Workshop "New Scientific Capabilities at European XFEL"
DESY, Hamburg
2019-03-26

Watching and steering electrons with intense optical and free-electron lasers

Thomas Pfeifer
MPIK Heidelberg, Germany

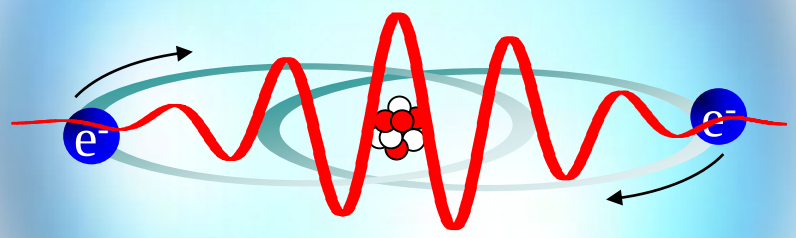
few-body quantum dynamics

a fundamental scientific question:

"how do two or more excited electrons move and interact in atoms and molecules?"

spatial scale
R ~ sub/few Å

temporal scale
T ~ sub/few fs



The
"**quantum few-body problem**"
in strong fields

Scientific goal:

measure / understand / control
*the **quantum dynamics** of*
few-body systems
in strong fields

(x-ray) movies of single molecules

Petahertz-clocked computing

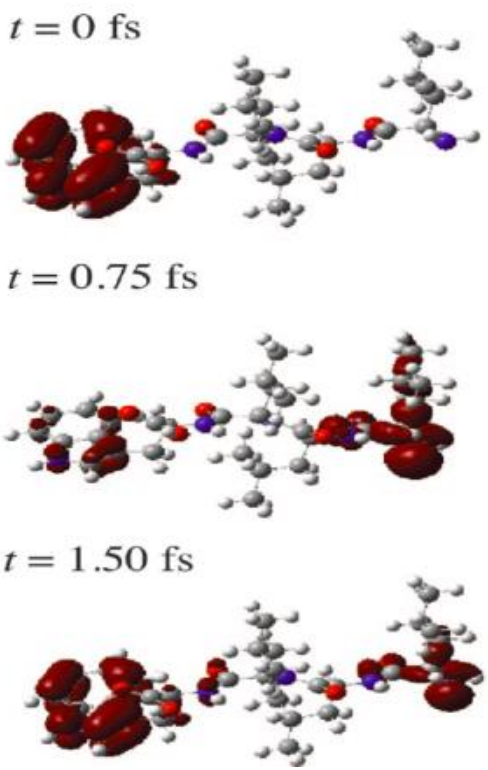
x-ray precision spectroscopy

Laser control of chemical reactions

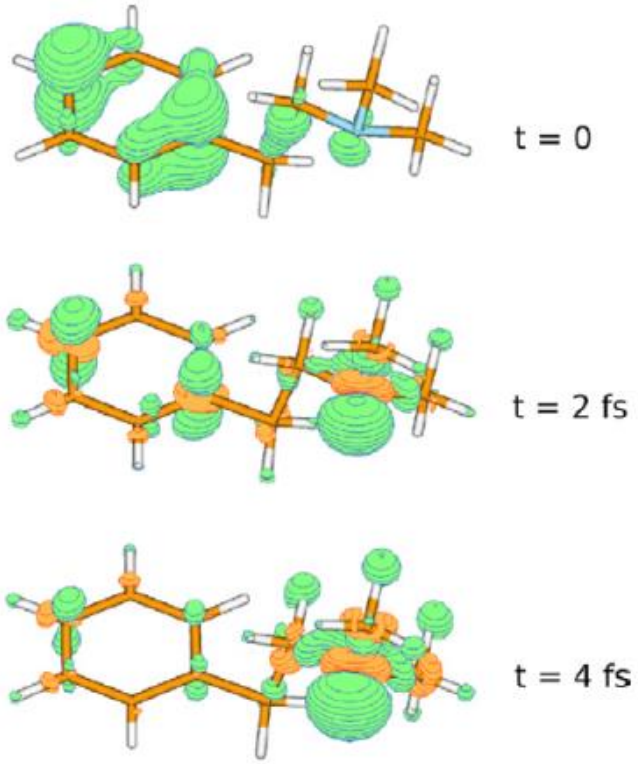
Motion of electrons in molecules

… it's fast…

… and it‘s (even more) complex!

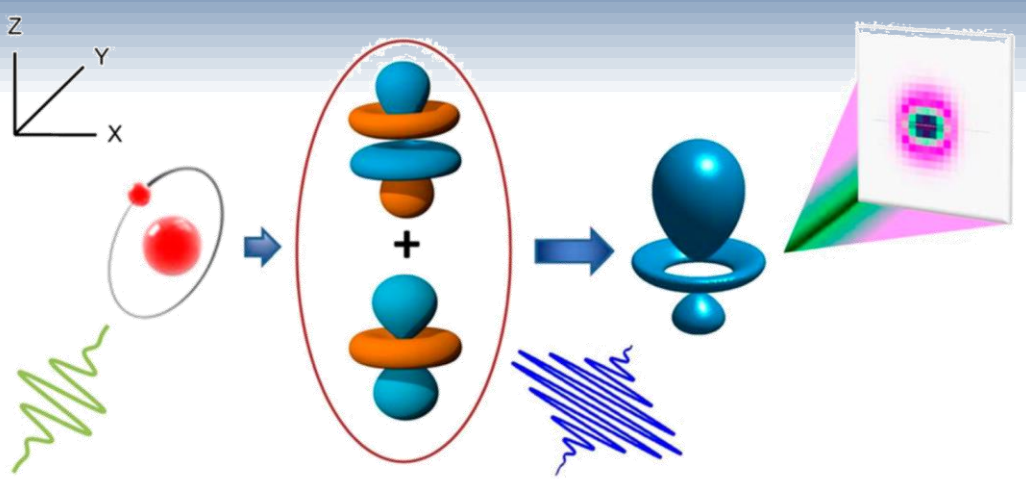


Remacle, Levine
PNAS 103, 6793 (2006)



Lünnemann, Kuleff, Cederbaum
CPL **450**, 232 (2008)

Imaging of electron dynamics

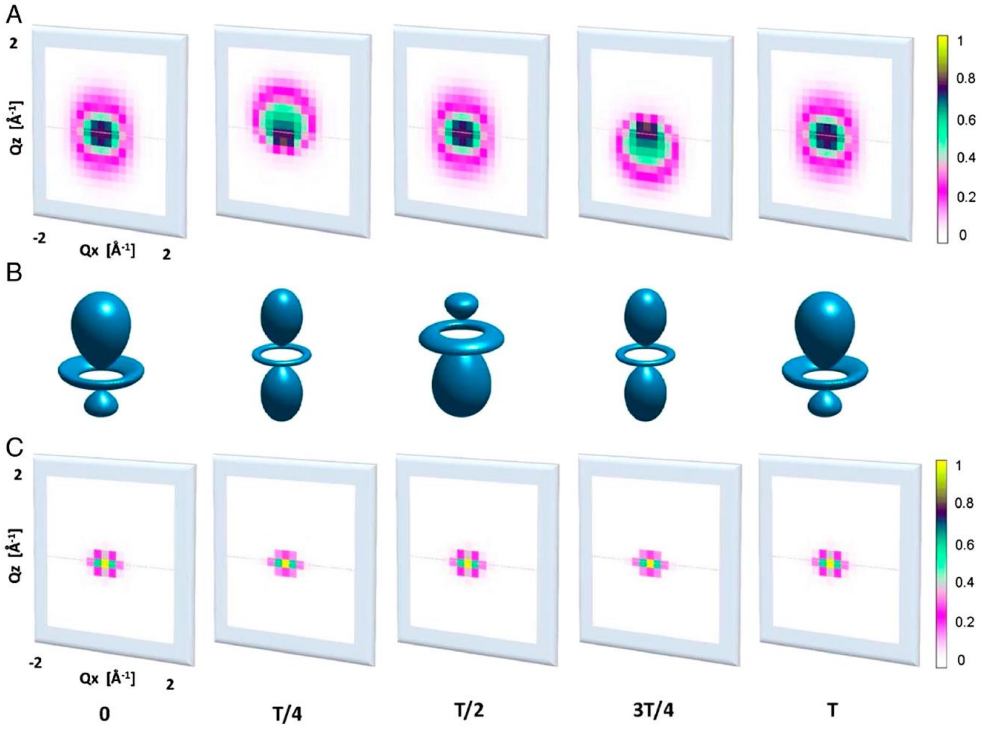


here:
single
electrons

Dixit, Vendrell, Santra PNAS **109**, 11636 (2012)

QED
theory

"classical"
theory

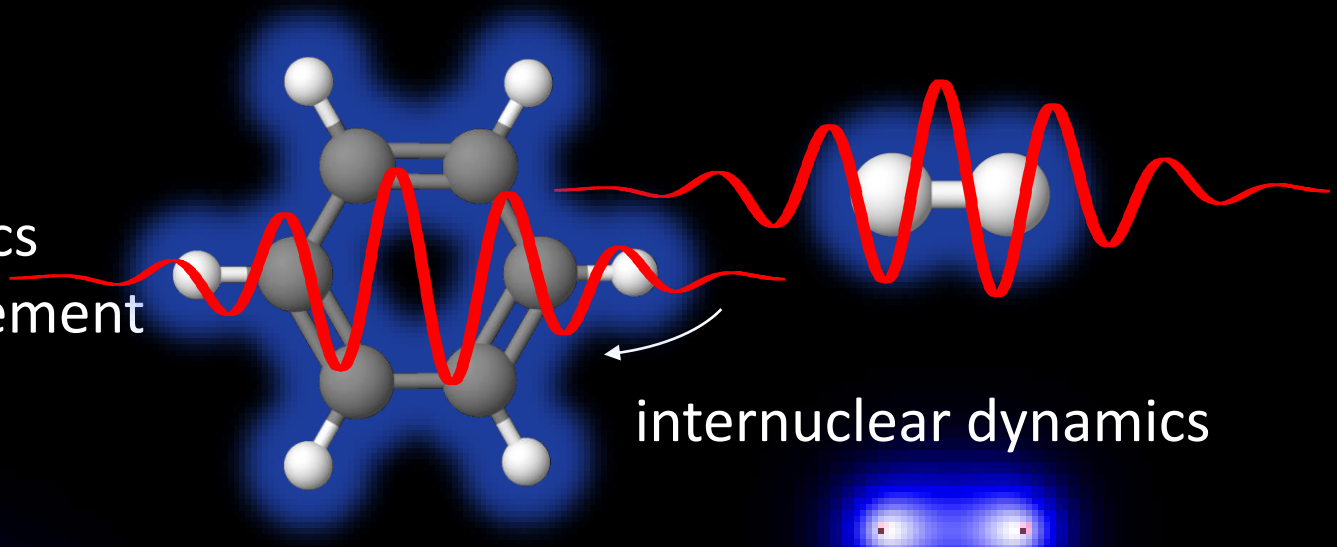


molecule images from molview.org

quantum dynamics in intense laser fields

… in molecules

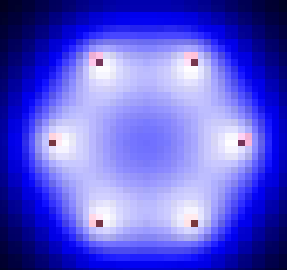
electron dynamics
e.g. charge rearrangement
 asec

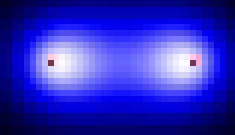


internuclear dynamics

e.g. dissociation

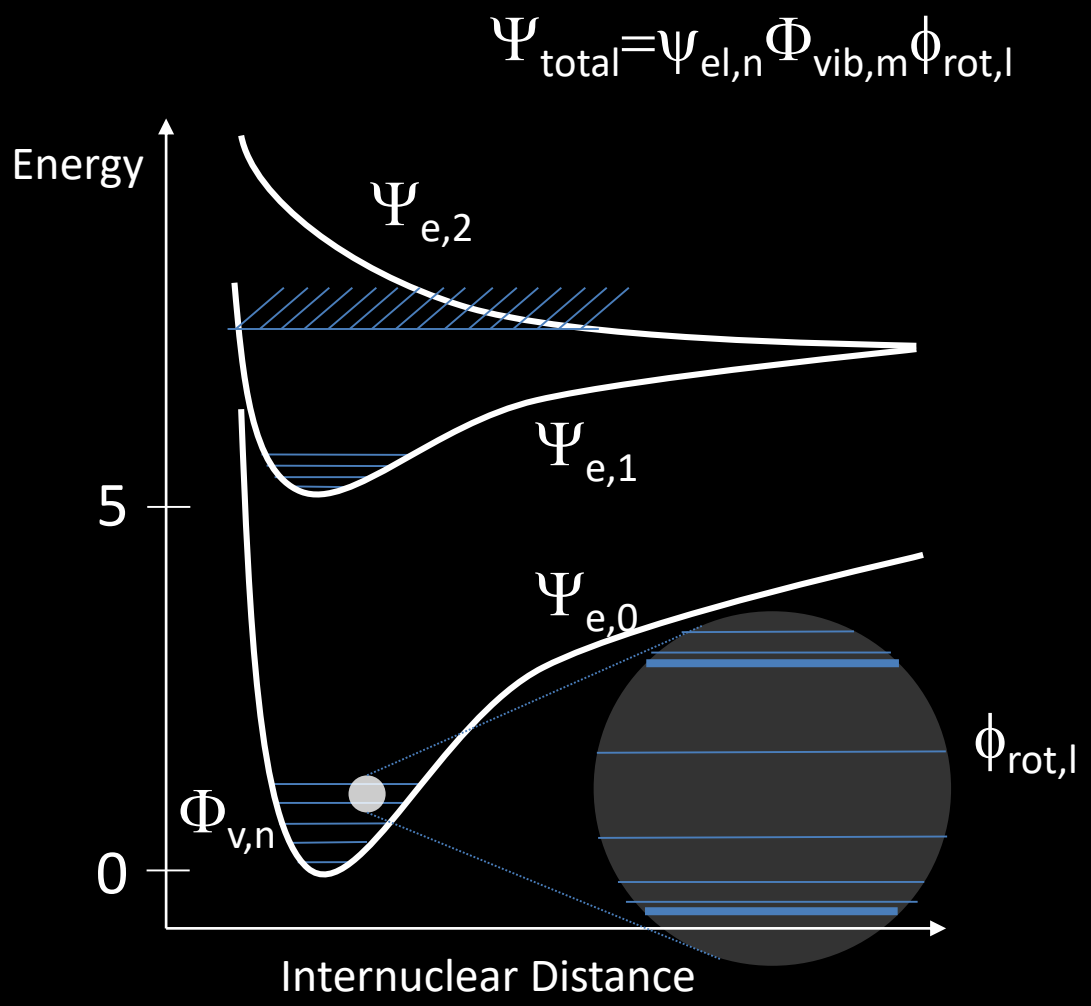
+ charge rearrangement





But what is a molecule, really...

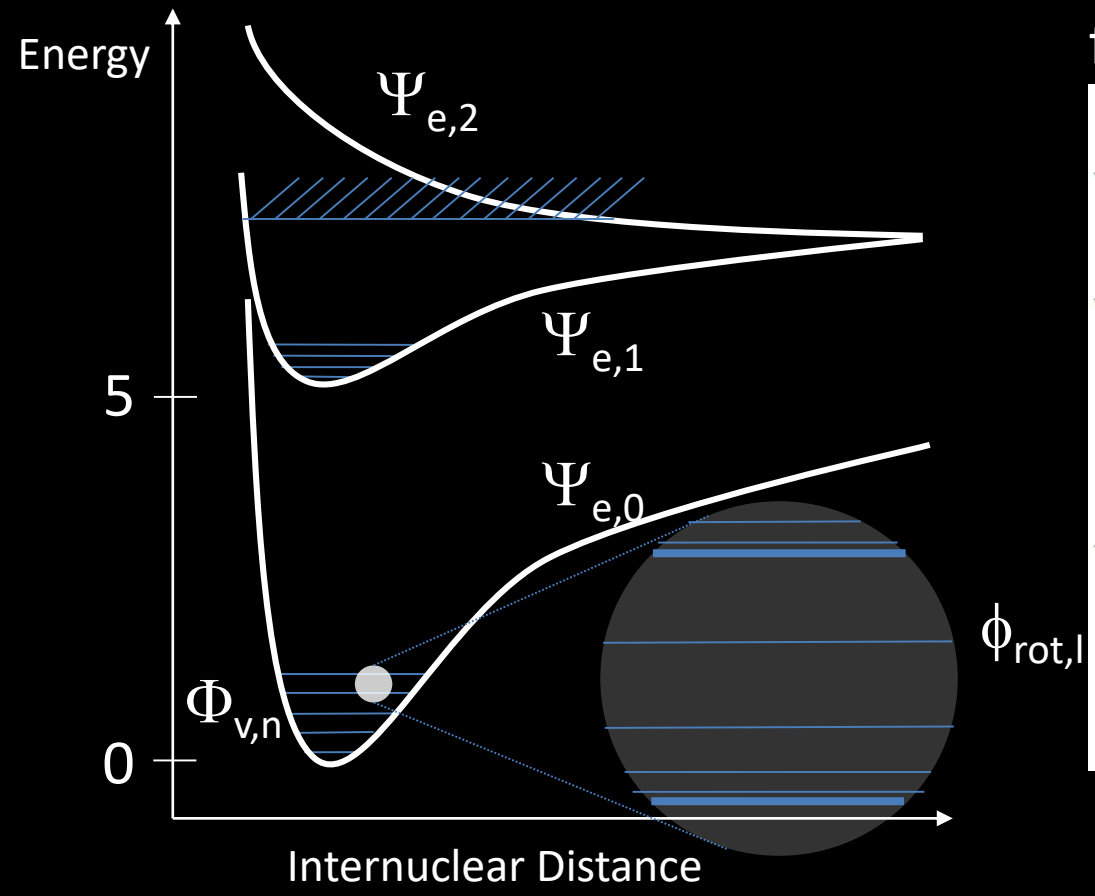
Separation: Electronic, Vibrational, Rotational



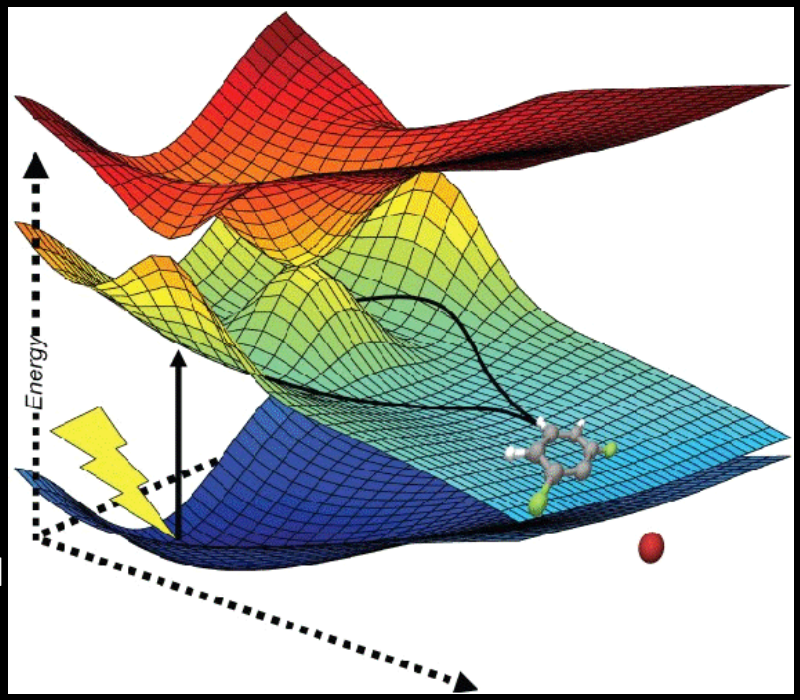
But what is a molecule, really...

Separation: Electronic, Vibrational, Rotational

$$\Psi_{total} = \psi_{el,n} \Phi_{vib,m} \phi_{rot,l}$$



for more than two atoms...



