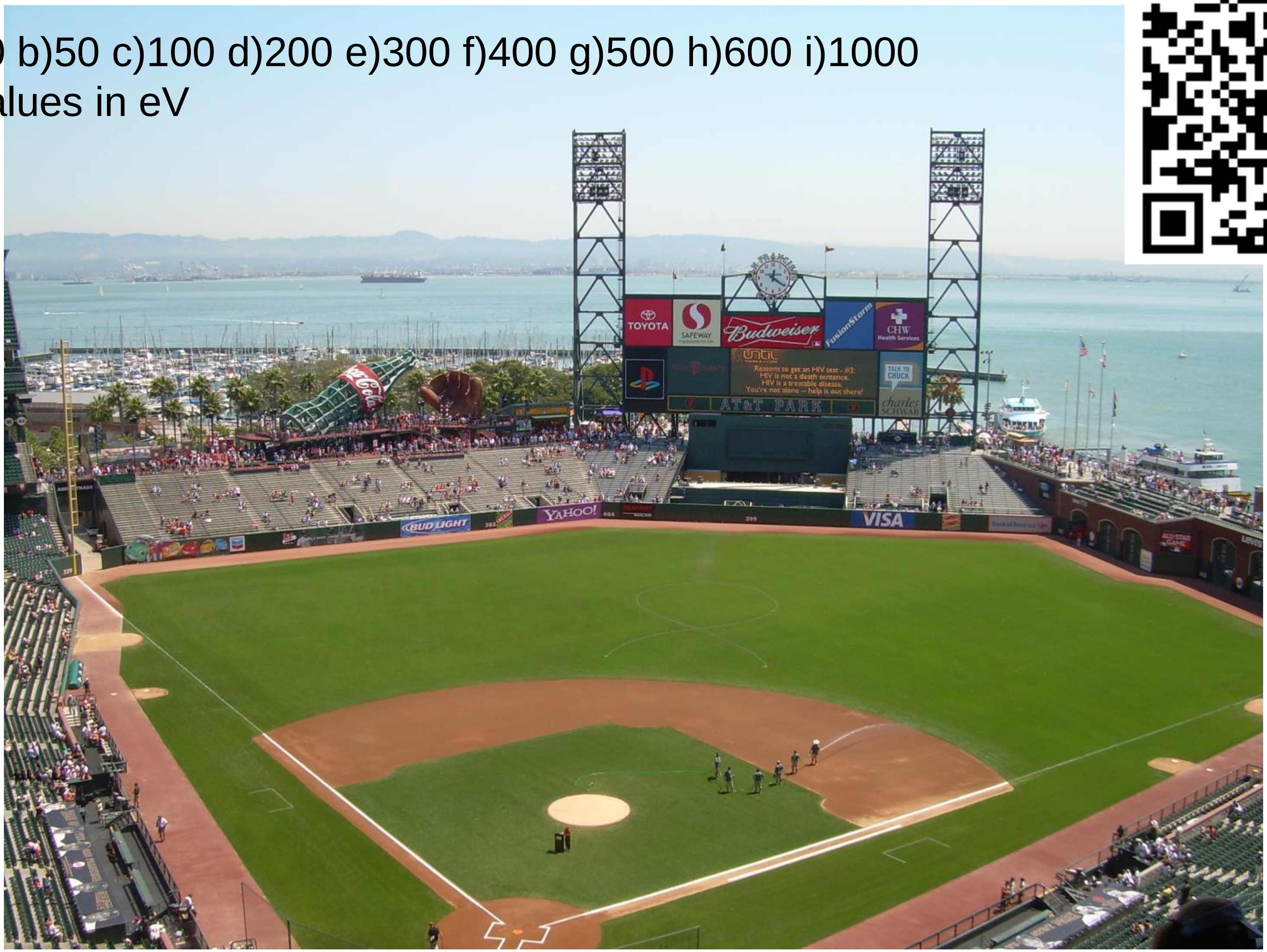
X-ray matter interaction

Two examples for ultrafast x-ray spectroscopy
resonant absorption
photoelectron spectroscopy



Quiz: Binding energy of a 1s carbon – just ballpark

10 b)50 c)100 d)200 e)300 f)400 g)500 h)600 i)1000
Values in eV

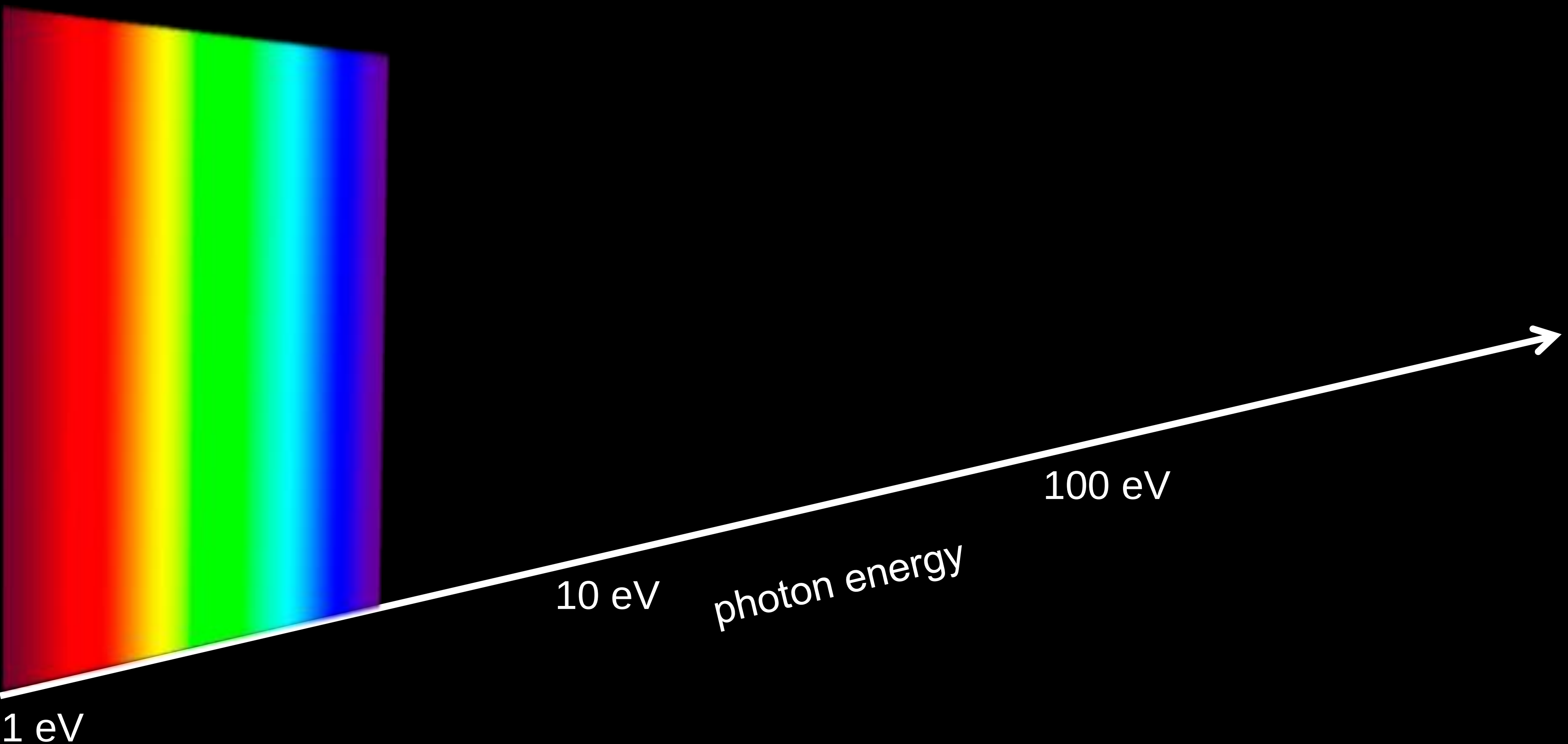


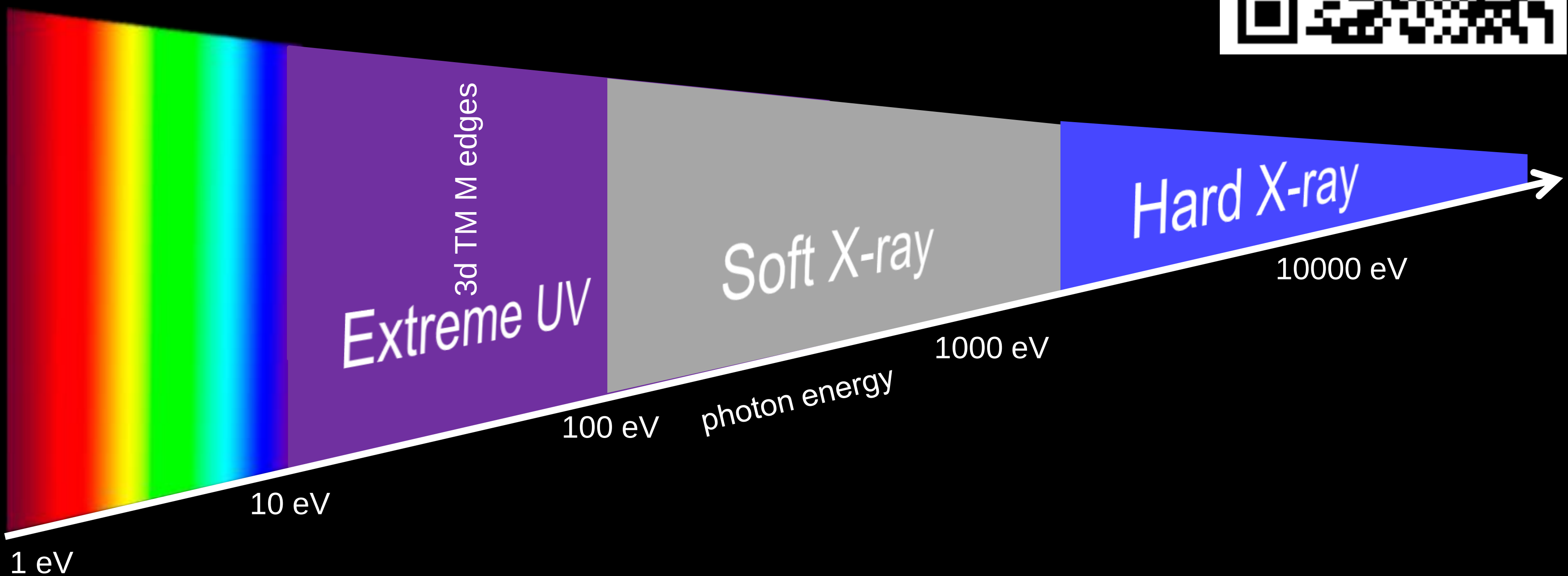
Quiz: Binding energy of a 1s carbon – just ballpark

E_n=13.6 [eV] Z^2 $(1/n^2)$ for H, He^+, Li^{++}, Be^{3+}, B^{4+}, C^{5+}

Binding energies in eV

He+	54.4	He	24.6
Li++	122.4	Li	54.7
Be3+	217.6	Be	111.5
B4+	340	B	188
C5+	489	C	284


100 eV
10 eV
photon energy
1 eV

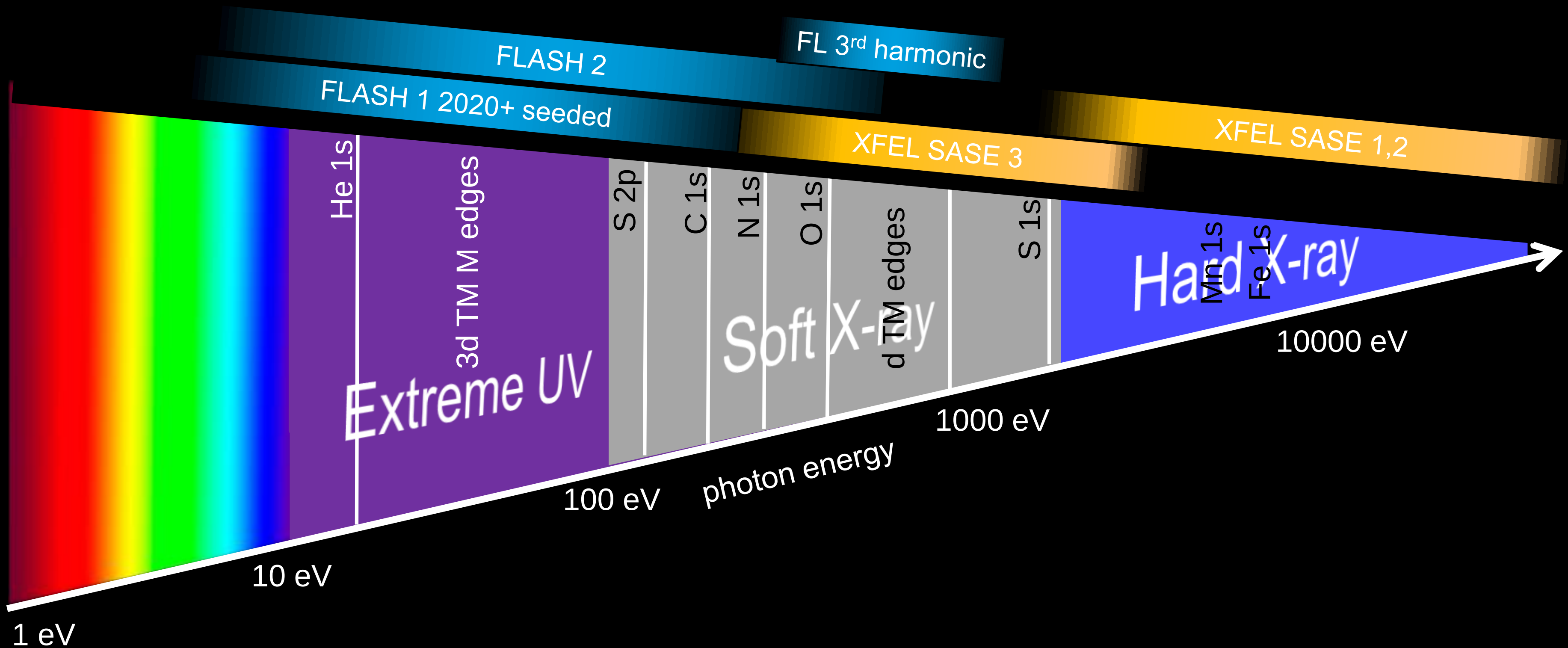


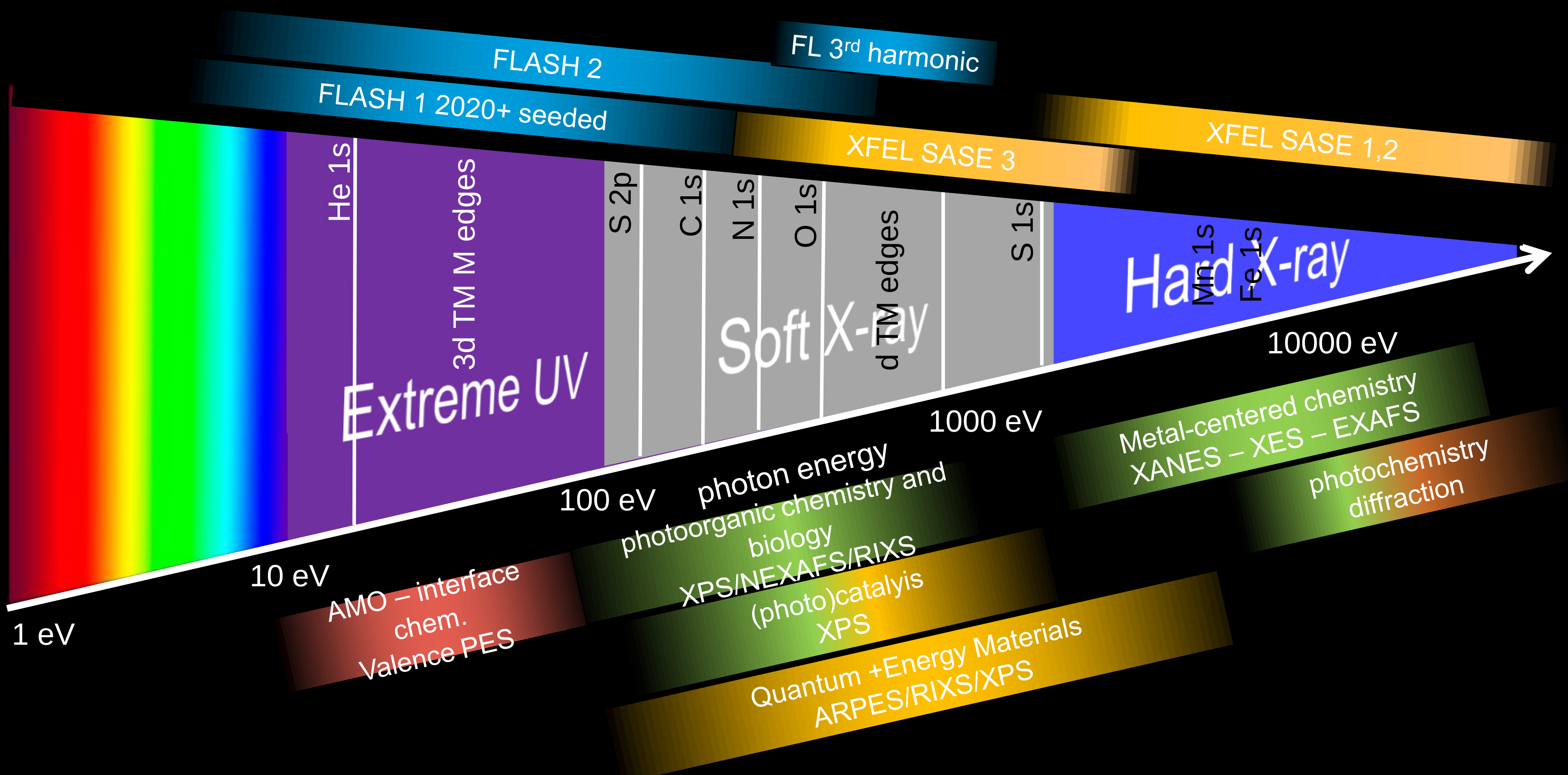
Please….order the 1s binding energies $E_{bind,1s}$ of Nitrogen, Oxygen and Carbon

a) $E_{bind,1s}$ Carbon > $E_{bind,1s}$ Nitrogen > $E_{bind,1s}$ Oxygen

b) $E_{bind,1s}$ Oxygen > $E_{bind,1s}$ Nitrogen > $E_{bind,1s}$ Carbon

c) $E_{bind,1s}$ Nitrogen > $E_{bind,1s}$ Carbon > $E_{bind,1s}$ Oxygen

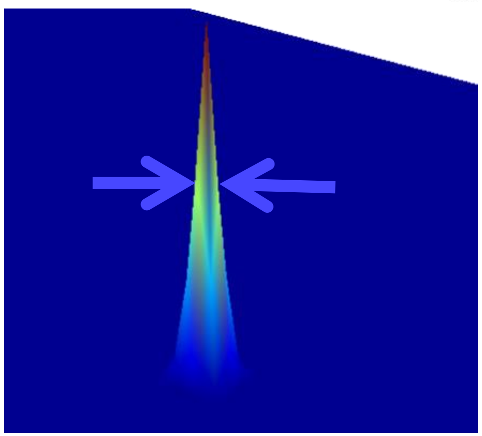

FLASH 2
FL 3rd harmonic
FLASH 1 2020+ seeded
XFEL SASE 3
XFEL SASE 1,2
He 1s
3d TM M edges
S 2p
C 1s
N 1s
O 1s
d TM edges
S 1s
Mn 1s
Fe 1s
Extreme UV
Soft X-ray
Hard X-ray
1 eV
10 eV
100 eV
photon energy
1000 eV
10000 eV

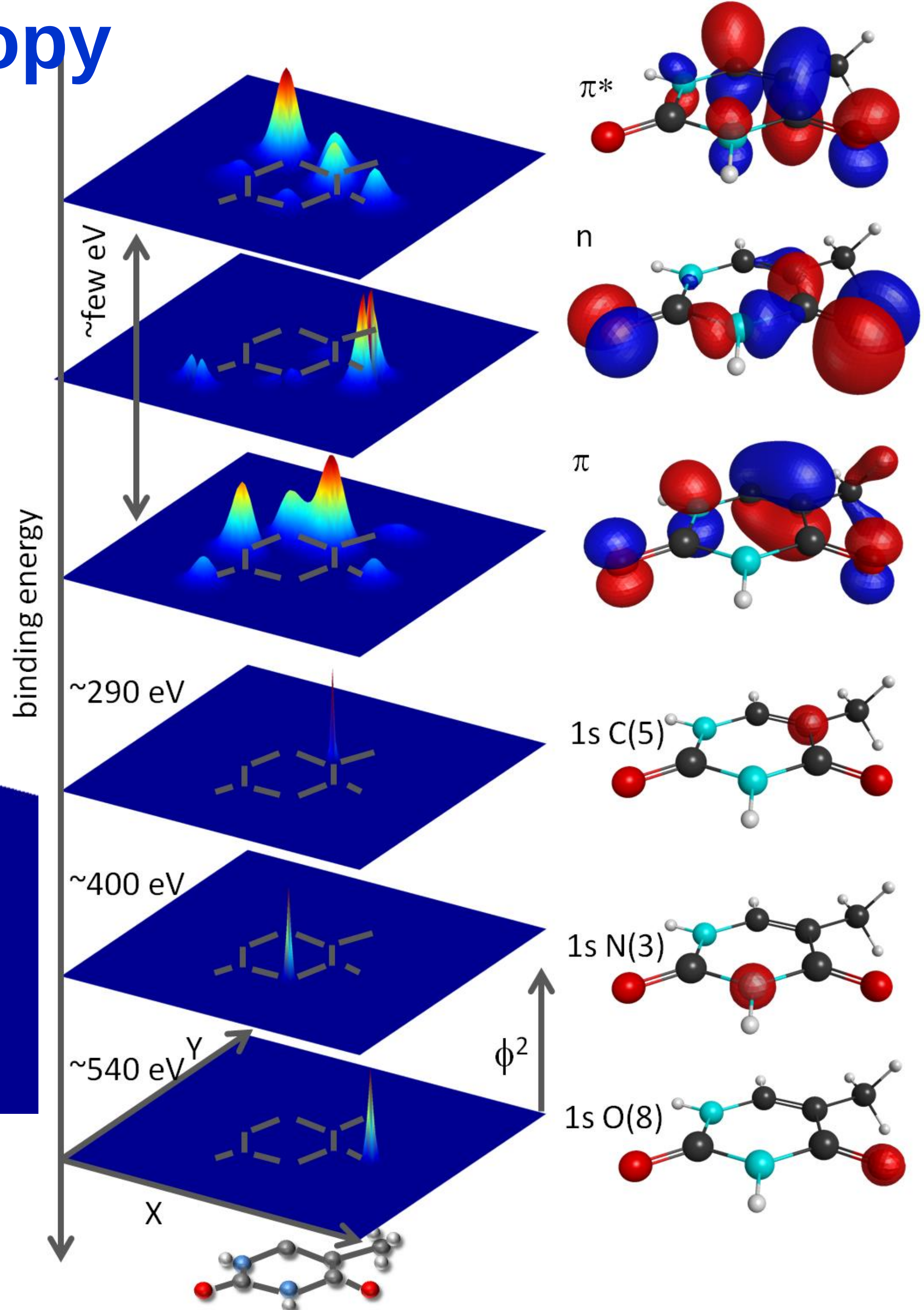

FLASH 2
FL 3rd harmonic
FLASH 1 2020+ seeded
XFEL SASE 3
XFEL SASE 1,2
He 1s
3d TM M edges
S 2p
C 1s
N 1s
O 1s
d TM edges
S 1s
Mn 1s
Fe 1s
Extreme UV
Soft X-ray
Hard X-ray
1 eV
10 eV
100 eV
1000 eV
10000 eV
photon energy
AMO – interface chem. Valence PES
photoorganic chemistry and biology XPS/NEXAFS/RIXS
(photo)catalyis XPS
Quantum +Energy Materials ARPES/RIXS/XPS
Metal-centered chemistry XANES – XES – EXAFS
photochemistry diffraction

X-ray spectroscopy is element sensitive

X-ray spectroscopy is local

~10pm

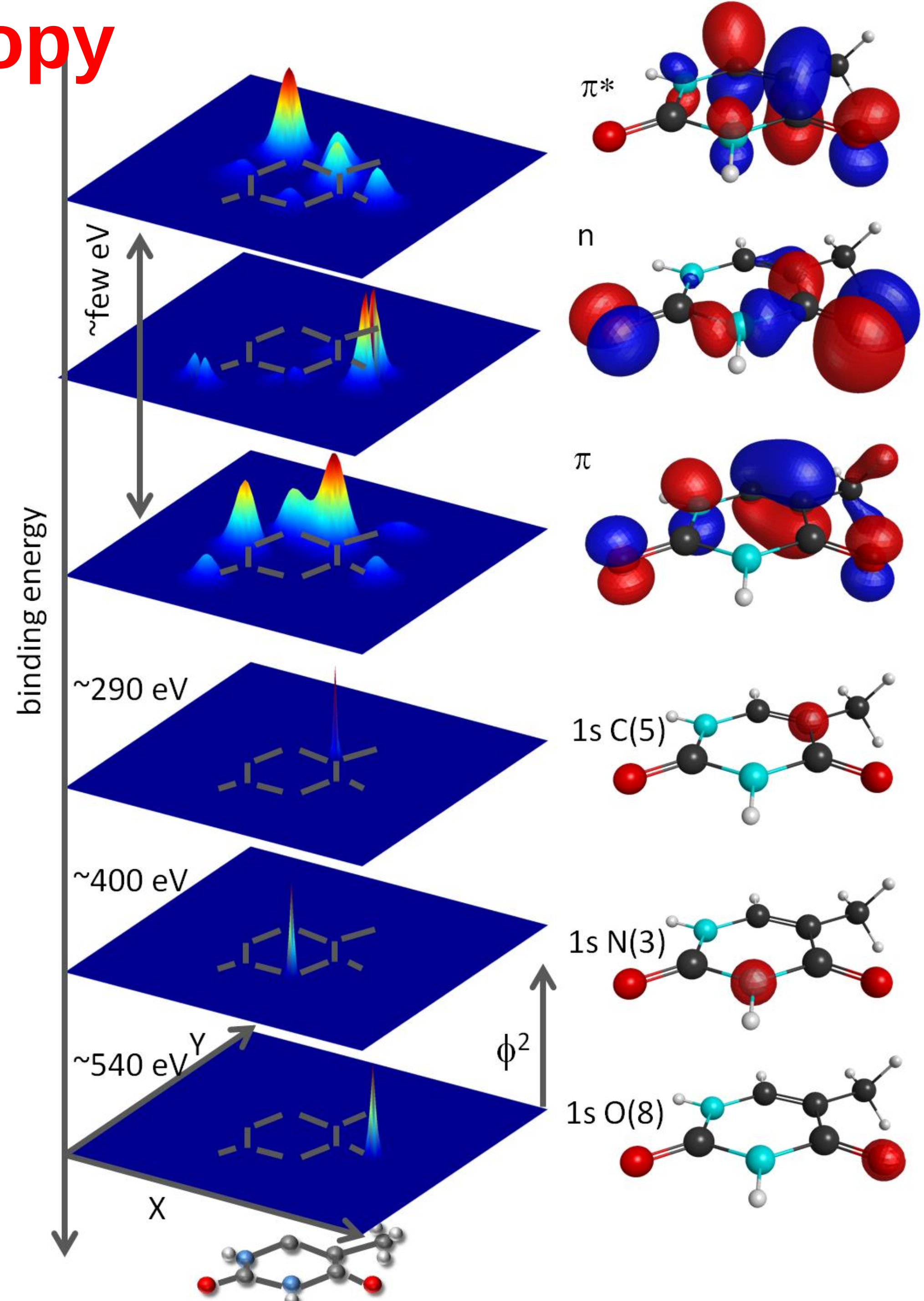




X-ray spectroscopy is local

Which empty valence orbital would yield the largest absorption?

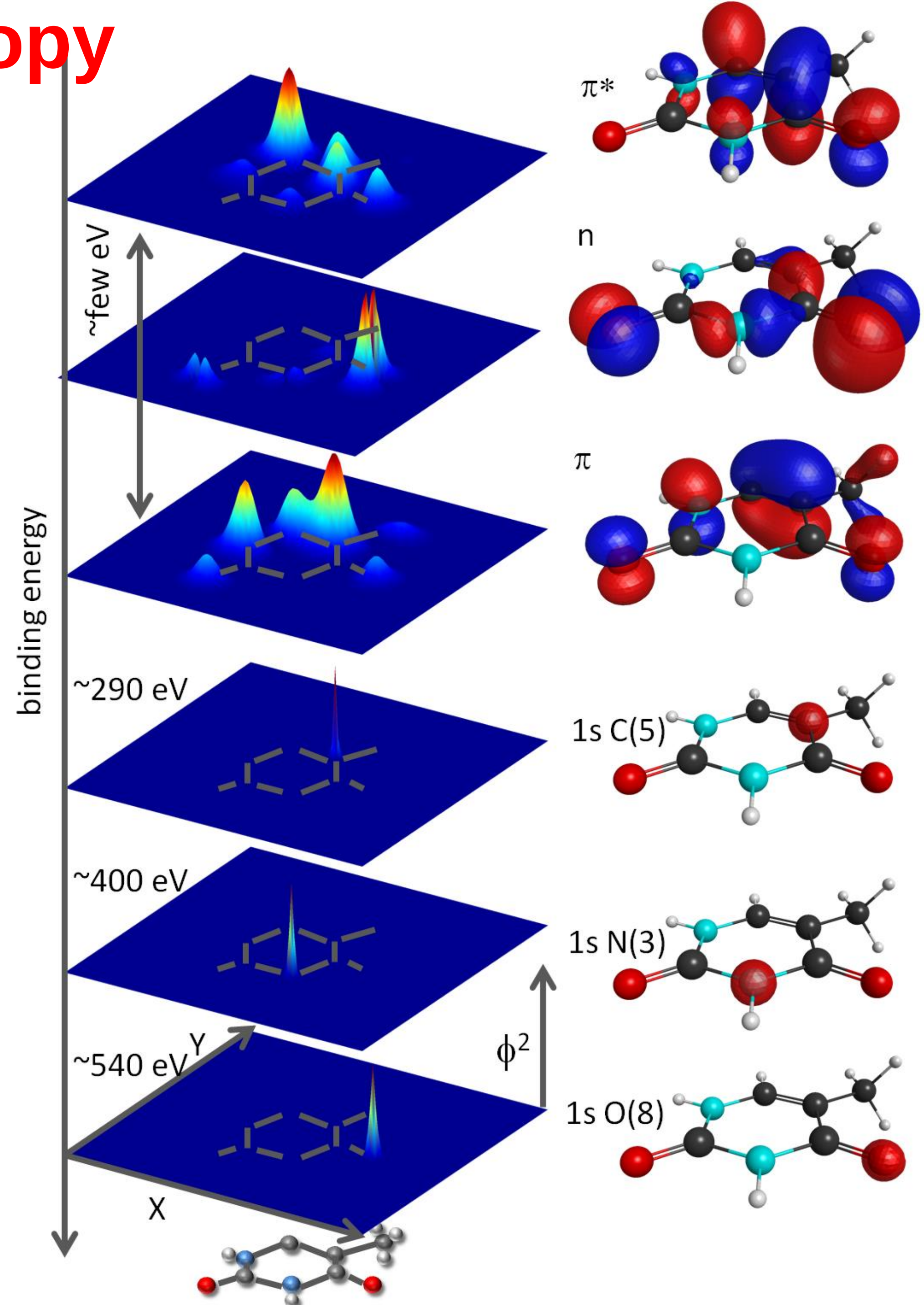
a) π*
b) n
c) π



X-ray spectroscopy is local

Which empty valence orbital would yield the largest absorption?

a) π*
b) n
c) π



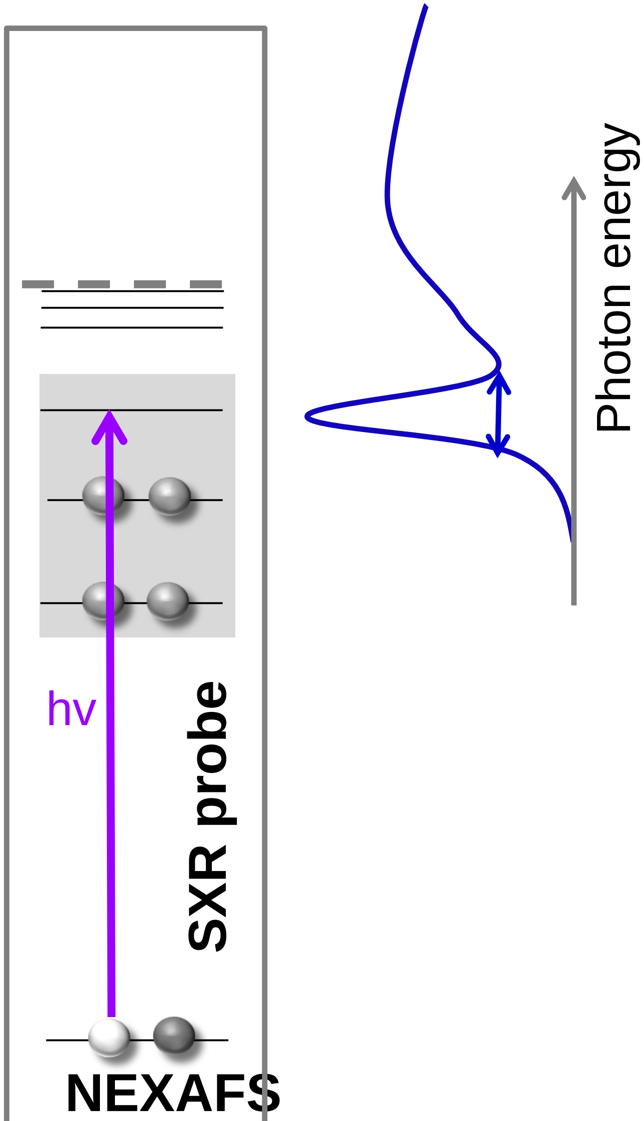
Beloved Children have many names

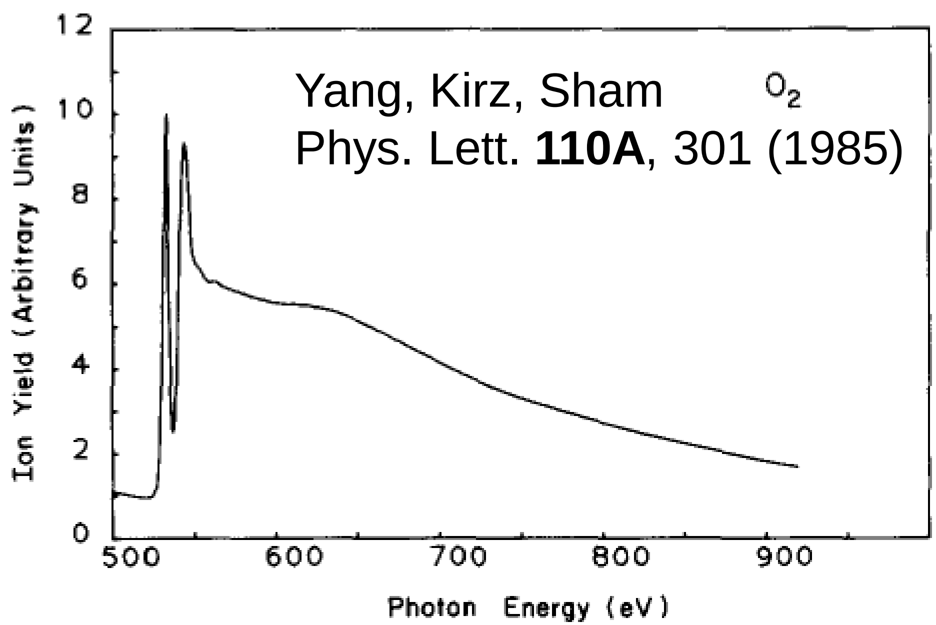
NEXAFS: Near Edge X-ray Absorption Fine Structure

XANES: X-ray Absorption Near Edge Structure

Linewidth ~1/core hole lifetime

few femtoseconds core hole lifetime
~100 meV linewidth

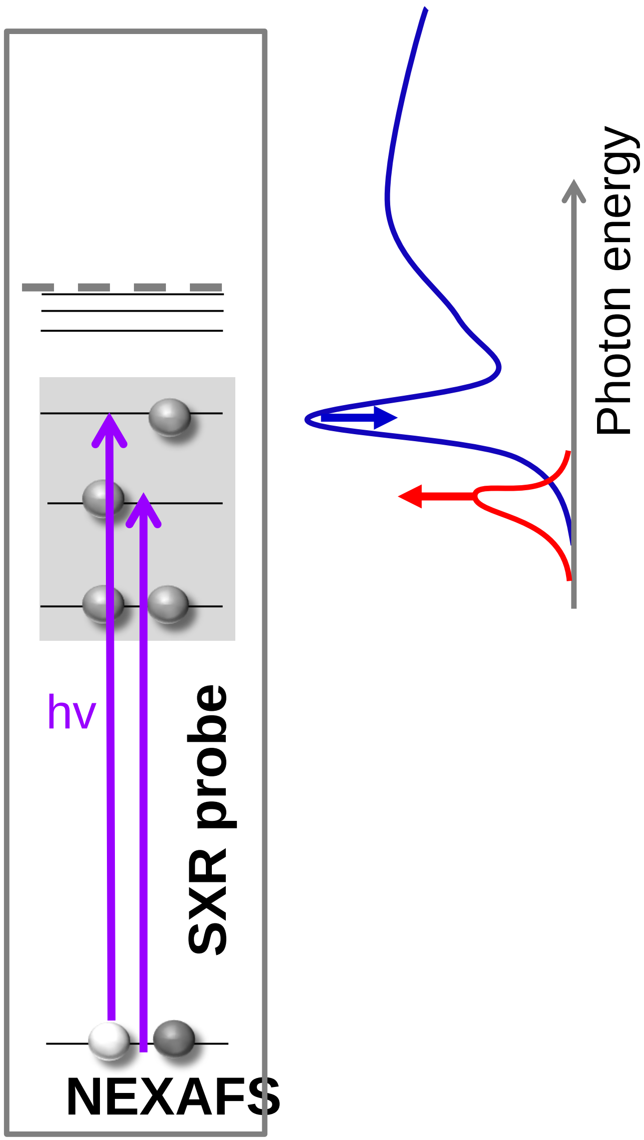


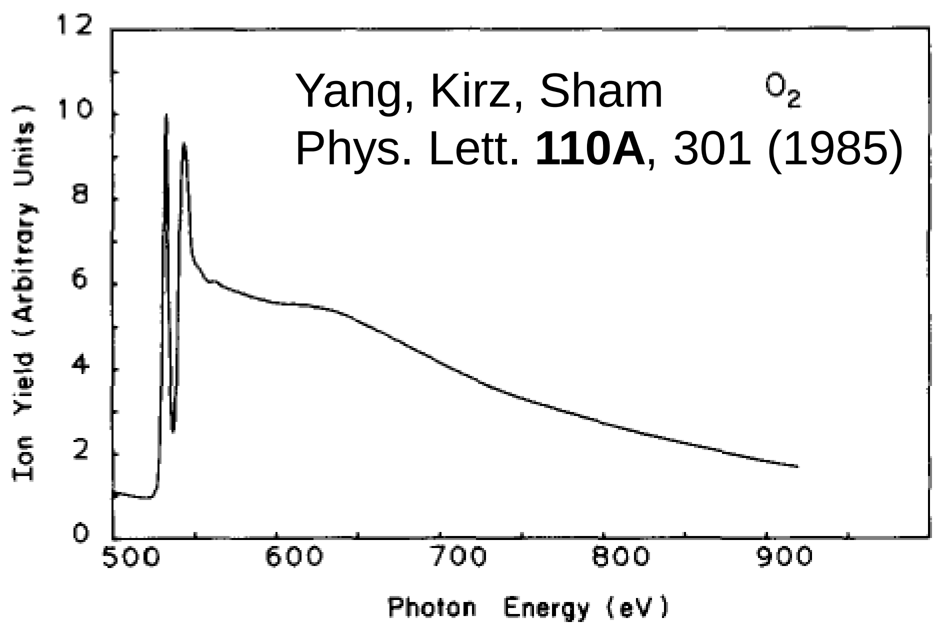


Absorption spectroscopy: NEXAFS

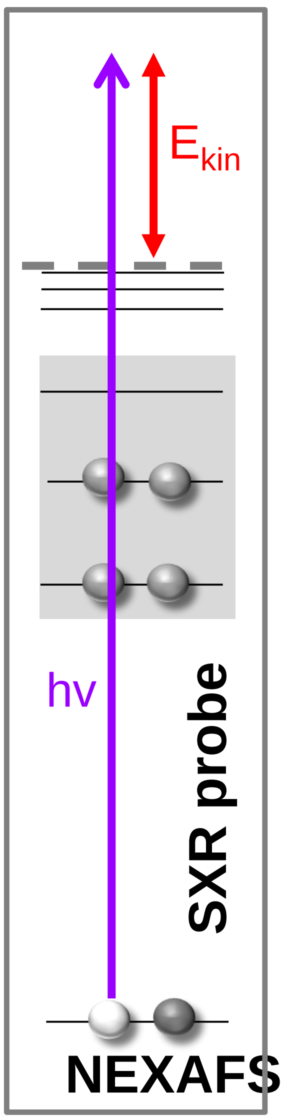
NEXAFS: Near Edge X-ray Absorption Fine Structure

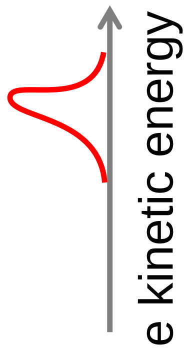
XANES: X-ray Absorption Near Edge Structure





X-ray photoemission (XPS)





Kinetic energy=photon energy-binding energy

A. Einstein

Bern, den 17. März 1905.

A deeper look: Site selectivity - chemical shift

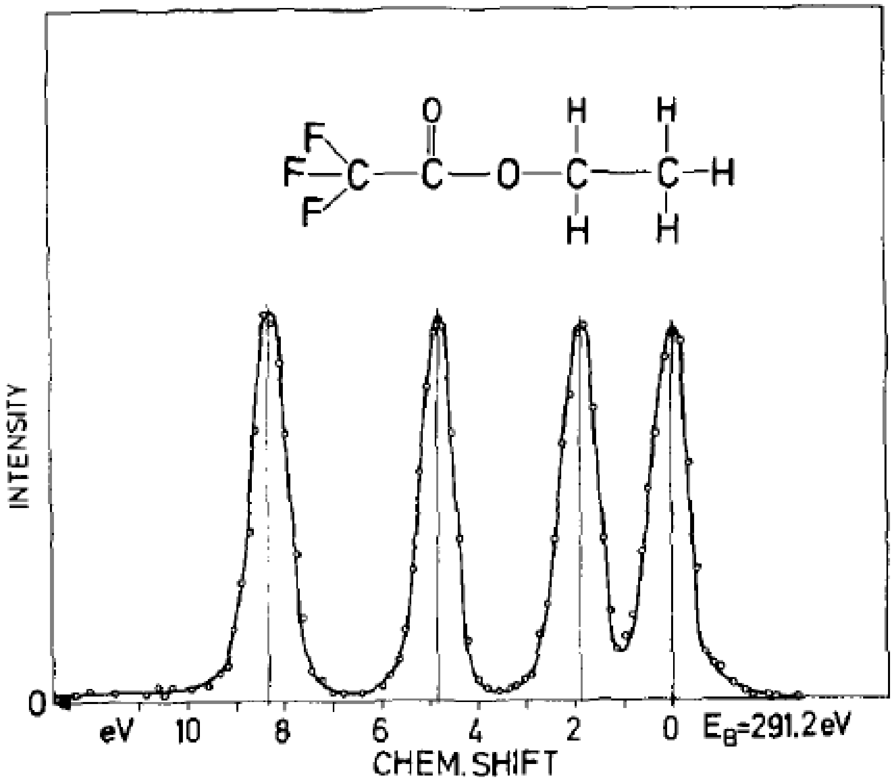
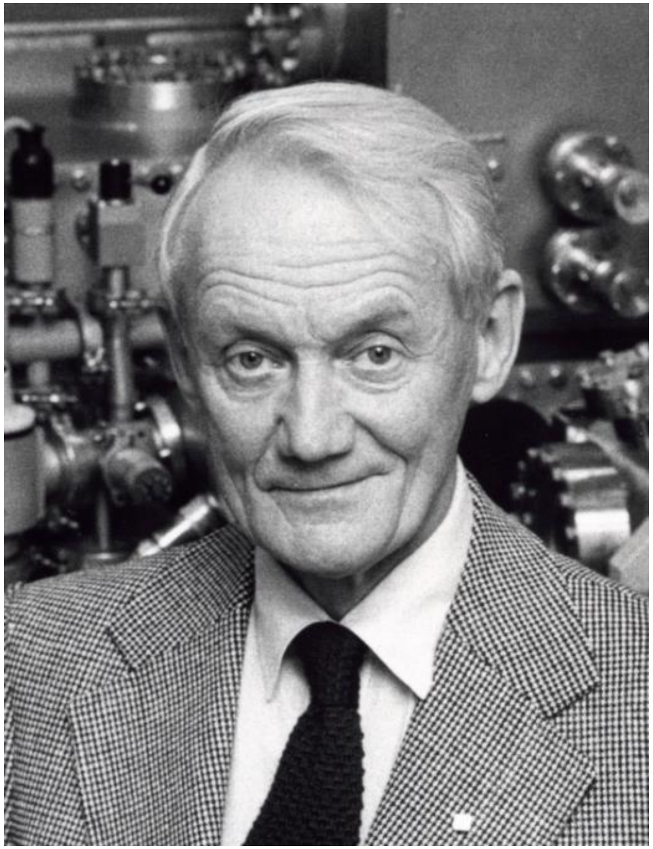


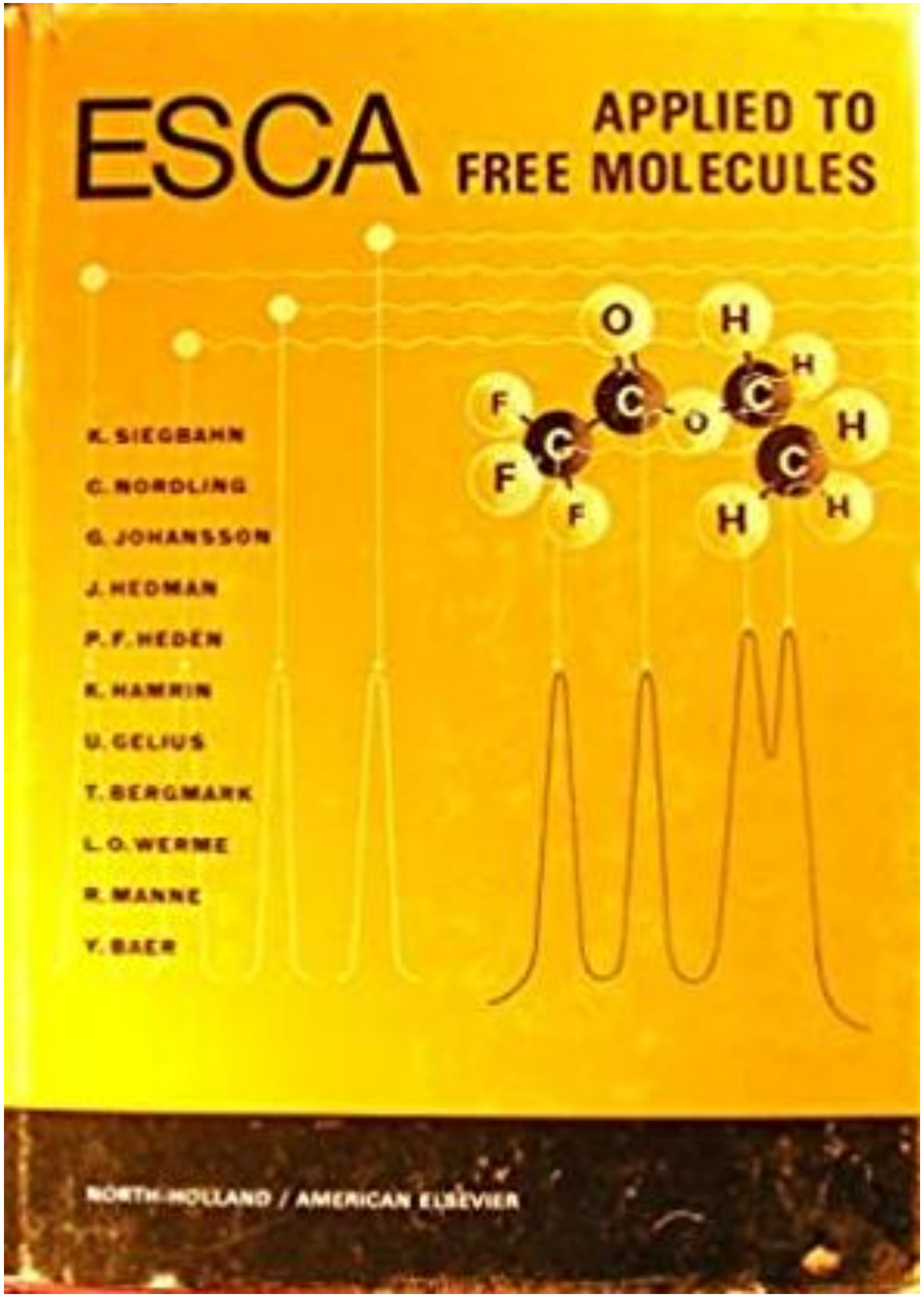
Figure 18. The carbon 1s electron lines in ethyl trifluoroacetate.



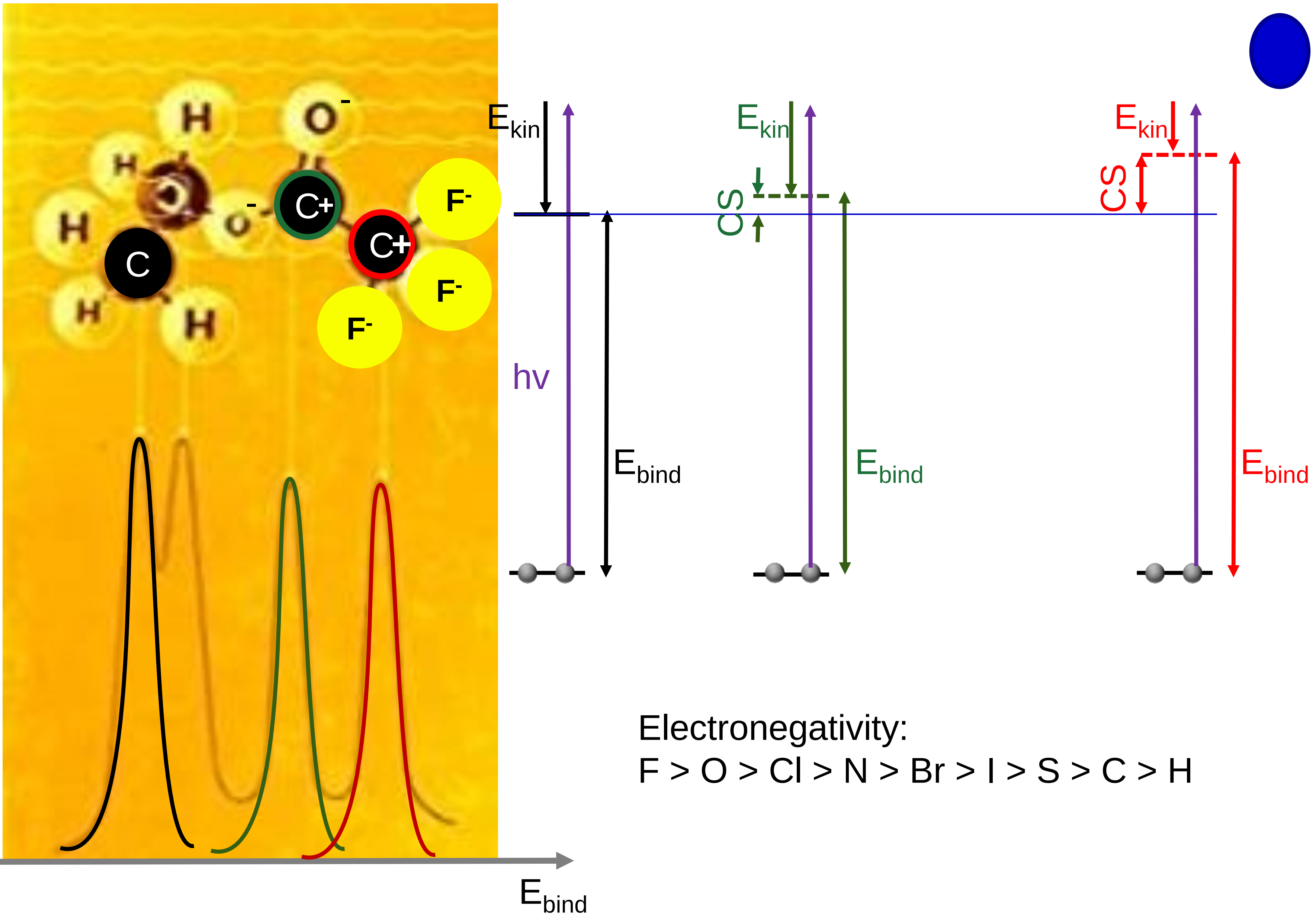
K. Siegbahn

Gelius *et al*, J. Electr. Spectr. Rel. Phen. **2**, 405 (1974)

Chemical shift and local charge

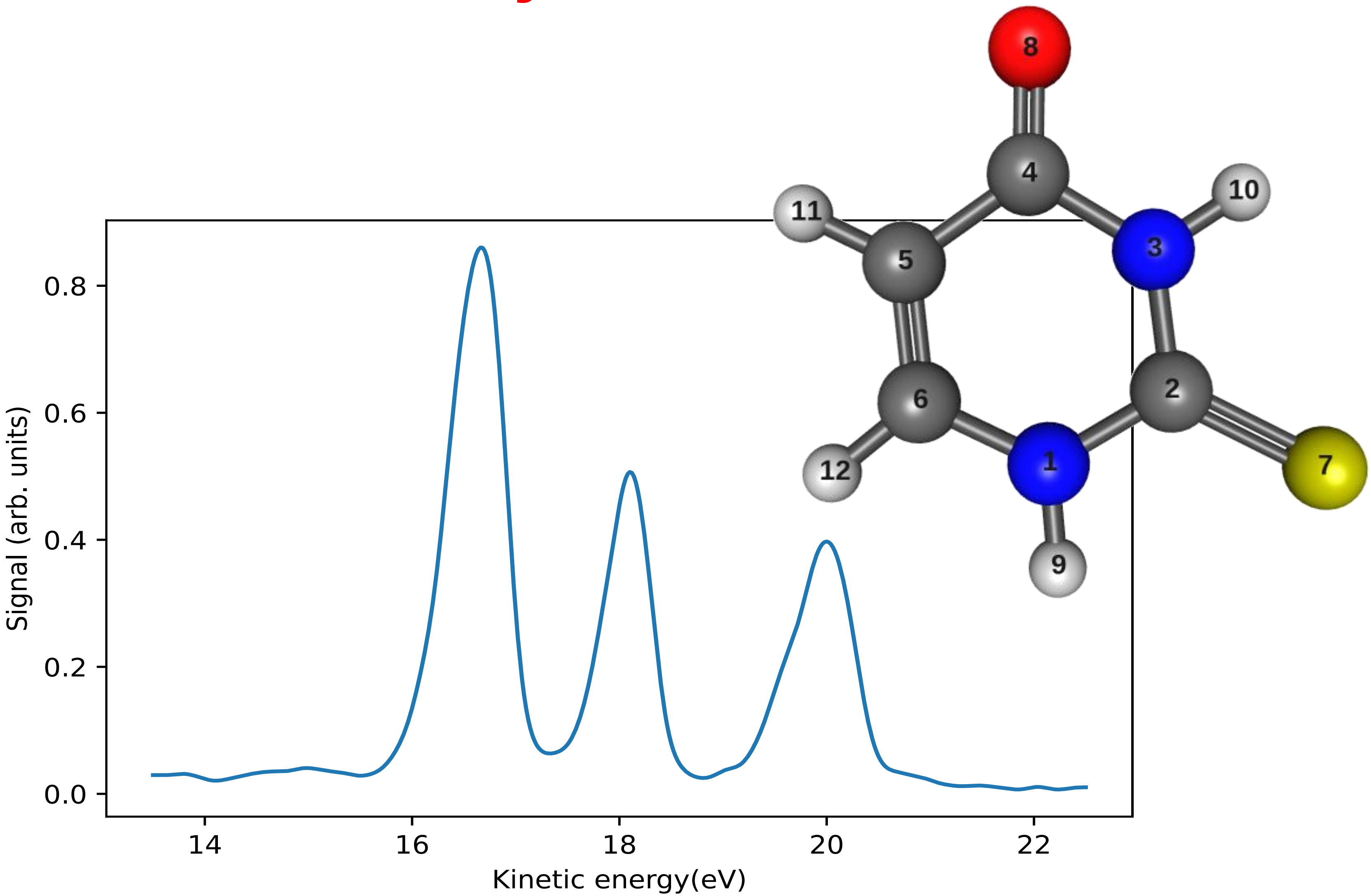


Chemical shift and local charge



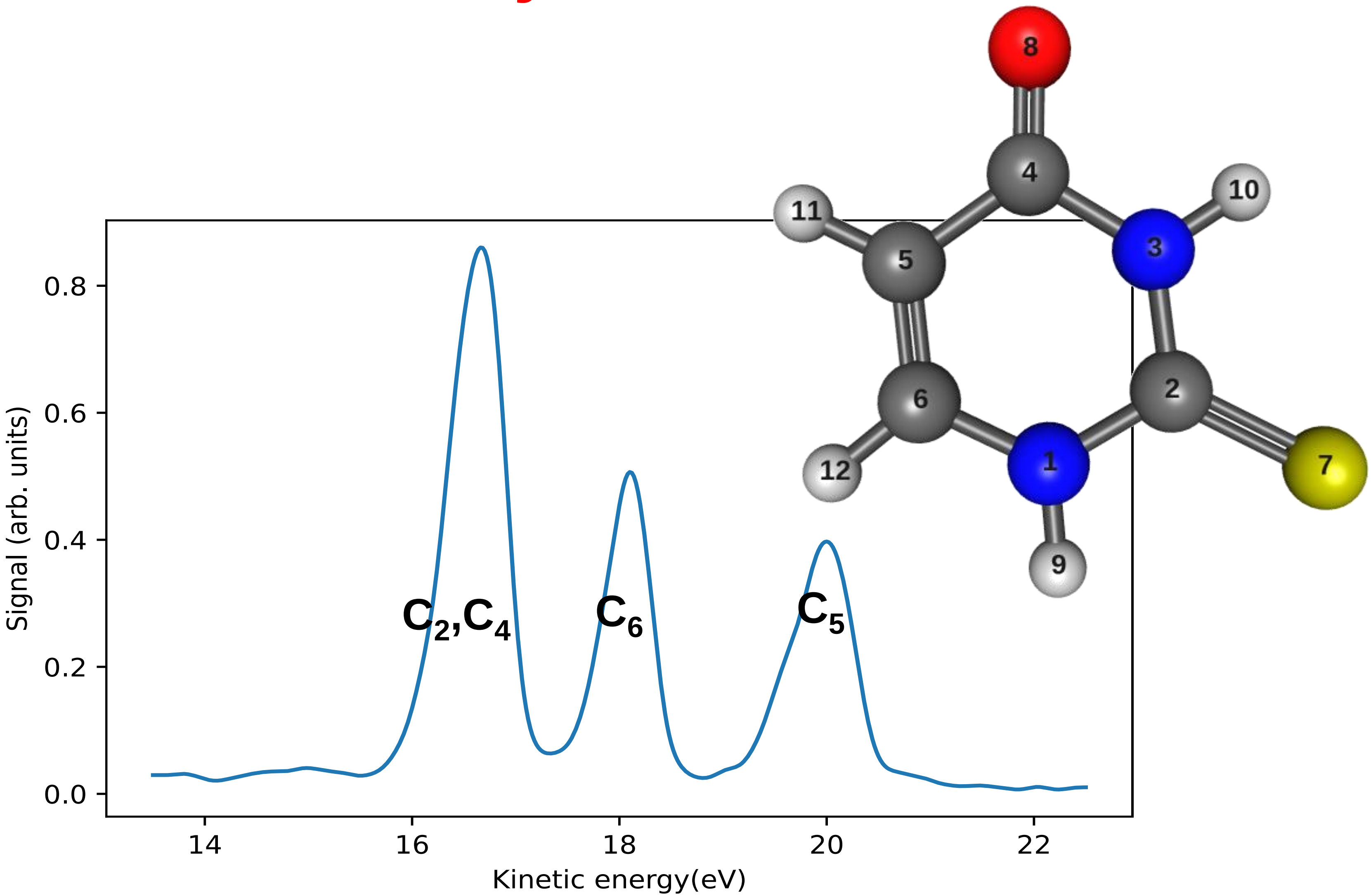
Electronegativity:
F > O > Cl > N > Br > I > S > C > H