Tuckerman lecture, NYU

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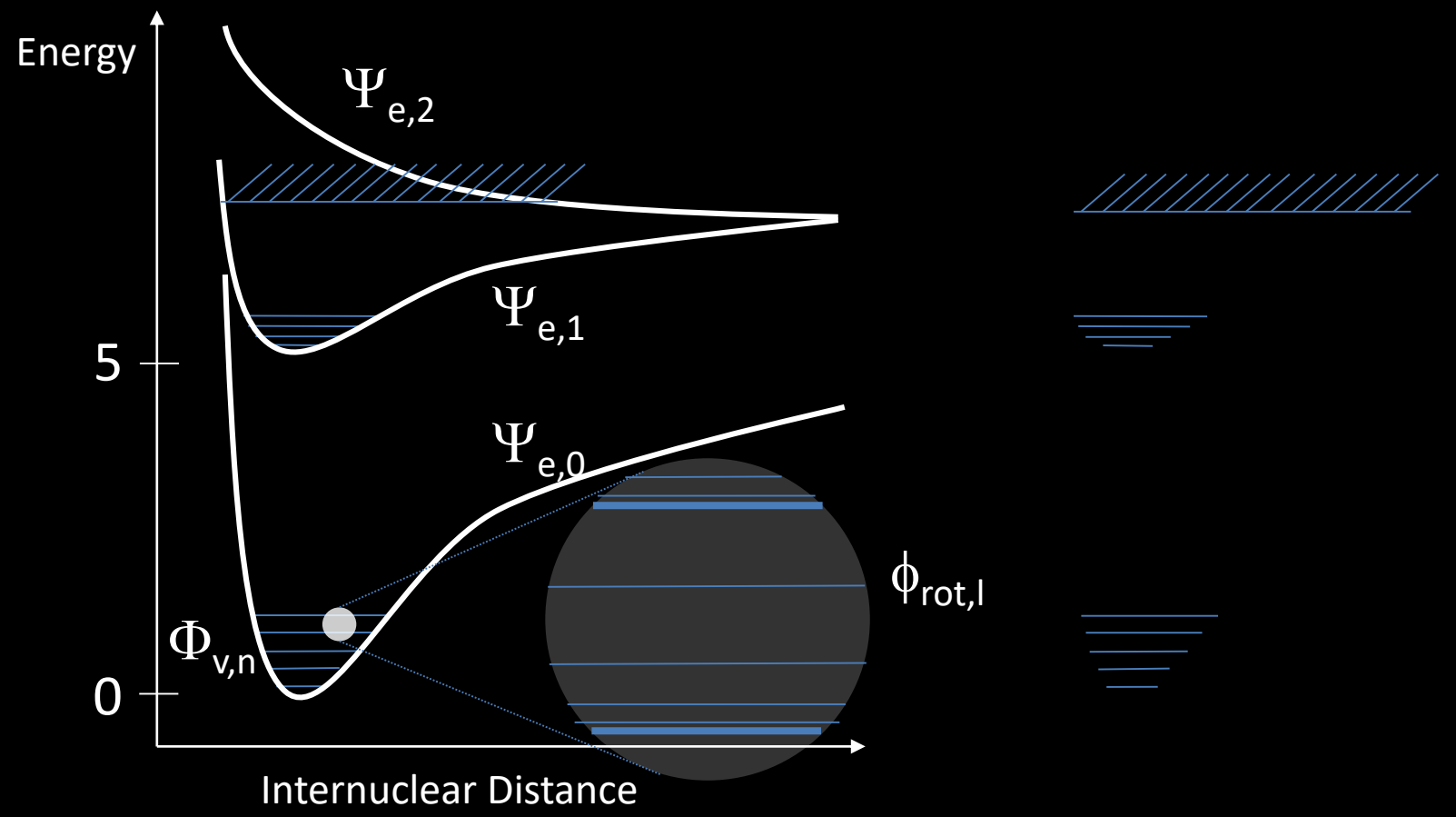

But what is a molecule, really...

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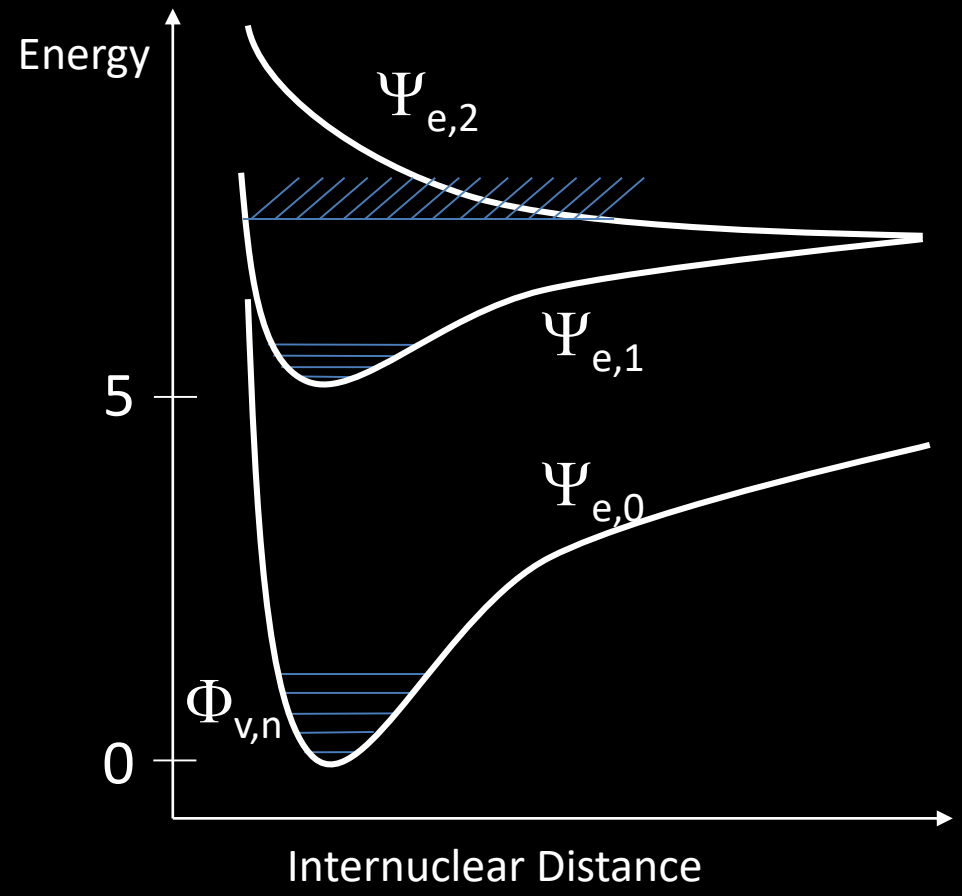
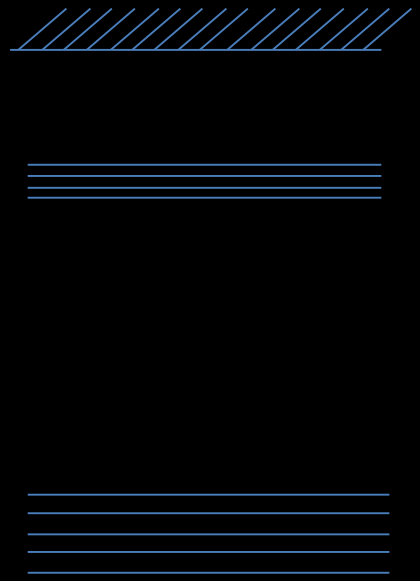
... just a set of levels ...





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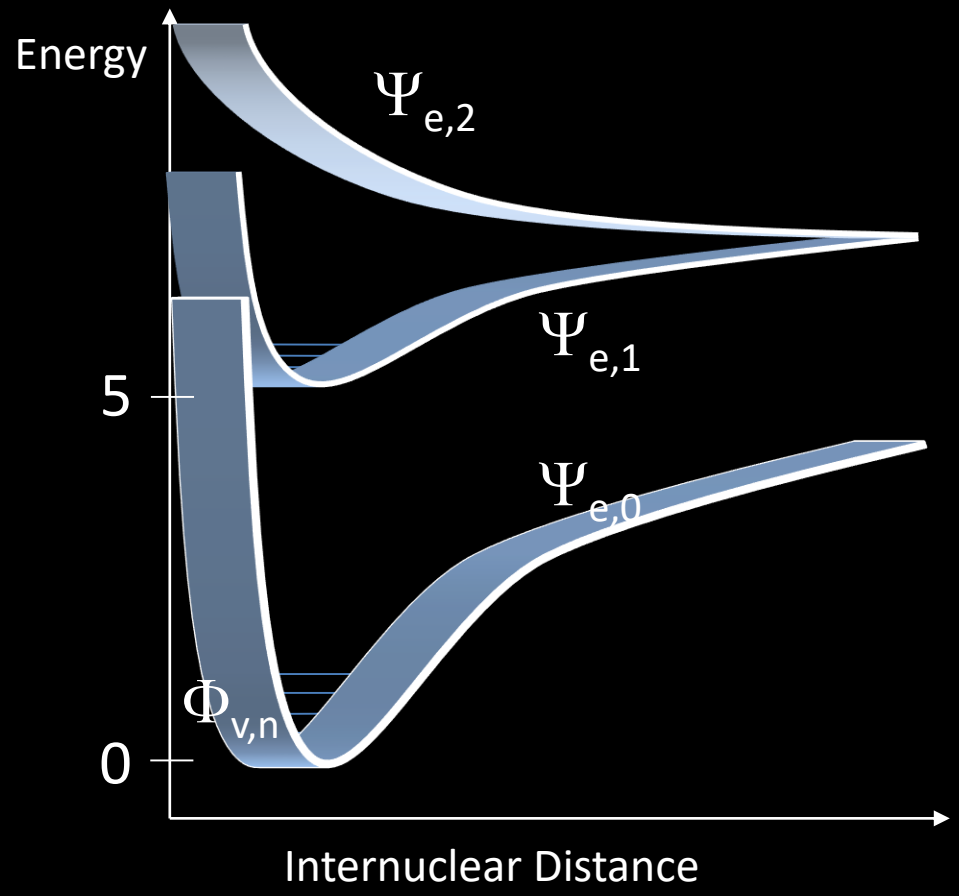

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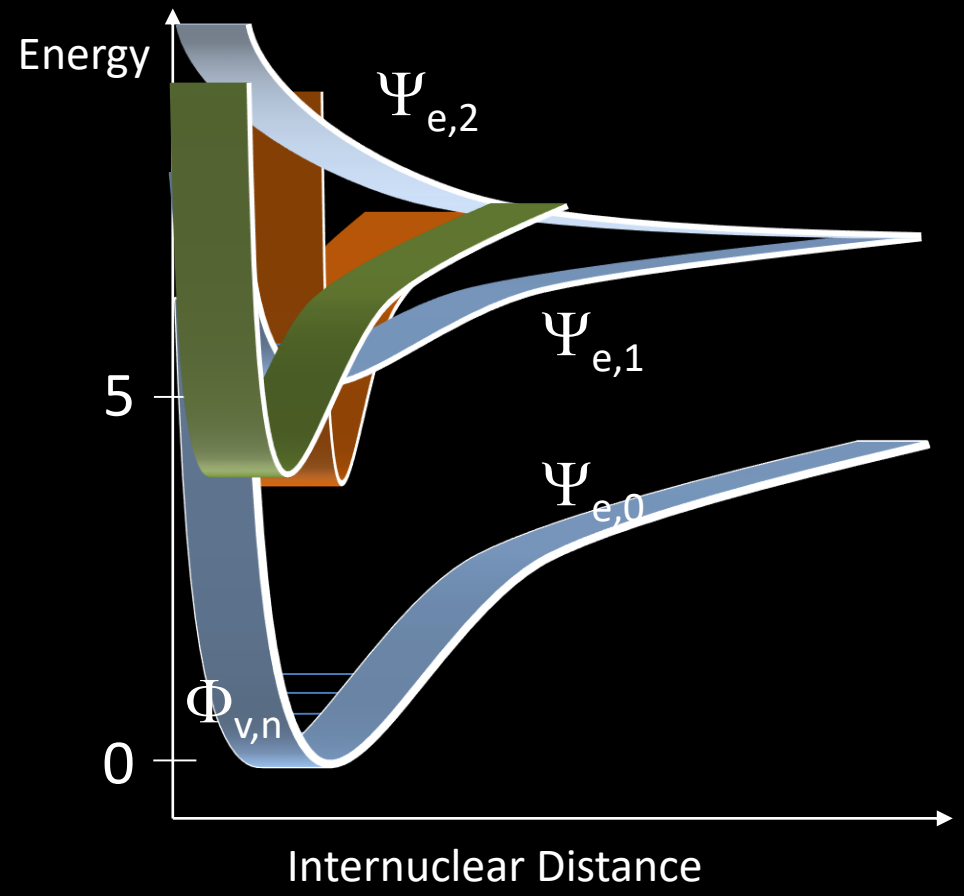
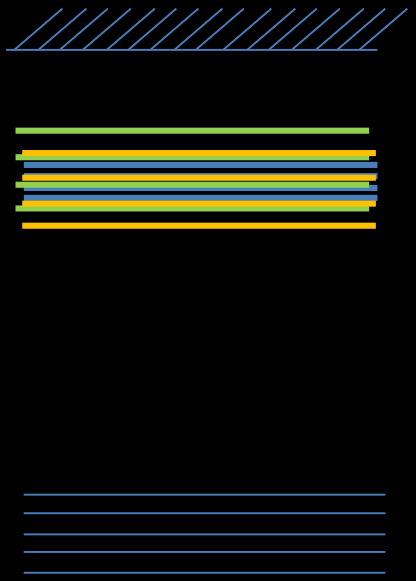
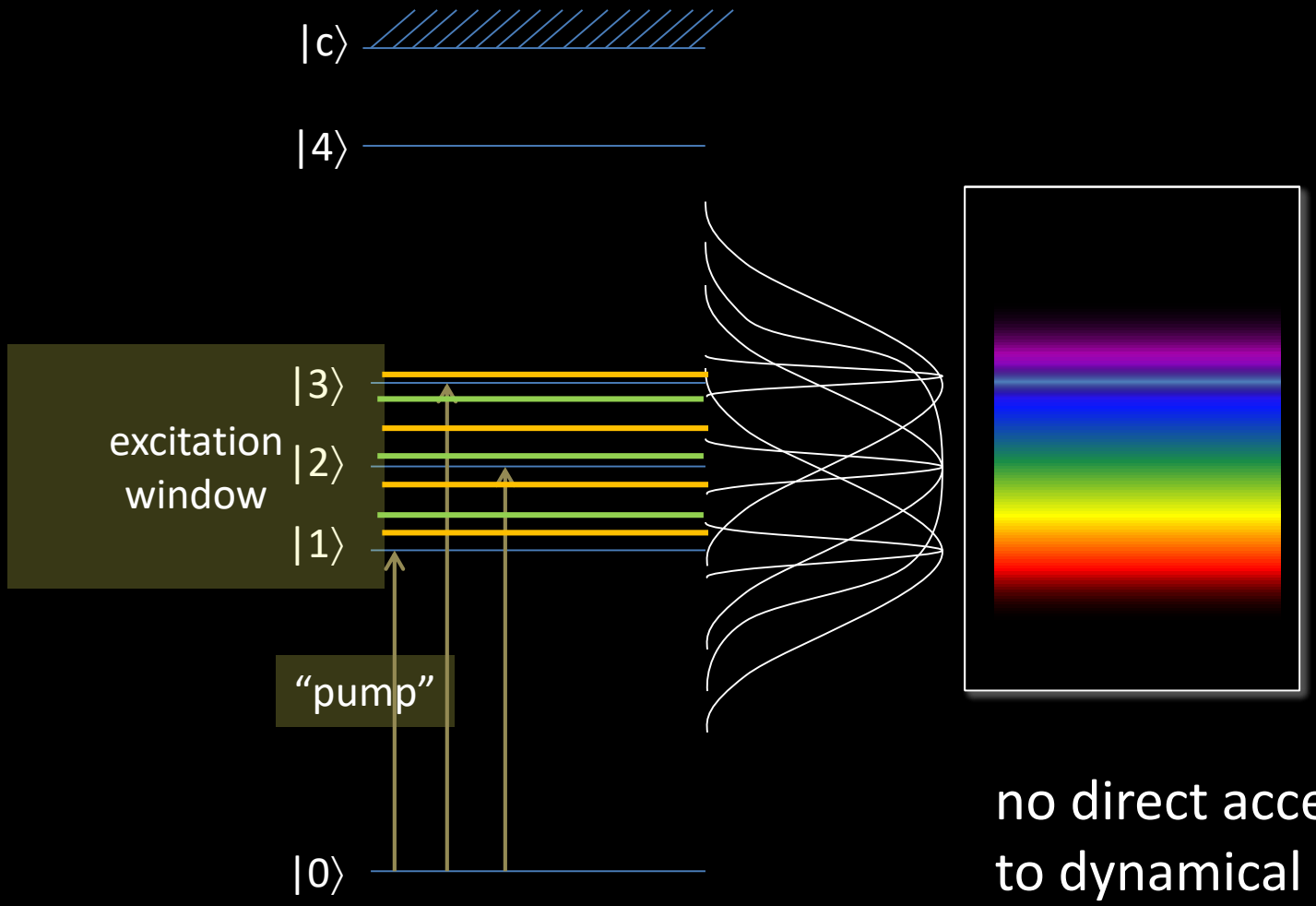


... just a set of levels ...

... <u>many levels</u>



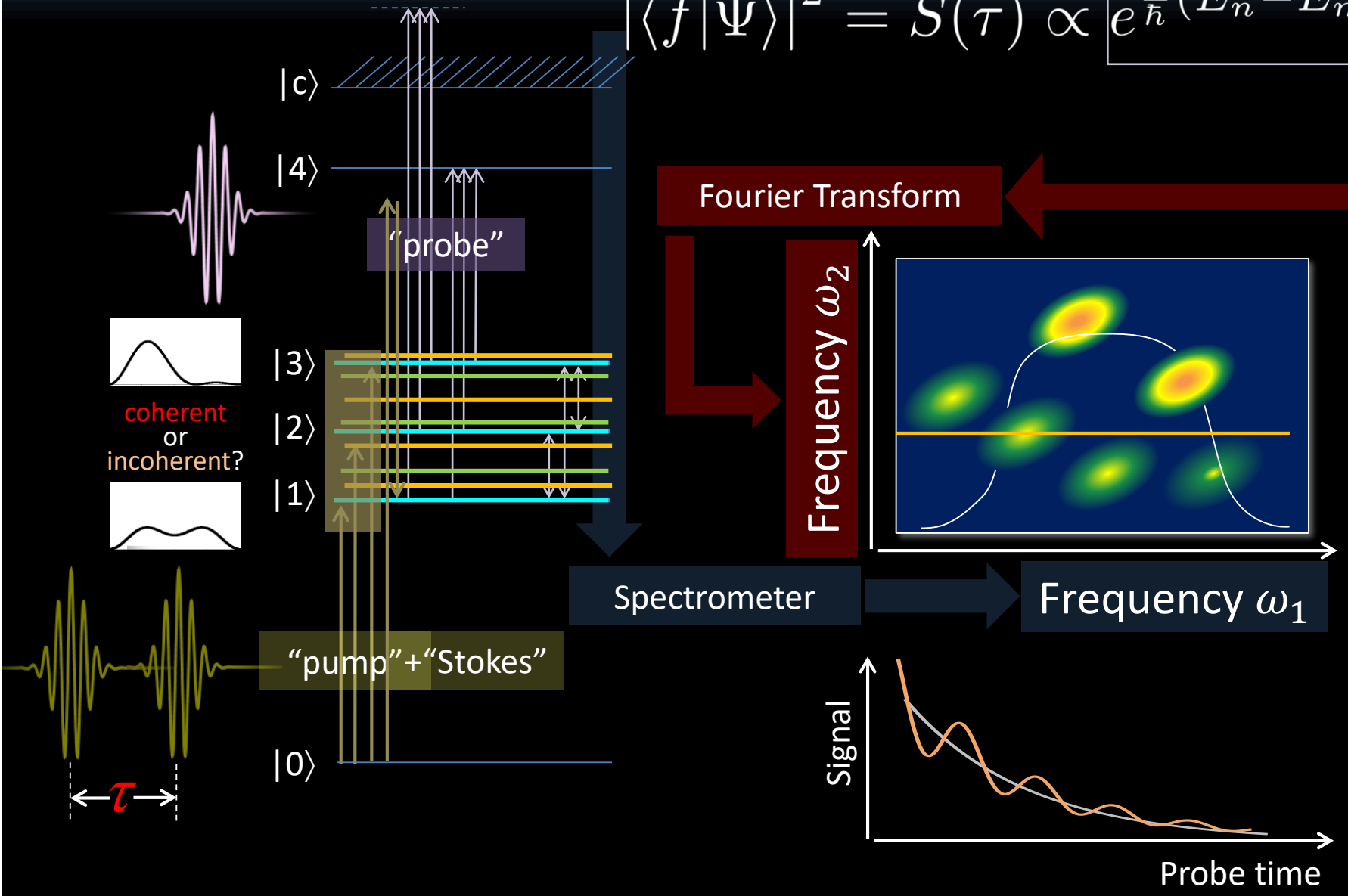
traditional (linear) spectroscopy limitations



no direct access
to dynamical information

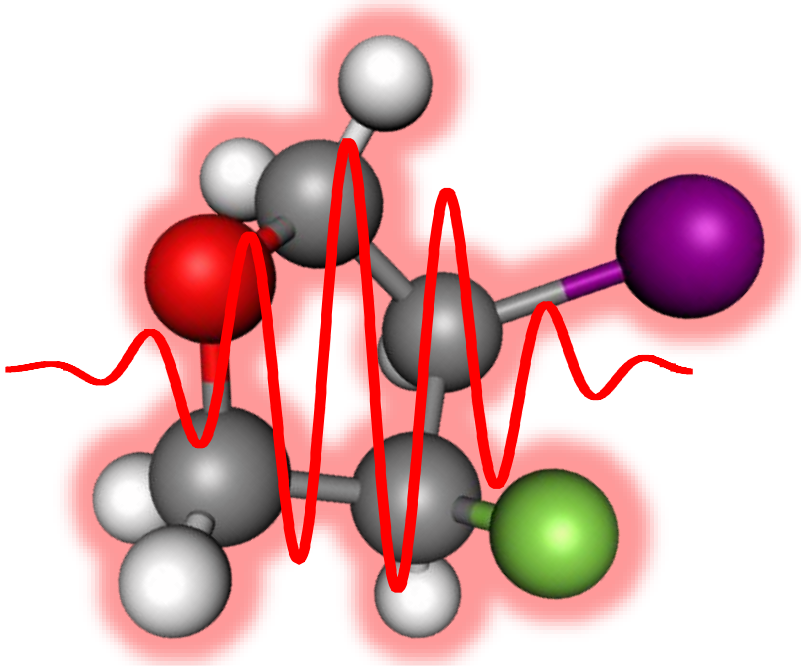
From pump-probe ...
to "two-dimensional" spectroscopy

$$|\langle f|\Psi\rangle|^2 = S(\tau) \propto e^{\frac{i}{\hbar}(E_n - E_m)\tau}$$