Quiz: site sensitivity – chemical shift



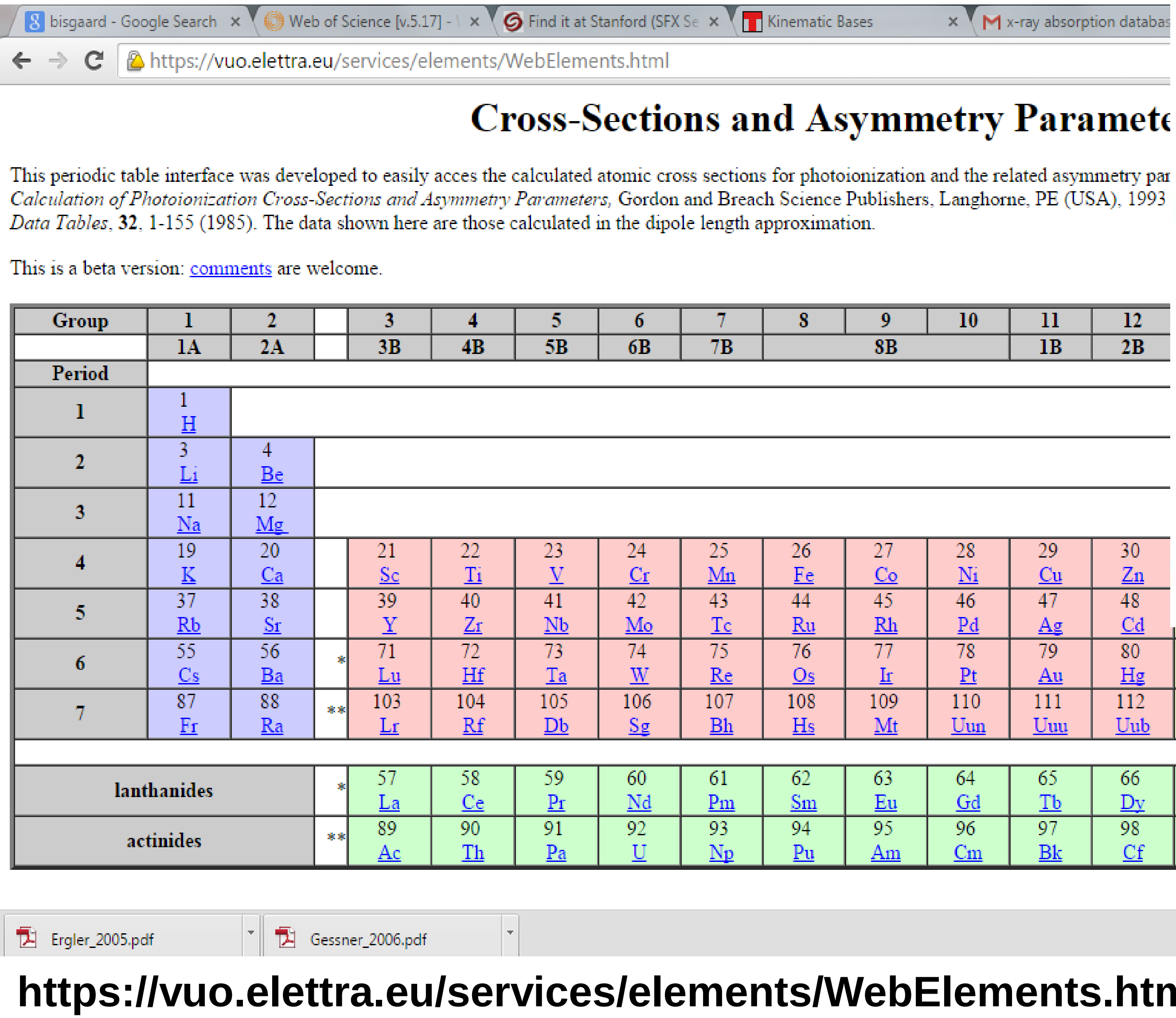
Electronegativity:
F > O > Cl > N > Br > I > S > C > H

Quiz: site sensitivity – chemical shift



Electronegativity:
F > O > Cl > N > Br > I > S > C > H

Absorption spectra – useful help

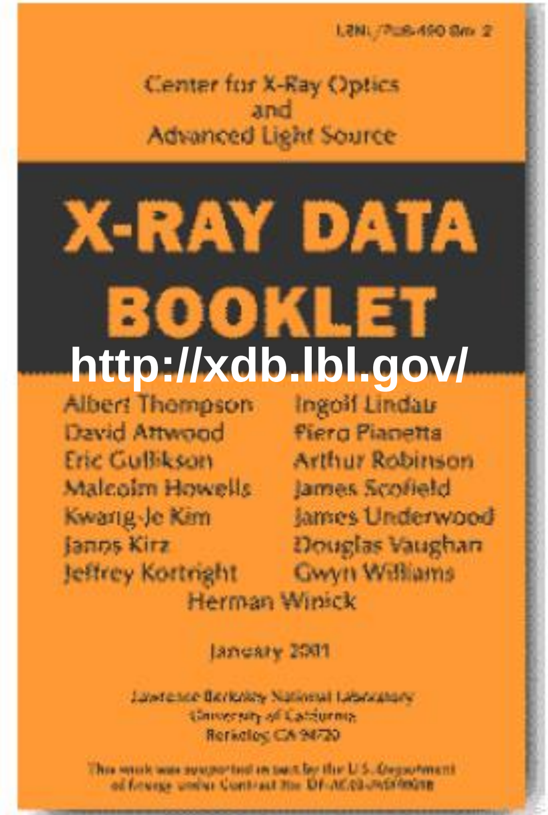


Cross-Sections and Asymmetry Paramete

This periodic table interface was developed to easily acces the calculated atomic cross sections for photoionization and the related asymmetry par
Calculation of Photoionization Cross-Sections and Asymmetry Parameters, Gordon and Breach Science Publishers, Langhorne, PE (USA), 1993
Data Tables, **32**, 1-155 (1985). The data shown here are those calculated in the dipole length approximation.

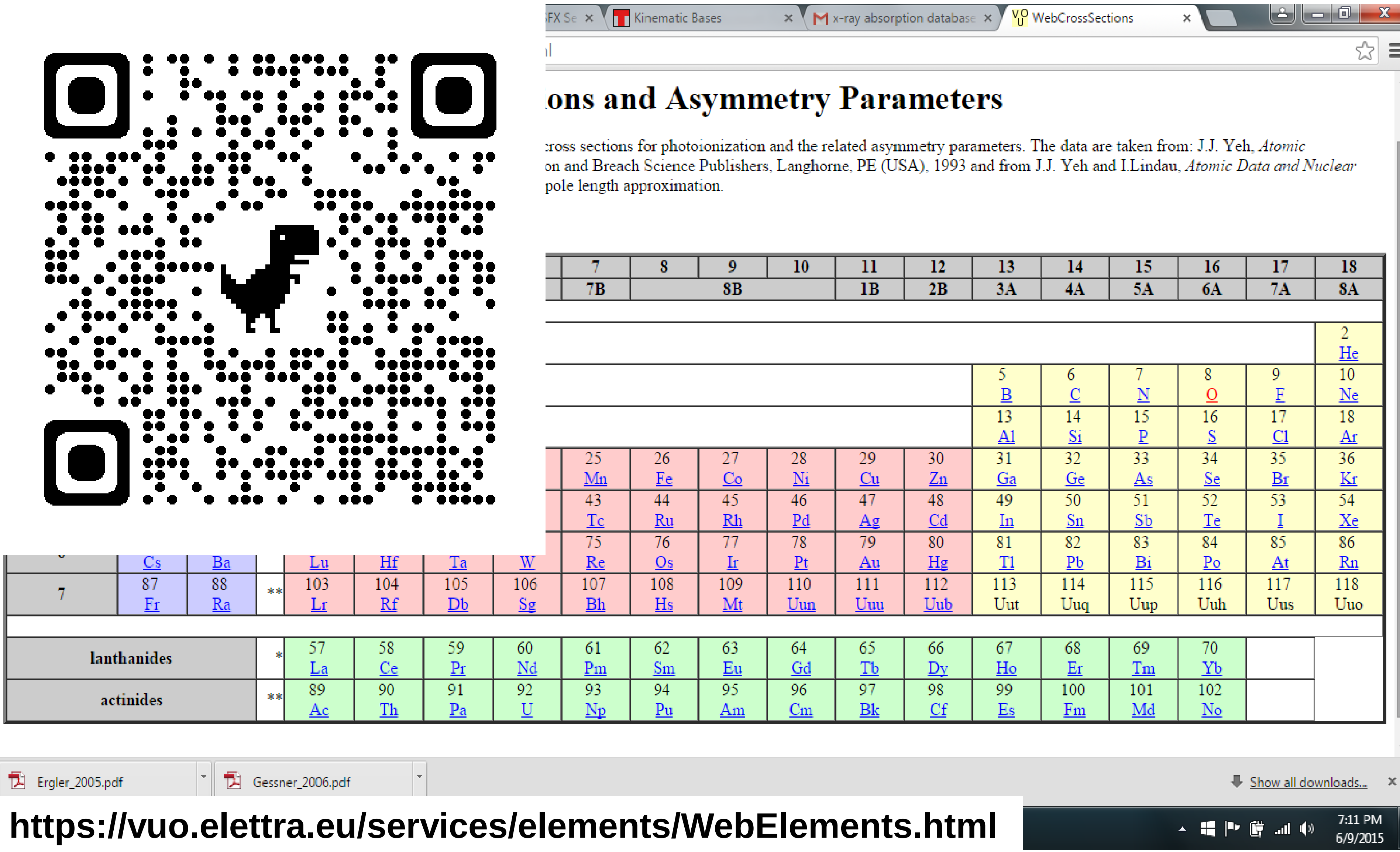
This is a beta version: comments are welcome.

Group	1	2		3	4	5	6	7	8	9	10	11	12						
	1A	2A		3B	4B	5B	6B	7B		8B		1B	2B						
Period																			
1	1 H																		
2	3 Li	4 Be																	
3	11 Na	12 Mg																	
4	19 K	20 Ca		21 Sc	22 Ti	23 V	24 Cr	25 Mn	26 Fe	27 Co	28 Ni	29 Cu	30 Zn						
5	37 Rb	38 Sr		39 Y	40 Zr	41 Nb	42 Mo	43 Tc	44 Ru	45 Rh	46 Pd	47 Ag	48 Cd						
6	55 Cs	56 Ba	*	71 Lu	72 Hf	73 Ta	74 W	75 Re	76 Os	77 Ir	78 Pt	79 Au	80 Hg	81 Tl	82 Pb	83 Bi	84 Po	85 At	86 Rn
7	87 Fr	88 Ra	**	103 Lr	104 Rf	105 Db	106 Sg	107 Bh	108 Hs	109 Mt	110 Uun	111 Uuu	112 Uub	113 Uut	114 Uuq	115 Uup	116 Uuh	117 Uus	118 Uuo
lanthanides			*	57 La	58 Ce	59 Pr	60 Nd	61 Pm	62 Sm	63 Eu	64 Gd	65 Tb	66 Dy	67 Ho	68 Er	69 Tm	70 Yb		
actinides			**	89 Ac	90 Th	91 Pa	92 U	93 Np	94 Pu	95 Am	96 Cm	97 Bk	98 Cf	99 Es	100 Fm	101 Md	102 No		



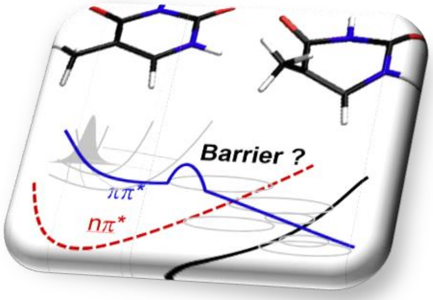
https://vuo.elettra.eu/services/elements/WebElements.html

Quiz: Absorption spectroscopy

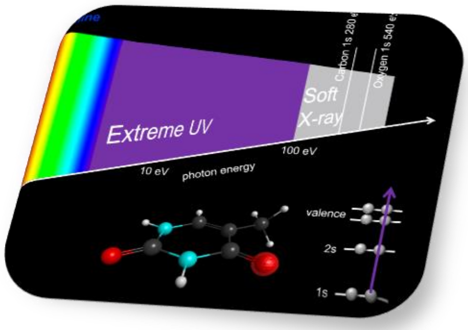


https://vuo.elettra.eu/services/elements/WebElements.html

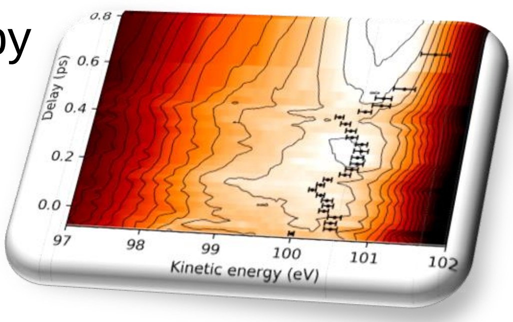
Outline



Basics: Coupled electronic and nuclear dynamics in molecules



X-ray matter interaction



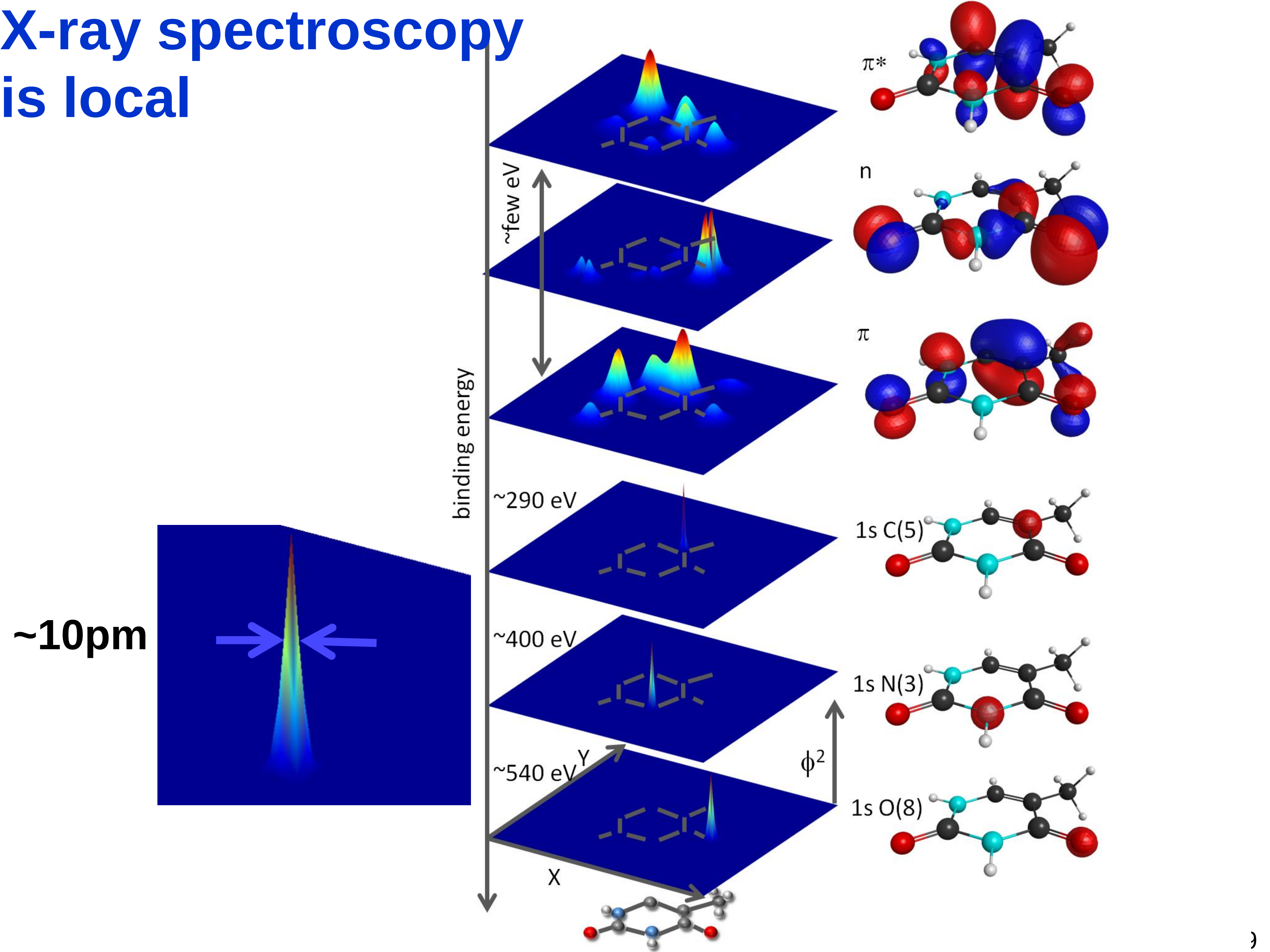
Two examples for ultrafast x-ray spectroscopy

resonant absorption

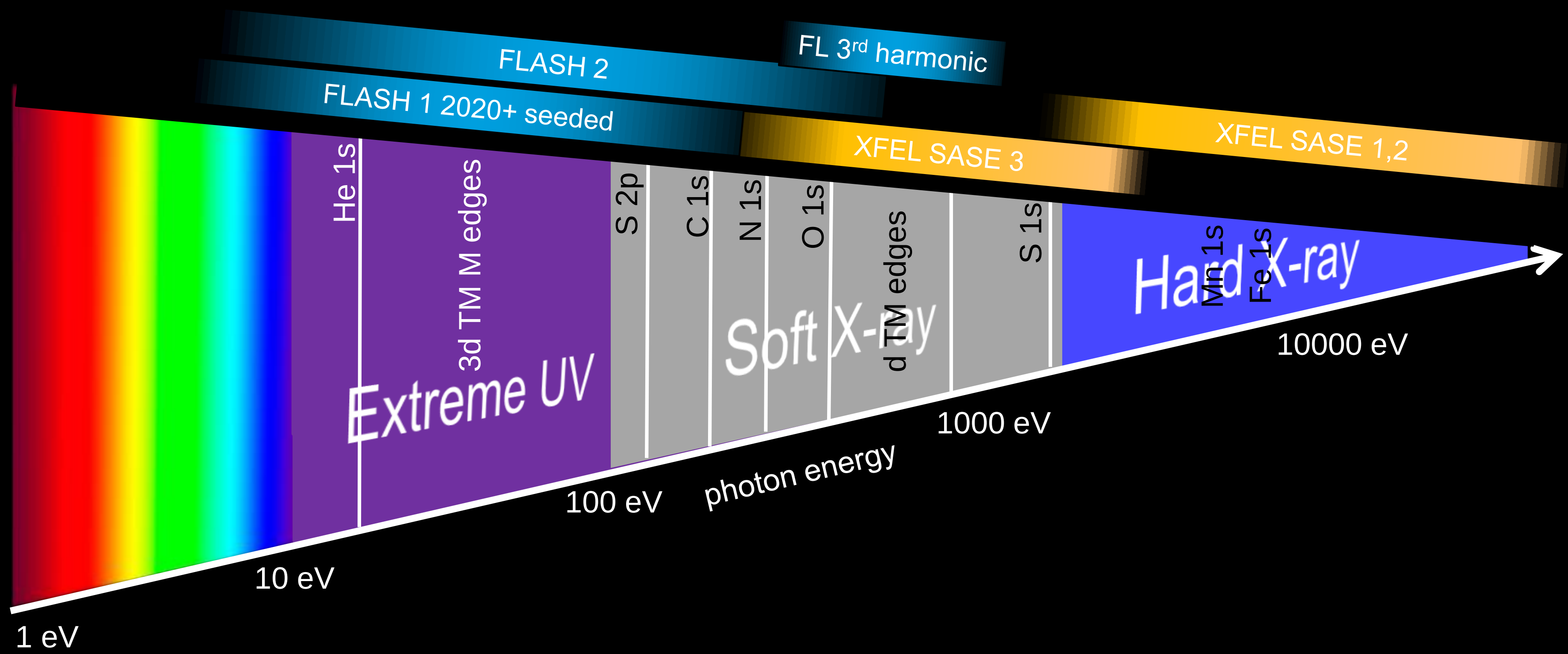
photoelectron spectroscopy

Part II

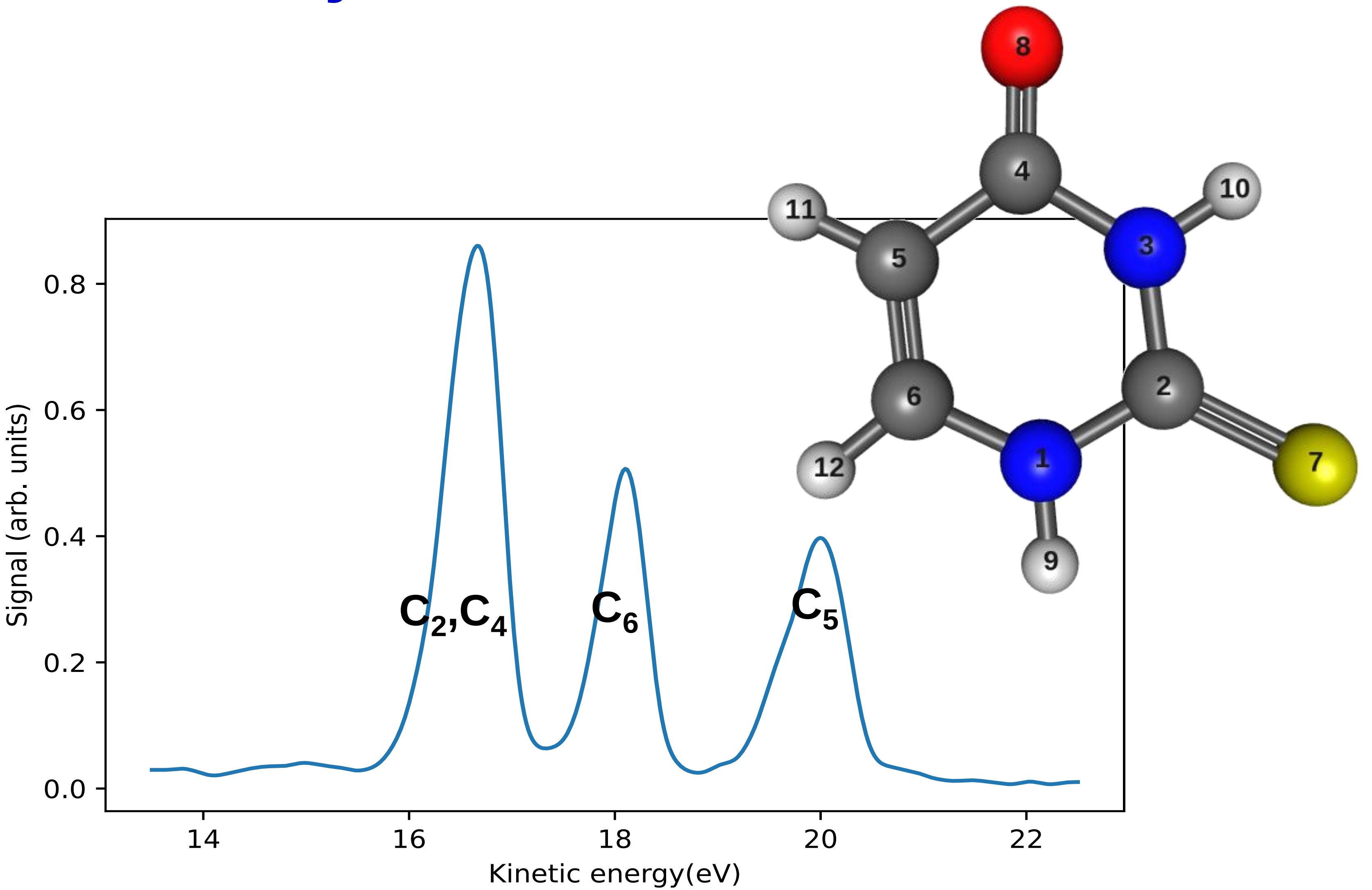
X-ray spectroscopy is local



X-ray spectroscopy is element specific



Site sensitivity – chemical shift

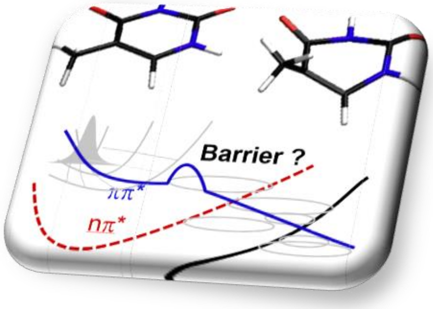


Electronegativity:

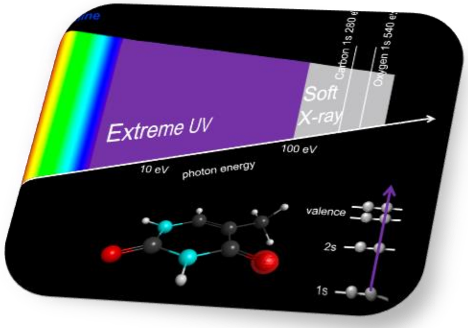
F > O > Cl > N > Br > I > S > C > H

Outline

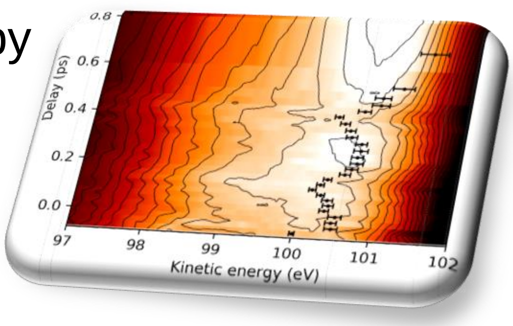
Basics: Coupled electronic and nuclear dynamics in molecules



X-ray matter interaction



Two examples for ultrafast x-ray spectroscopy
resonant absorption
photoelectron spectroscopy