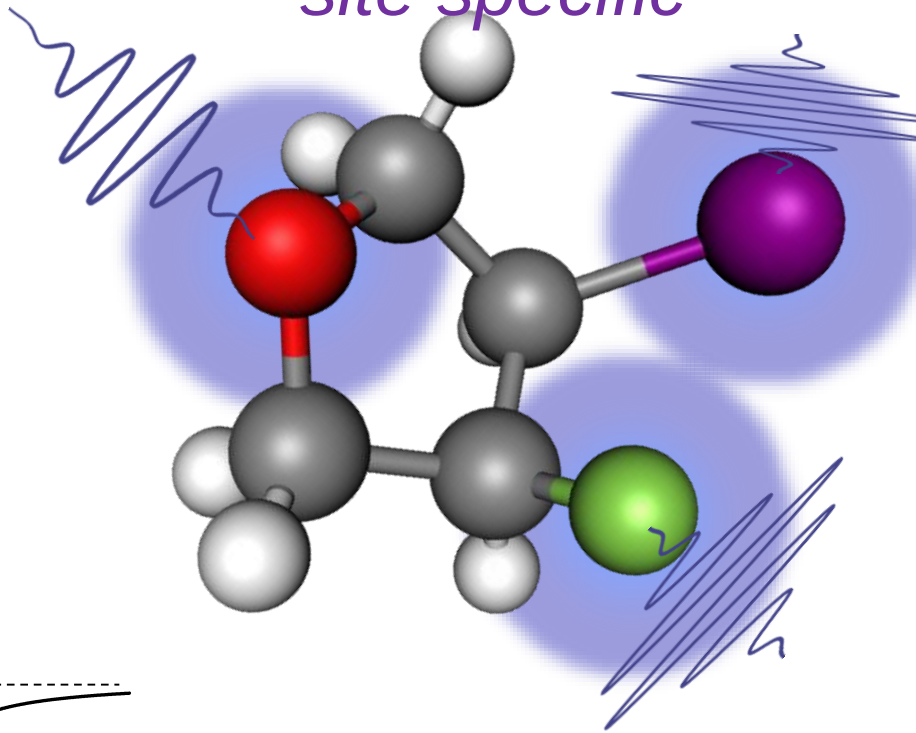
Interactions of molecules with light

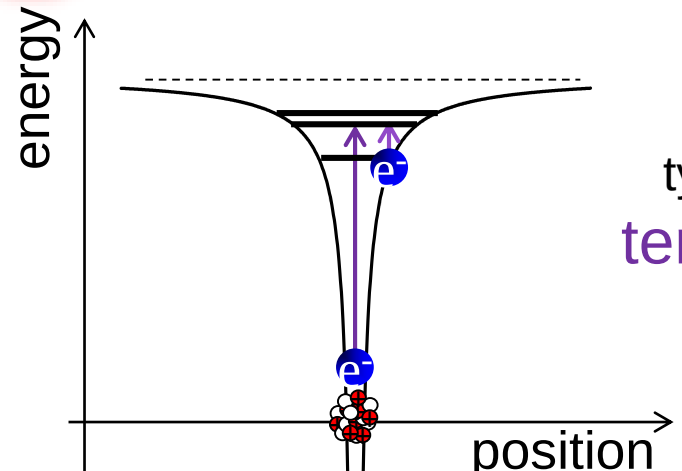
global excitation



local excitation

site specific





typical ΔE for core transitions:

ten(s) of **eV** to **few keV**

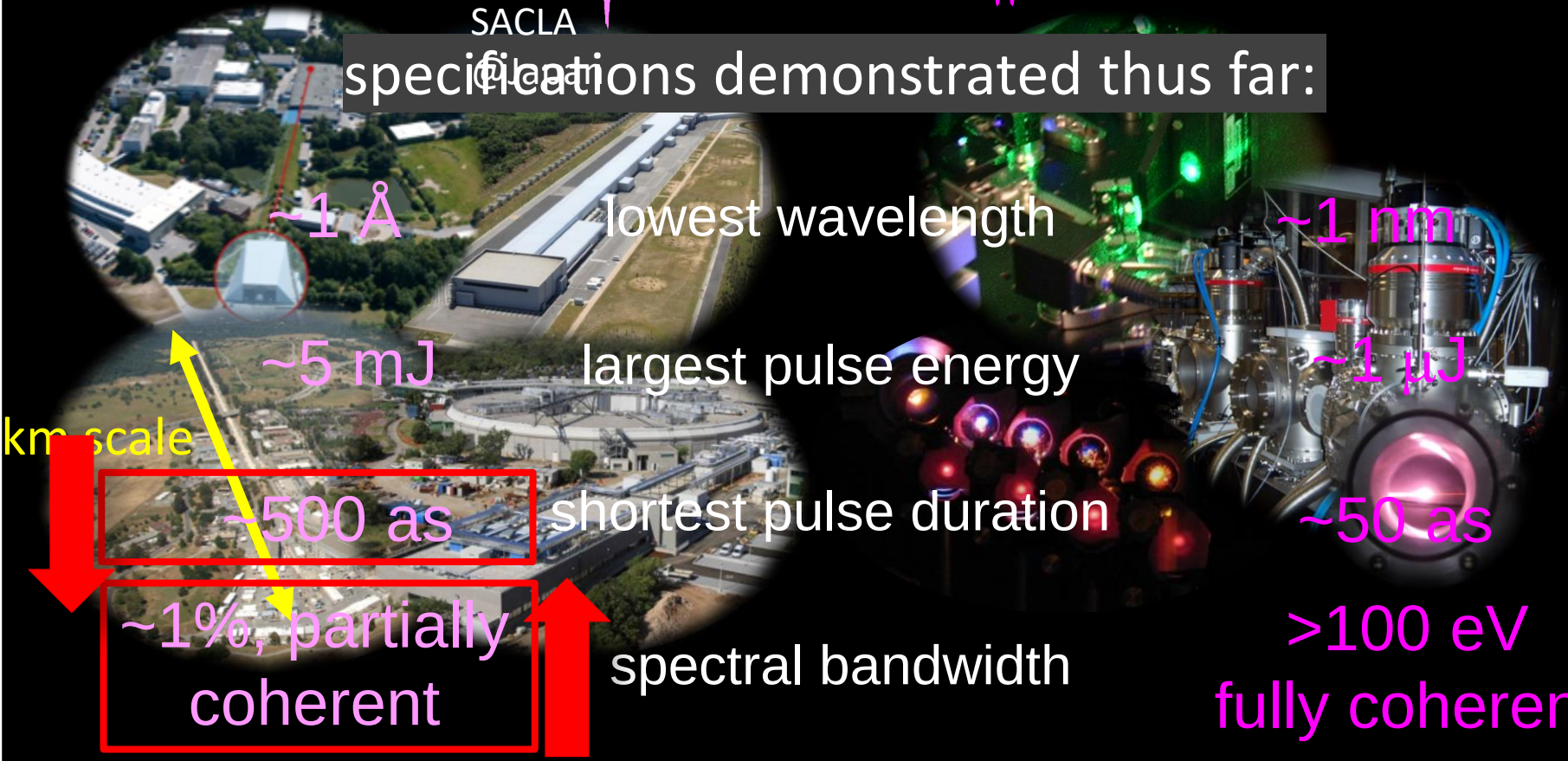
XUV to **x-ray**

molecule images: molview.org

The tools: Provided by two parallel revolutions in ultrashort x-ray/XUV laser science

Free Electron Lasers

High Harmonic Generation



specifications demonstrated thus far:

~1 Å	lowest wavelength	~1 nm
~5 mJ	largest pulse energy	~1 μJ
~500 as	shortest pulse duration	~50 as
~1%, partially coherent	spectral bandwidth	>100 eV fully coherent

Our Experimental Approach

IR/VIS Lasers

Free Electron Lasers

High Harmonic Generation

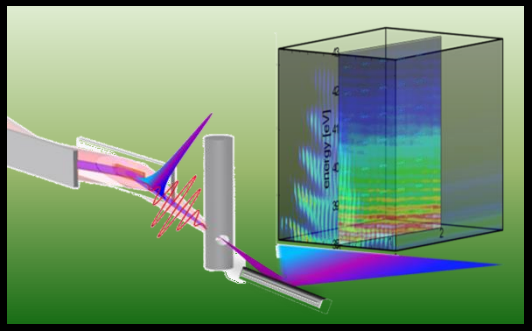
atom or molecule

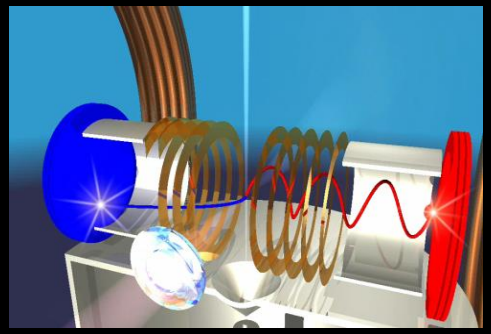
"listening"

"imaging"

Multidim. optical spectroscopy

Reaction microscope/COLTRIMS





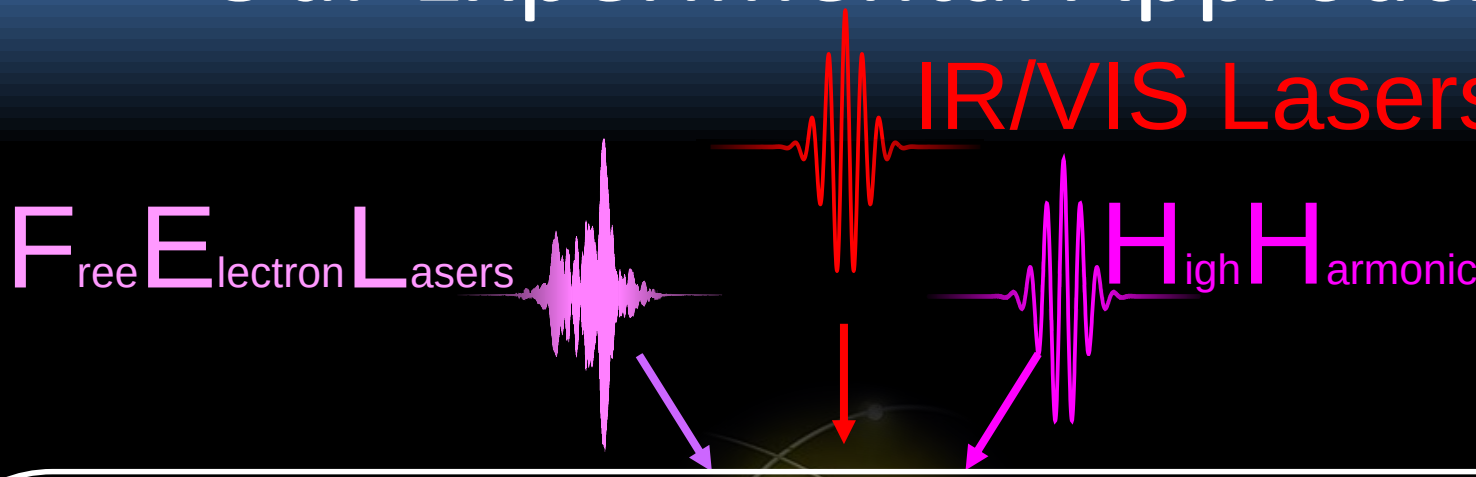
time&energy-resolved detection of **photons**

extract complete information

coincidence detection of **electrons/ions**

atom image from: http://startswithabang.com/?p=1795

Our Experimental Approach



atom or molecule

"listening"

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Multidim. optical spectroscopy

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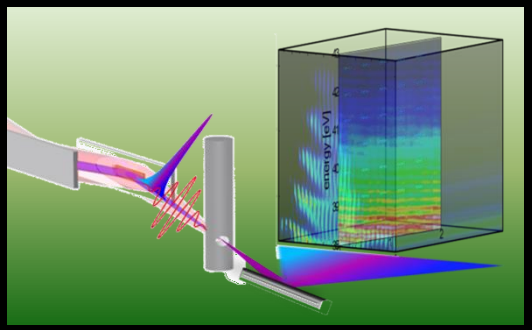
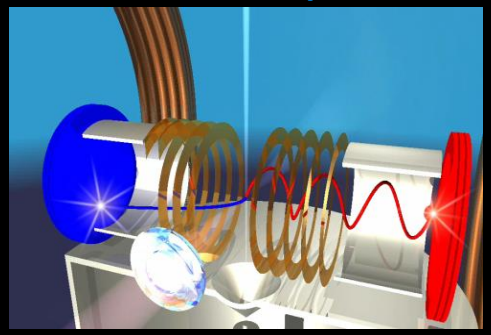
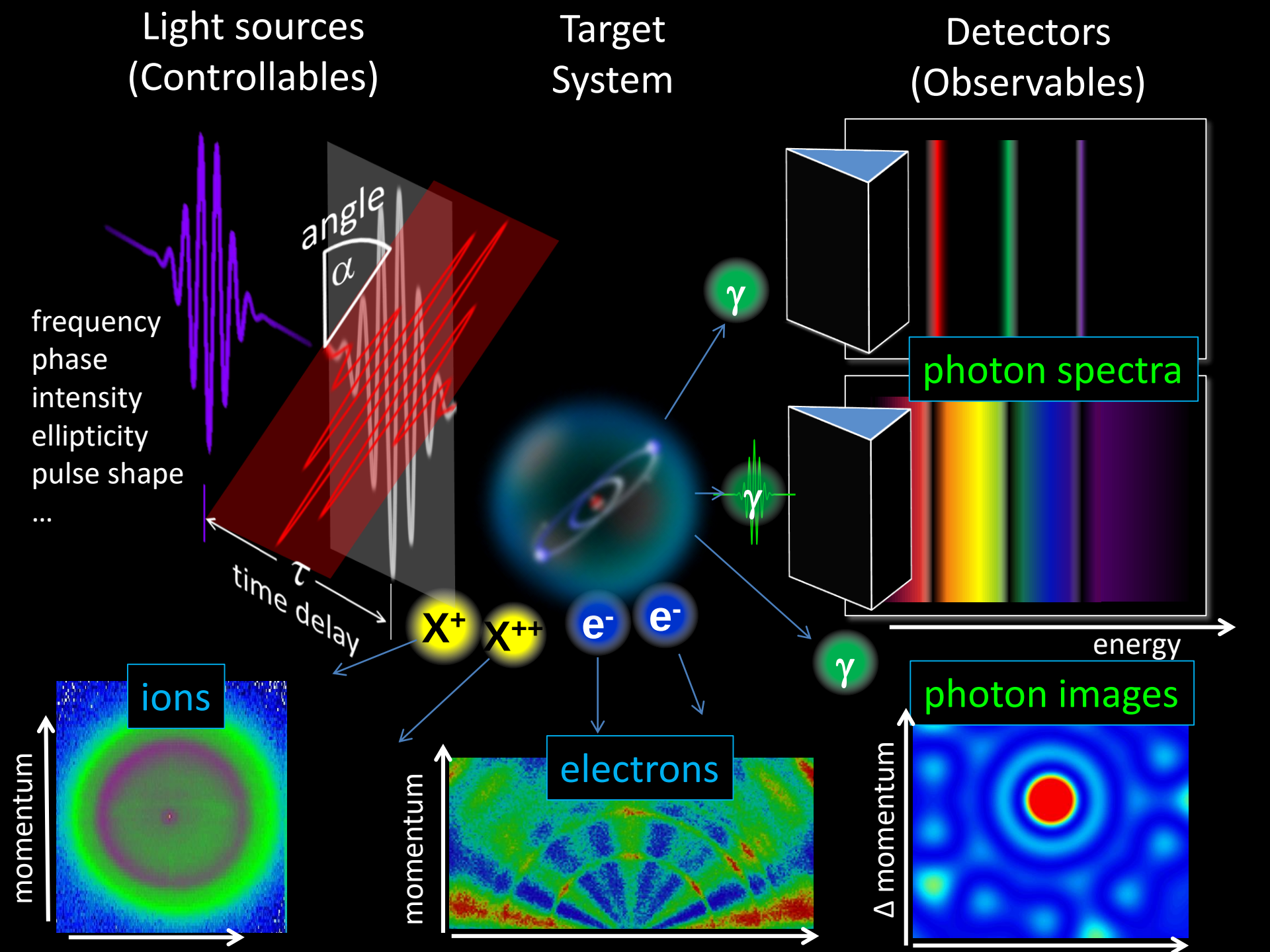




time&energy-resolved detection of **photons**

extract complete information

coincidence detection of **electrons/ions**

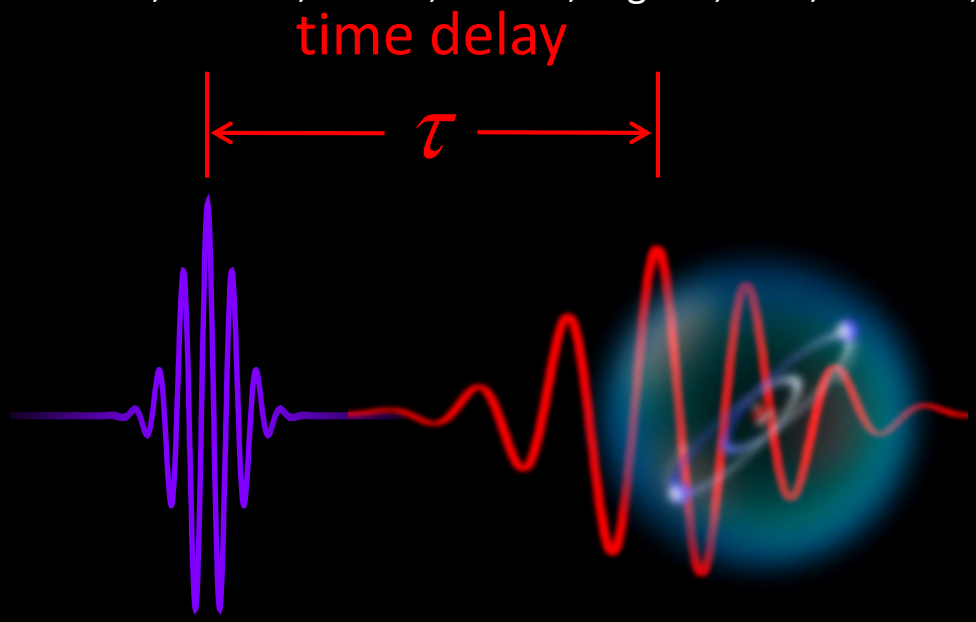

Light sources
(Controllables)
Target
System
Detectors
(Observables)
frequency
phase
intensity
ellipticity
pulse shape
…
angle
α
τ
time delay
γ
γ
γ
photon spectra
energy
X+
X++
e-
e-
ions
momentum
electrons
momentum
photon images
Δ momentum

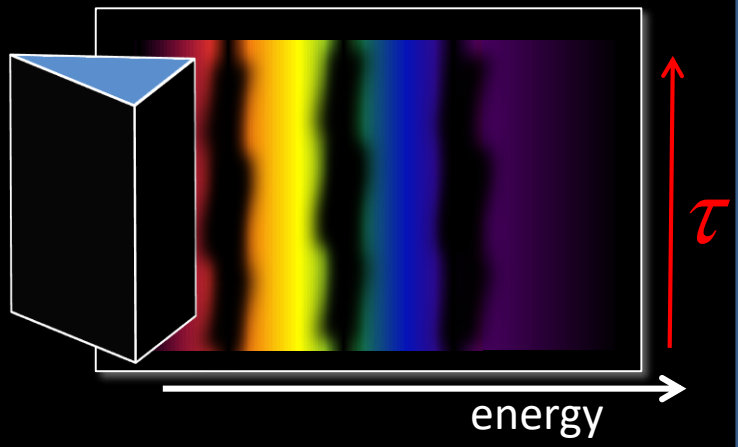
time-dependent XUV absorption spectroscopy

HHG-based applications, active groups:

Exp.: Leone, Neumark, Keller, Gallmann, Chang, Krausz, Schultze, Sansone, Kim, Sandhu, Mauritsson, Vrakking, Wörner, Biegert ...

Theory: Schafer, Gaarde, Santra, Martín, Argenti, Rost, Madsen, Rohringer, Greene, Keitel, Stockman, ...



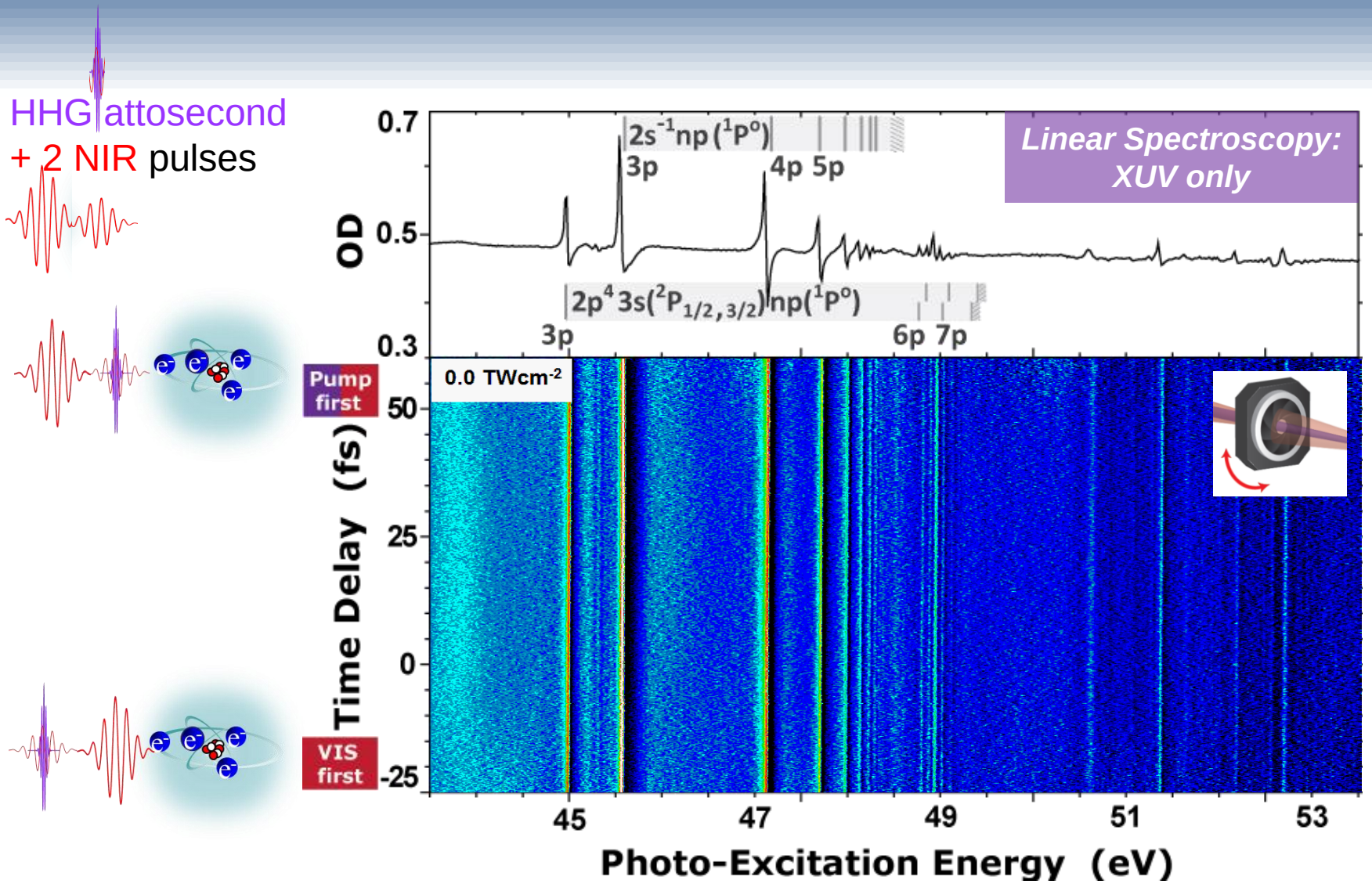


$$\psi(x,t) \propto a_1 \psi_1(x) e^{-\frac{i}{\hbar}E_1 t} + a_2 \psi_2(x) e^{-\frac{i}{\hbar}E_2 t}$$

Phase φ_1 Phase φ_2

Time-resolved inner-shell dynamics in Neon

HHG attosecond
+ 2 NIR pulses

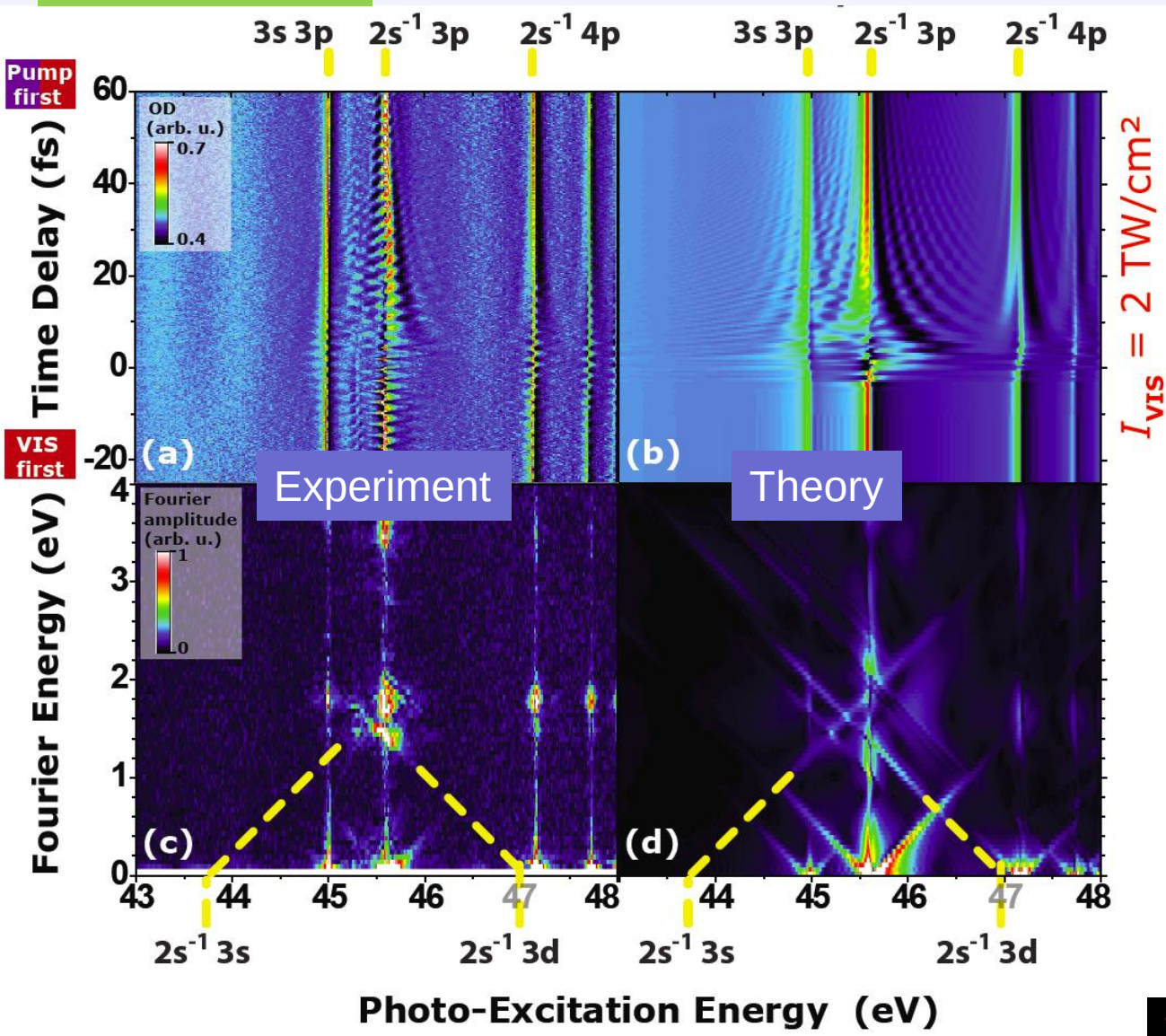


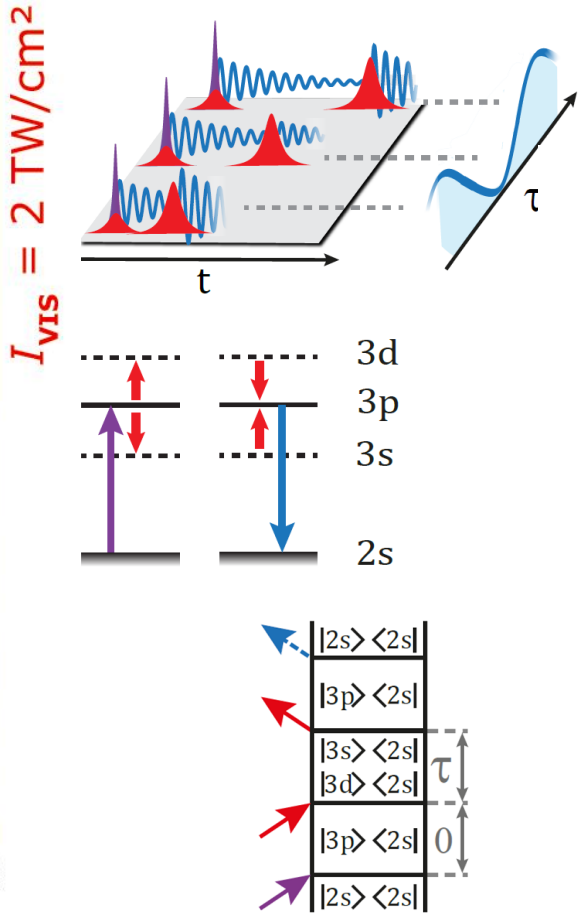
note: HHG (coherent) spectral bandwidth: 10 eV

2D-spectroscopy of inner-valence excitations

in Neon

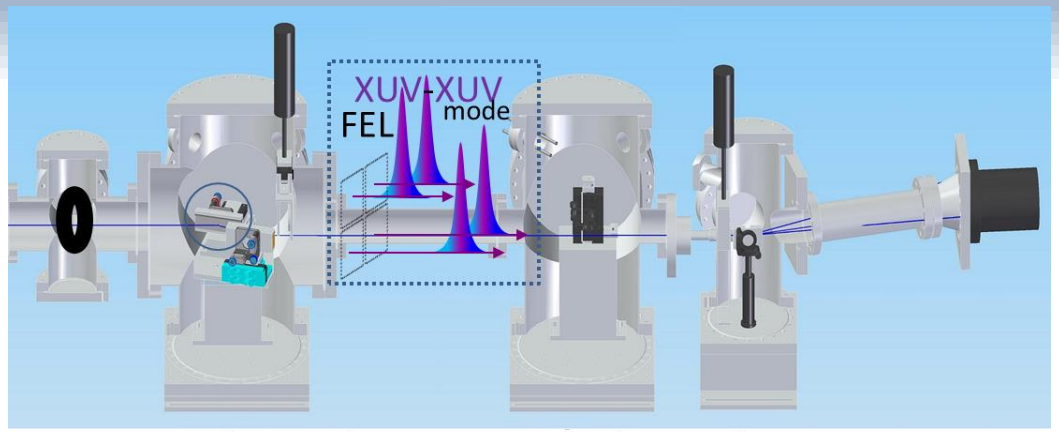
cooperation with H. van der Hart, A. Brown (Theory, QUB)

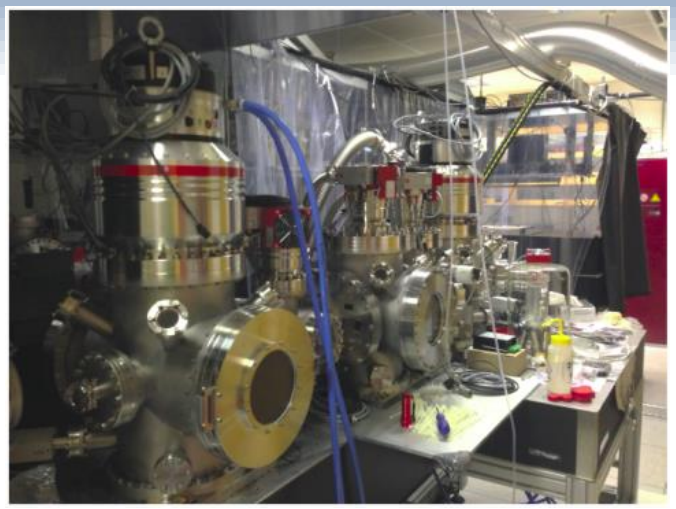


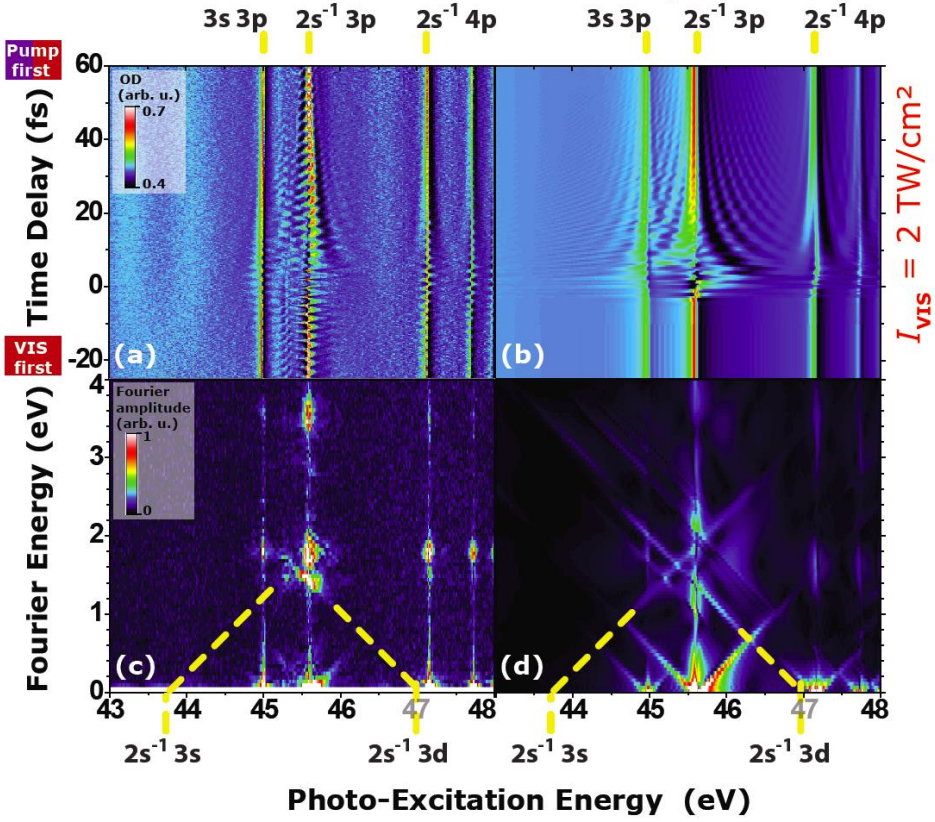


T. Ding *et al.* Opt. Lett. (2016)

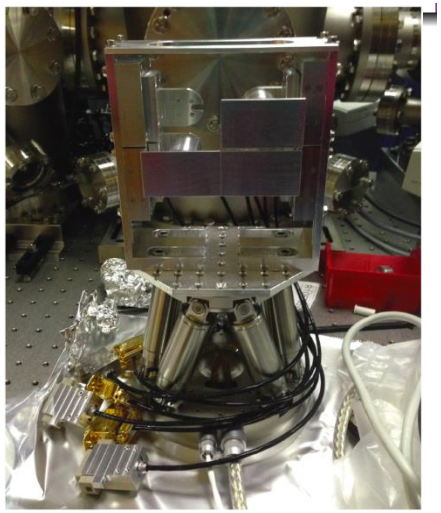
XUV 2D spectroscopy@FELs

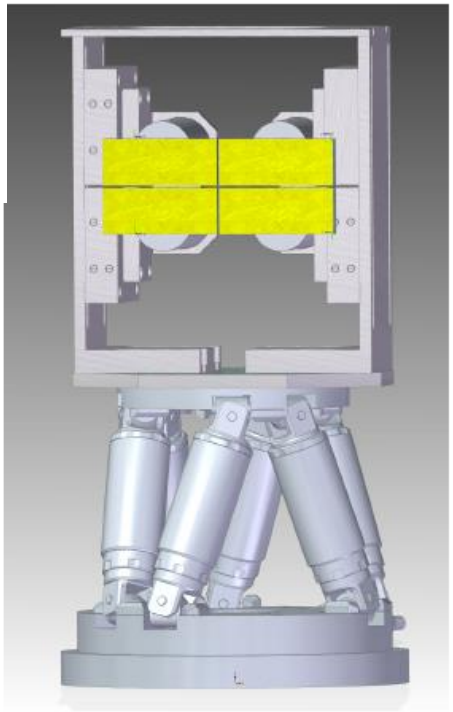




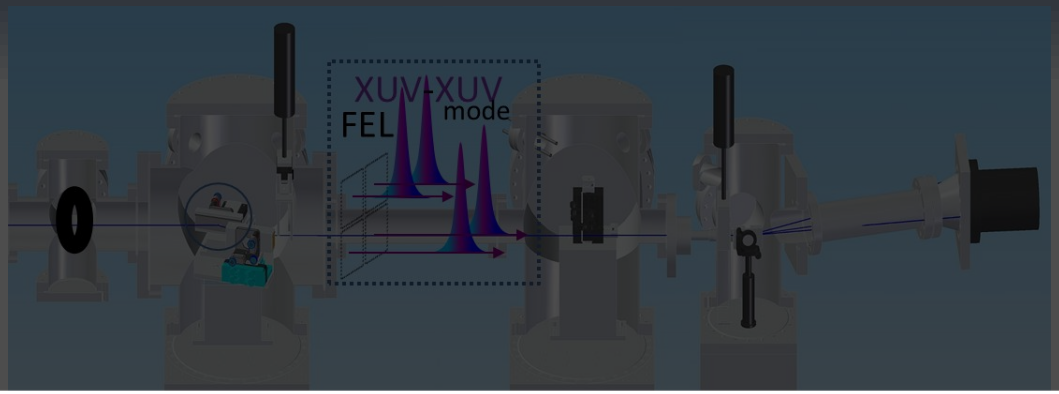


4-quadrant mirror split-and-delay unit

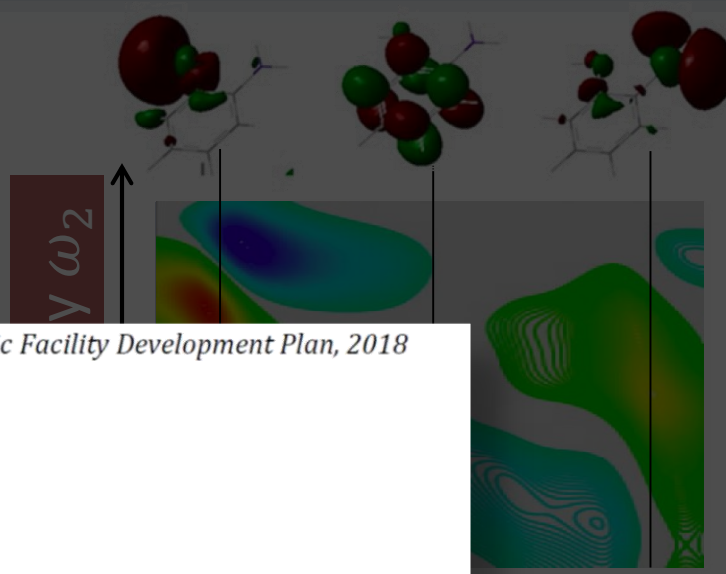




Site-selective 2D-Spectroscopy with (soft) x-rays



example: aminophenol



LCLS Strategic Facility Development Plan, 2018

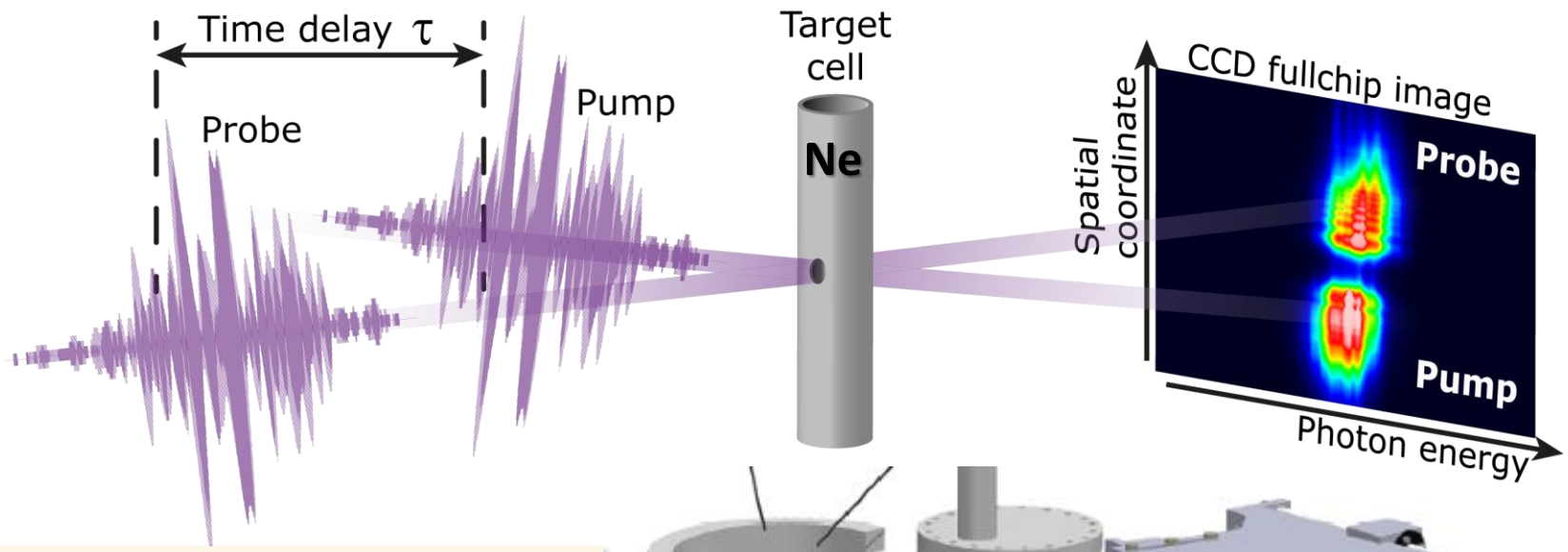
2 SCIENTIFIC DRIVERS

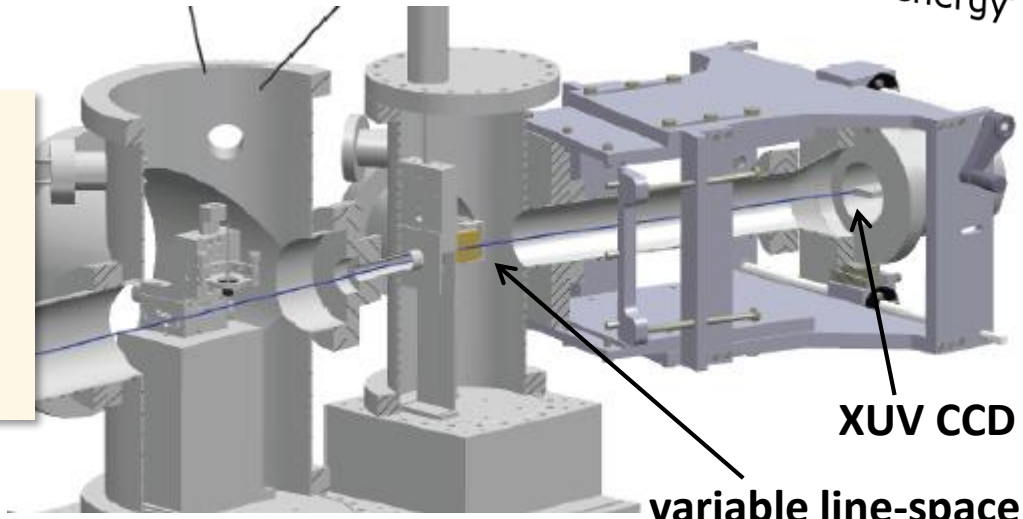
2.1 Example Scientific Drivers for LCLS-II