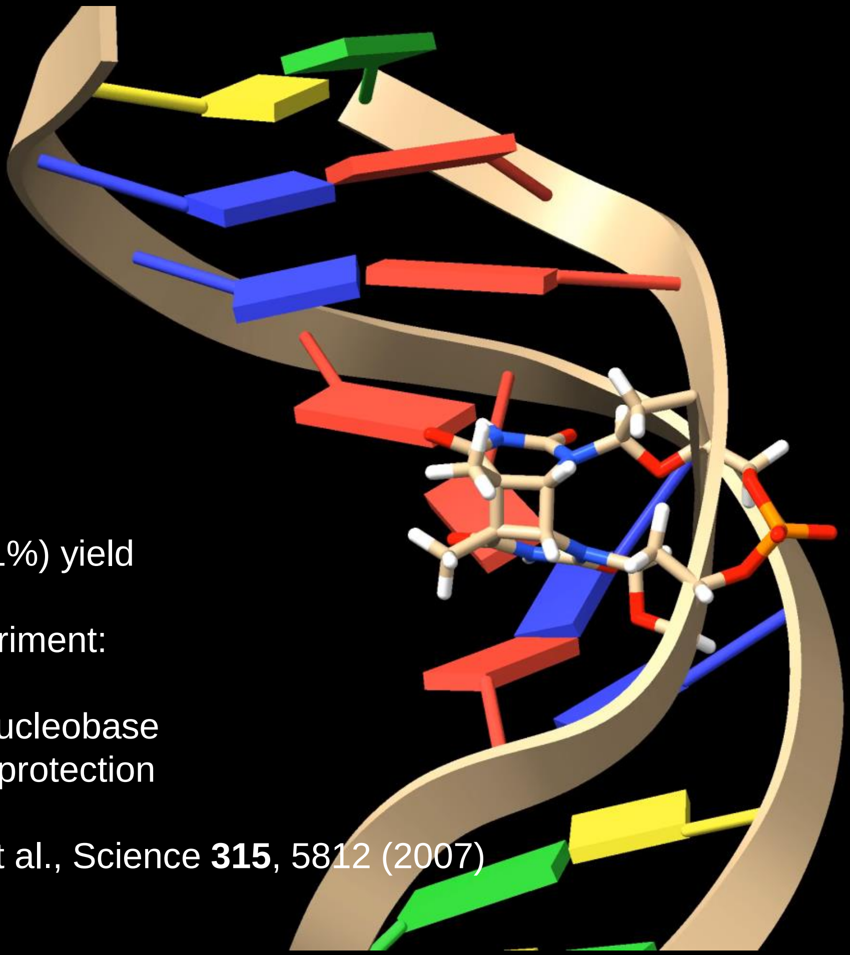
UV Photoläsion

Low (sub 1%) yield

RNA Experiment:

Ultrafast nucleobase
relaxation protection

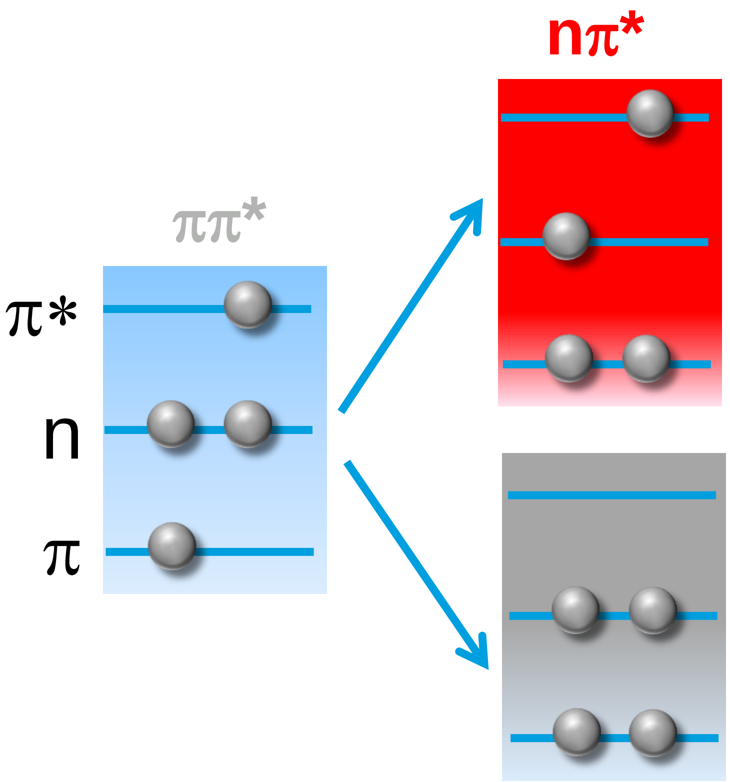
Schreier et al., Science **315**, 5812 (2007)



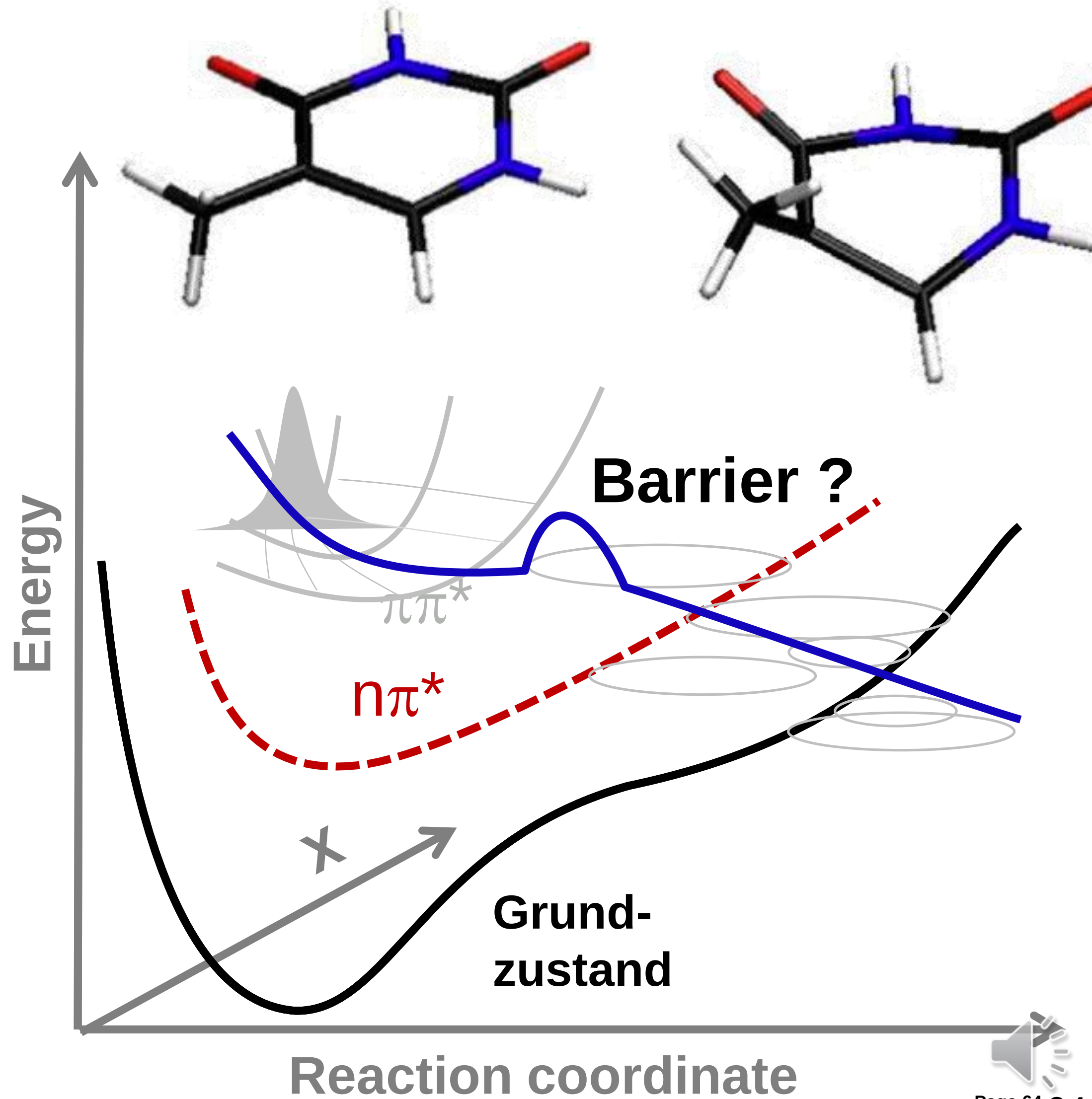
Cyclobutane
Pyrimidine
Dimer (CPD)

Two sides of the problem:

Electronic structure



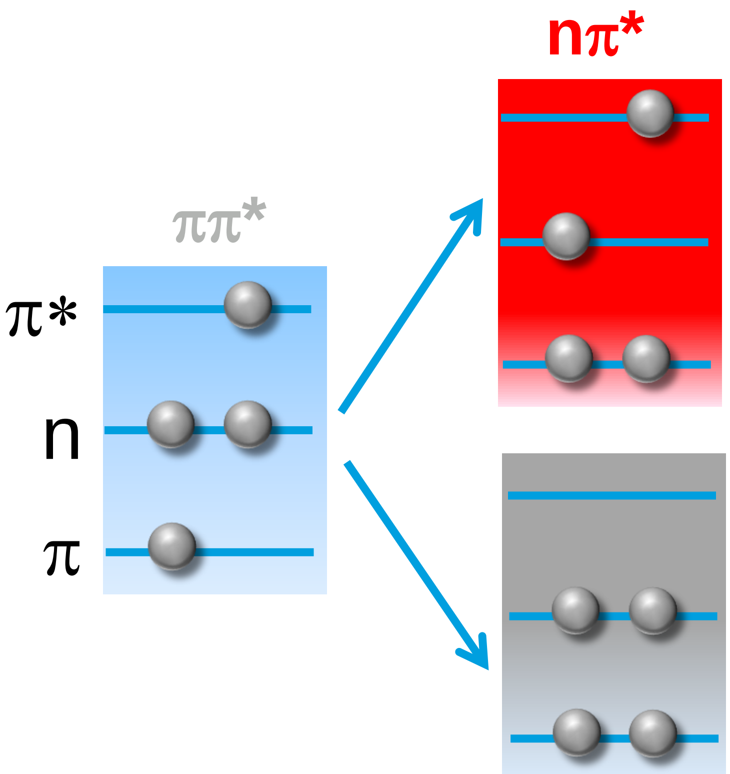
Nuclear geometry



Asturiol et al.,
J. Phys. Chem. A,**113**, 10211 (2009)
Hudock et al.,
J. Phys. Chem. A,**111**, 85 (2007)

Quiz: Which probe method would you apply here?

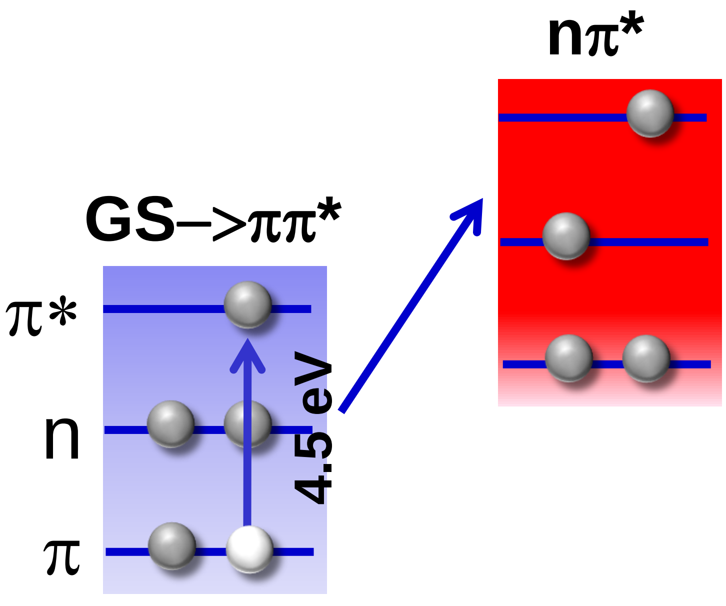
Electronic structure

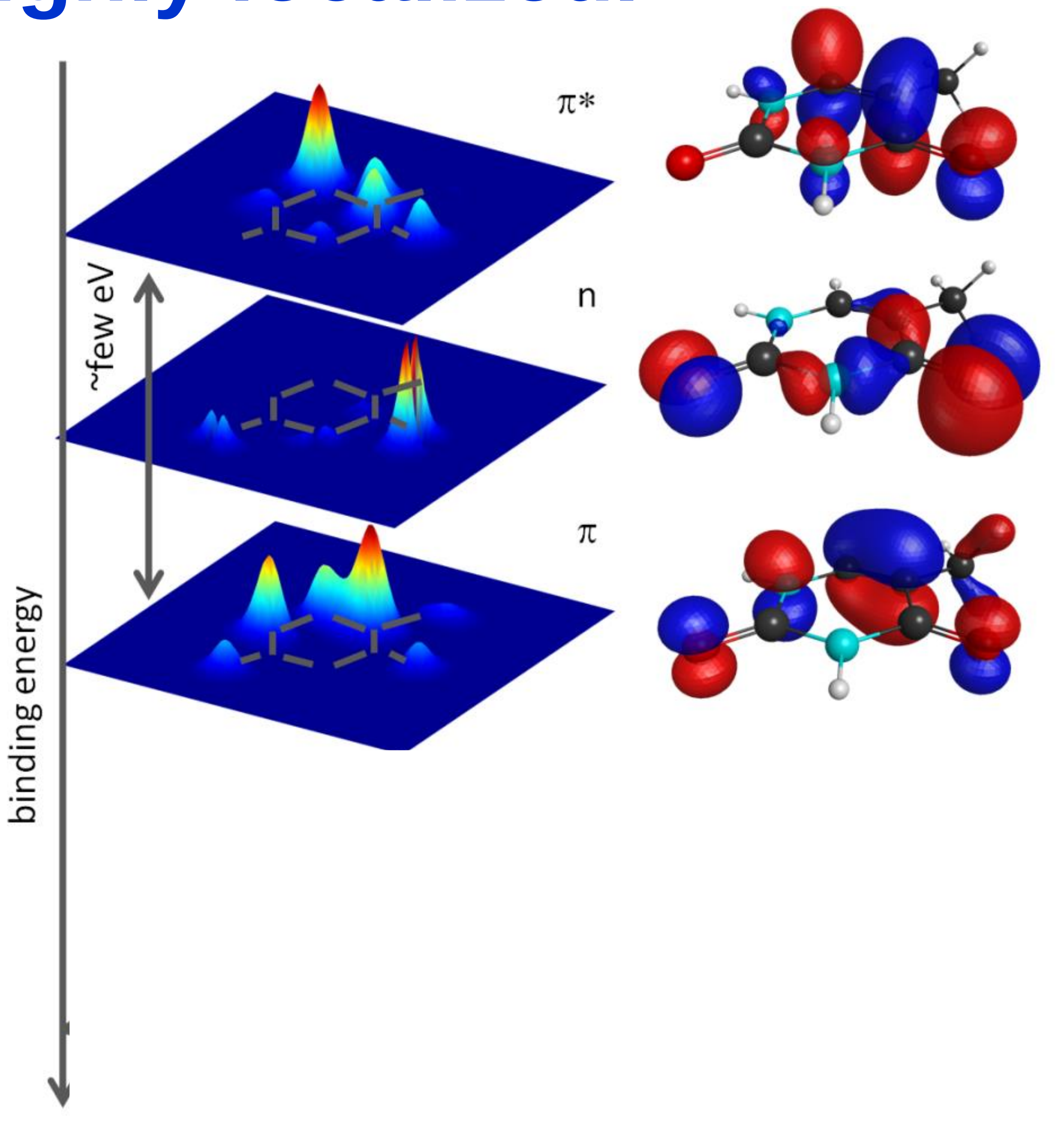


a)NEXAFS
b)XANES
c)Hard X-ray diffraction

Asturiol et al.,
J. Phys. Chem. A,**113**, 10211 (2009)
Hudock et al.,
J. Phys. Chem. A,**111**, 85 (2007)

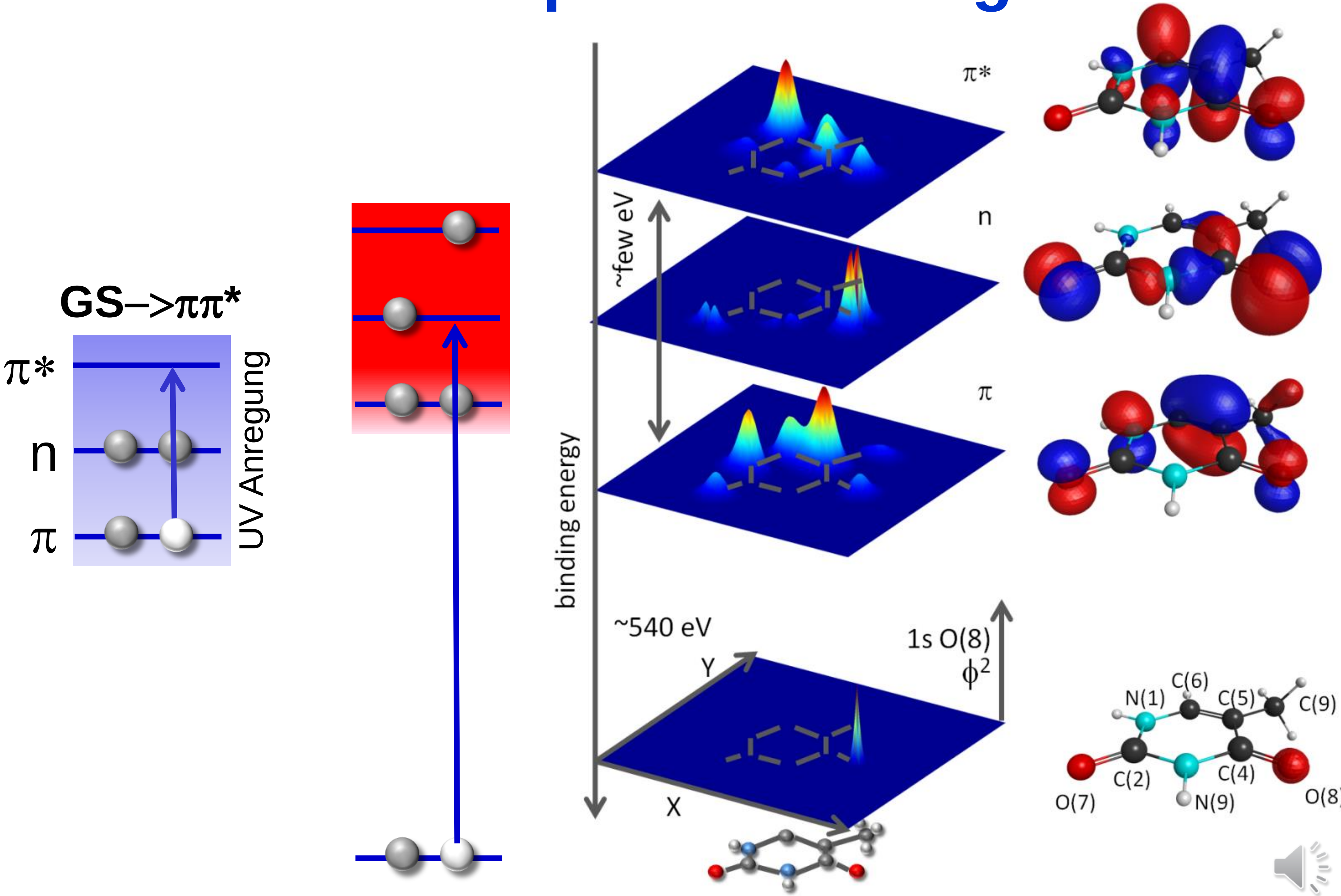
The n orbital is highly localized.

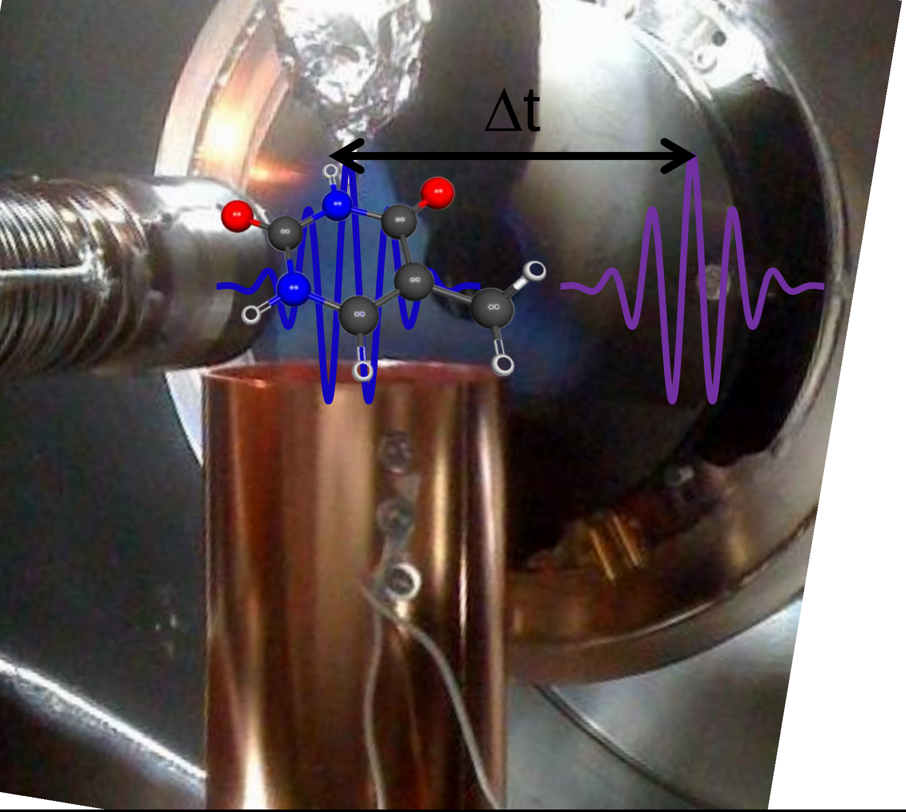


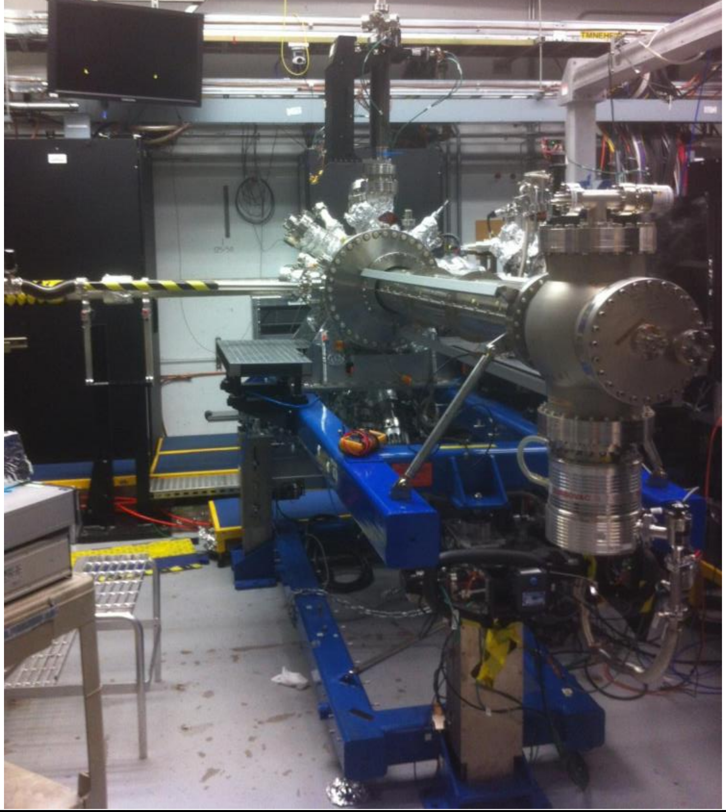


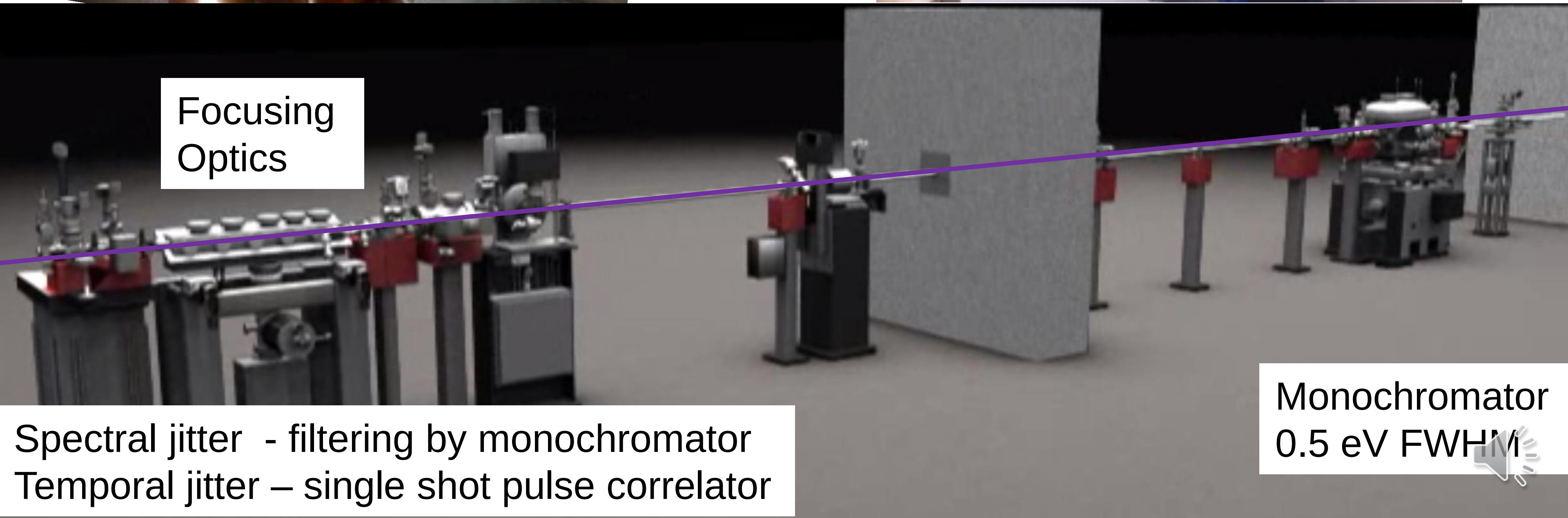
Previous work: McFarland *et al.*, Nature Comm. **5**, 4235 (2014)

The1s to n absorption is strong.