LCLS-II will be a transformative tool for energy science, qualitatively changing the way that X-ray imaging, scattering and spectroscopy can be used to study how natural and artificial systems function. It will enable new ways to capture rare chemical events, characterize fluctuating heterogeneous complexes, and reveal quantum phenomena in matter, using nonlinear, multidimensional and coherent X-ray techniques that are possible only with X-ray lasers. This facility will provide access to the "tender X-ray" regime (2 to 5 keV) that is largely inaccessible today, and will use seeding technologies to provide fully coherent X-rays in a uniformly spaced series of pulses with programmable repetition rate and rapidly tunable photon energies.

part of LCLS Strategic Facility Development Plan (2018)

XUV-Pump XUV-Probe Absorption Spectroscopy in Neon gas

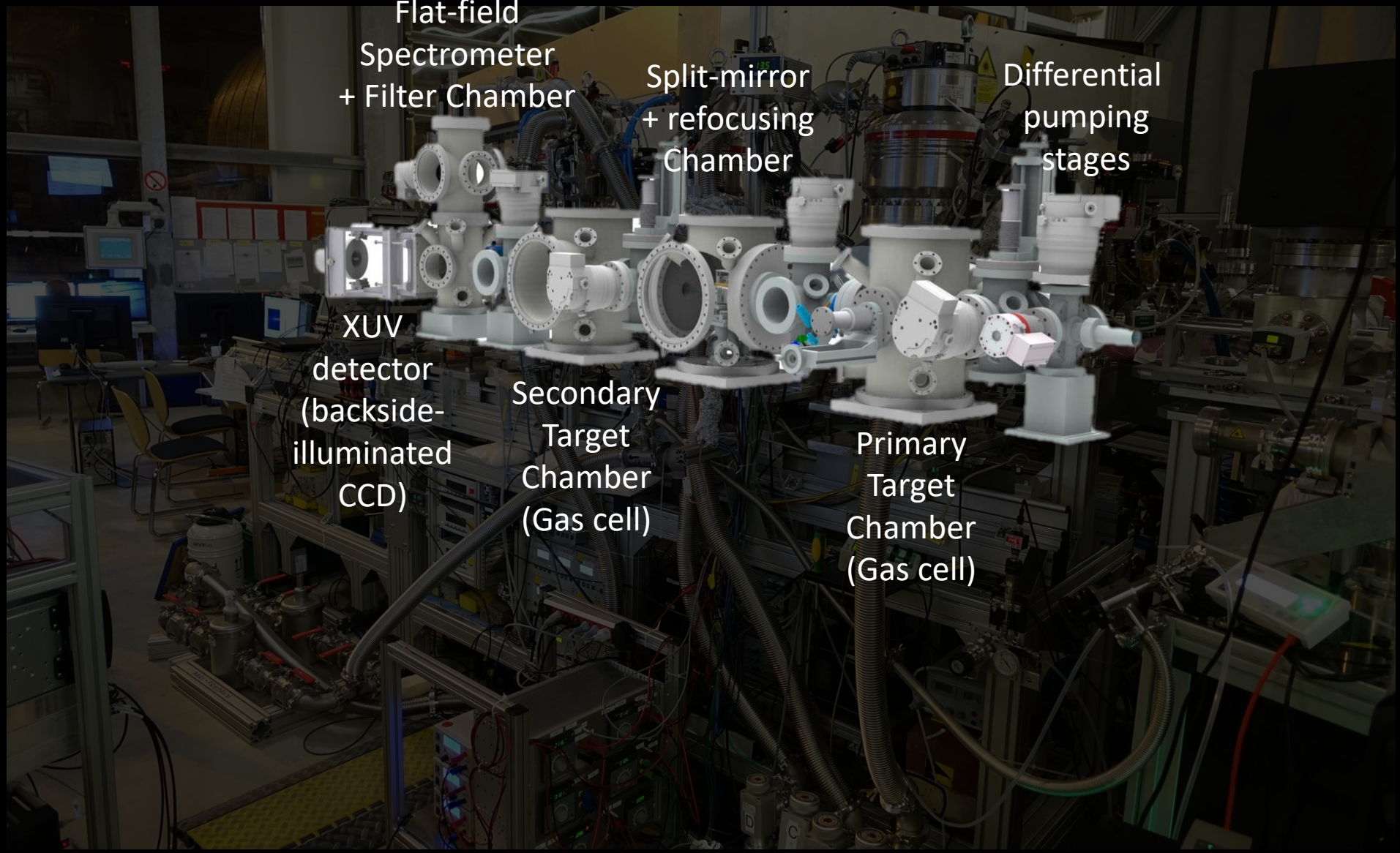




Cooperation with:

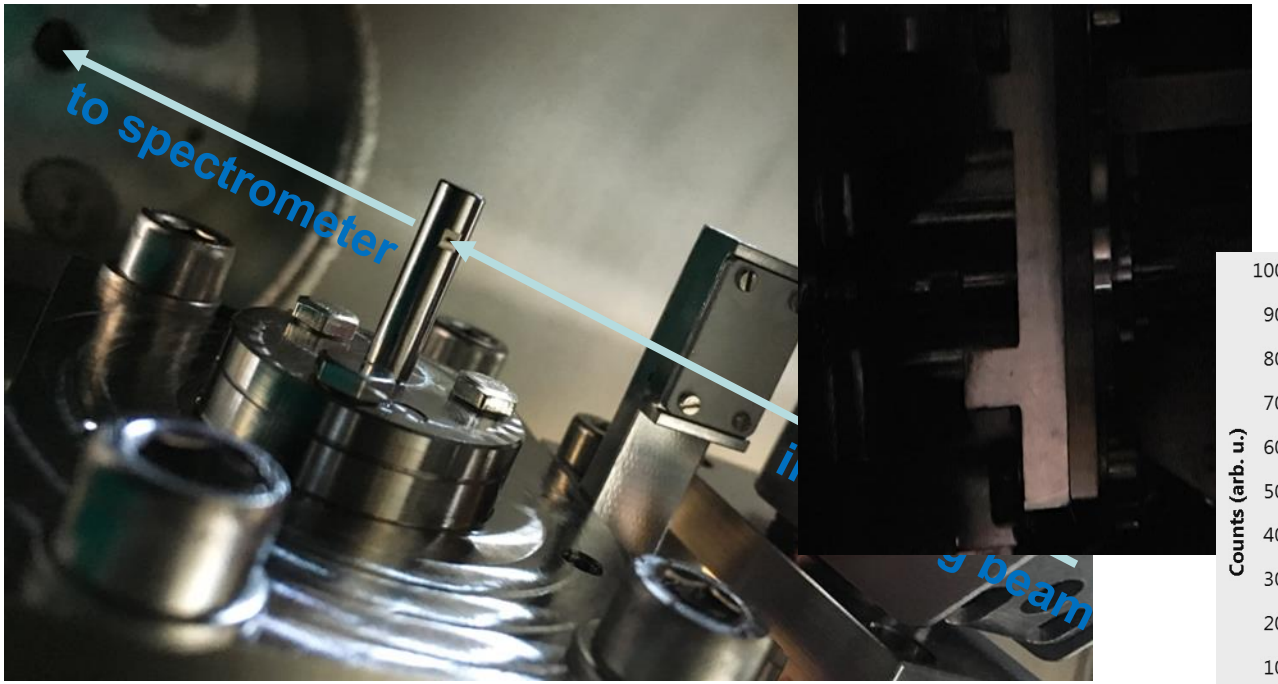
S. Roling, H. Zacharias (Univ. Münster)
S. Düsterer, R. Treusch (DESY)
Z.-H. Loh (NTU Singpore)
A. R. Attar (UC Berkeley)
T. Gaumnitz (ETH Zürich)

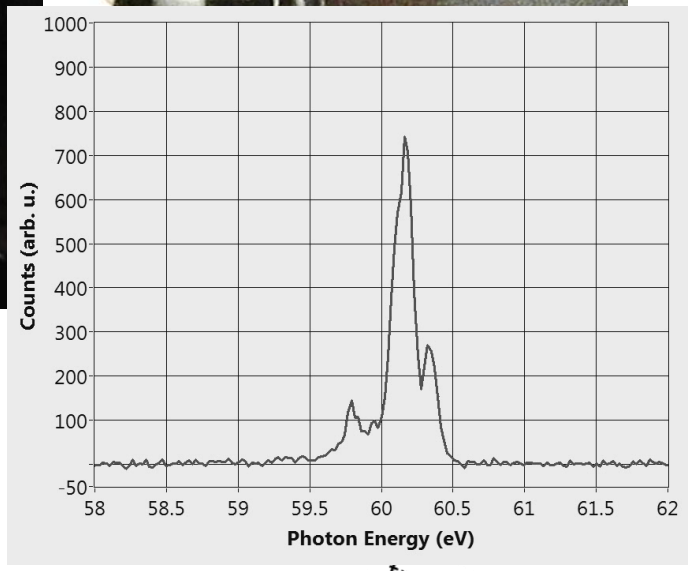
X-MuSiC Beamline Design & Setup

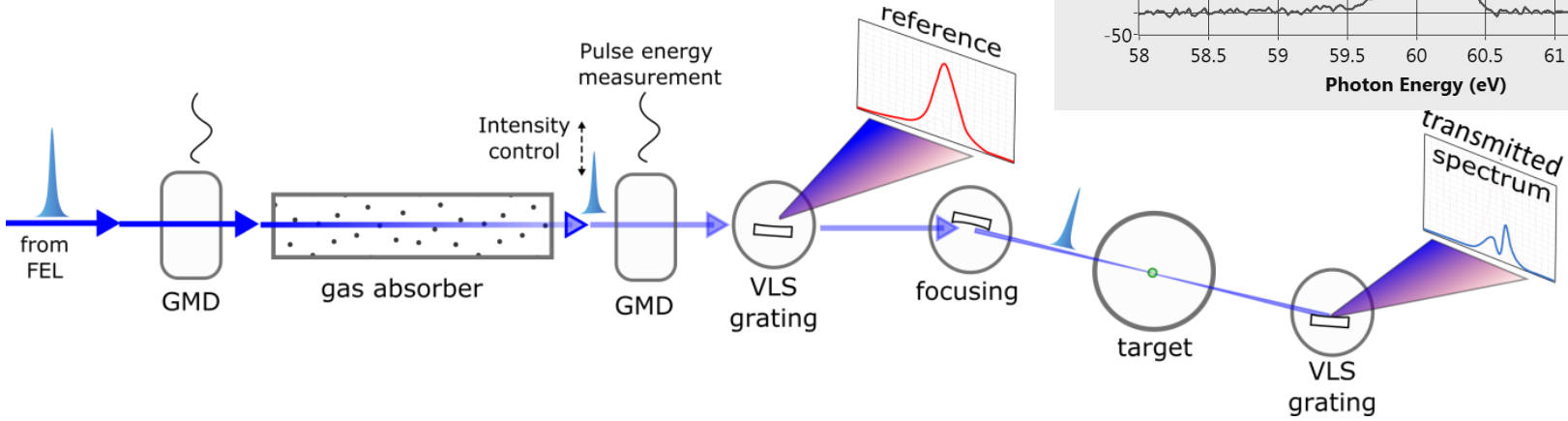


Experimental Setup

200 µm machine drilled holes; interaction length: ~3 mm



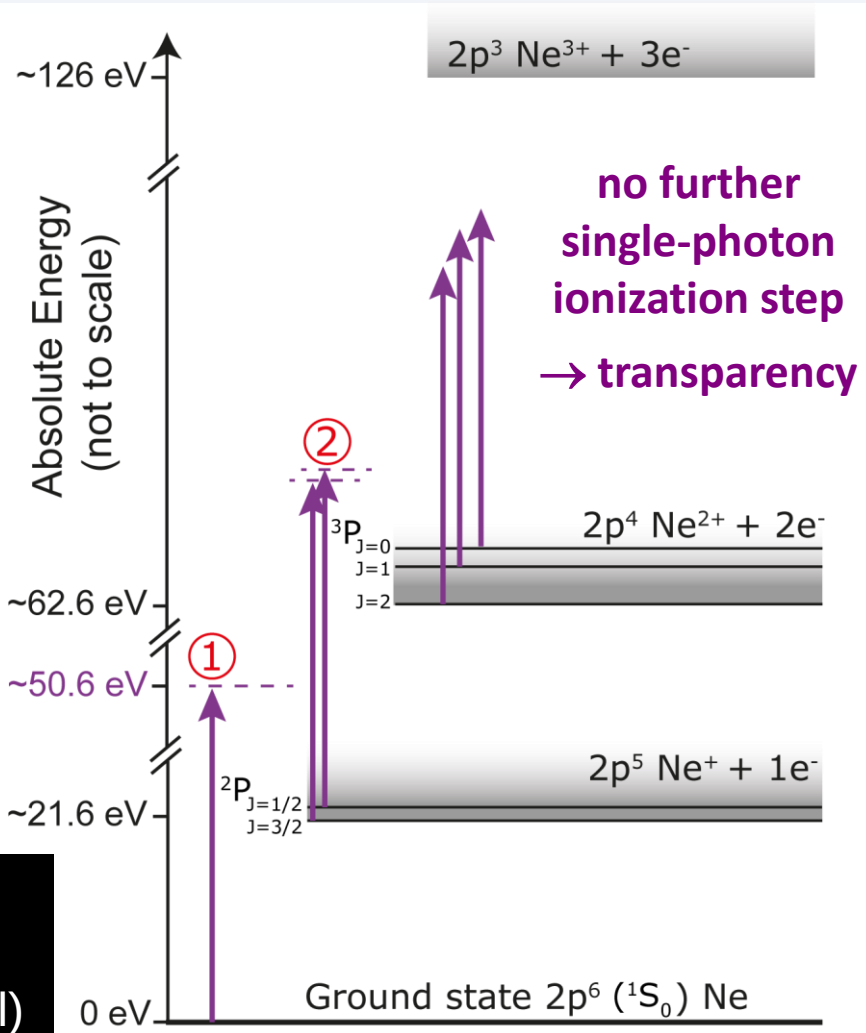




Time-resolved transmission changes through neon

for Pump vice versa

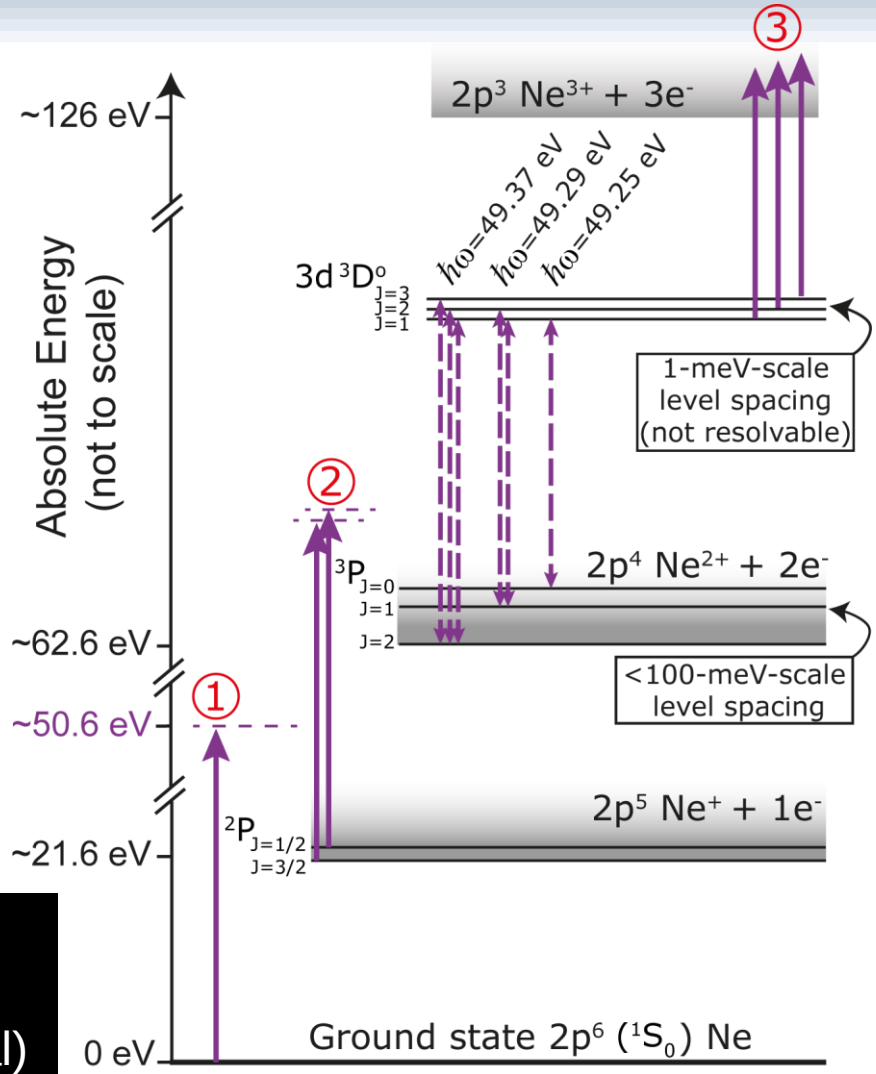
1-2 eV bandwidth



***In-situ* time-resolved probing of sequential ionization**
... as opposed to after the pulse (ion signal)

Time-resolved transmission changes through neon

for Pump vice versa



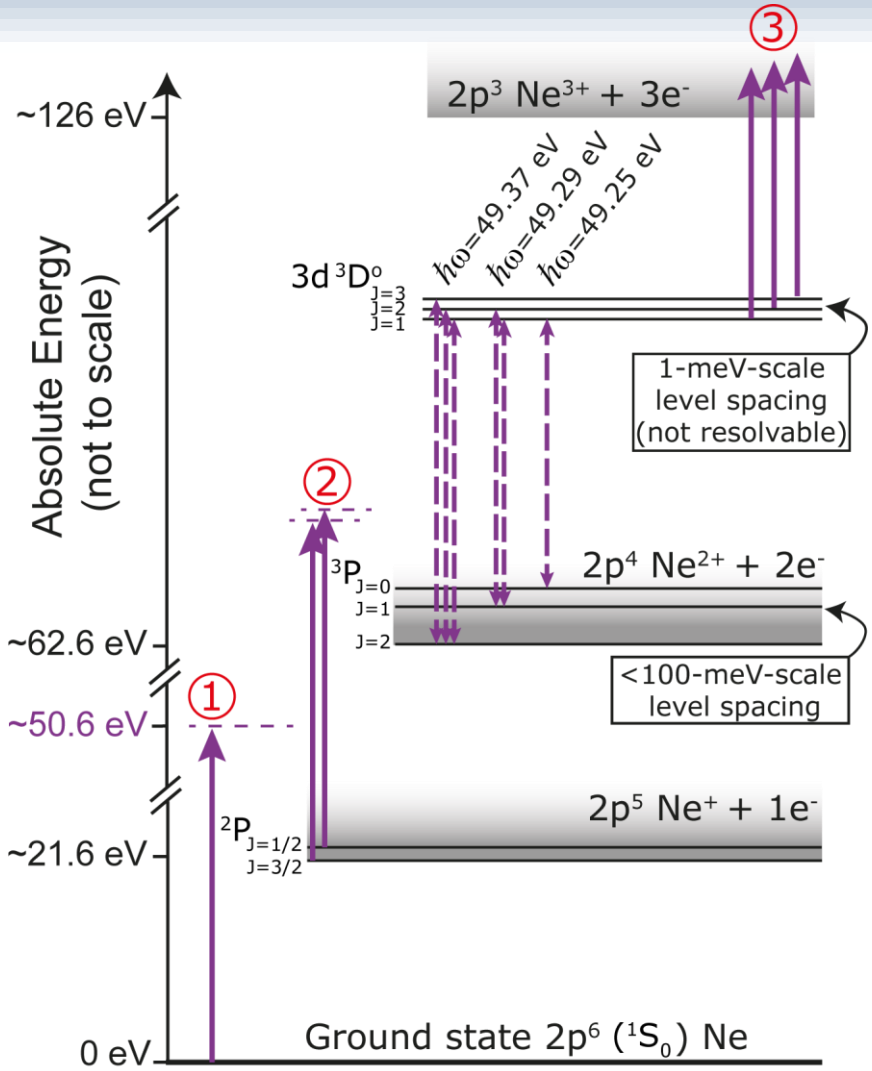
***In-situ* time-resolved probing of sequential ionization**

... as opposed to after the pulse (ion signal)

Time-resolved spectroscopy of Ne^{2+} transitions

Optical density (absorbance)