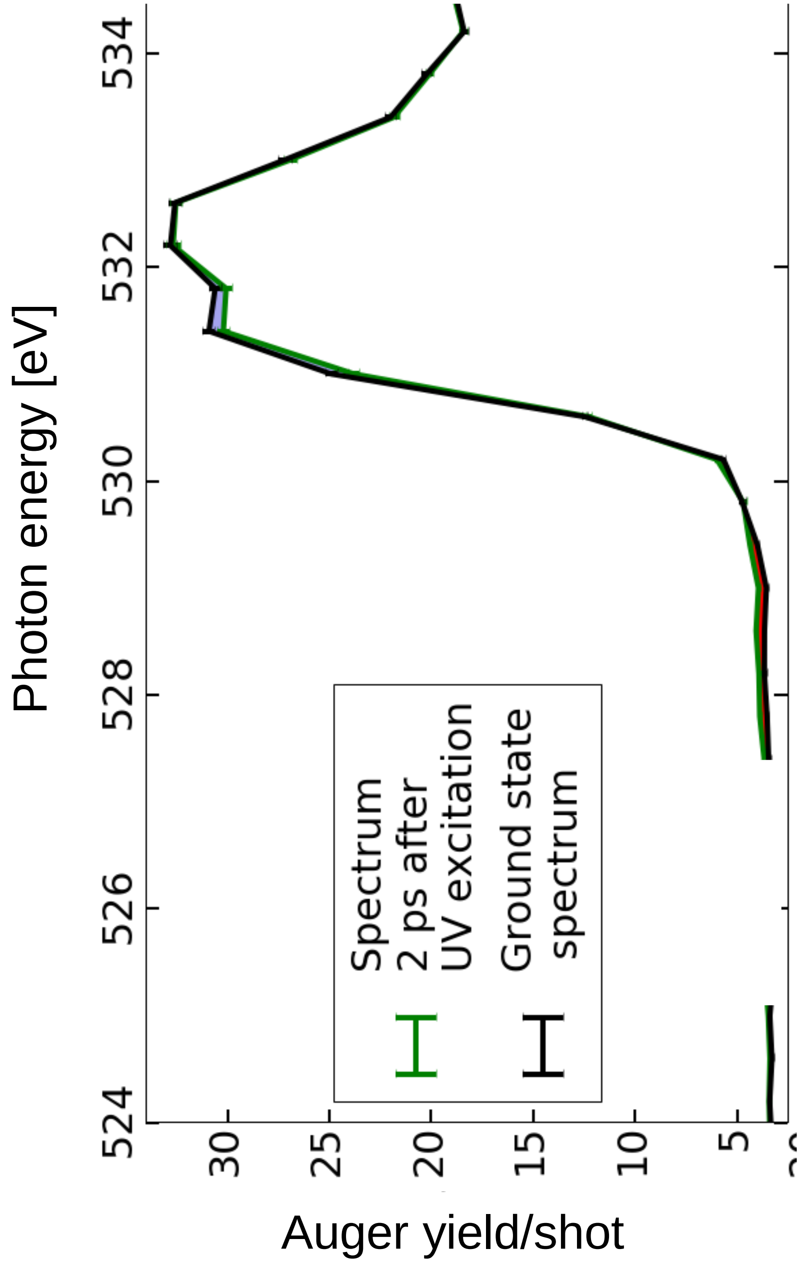


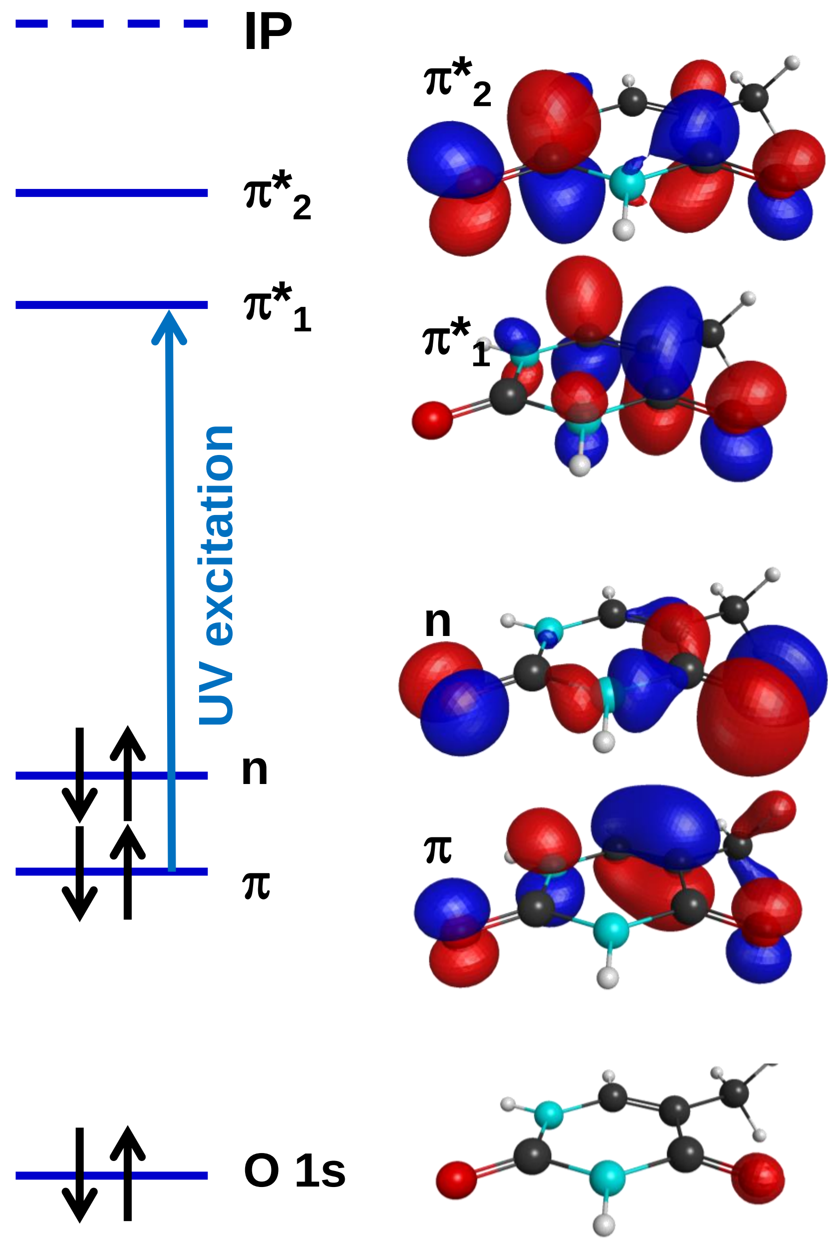




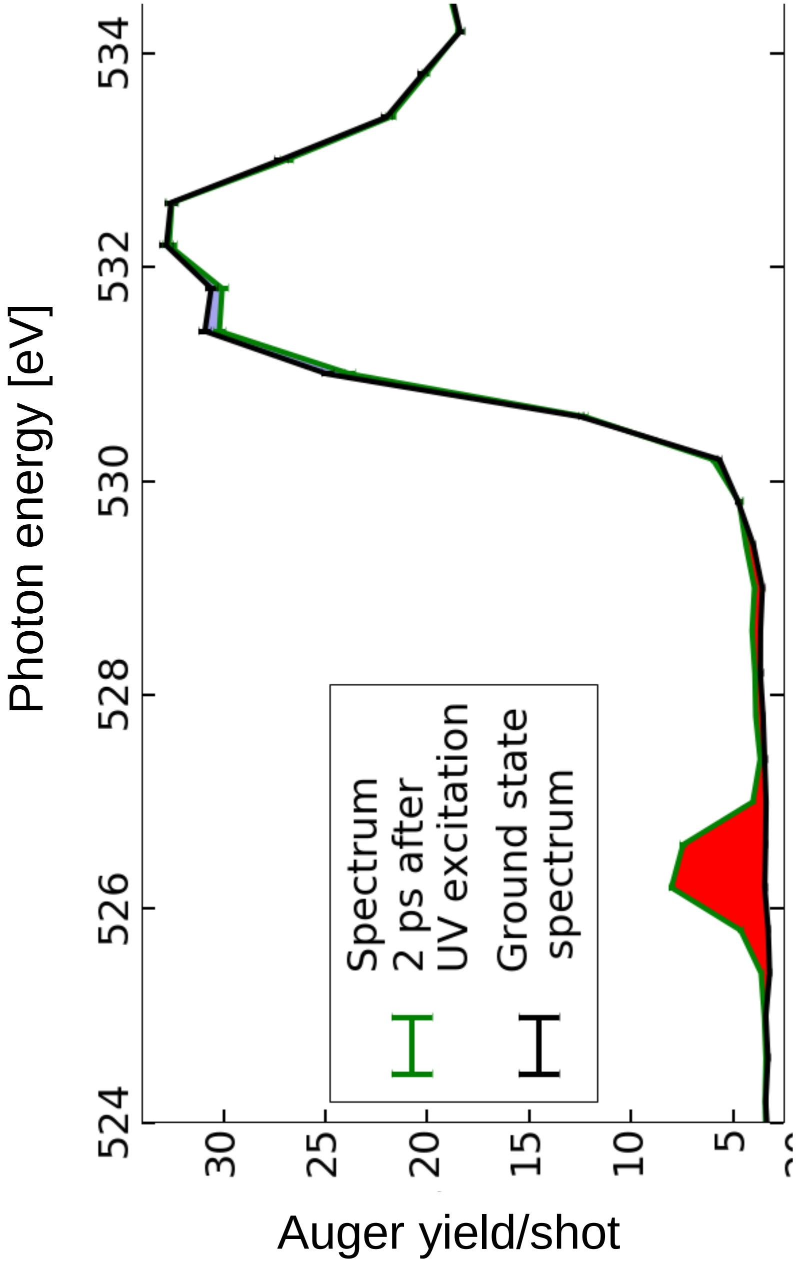
Spectral jitter - filtering by monochromator
Temporal jitter – single shot pulse correlator

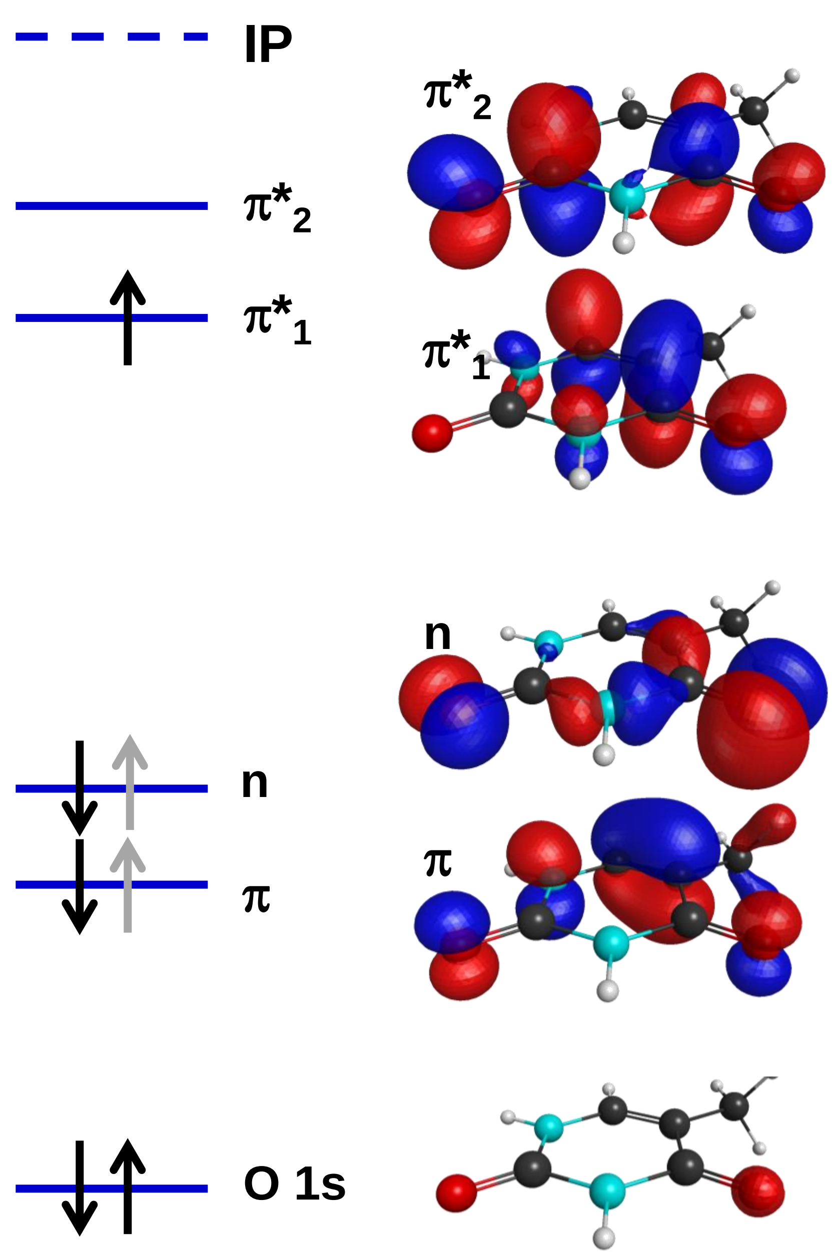
NEXAFS shows resonances

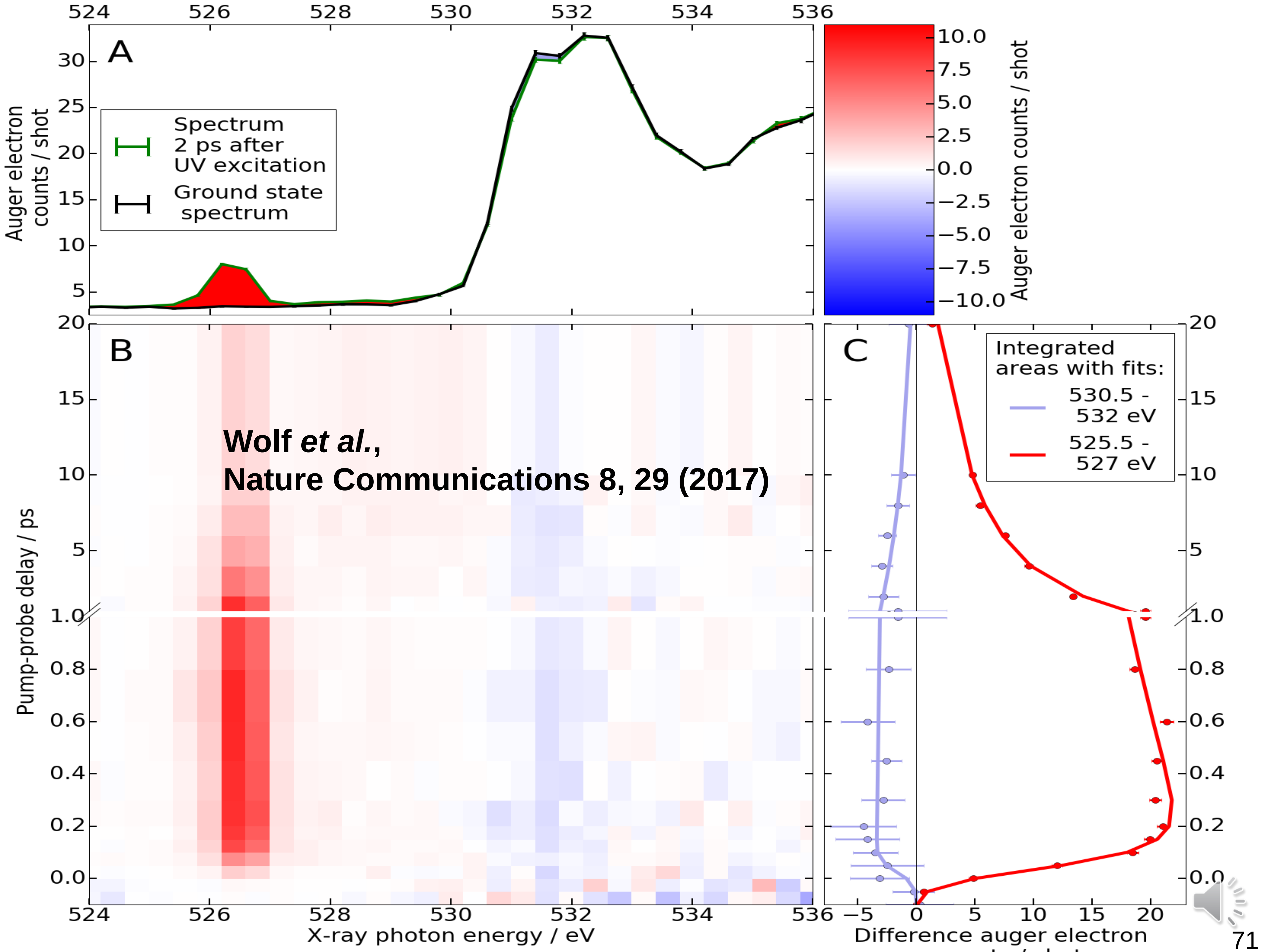




NEXAFS shows resonances





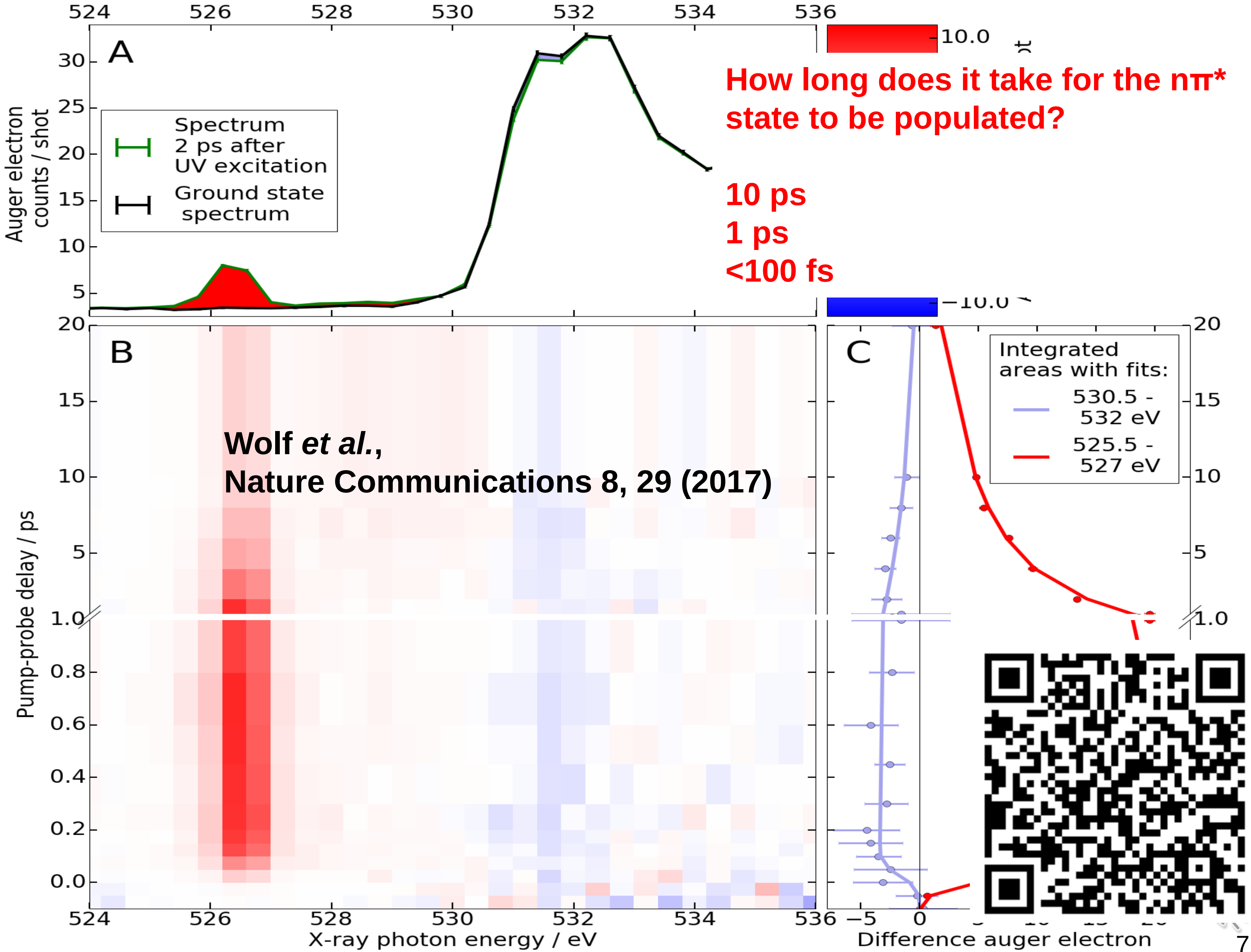

A
Spectrum
2 ps after
UV excitation
Ground state
spectrum
Auger electron counts / shot
Auger electron
counts / shot
B
C
Integrated
areas with fits:
530.5 -
532 eV
525.5 -
527 eV
Pump-probe delay / ps
X-ray photon energy / eV
Difference auger electron
Wolf *et al.*,
Nature Communications 8, 29 (2017)

How long does it take for the nπ* state to be populated?

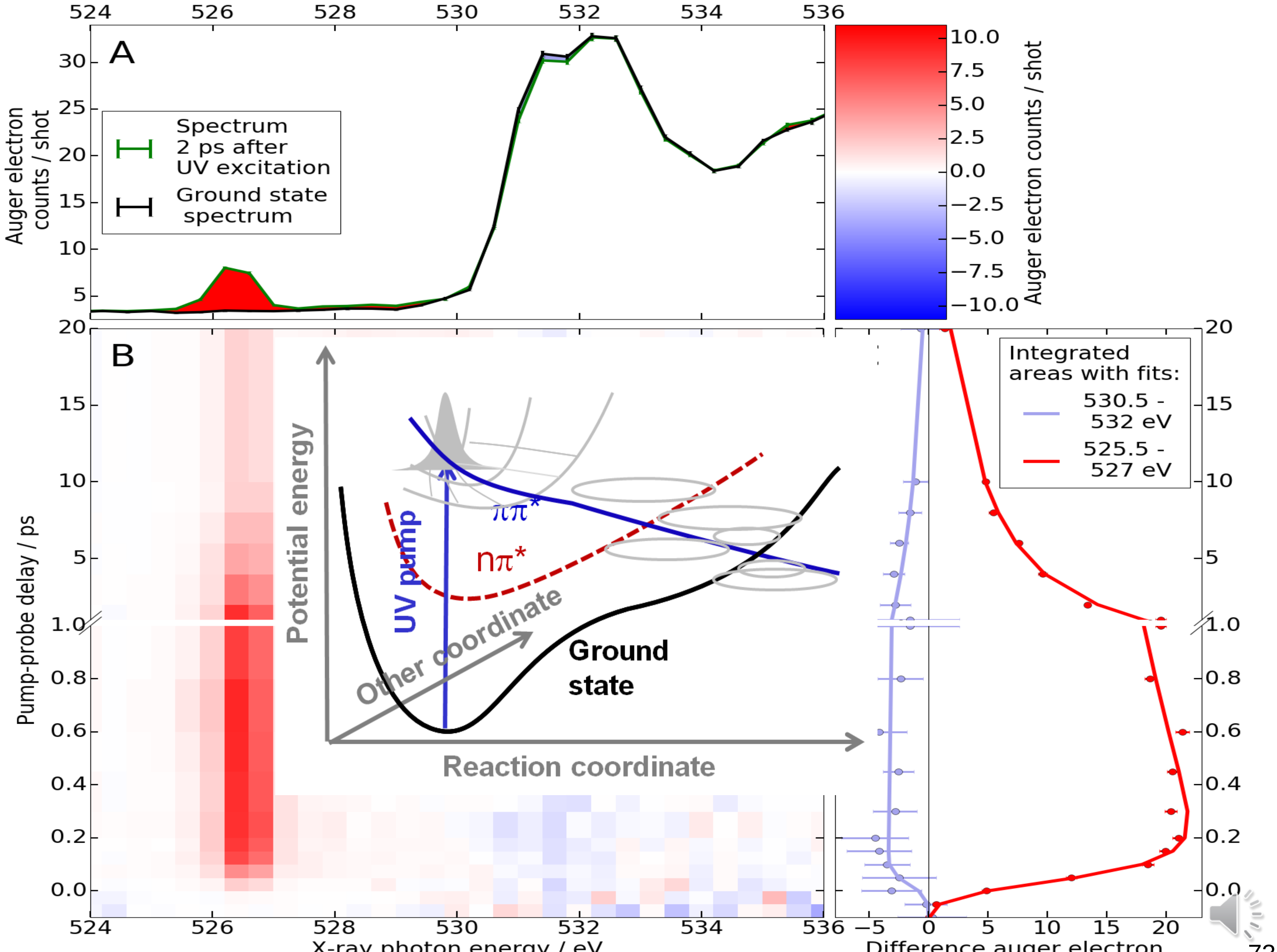
10 ps

1 ps

<100 fs

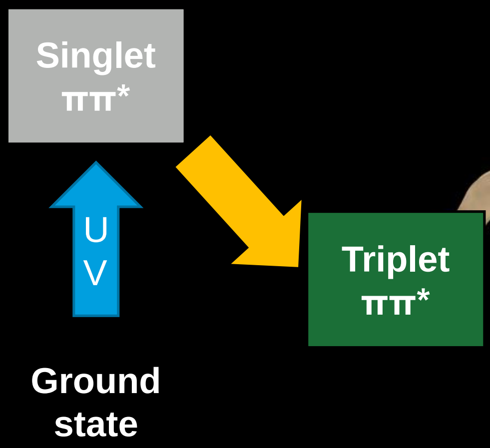


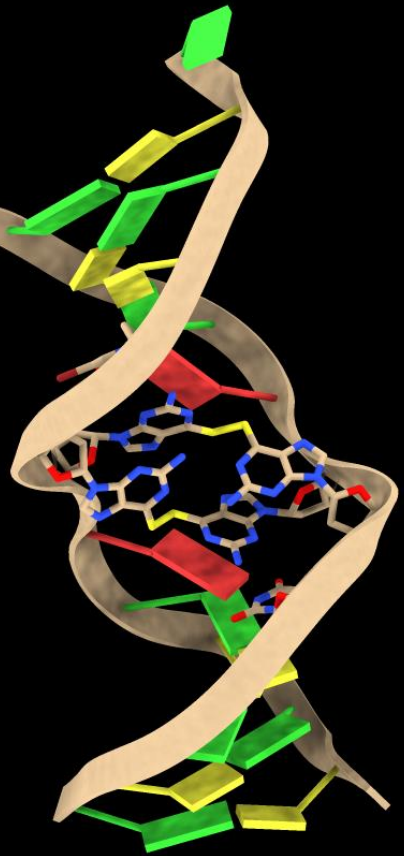
Wolf *et al.*,
Nature Communications 8, 29 (2017)



A
Spectrum
2 ps after
UV excitation
Ground state
spectrum
Auger electron
counts / shot
Auger electron counts / shot
B
Pump-probe delay / ps
Potential energy
UV pump
ππ*
nπ*
Other coordinate
Ground
state
Reaction coordinate
Integrated
areas with fits:
530.5 -
532 eV
525.5 -
527 eV
X-ray photon energy / eV
Difference auger electron

Thionukleobasen – Alles wird schlimmer!



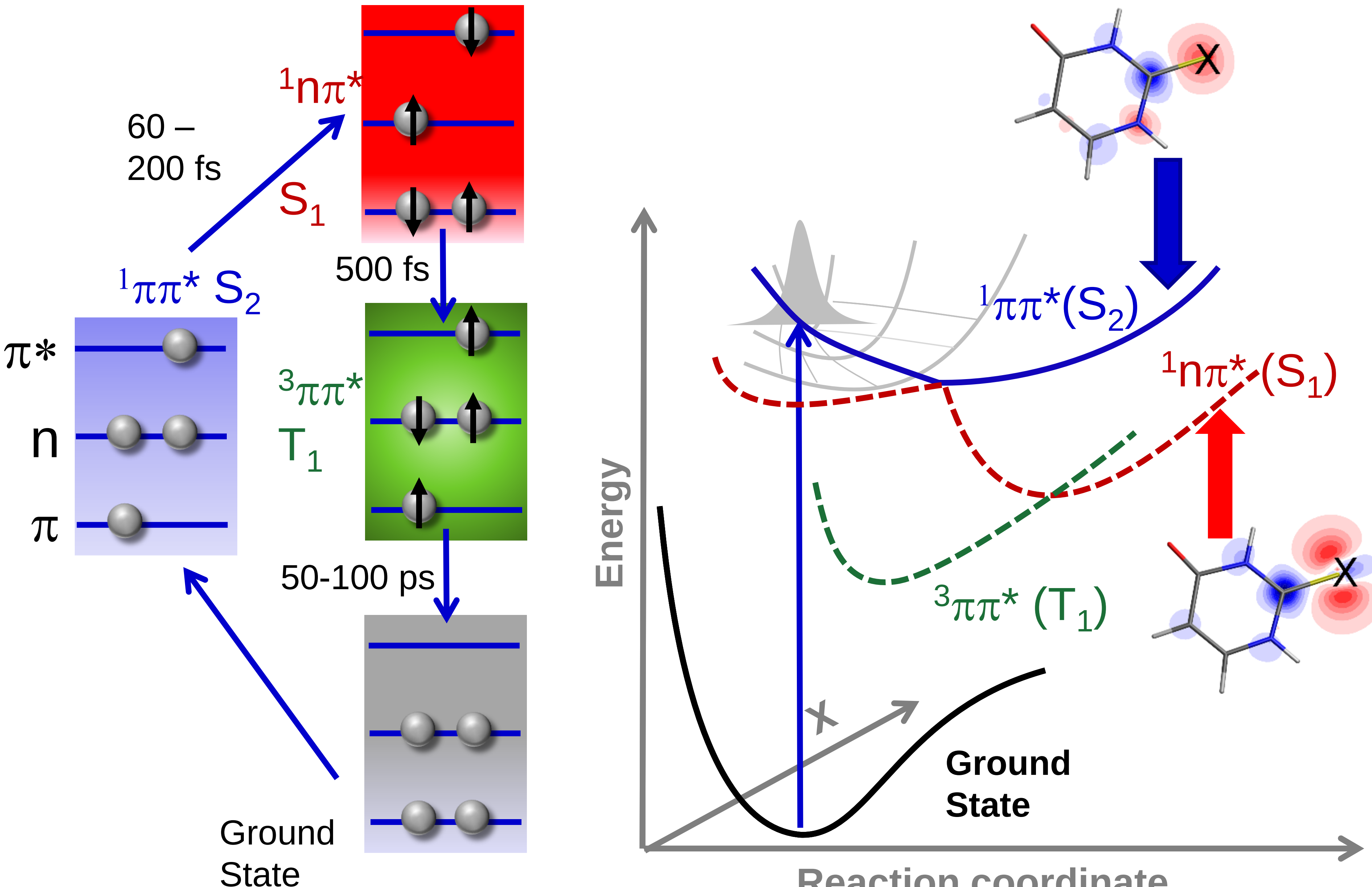


Interstrand
Cross-linking

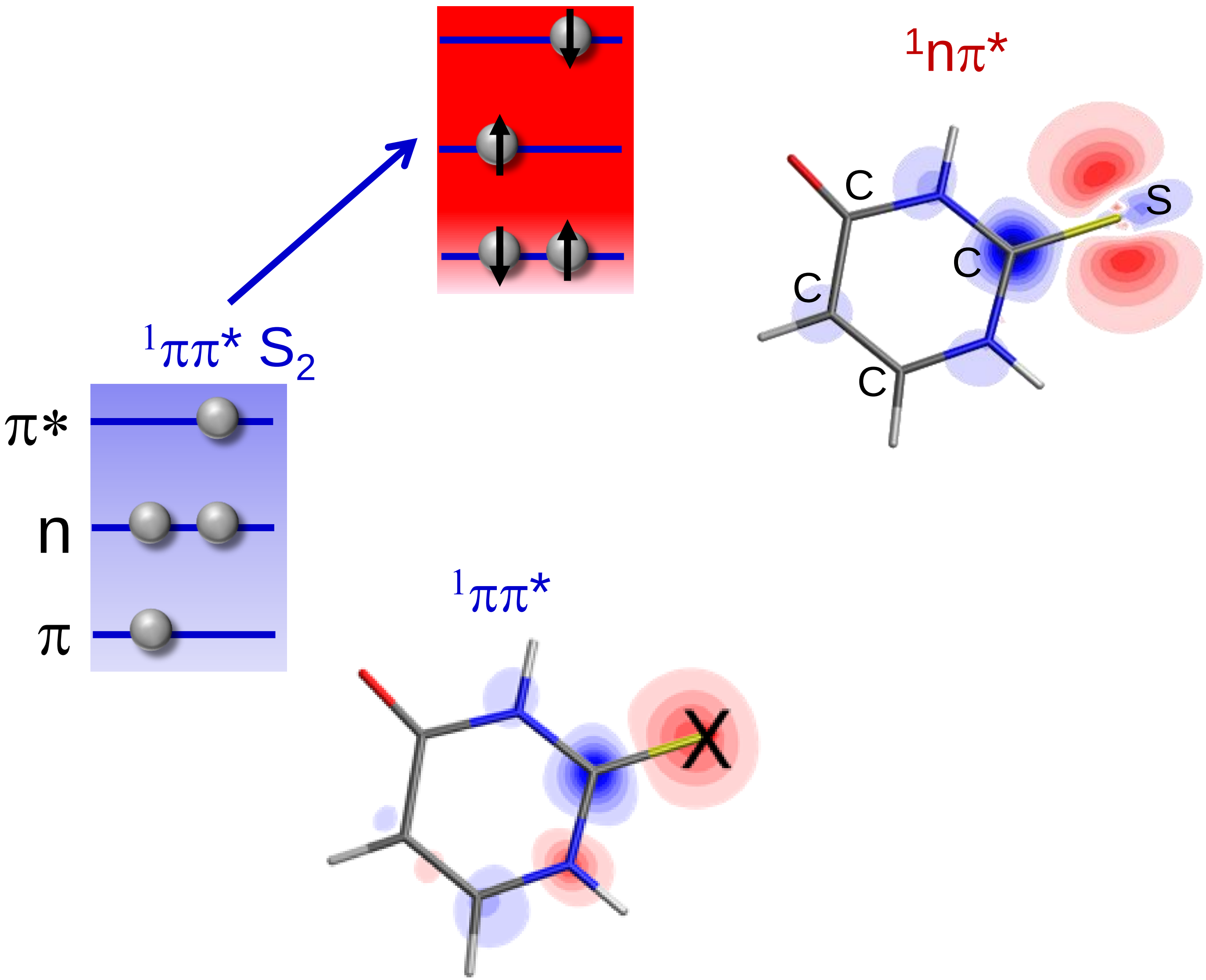
Way worse than usual nucleobases:
Thionucleobases

B. Ashwood, M. Pollum, C. E. Crespo-Hernández, Photochem. Photobiol. **95**, 33 (2019)
S. Bai, M. Barbatti, PCCP **19**, 12674 (2017)
Arslancan, Martínez-Fernández, Corral, Molecules **22**, 998 (2017).