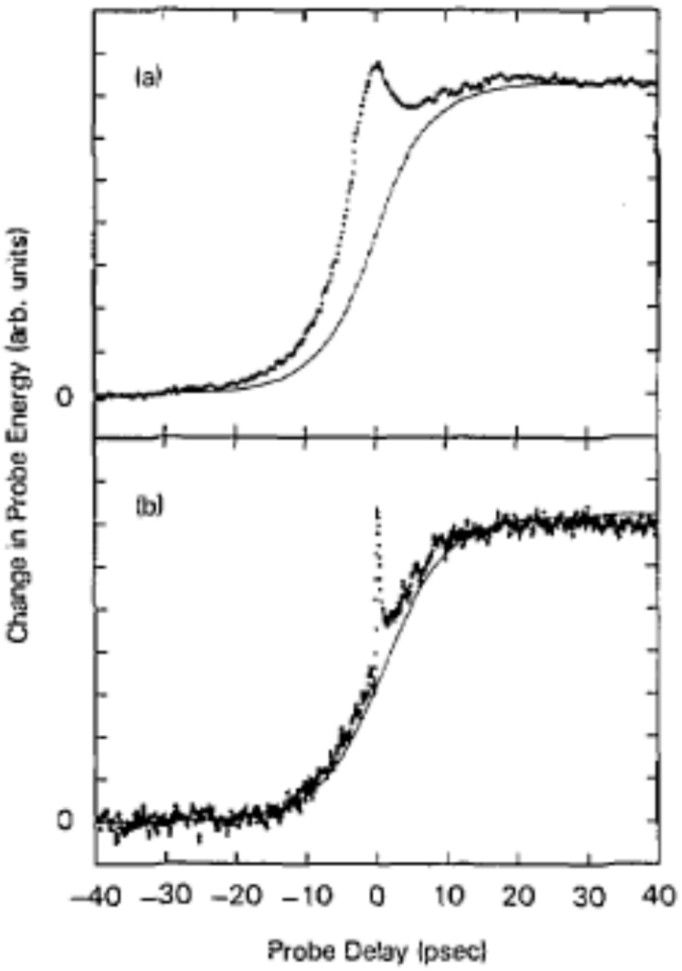
Resolve and identify <100-meV-spaced transitions (^3P – ^3D spin-orbit multiplet)



Coherence effects around $\tau = 0$

zooming on "spectral dip"

Optical density (absorbance)



Resolve and identify <100-meV-spaced transitions (^3P – ^3D spin-orbit multiplet)

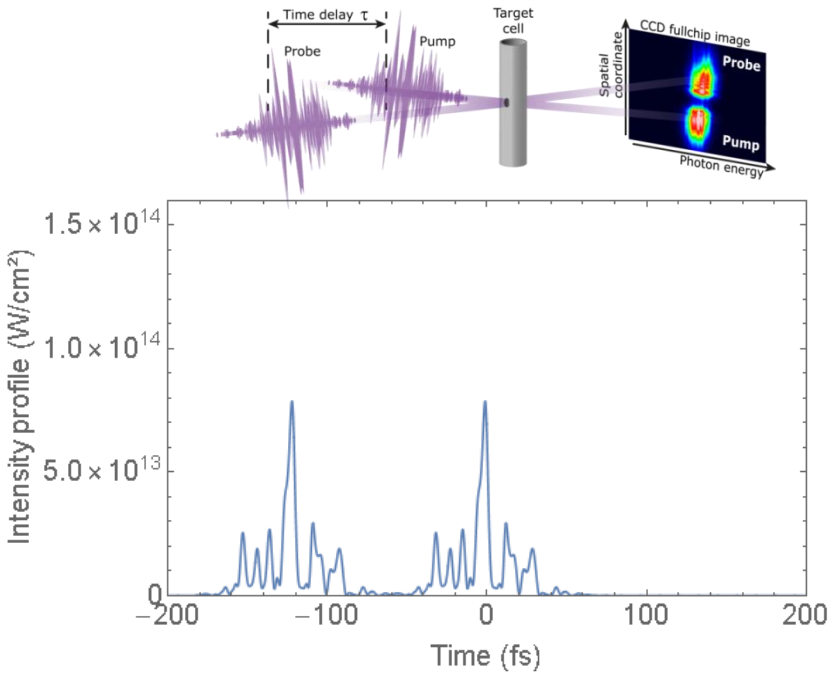
T.F. Heinz, S.L. Palfreq, K.B. Eisenthal
Opt. Lett. 9, 359 (1984)

Coherence effects around $\tau = 0$

zooming on "spectral dip"

Optical density (absorbance)

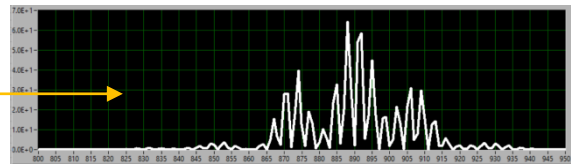
Local enhancement due to interfering SASE spikes

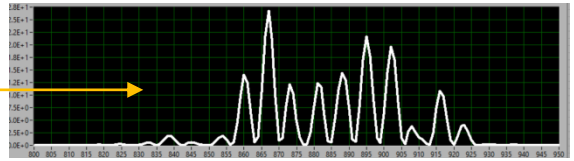


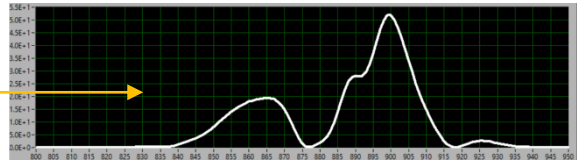
- **Most prominent in vicinity of resonances (reduced absorbance)**
- **Time scale : ca. 2-3 fs width (for 0.8 eV FEL bandwidth @ 50.6 eV)**

Interferometric Information from Experiment(!)

Illustration from simulation







Coherence effects around $\tau = 0$

zooming on "spectral dip"

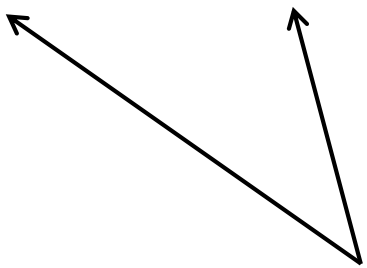
Experiment

Few-level simulation of $^3P - {}^3D$ spin-orbit multiplet

- non-perturbative
- stochastic pulses

Coherence effects around $\tau = 0$

zooming on "spectral dip"



$$\Delta\omega_{coh} \propto {}^{1}/_{\Delta\tau_{coh}}$$

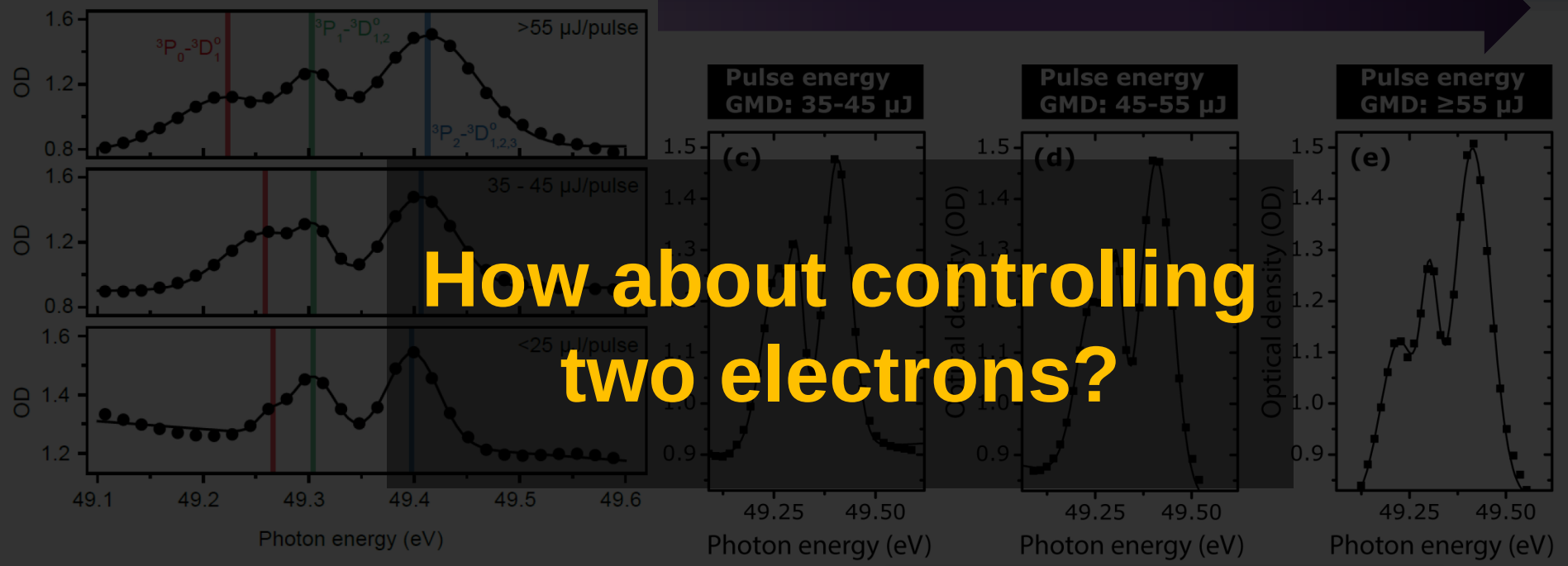
$\rightarrow$ slight overestimation of coherence bandwidth in simulation

Lineout experiment: $\Delta\tau_{exp}$= (2.4 ± 0.3) fs

Lineout simulation: $\Delta\tau_{sim}$= (1.2 ± 0.1) fs

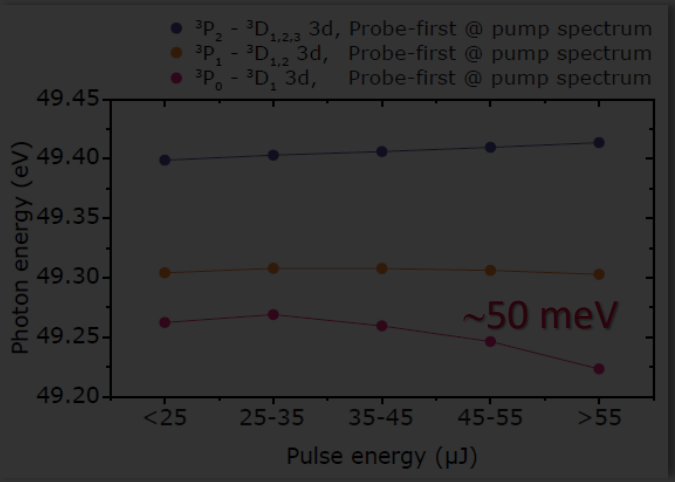
Intensity-dependent shifts of resonances

increasing FEL intensity



How about controlling two electrons?

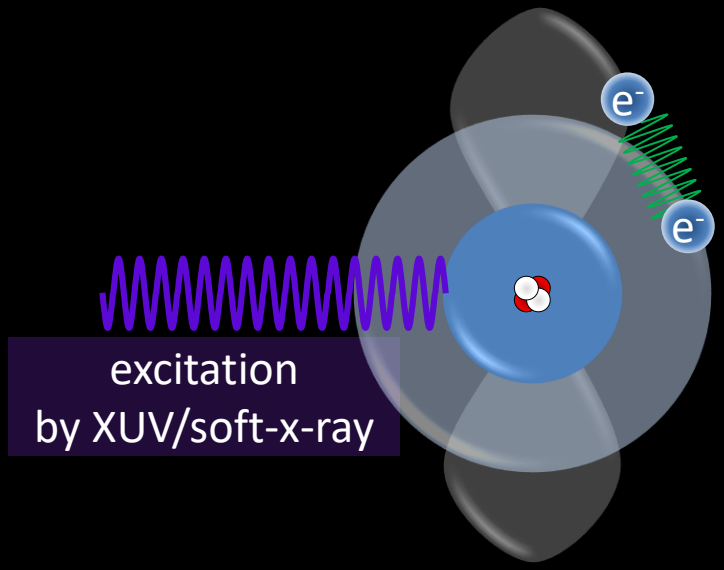
- **Observe intensity-dependent resonance shifts (Stark effect)**
- **XUV-induced strong coupling of ^3P – ^3D spin-orbit multiplet in Ne^{2+}**



doubly-excited helium, in a strong field

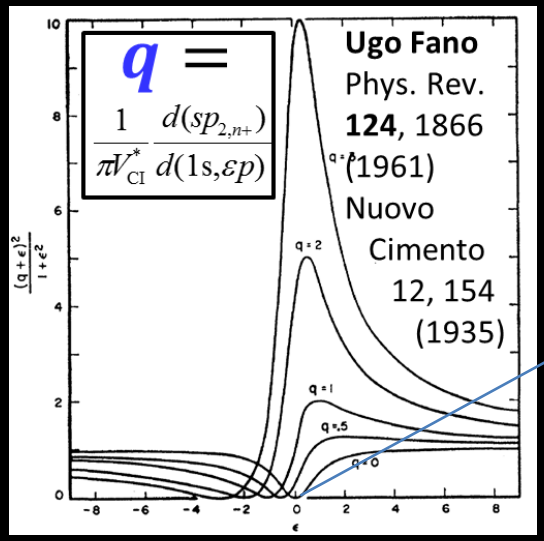
a prototype system
for electron correlation

intense XUV laser (FEL)

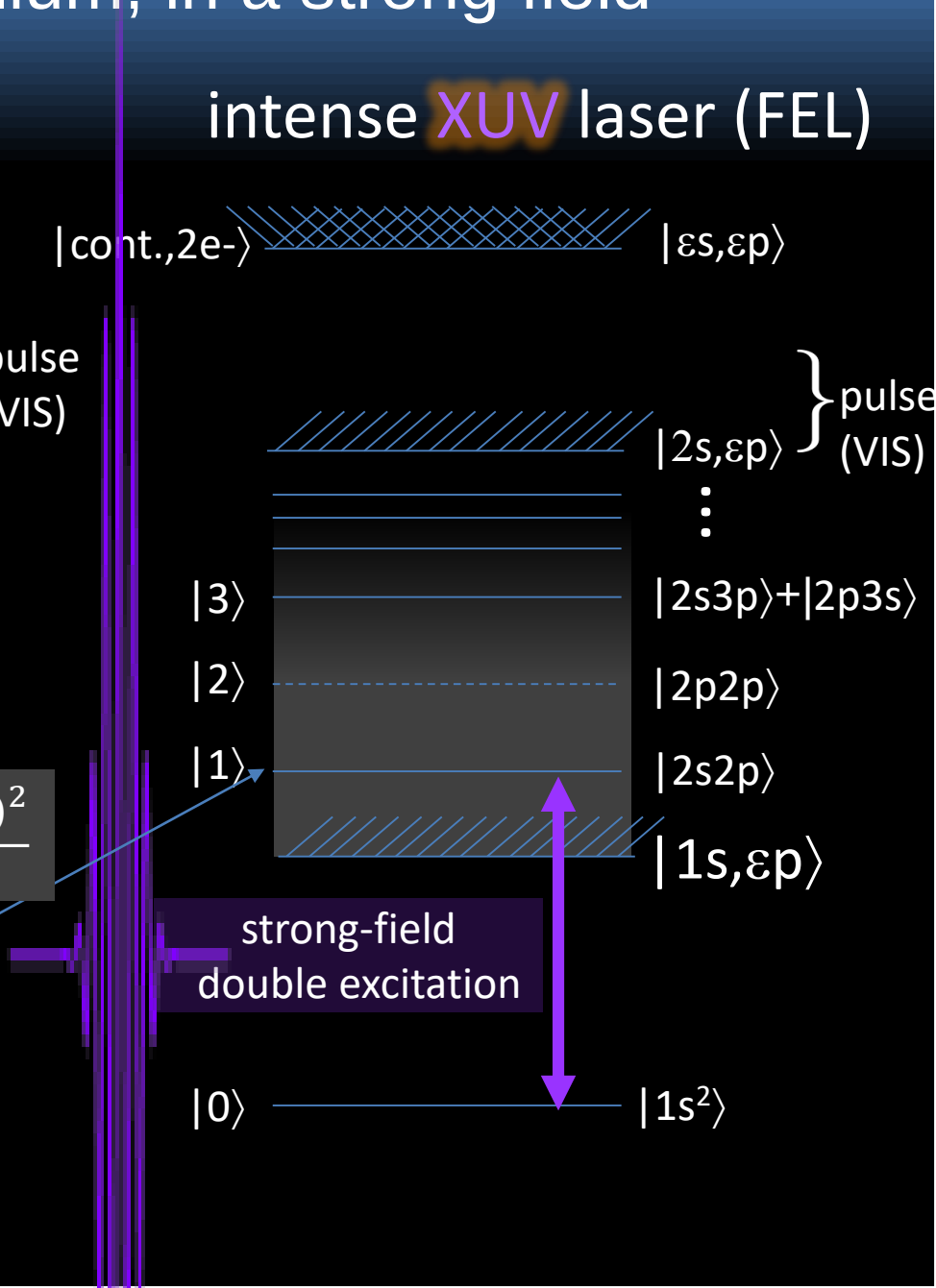


excitation
by XUV/soft-x-ray

pulse
(VIS)

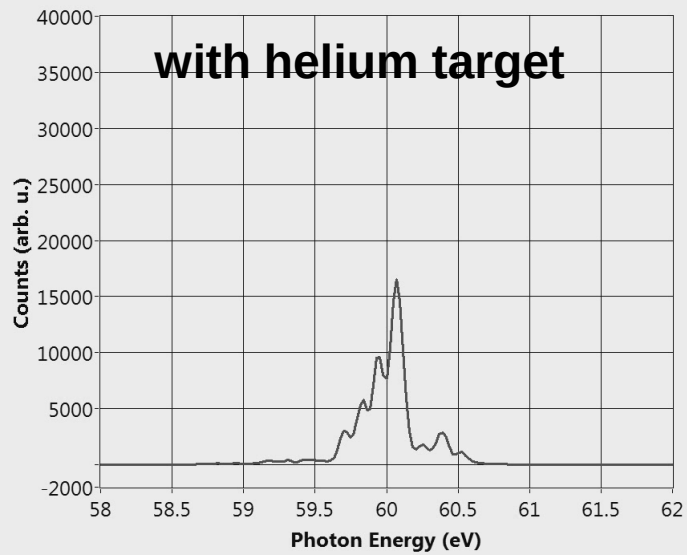


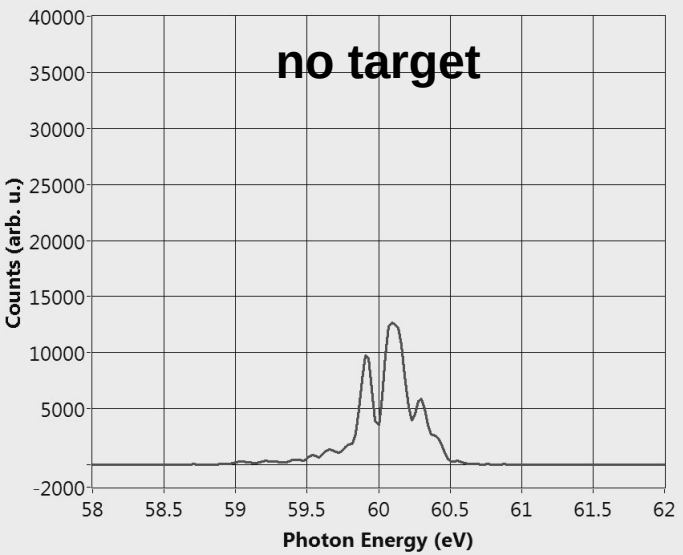
$$\sigma_{Fano} \sim \frac{(q+\varepsilon)^2}{1+\varepsilon^2}$$



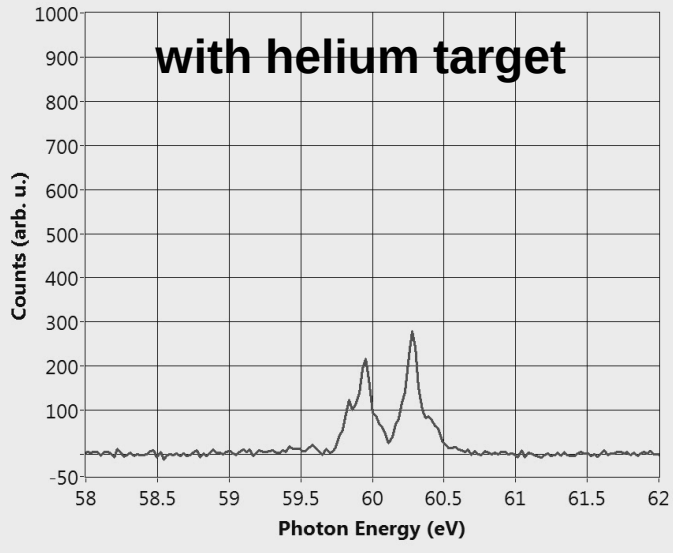
Single-shot FEL spectra

high FEL intensity





low **FEL intensity**



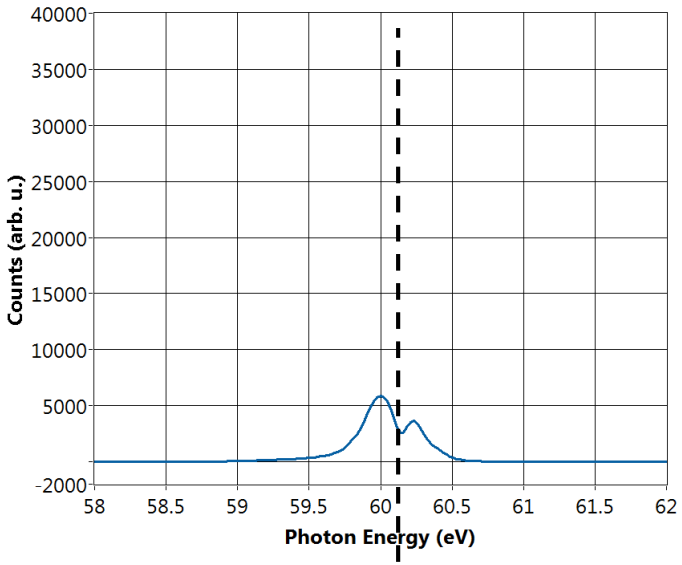
Averaged FEL spectra

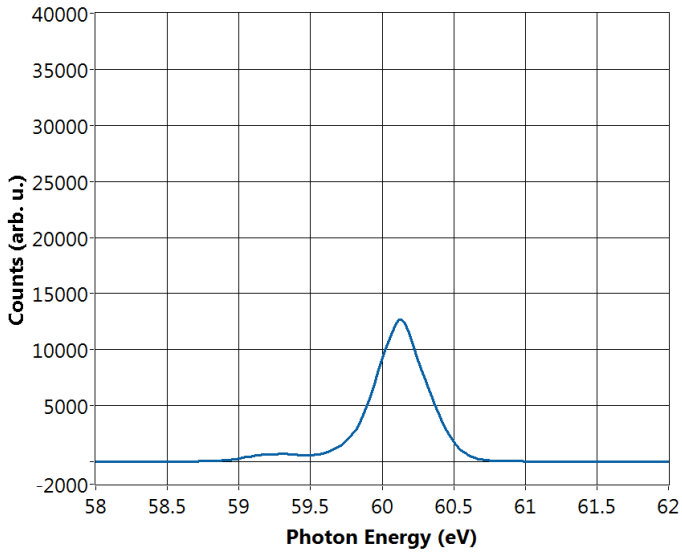
with helium target

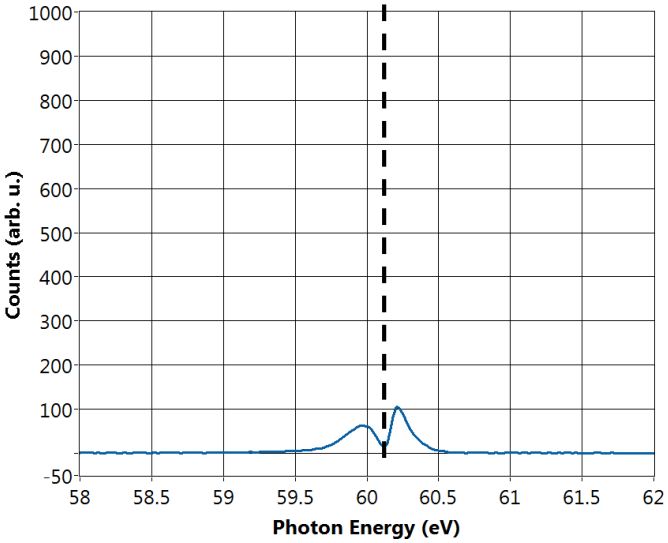
no target

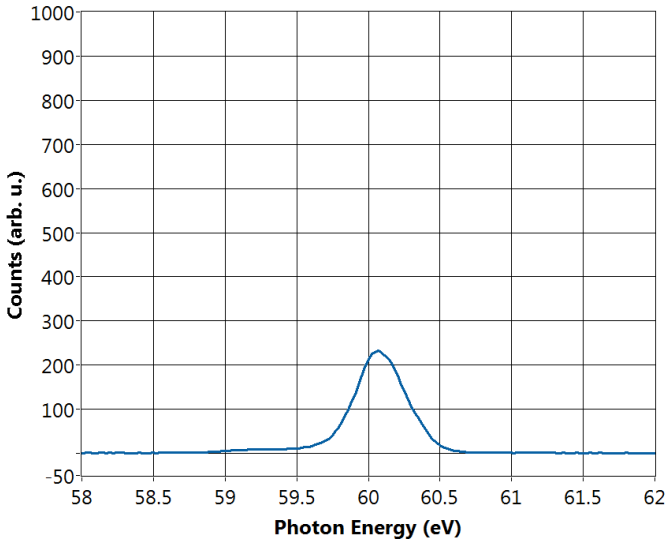
high FEL intensity

low **FEL intensity**





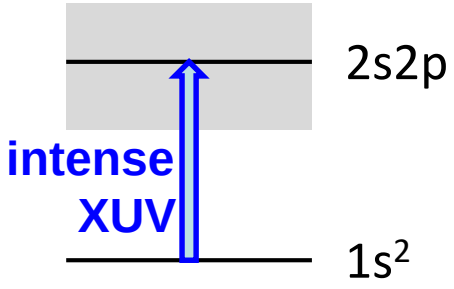


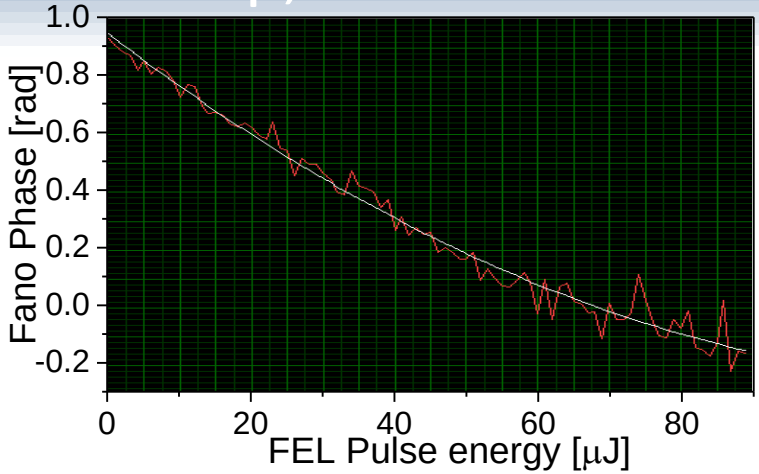


XUV *strong-field* effects in Helium@FLASH

He 2s2p, 60.1 eV Fano resonance

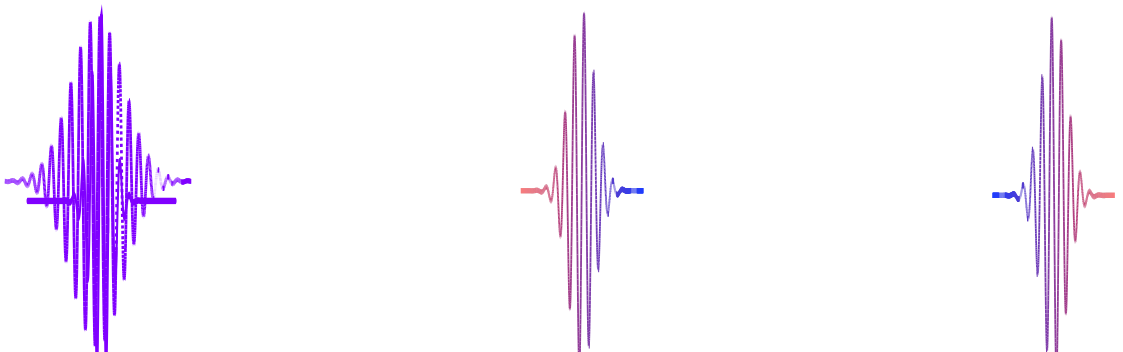
XUV-intensity-dependent phase shift





What is happening inside the Helium atom?

depends on the XUV pulse shape…



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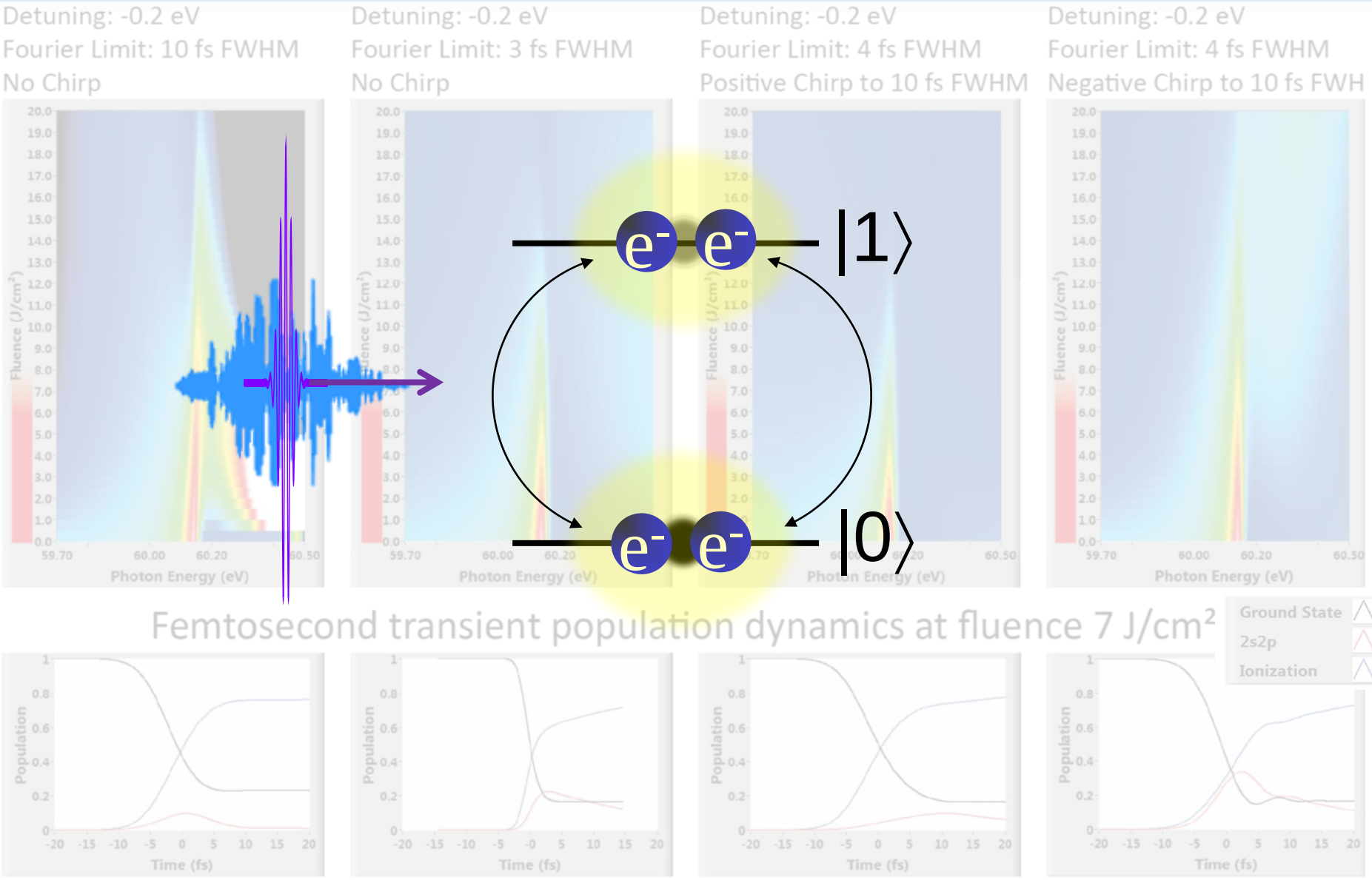
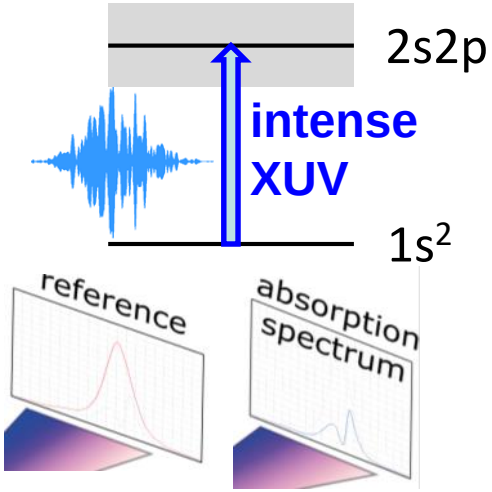
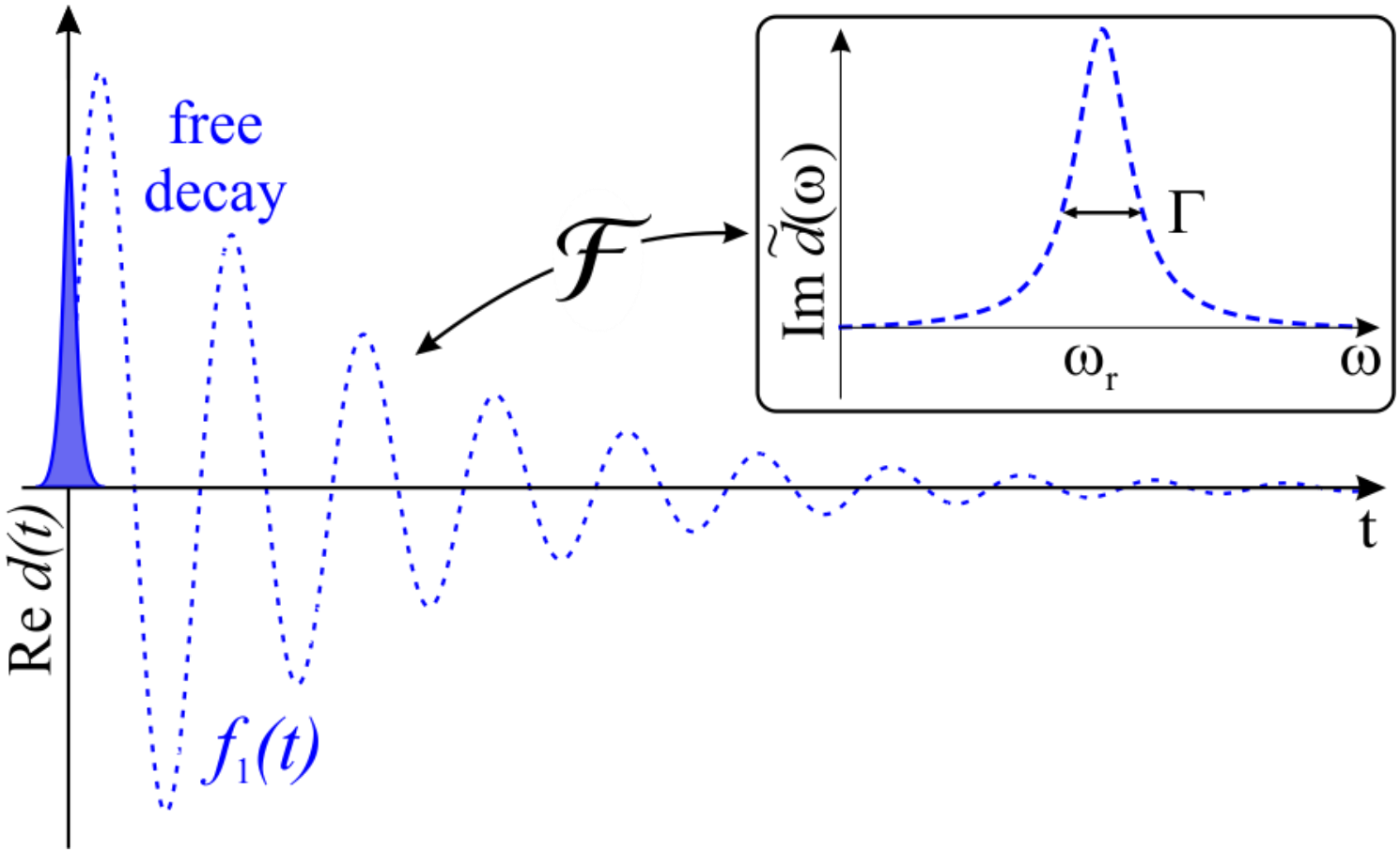


Strongly driven double excitation?



Two-level system interacting with typical noisy SASE FEL pulses, avg. over ~100 shots

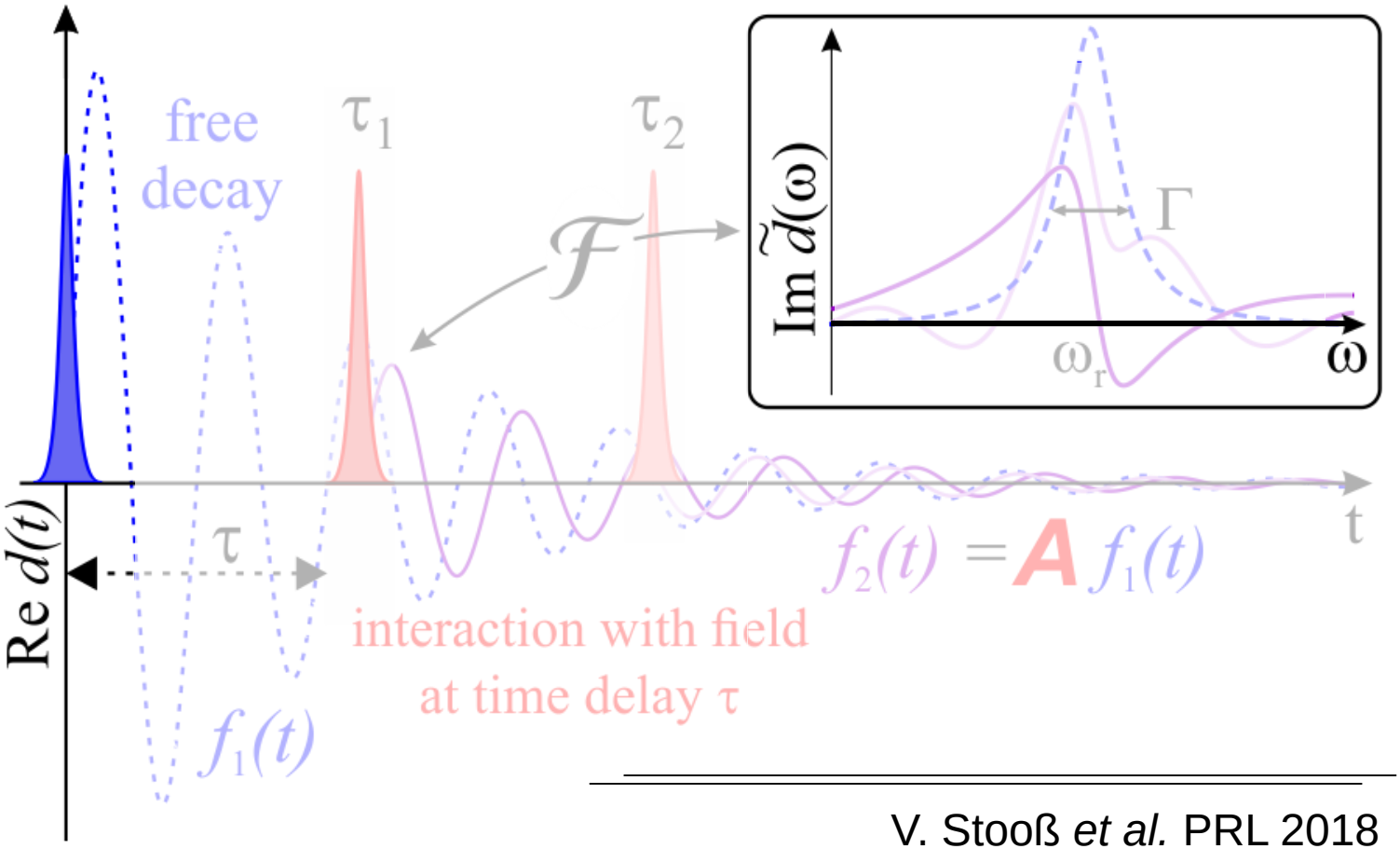
The dipole control model (DCM)



Reconstruction of complex dipole response functions of strong-field driven states

Analytical result: Blättermann *et al.* J. Phys. B: At. Mol. Opt. Phys. **47** 124008 (2014)

$$\tilde{d}_\tau(\omega,\tau) \propto -\mathrm{i}\frac{1-\mathrm{e}^{\mathrm{i}(\omega_\mathrm{r}-\omega)\tau-\frac{\Gamma}{2}\tau}\left(1-\boldsymbol{A}(\tau)\right)}{\mathrm{i}(\omega_\mathrm{r}-\omega)-\Gamma/2}$$