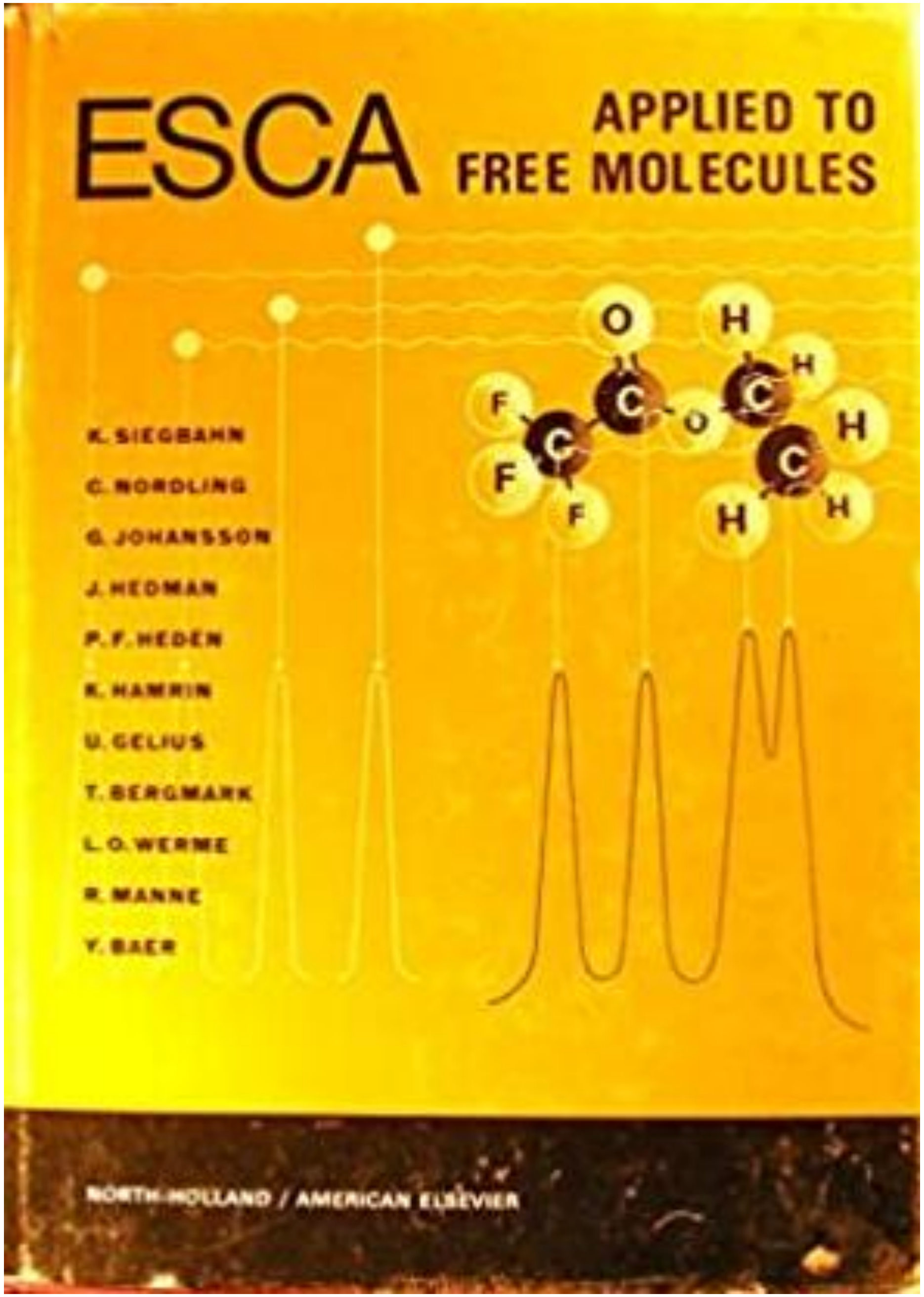
Nuclei couple to electrons and their spin.



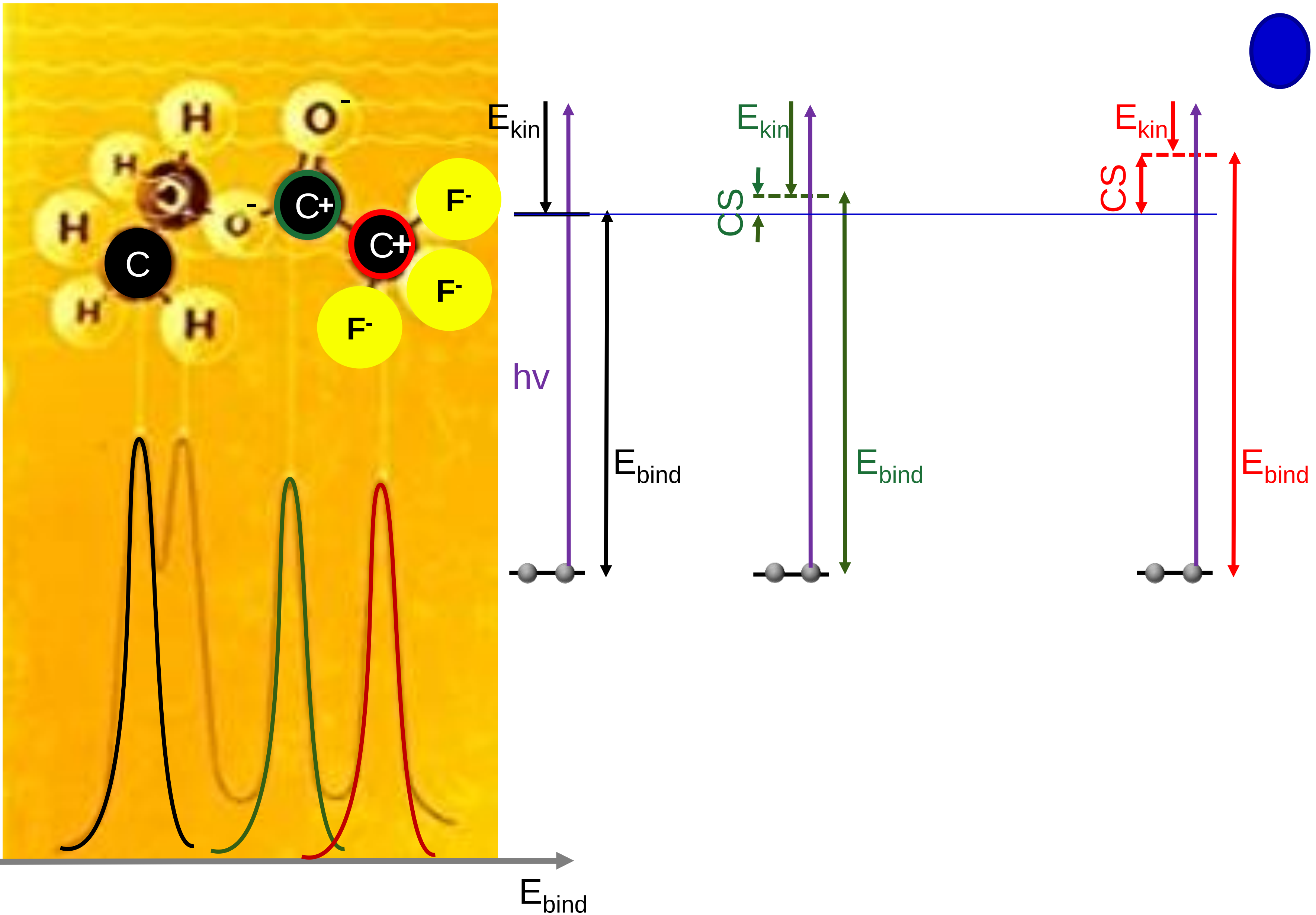
FLASH makes electronic molecular movies.

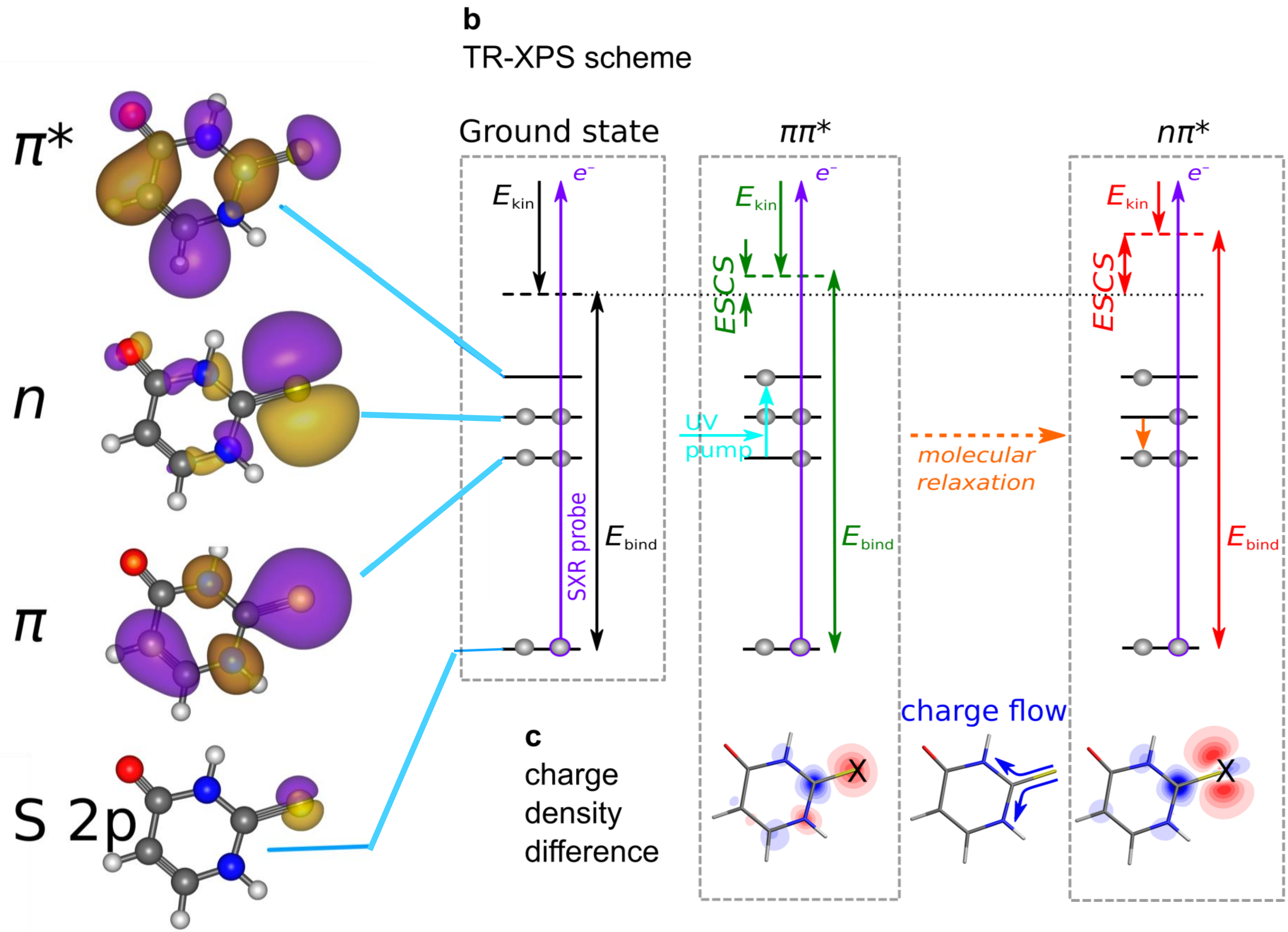


Chemical shift and local charge

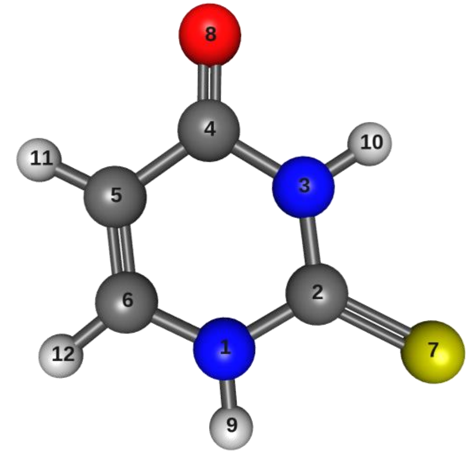


Chemical shift and local charge




b
TR-XPS scheme
π*
n
π
S 2p
Ground state
ππ*
nπ*
e⁻
E kin
E bind
SXR probe
ESCS
UV
pump
molecular
relaxation
charge flow
X
c
charge
density
difference

Quiz: convince your advisor



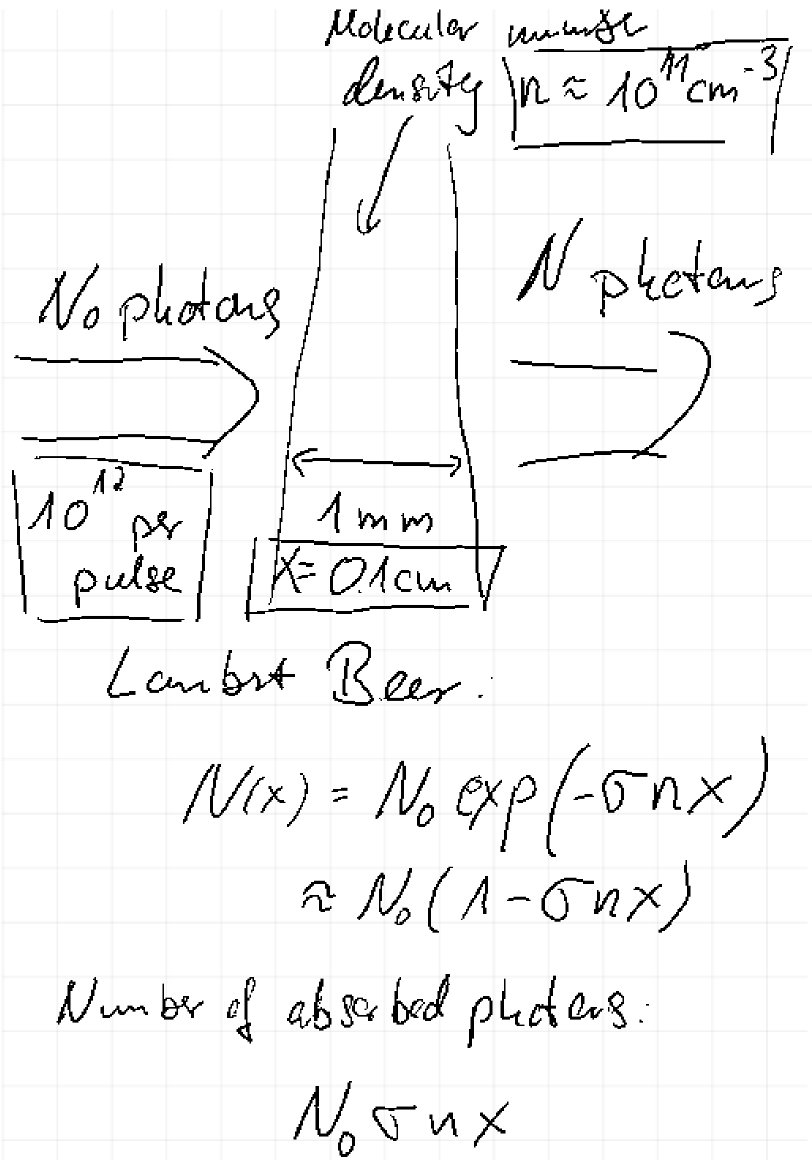
You have an idea for your thesis project:

XPS on the sulfur 2p electrons of a gas target of thiouracil. You choose 300 eV as your probe photon energy. So you prepare a proposal for SwissFEL/FERMI or FLASH, the call closes in 5 min!

Your thesis advisor finally has read the proposal and tells you that this is a bad idea, because you will never get a reasonable amount of photoelectrons from a thin gas jet.

You are How do you convice your advisor?

Quiz: convince your advisor

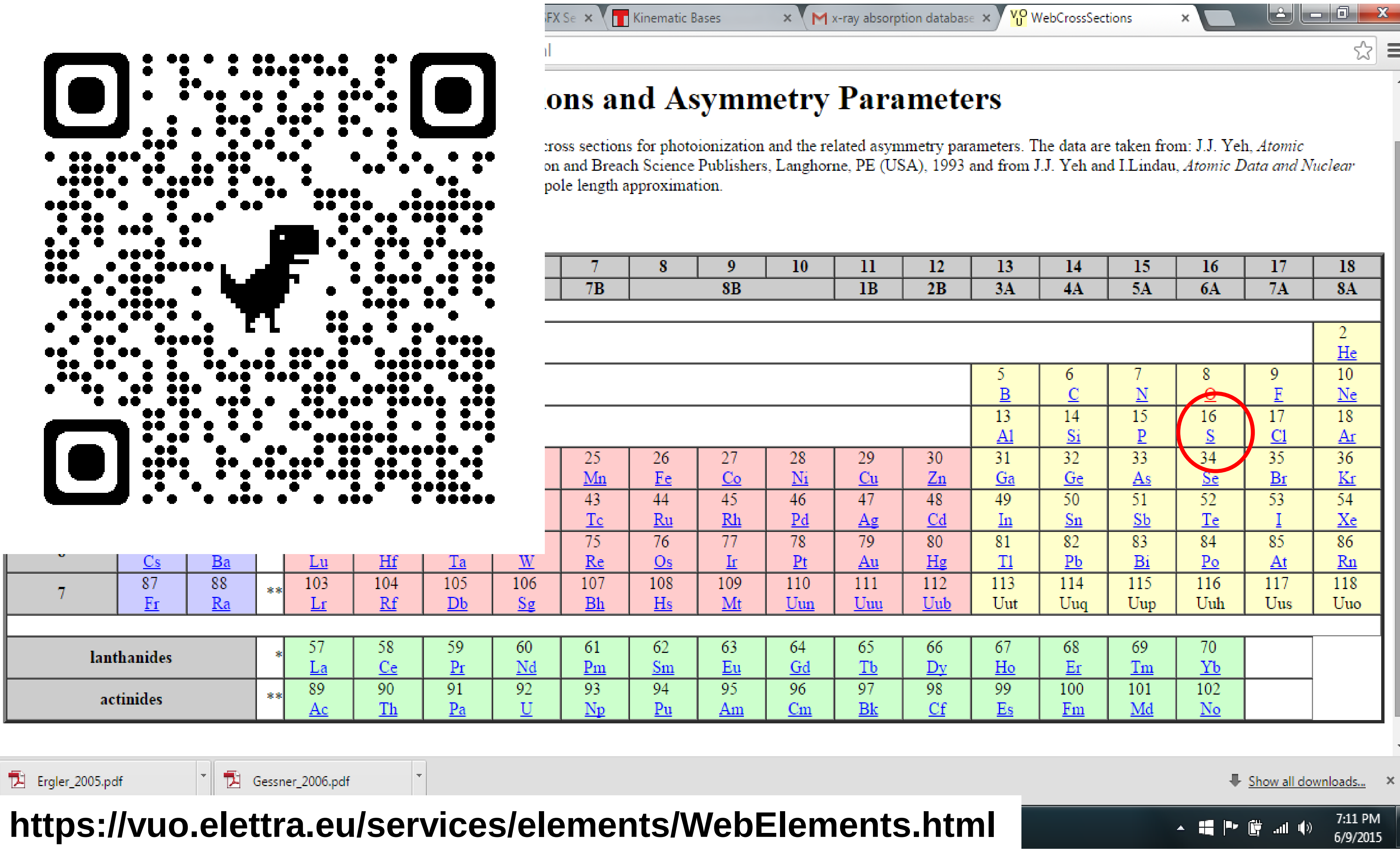


Absorption:

How many electrons per pulse?

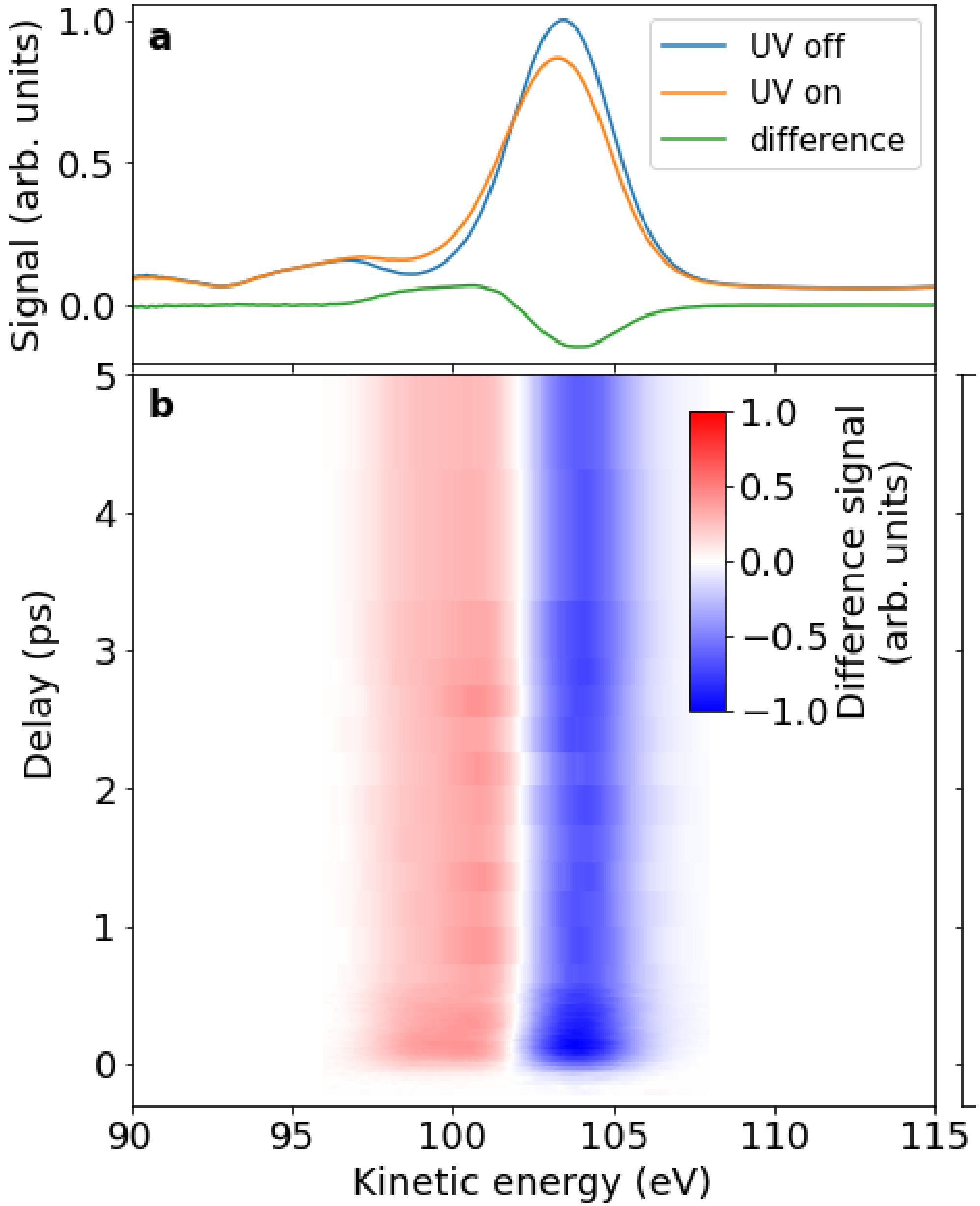
a) 1
b) 10
c) 100
d) 1000
e) 10000
f) 100000

Absorption spectra – useful help



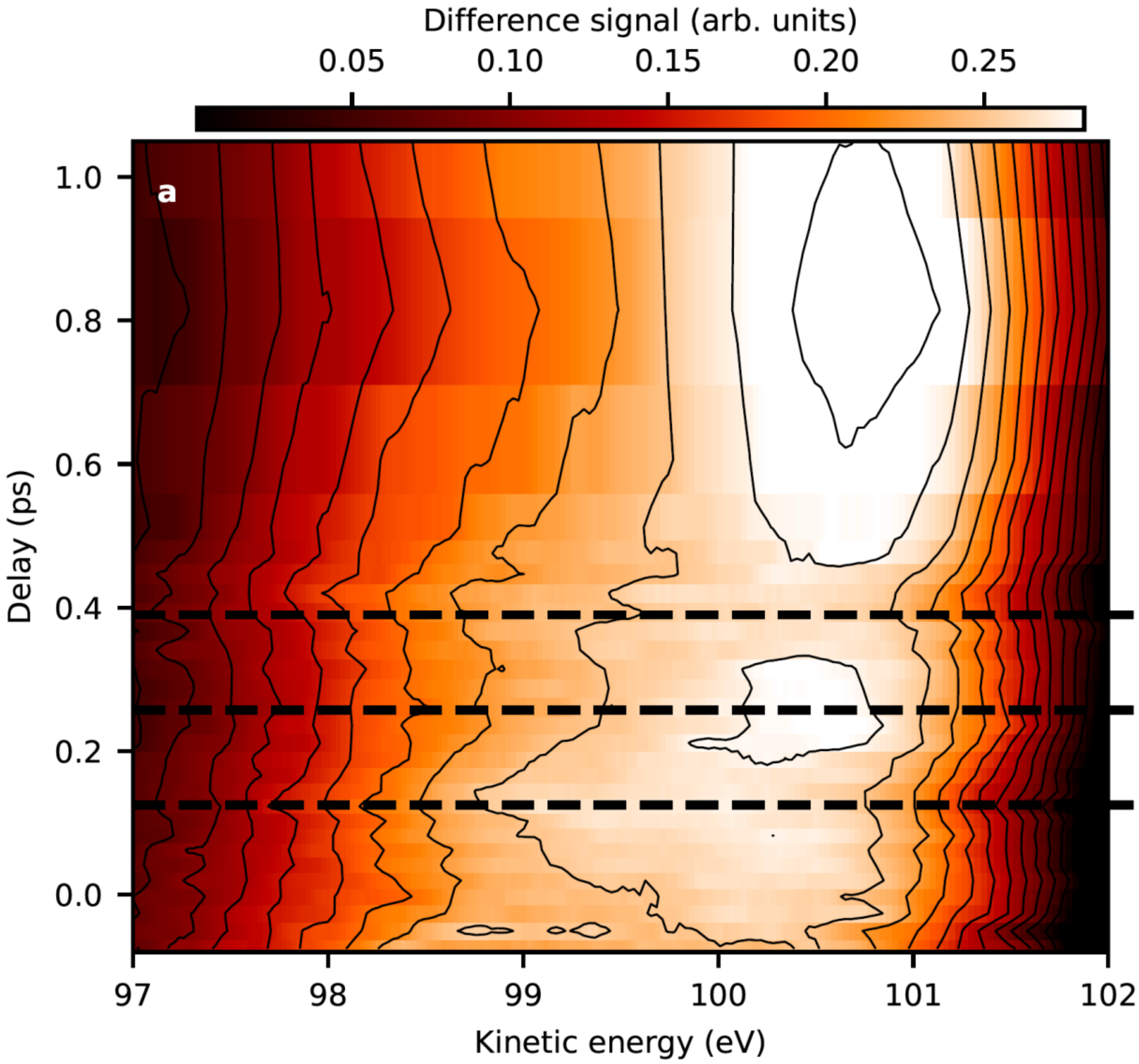
https://vuo.elettra.eu/services/elements/WebElements.html

Long lasting photoelectron shifts



Mayer, Lever, Picconi et al.
Nature Comm. **13**, 198 (2022)

250 fs osciallations in the photoelectron spectra



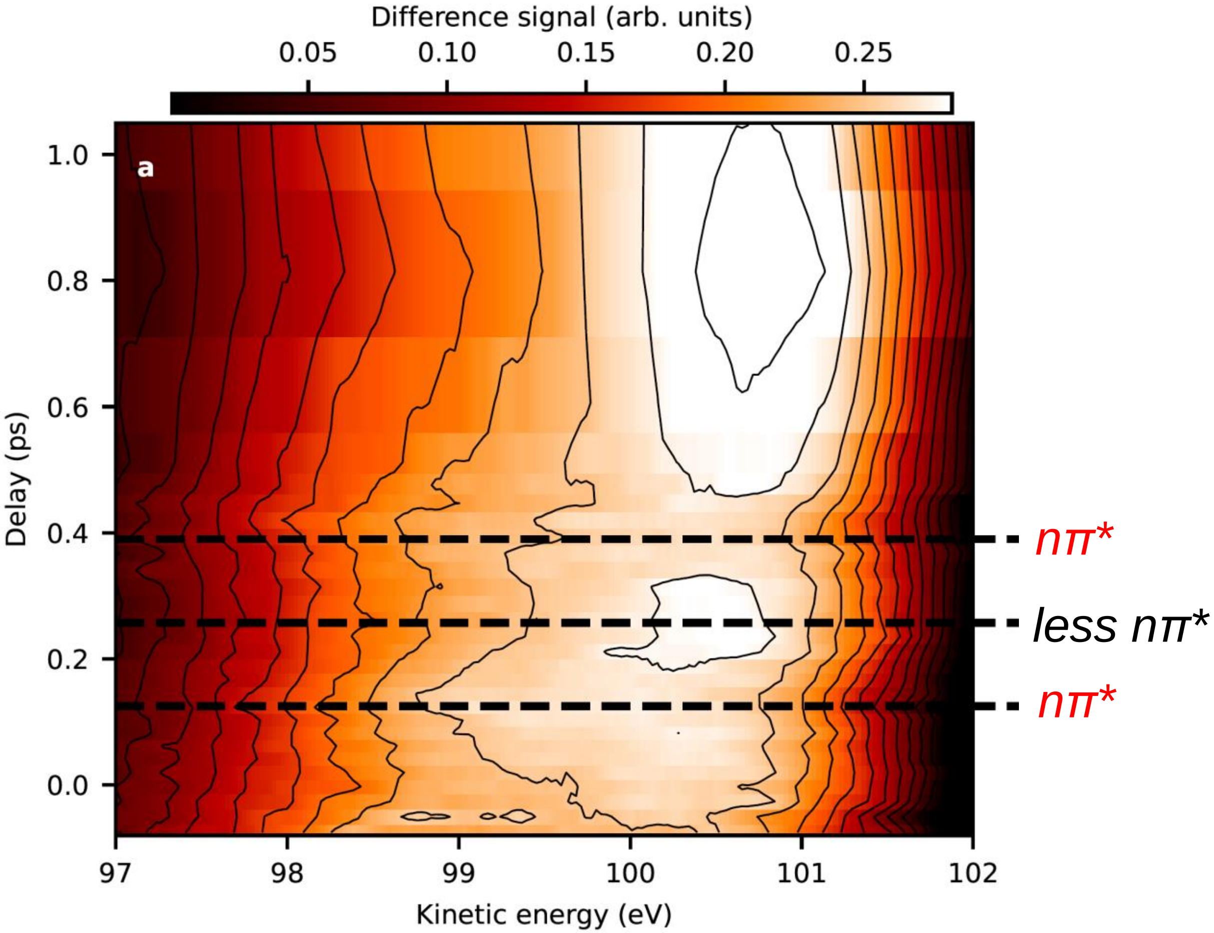
Mayer, Lever, Picconi et al.
Nature Comm. **13**, 198 (2022)

Photoelectron spectrum
normalized on maximum

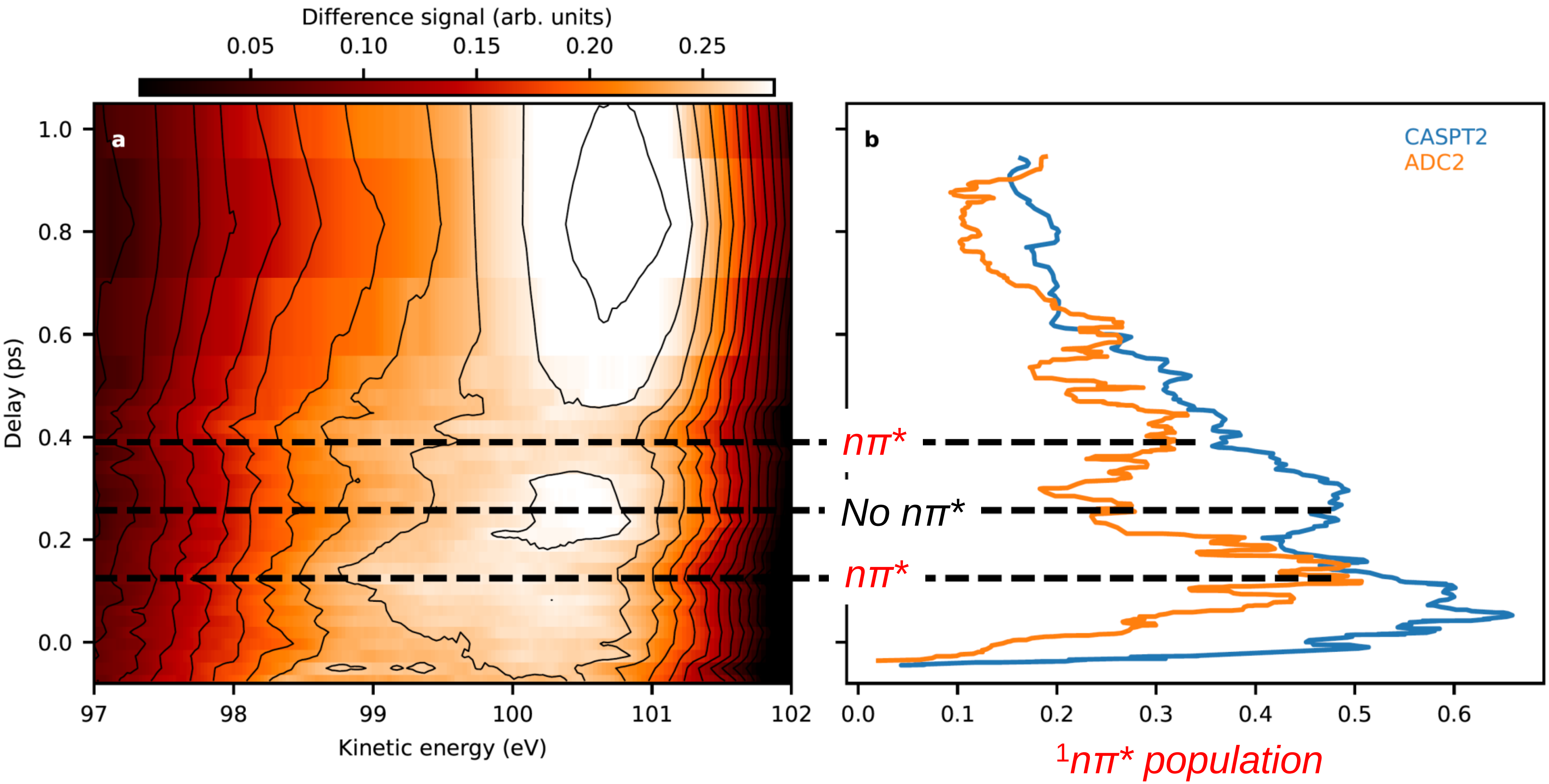
Oscillation in the
excited state chemical shift

250 fs osciallations in the photoelectron spectra

Mayer, Lever, Picconi et al.
Nature Comm. **13**, 198 (2022)

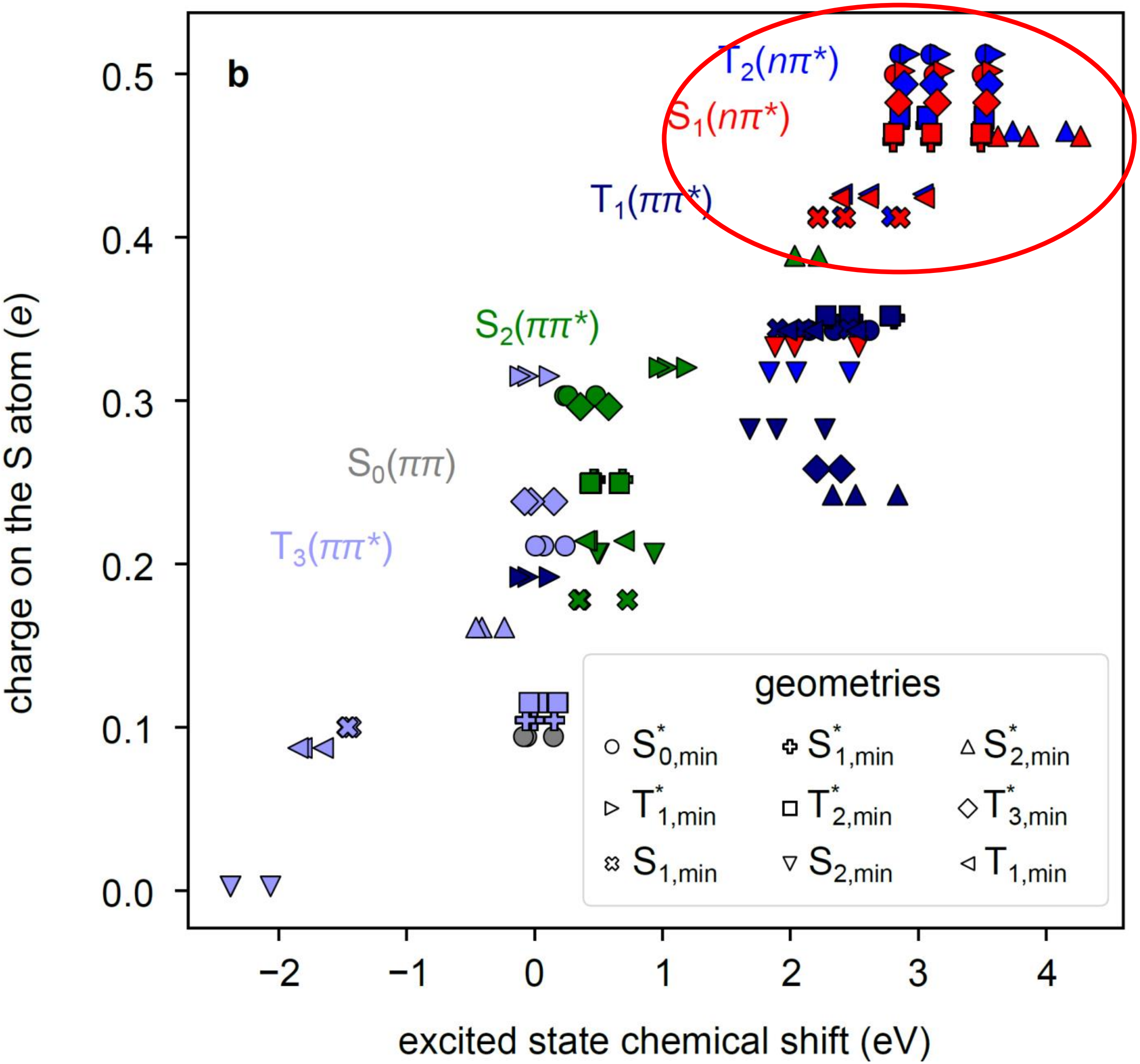


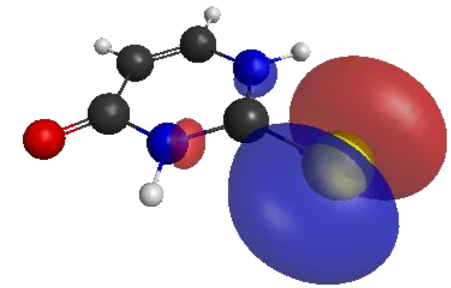
250 fs osciallations in the photoelectron spectra



Simulations:
Mai, Marquetand, González, J. Phys. Chem. Lett. **7**, 1978–1983 (2016)
Mai *et al.* J. Chem. Phys. **147**, 184109 (2017)

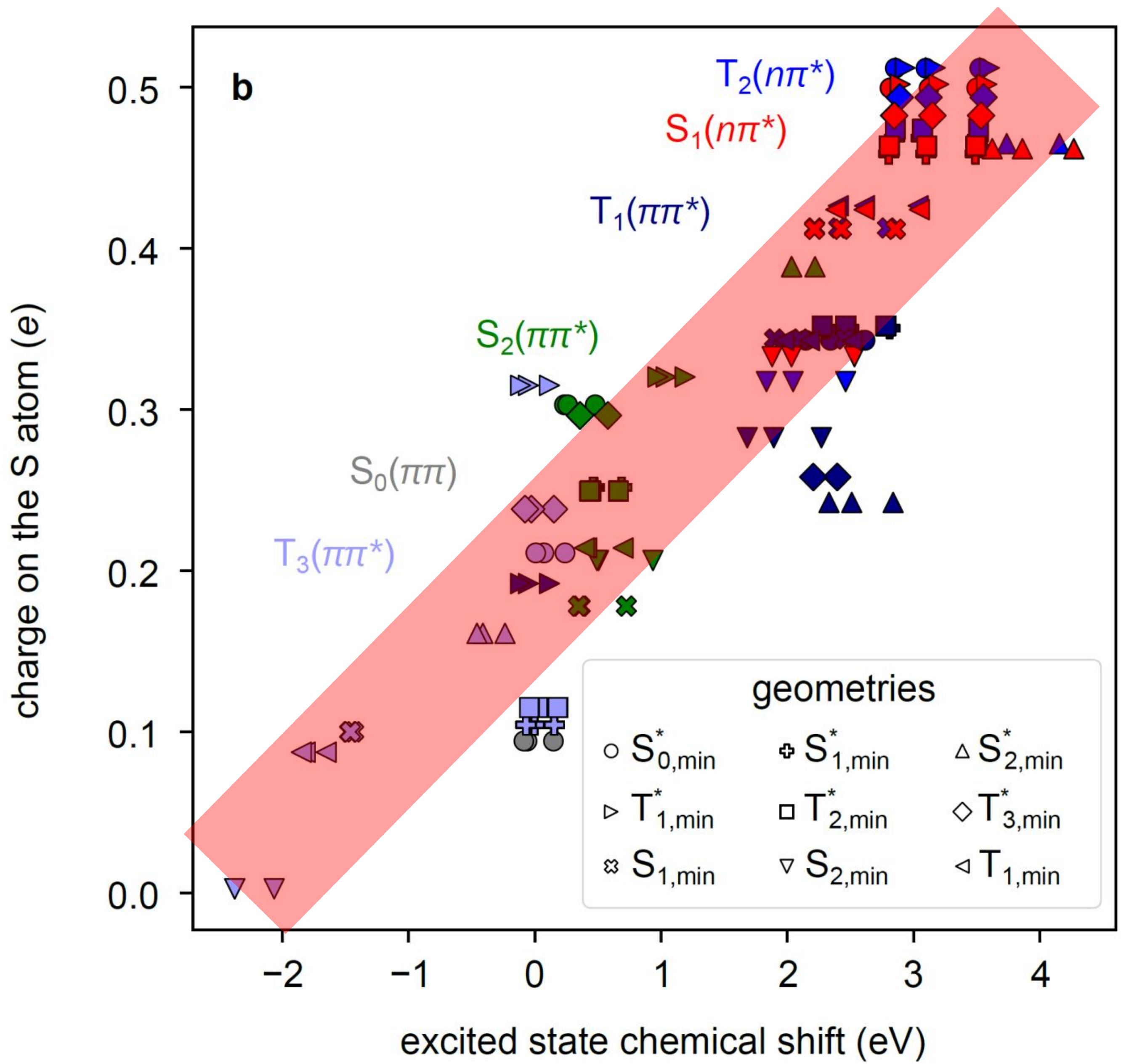
ESCS depends strongly on electronic state





David Picconi
Potsdam
Now Groningen

Linear relation between charge and binding energy



Very similar to a chemical shift potential model

Gelius, Phys. Scr. **9**, 133 (1974)

nature COMMUNICATIONS

ARTICLE

https://doi.org/10.1038/s41467-021-27908-y OPEN

Check for updates

Following excited-state chemical shifts in molecular ultrafast x-ray photoelectron spectroscopy

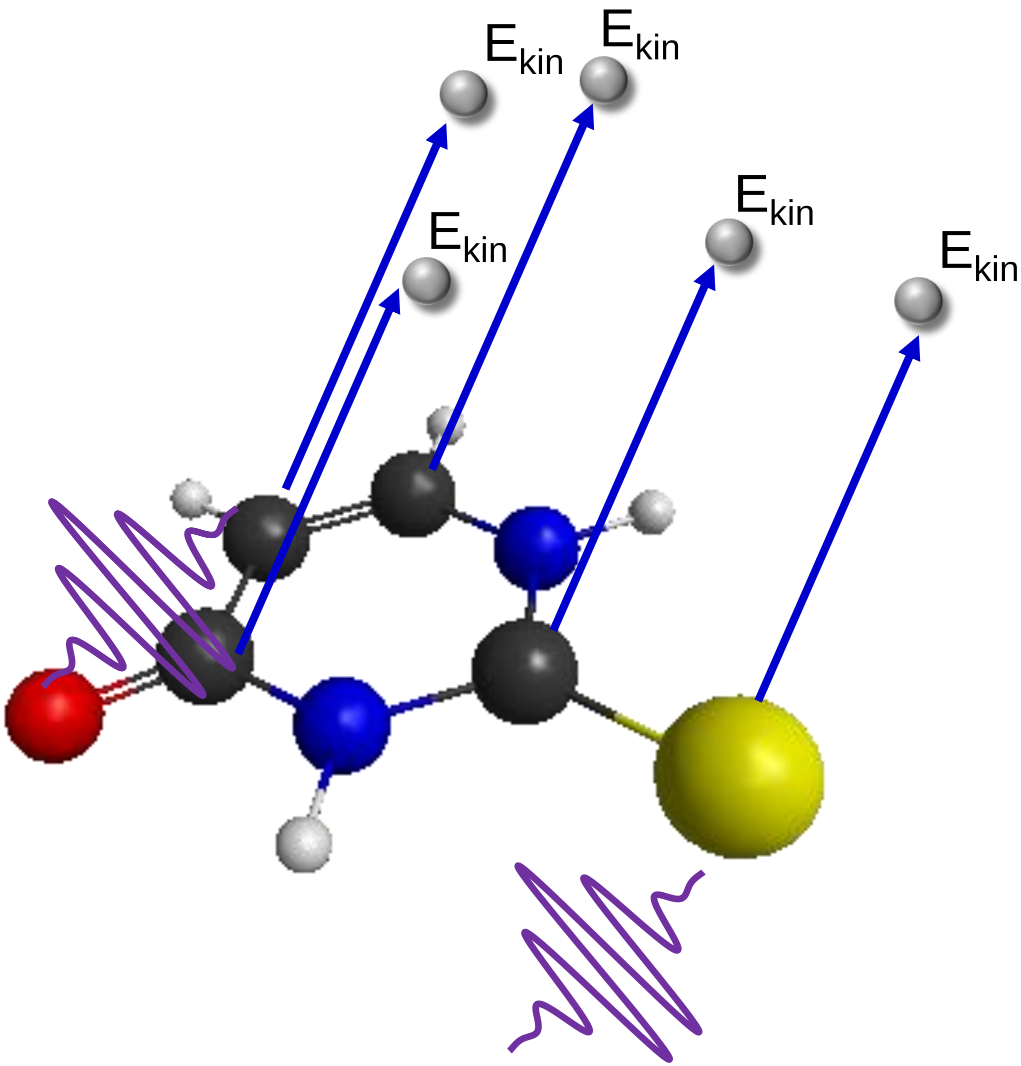
D. Mayer[1,11], F. Lever[1,11], D. Picconi[2✉], J. Metje[1], S. Alisauskas[3], F. Calegari[4,5,6], S. Düsterer[3], C. Ehlert[7], R. Feifel[8], M. Niebuhr[1], B. Manschwetus[3], M. Kuhlmann[3], T. Mazza[9], M. S. Robinson[1,4,5], R. J. Squibb[8], A. Trabattoni[4], M. Wallner[8], P. Saalfrank[2], T. J. A. Wolf[10] & M. Gühr[1✉]

You can use that instrument a FLASH

Potsdam, FLASH Feb. 21st 2019

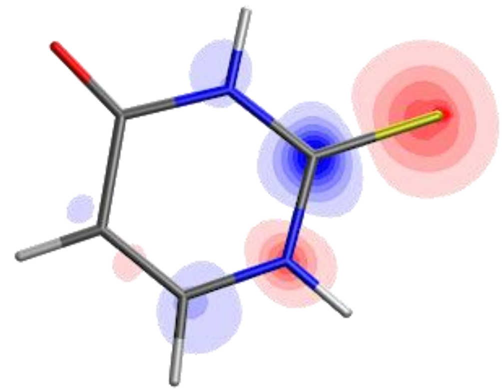
Hamburg, FLASH Feb. 21st 2019

Now: Deduce valence charge changes at Carbon sites

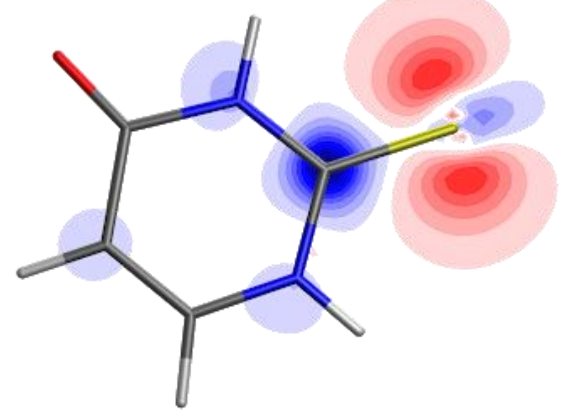


Charge density change

$\pi\pi^*$



$n\pi^*$



Now: Deduce valence charge changes at Carbon sites

We have PhD and PostDoctoral Positions available!

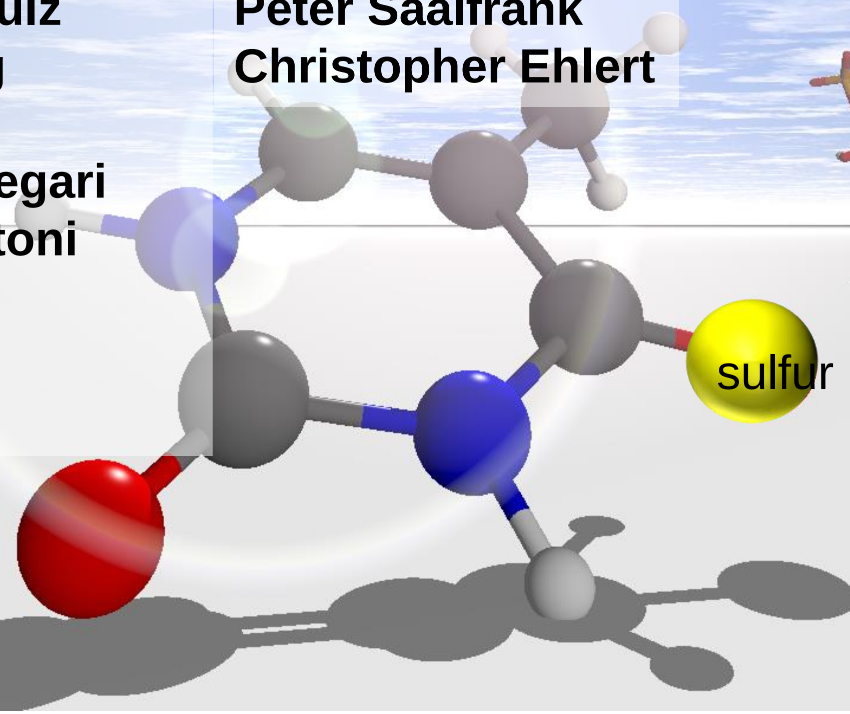
Contact Dennis or Fabiano or me (markus.guehr@desy.de)

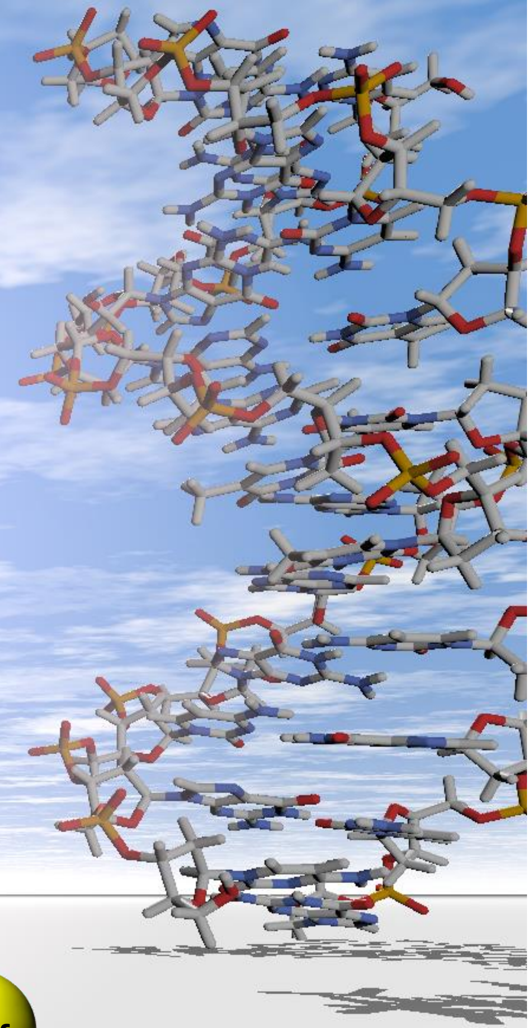
Thionucleobase collaboration FLASH

DESY
Stefan Düsterer
Skirmantas Alisauskas
Marion Kuhlmann
Giovanni Cirmi
Sebastian Schulz
Ulrike Frühling
Atia Tul Noor
Francesca Calegari
Andrea Trabattoni
Ingmar Hartl
Agata Azzolin
Markus Gühr

SLAC:
Alice Green

Potsdam Theory:
David Picconi
Peter Saalfrank
Christopher Ehlert





Bundesministerium
für Bildung
und Forschung

FLASH 2

20-400 eV in fundamental
3rd harmonic

3 Beamlines – 1 is Monochromatic
Variable gap undulators

X-ray split and Delay

Optical laser

Fixed REMI instrument for AMO

Facility operated instruments for AMO, Chem, CM, HED

FLASH 1

20-300 eV in fundamental
3rd harmonic

3 Beamlines – 1 is Monochromatic
Fixed gap undulators

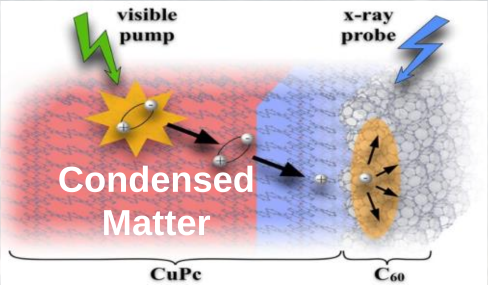
X-ray split and Delay

Optical laser

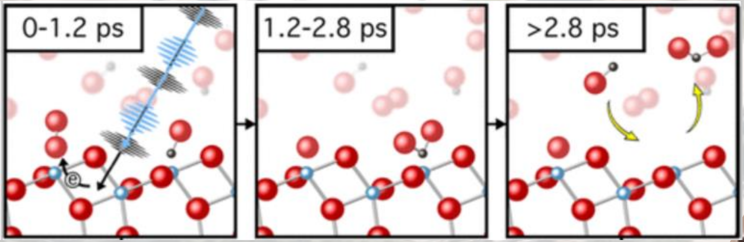
Fixed RIXS instrument for CM
Fixed CAMP instrument for AMO

Facility operated instruments for AMO, Chem, CM, HED

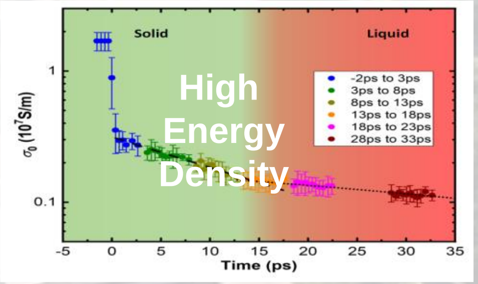
J. W. L. Lee, et al.; Nature Communications, 2021



F. Roth et al.; Nature Communications, 2021



Wagstaffe et al., ACS Catal. 2020, 10, 13650−13658

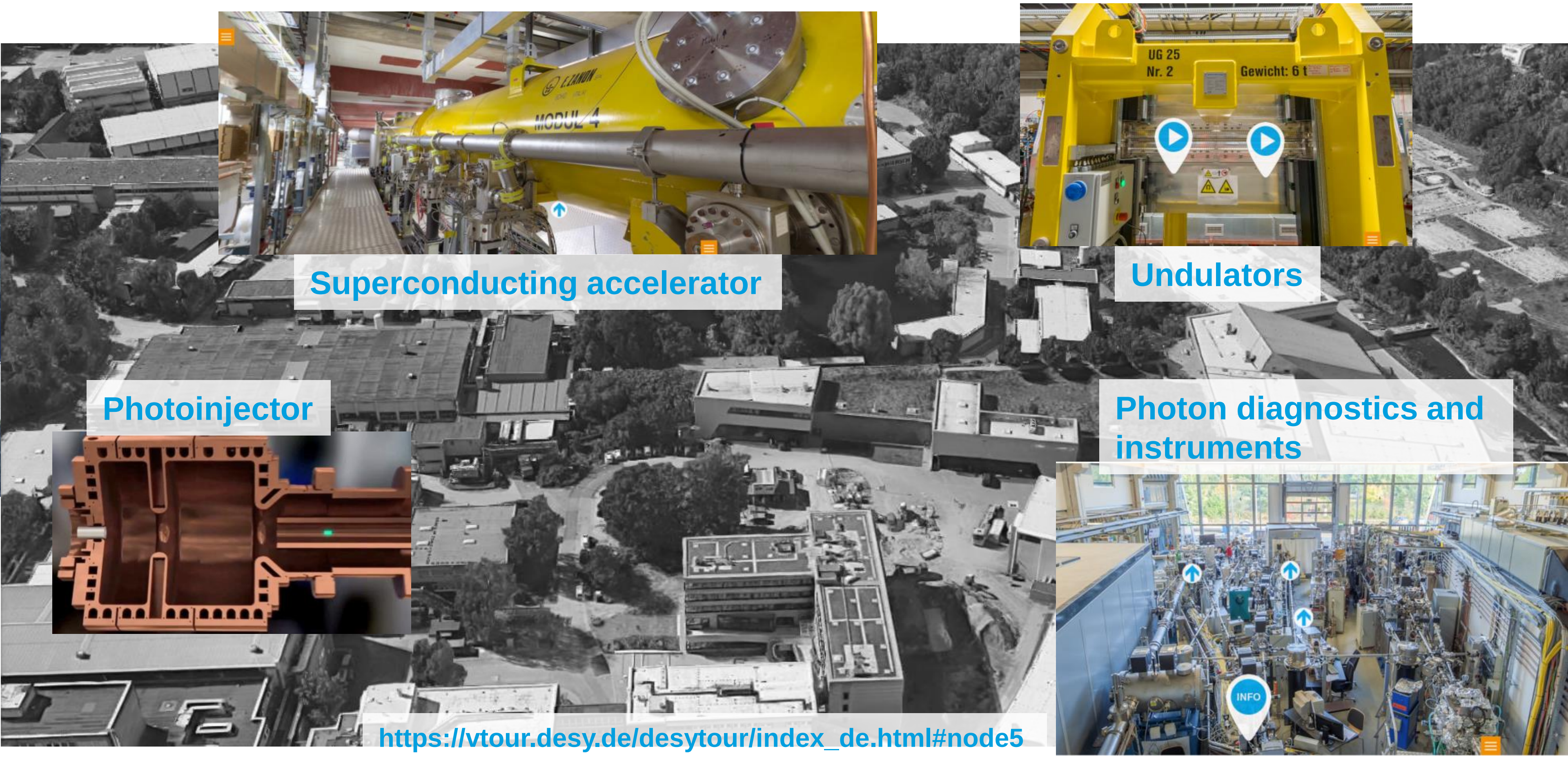


Z. Chen et al.; Nature Communications, 2021

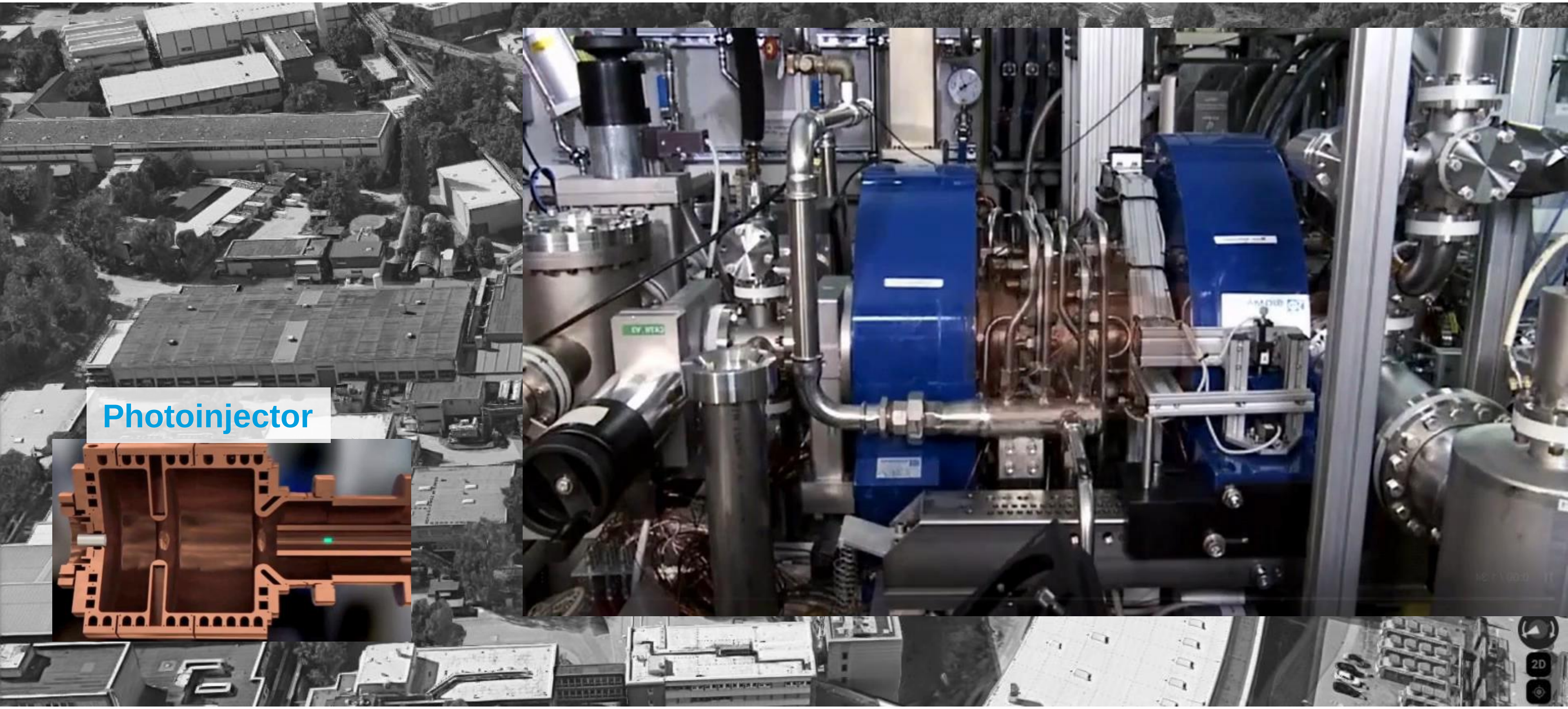
FLASH overview

Google Maps

FLASH overview

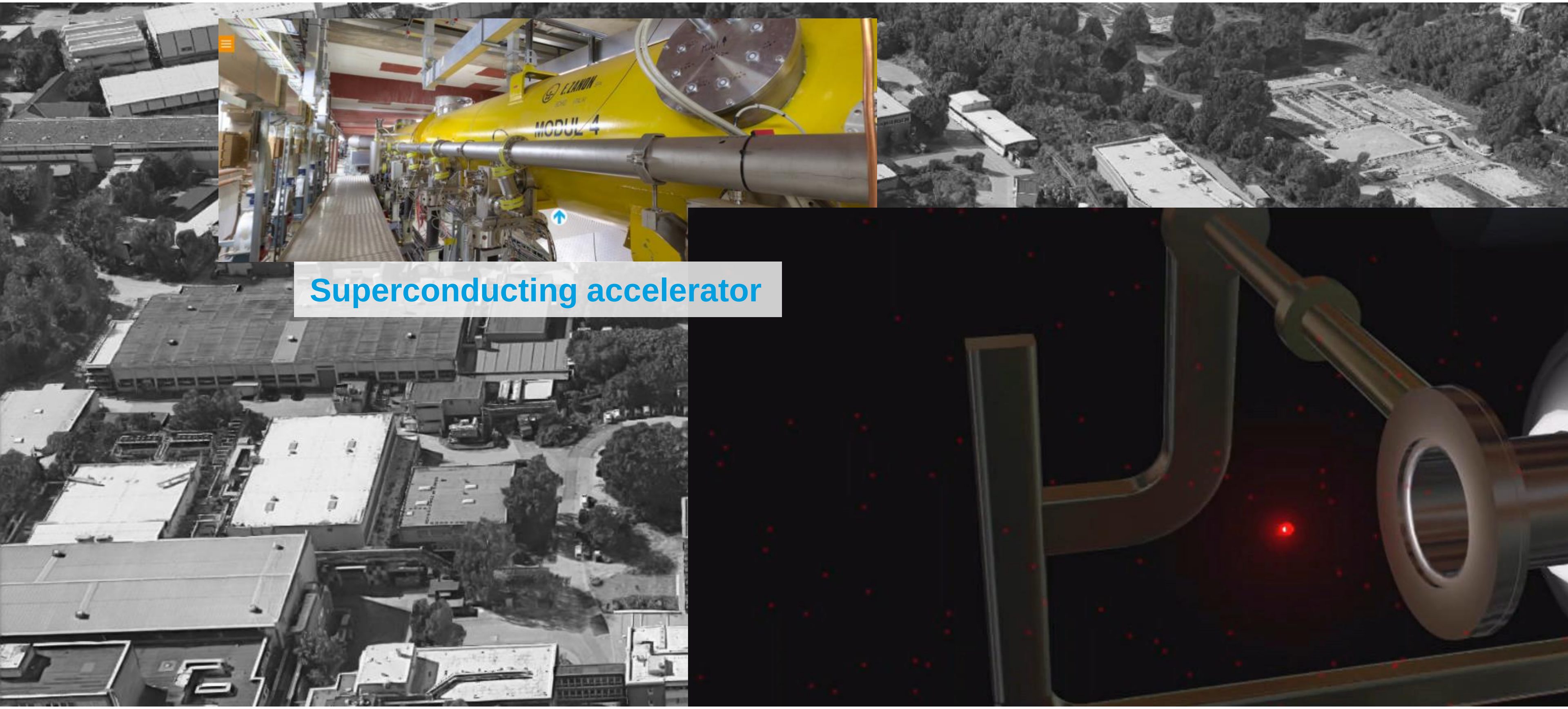


FLASH overview



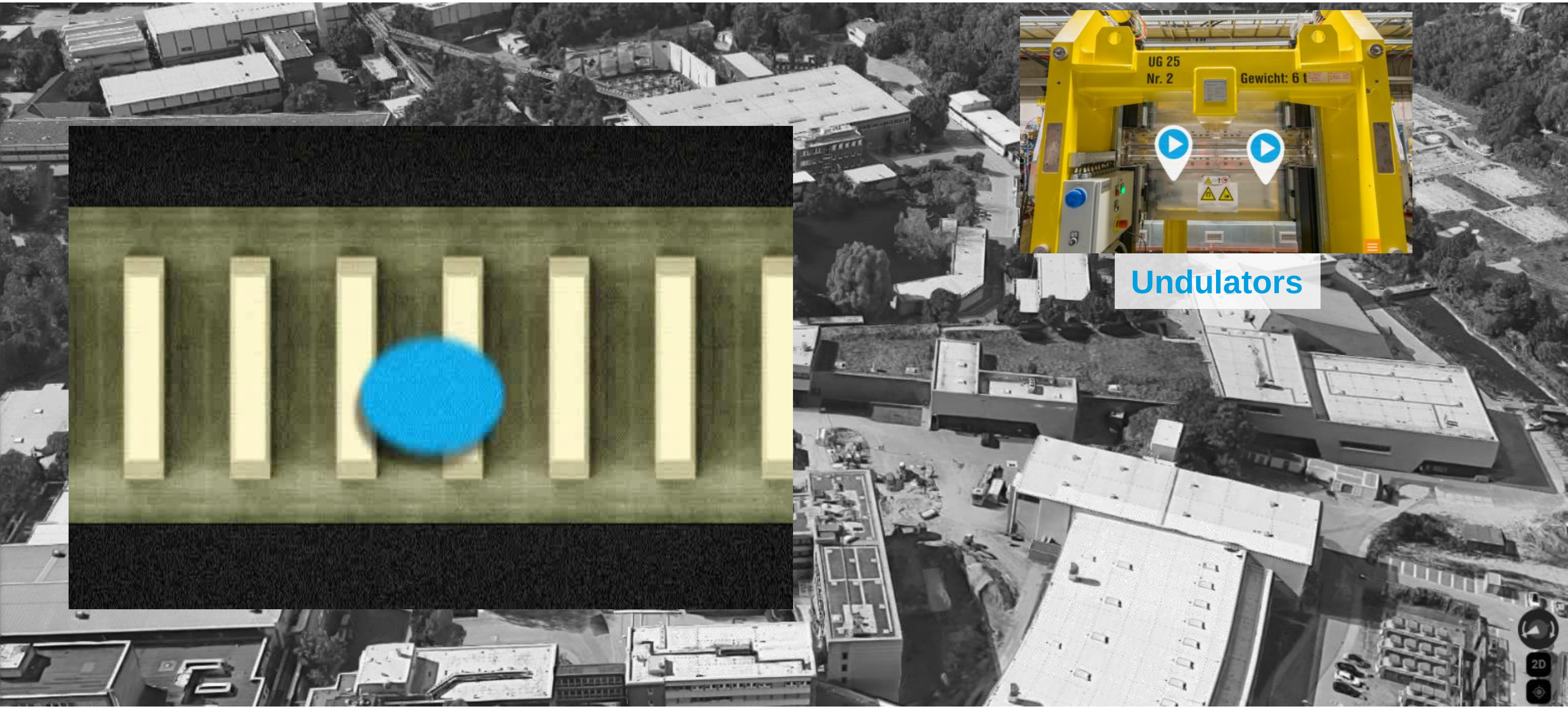
https://vtour.desy.de/desytour/index_de.html#node5

FLASH overview



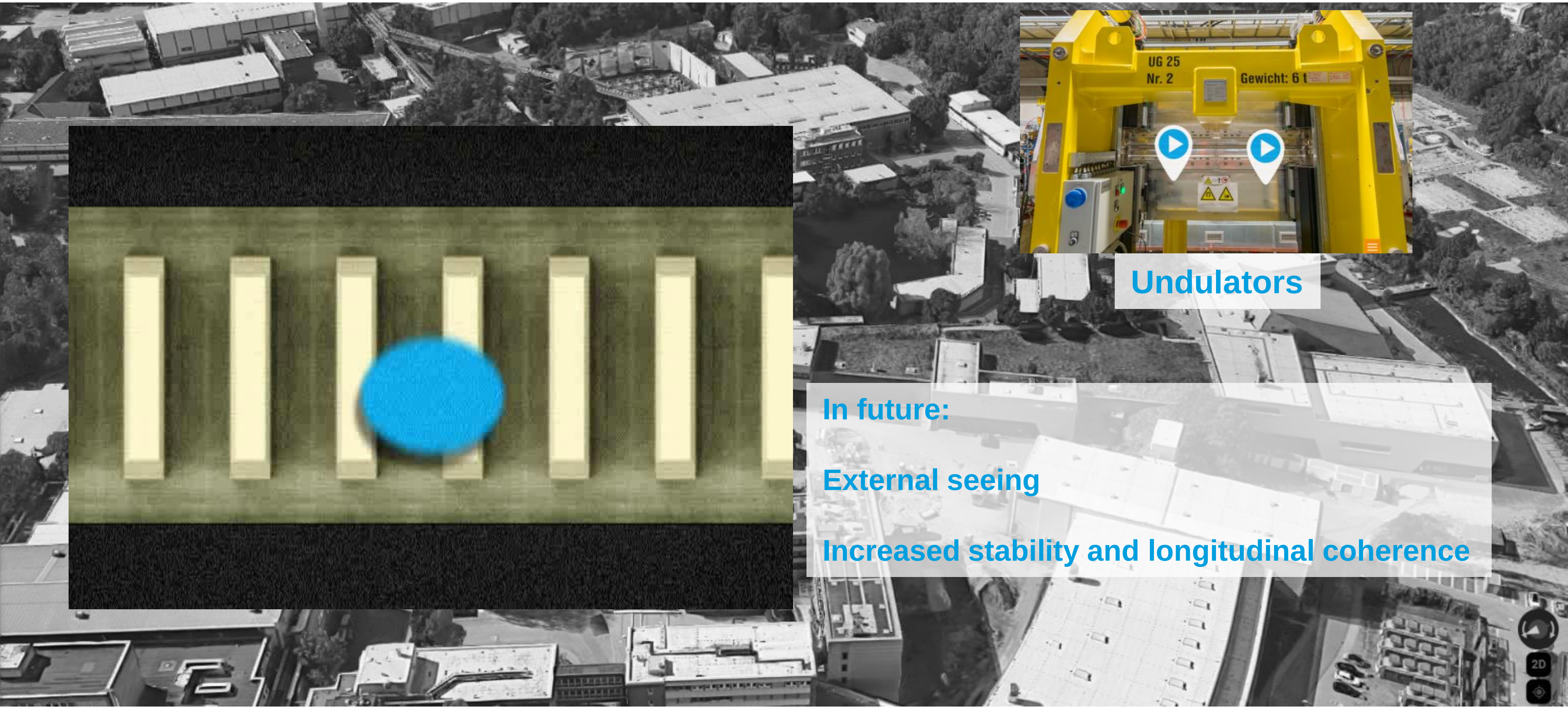
https://vtour.desy.de/desytour/index_de.html#node5

FLASH overview



https://www.youtube.com/watch?v=RG-PYmeq2XE

FLASH overview



Undulators

In future:

External seeing

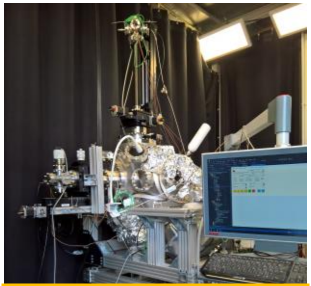
Increased stability and longitudinal coherence

https://www.youtube.com/watch?v=RG-PYmeq2XE

FLASH efficiency: facility operated instrumentation

REMI (AMO)
Ulrike Frühling, Markus Braune

CAMP (AMO/CHEM)
Benjamin Erk



URSA-PQ (AMO, CHEM) **Markus Gühr**

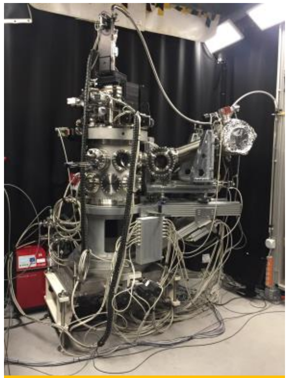
FLASH currently operates 3 fixed and 5 transportable instruments

5 more instruments are under construction

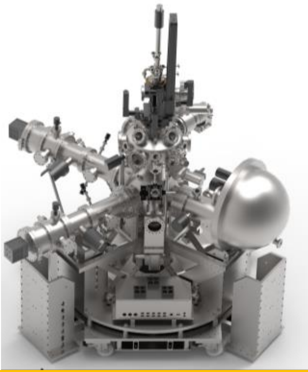
Transportable instruments can also be used in other onsite labs

Starting more unified controls and analysis suite for users

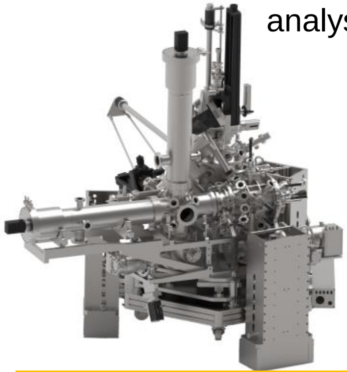
MULTIP (WDM, AMO)
Sven Toleikis



MUSIX (Materials)
Martin Beye



WESPE (Materials, Catal.)
Dima Kutnyakhov

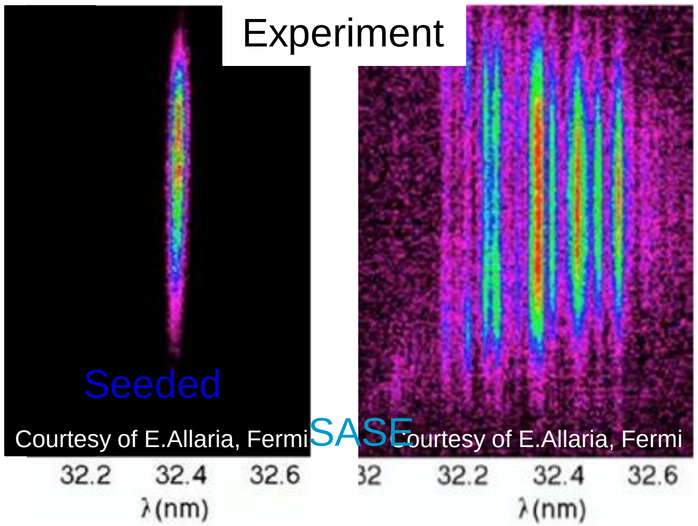


HEXTOF (Materials)
Dima Kutnyakhov

Coutesy: Rolf Treusch

Difference between seeded and SASE pulses
High repetition rate and external seeding in 2025

High repetition rate seeding of soft-X-ray pulses

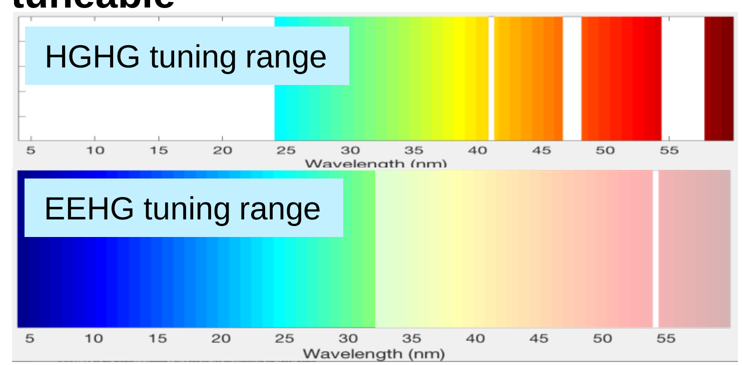


***Combination of HGHG and EEHG*:**

Fully coherent pulses with
variable wavelength (60 – 4 nm)
tens of fs duration and
1 MHz intra-bunch repetition rate

***Apple III undulators*:**

Variable polarization independently tuneable



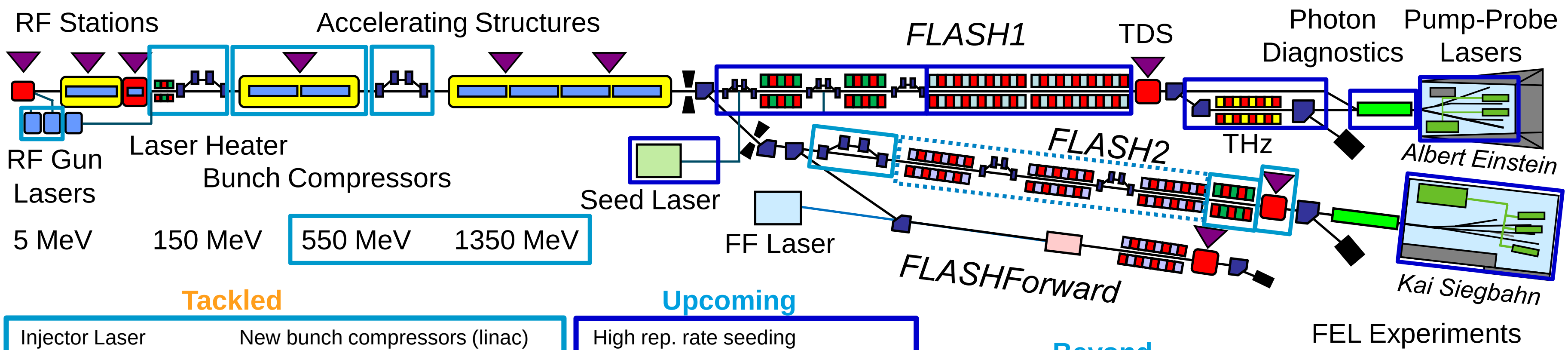
Seeded

- Narrow bandwidth
- Stability
- Longitudinal coherence
- Brilliance
- Laser controlled pulse properties
- Synchronisation to seed laser

DESY Photon Science Strategic Goals

FLASH2020+

FLASH: towards a high repetition rate seeded soft X-ray FEL



Tackled

Injector Laser	New bunch compressors (linac)
Energy upgrade	Variable gap undulators (FLASH2)
3rd BC FLASH2	interim pump-probe laser (FLASH1)
TDS (FLASH2)	
Laser heater	New beamline FL23 (FLASH2)
	Afterburner FLASH2

Upcoming

High rep. rate seeding (FLASH1)
Photon diagnostics (FLASH1)
Flexible pump-probe
New beamlines

Beyond

New undulator schemes (FLASH2)
New lasing concepts (FLASH2)

2021 2022 2023 2024 2025

nitrogen K-edge

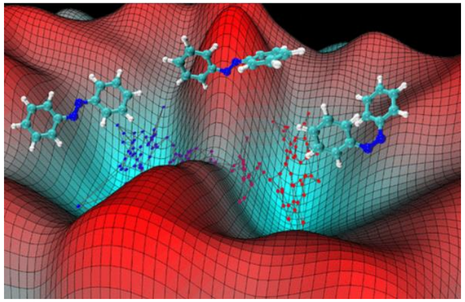
Variable polarization at third harmonic

Externally seeded operation up to the carbon K-edge

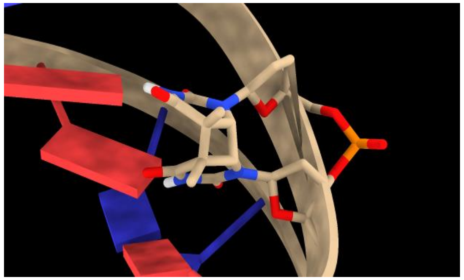
Markus Gühr

FLASH 2020+ Science challenges

Utilizing the unique properties of the new FLASH



Ground state isomerization of azobenzene
Tavadze et al. JACS 2018



UV – induced lesion in DNA

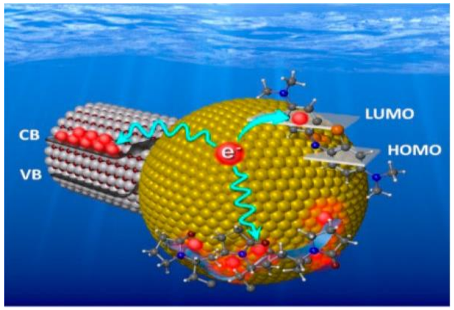
Examples from the FLASH strategy workgroups utilizing the high coherence/spectral brightness and repetition rate:

Ground state chemisty (rare events)
catch the rare events where a reaction happens

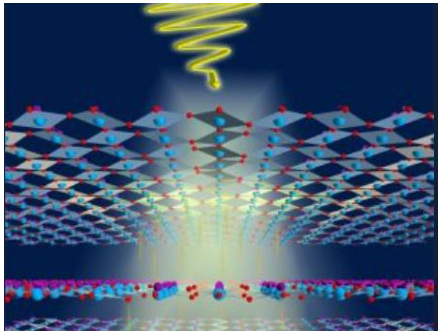
Life‘s photoprotection (dilute samples with traps)
Learning how nature protects itself from UV damage

Plasmonic photocatalysis (dilute samples, brightness)
Harvest sunlight via nanomaterials to accomplish green chemistry

Materials phase transition dynamics/control (brightness)
Understand material transformations in fields for future electronics



Zhang *et al.*, Chem. Rev. **118**, 2927 (2018)



http://qcmd.mpsd.mpg.de/index.php/research/research-science/Light-induced-SC-like-properties-in-cuprates.html

Markus Gühr