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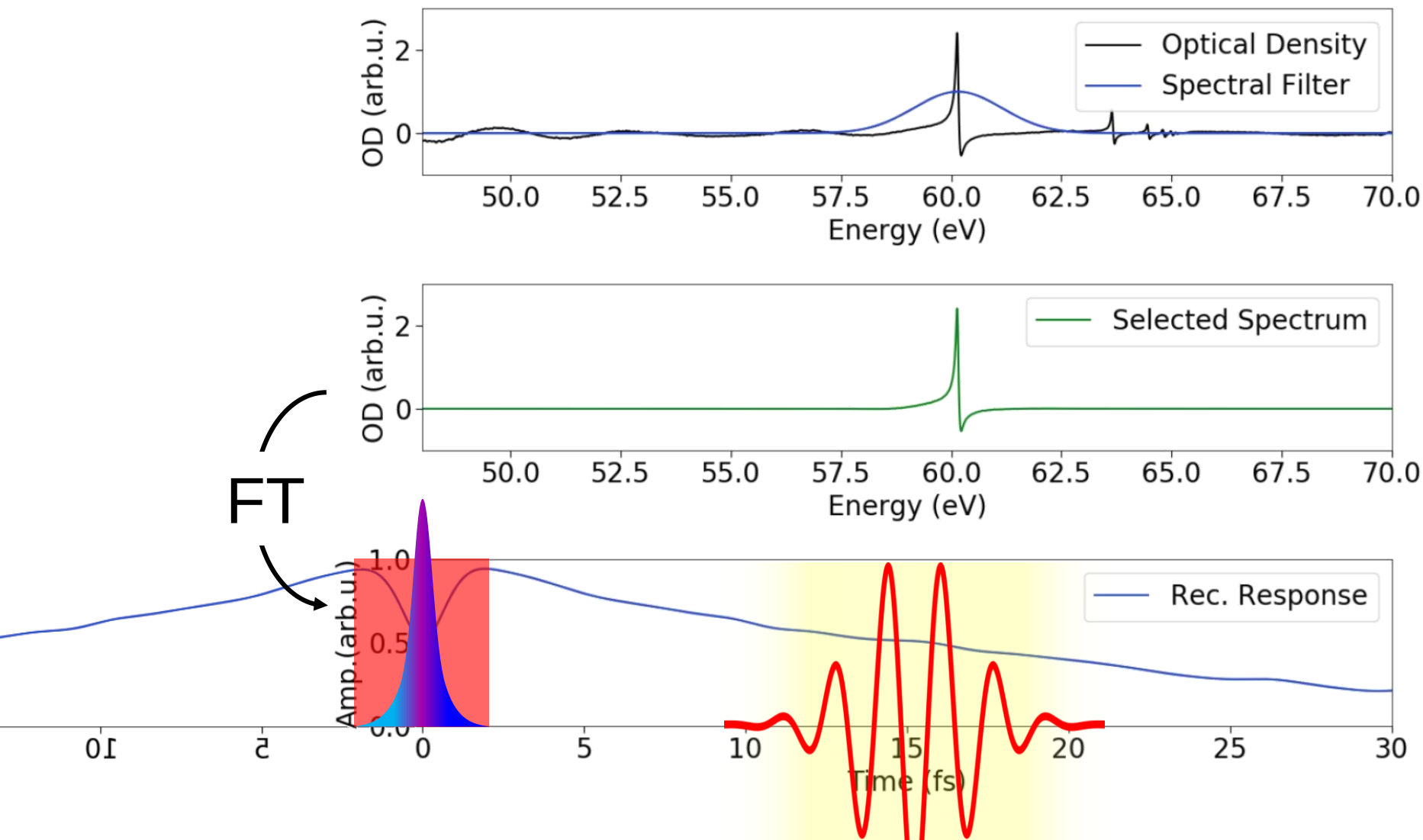
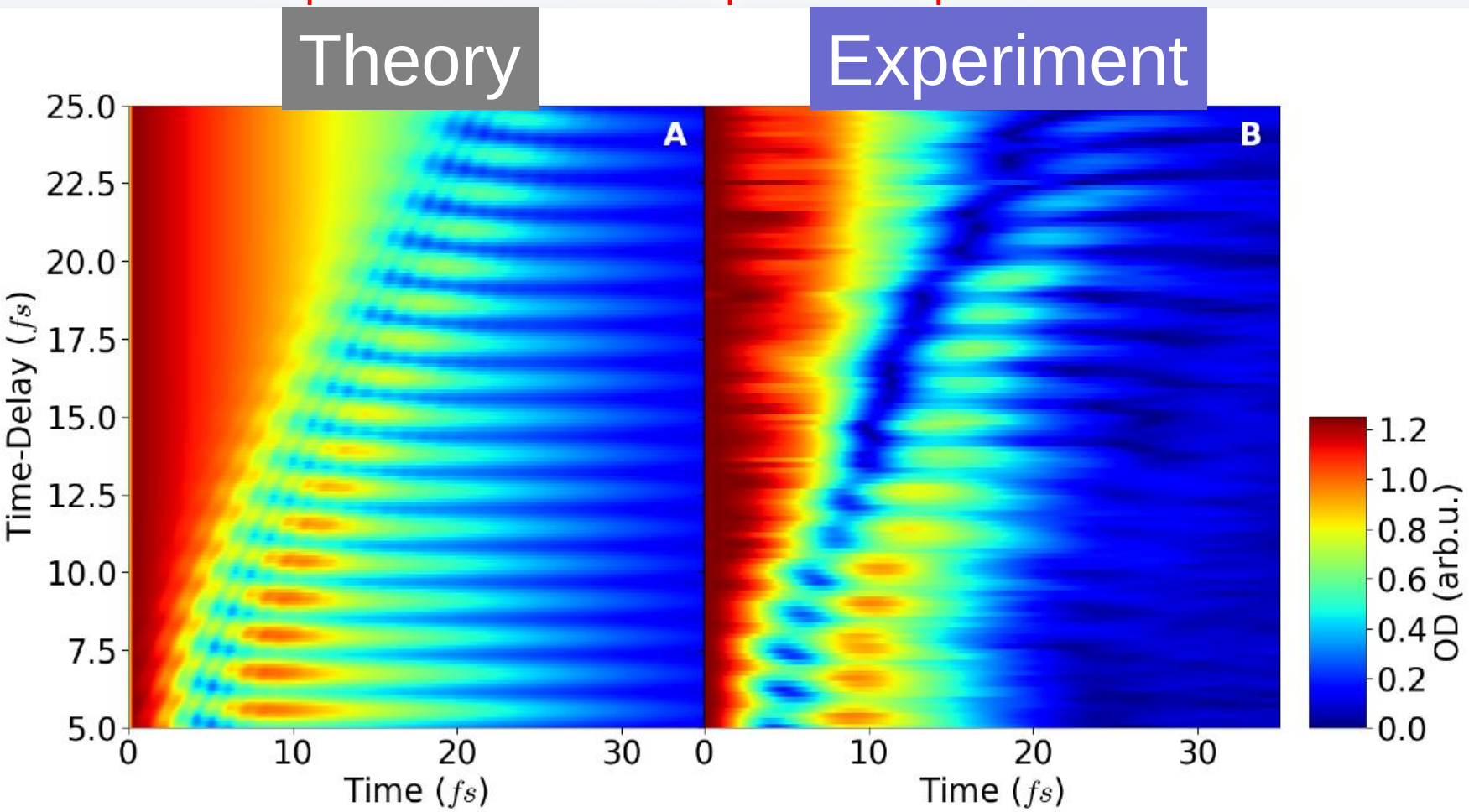
Time-domain reconstruction principle: the causal response



Time-delay dependence

the track of a strong laser pulse
imprinted in the time-dependent dipole moment



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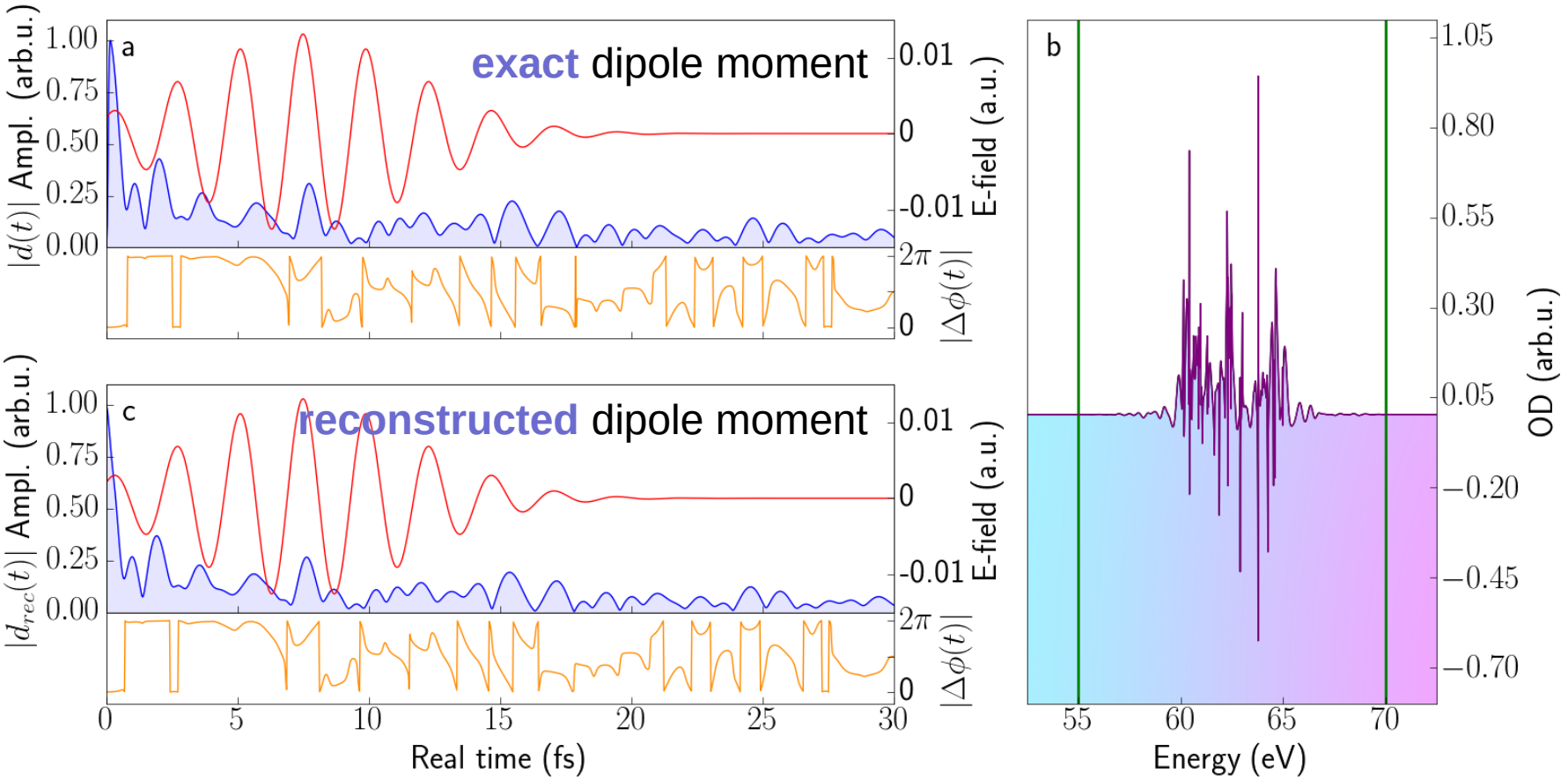
A general approach…

Here, a **complex 50-level system** strongly coupled to an intense laser field



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Christian Ott


Veit Stooß


Lennart Aufleger


Thomas Ding


Marc Rebholz


Patrick Rupprecht

Main Cooperators: Thanks to … and many more !

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C. Greene (Purdue Univ., USA): Fano Thory
L. Argenti, F. Martín (Univ. A. Madrid, Spain): 2e⁻ Theory
J. Madroñero (TUM&Univ. del Valle, Cali, Colombia): 2e⁻ Theory
S. Roling, H. Zacharias (Univ. Münster): FLASH Split and Delay
S. Düsterer, R. Treusch, G. Brenner (DESY): FLASH exp.
R. Pazourek, S. Donsa, I. Březinová
J. Burgdörfer (Vienna Univ.): Fano birth, TDDM
H. Wei, C.-D. Lin (Kansas State Univ., USA): Fano birth

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Grant #PF790

erc
X-MuSiC

Heidelberg Center for Quantum Dynamics

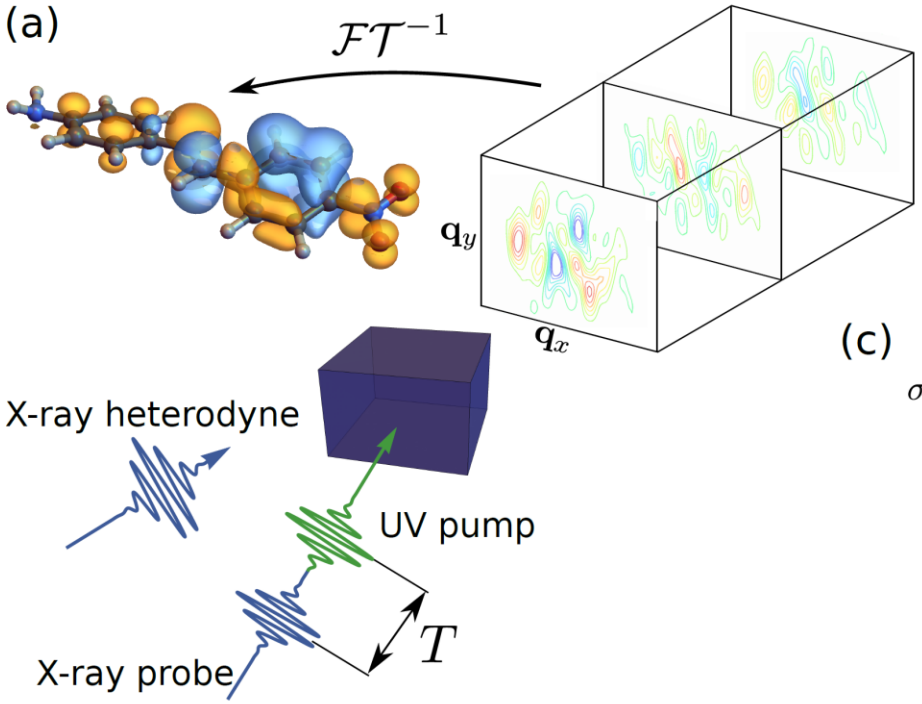
arXiv:1802.04421v1 [physics.chem-ph] 13 Feb 2018

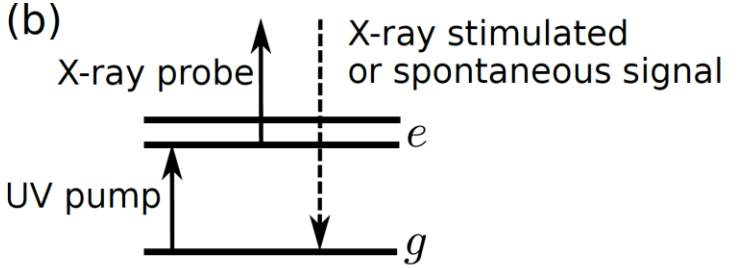
X-Ray sum frequency generation; direct imaging of ultrafast electron dynamics

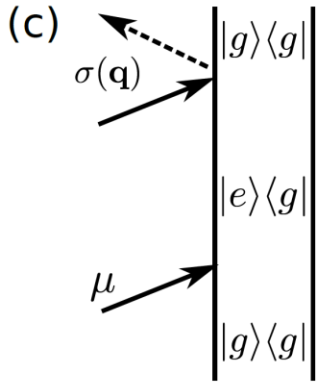
Jérémy R. Rouxel,* Markus Kowalewski, Kochise Bennett, and Shaul Mukamel†

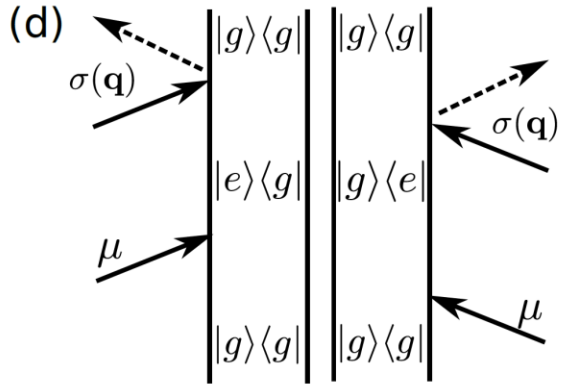
Department of Chemistry and Department of Physics and Astronomy, University of California,Irvine, CA 92697, USA

(Dated: February 14, 2018)



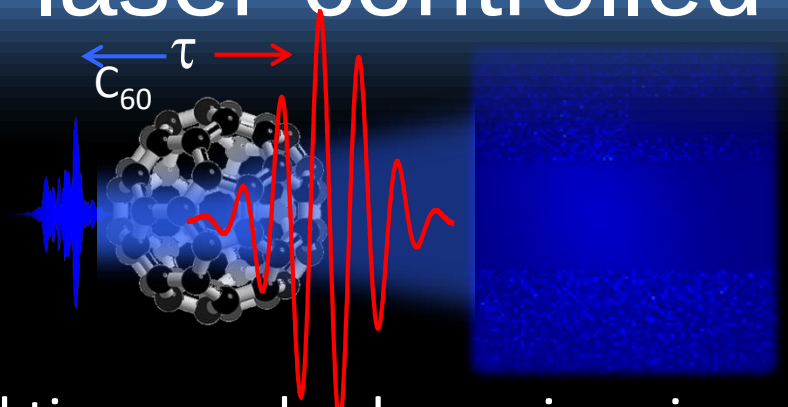






Imaging of laser-controlled molecules

only
~50 scattered ph/pulse
benefits from
high repetition rates



at LCLS, 2 keV

- femtosecond time-resolved x-ray imaging
at 10^{11} / cm^3 gas-phase (~molecular jet) densities
is possible (for ensemble measurements)

MPIK Heidelberg
K. Schnorr, S. Augustin, G. Schmidt, R. Moshammer, t.p.

Tohoku University, Japan
K. Motumura, Y. Kumagai, Y. Ito, K. Ueda

University of Connecticut, USA
R. Obaid, N. Berrah

CFEL/DESY Hamburg
R. Livingstone, J. Küpper

mpi-mf Heidelberg
L. Foucar, I. Schlichting

MBI Berlin
C. P. Schulz, A. Rouzee, M. Vrakking

Argonne National Lab, USA
A. AlHaddad, C. Bostedt

mpi-bpc Göttingen/DESY
K. Kubicek, S. Techert

TU Berlin
J. Zimmermann, T. Möller

PTB Braunschweig
J. Ullrich

LCLS/SLAC
T. Osipov, R. Coffee, D. Ray
AMO-, Comp., Detector, DAQ groups

Ohio State University
M. Lai, C. Blaga, L. DiMauro

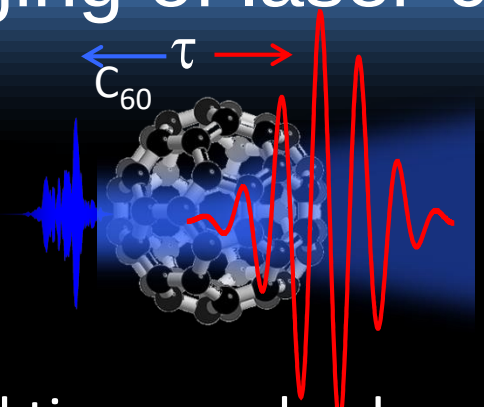
mpi-pks Dresden
U. Saalmann, J.M. Rost

TU Dresden
J. Handt, R. Schmidt

Kansas State University, USA
D. Rolles, A. Rudenko

Imaging of laser-controlled C_{60}

would benefit from high repetition rates



- femtosecond time-resolved x-ray imaging
  at 10^{11} / cm^3 gas-phase (~molecular jet) densities
  is possible (for ensemble measurements)
- in C_{60}: observation of different dynamical regimes
  depending on (optical) laser intensity

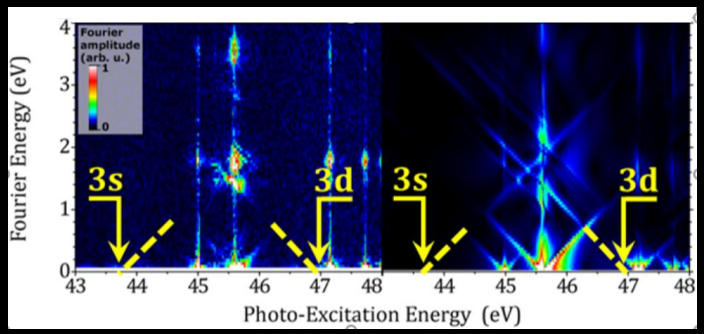
low intensity $10^{14} \frac{W}{cm^2}$ → high intensity $10^{15} \frac{W}{cm^2}$

Can we steer, and watch, **laser-driven dynamics** in real time?

Conclusion: A wish list...

Observing and steering electron motion in atoms and molecules with FELs

- Attosecond pulsed FELs, why:
  - **electrons are fast, and they are everywhere (localize)**
  - need to compete with ionization: **nonlinear processes win for short pulses**
- High intensity for
  - pump-probe
  - 2D spectroscopy
  - quantum control
- Controllable properties
  - Polarization
  - broadband (multi-eV) and/or multi color

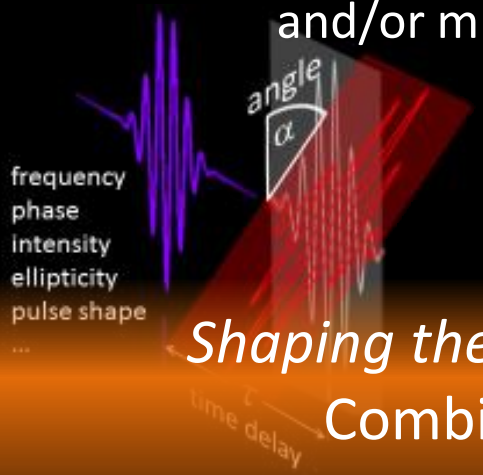


FEL-induced Stark shifts of resonance transitions in Neon

50 meV shifts at just 10^{14} W/cm^2

=> 50 eV shifts at 10^{20} W/cm^2

=> 500 as pulse with 100 μJ, focused to 0.5 μm



Shaping the Future: New scientific capabilities at EuXFEL:

Combine nonlinear spectroscopy with imaging

European XFEL GmbH
Holzkoppel 4
22869 Schenefeld
Germany

March 5, 2019

Technical Concepts Scope for SASE4/5

Scope of proposed concepts from the workshop "Shaping the future of the European XFEL: options for the SASE4/5 tunnels" held at the European XFEL, Schenefeld, December 6-7, 2018

Authors

Winfried Decking (DESY)

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Serguei Molodtsov (European XFEL)

Joachim Pflüger (European XFEL)

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Harald Sinn (European XFEL)

Mikhail Yurkov (DESY)

from Technical Concepts Scope SASE4/5

4. Soft X-ray FEL line with extended user capabilities

Scope

- Extended tunability range at a fixed electron energy (up to factor of 8 for upper boundary of the operating wavelength range with respect to SASE3).
- Extension of wavelength range towards lower photon energies.
- Extension of user capabilities with pump-probe experiments using independent FEL colors in the whole range.
- Full polarization control.
- Capabilities to generate ultimate radiation pulse energies in a Joule range.

Advanced option of radiation generation like Self-Seeding, Harmonic Lasing Self-Seeding (HLSS), maybe HGHG or EEHG (at longer wavelengths) can be implemented as well. These options allow for a significant increase of the coherence time.

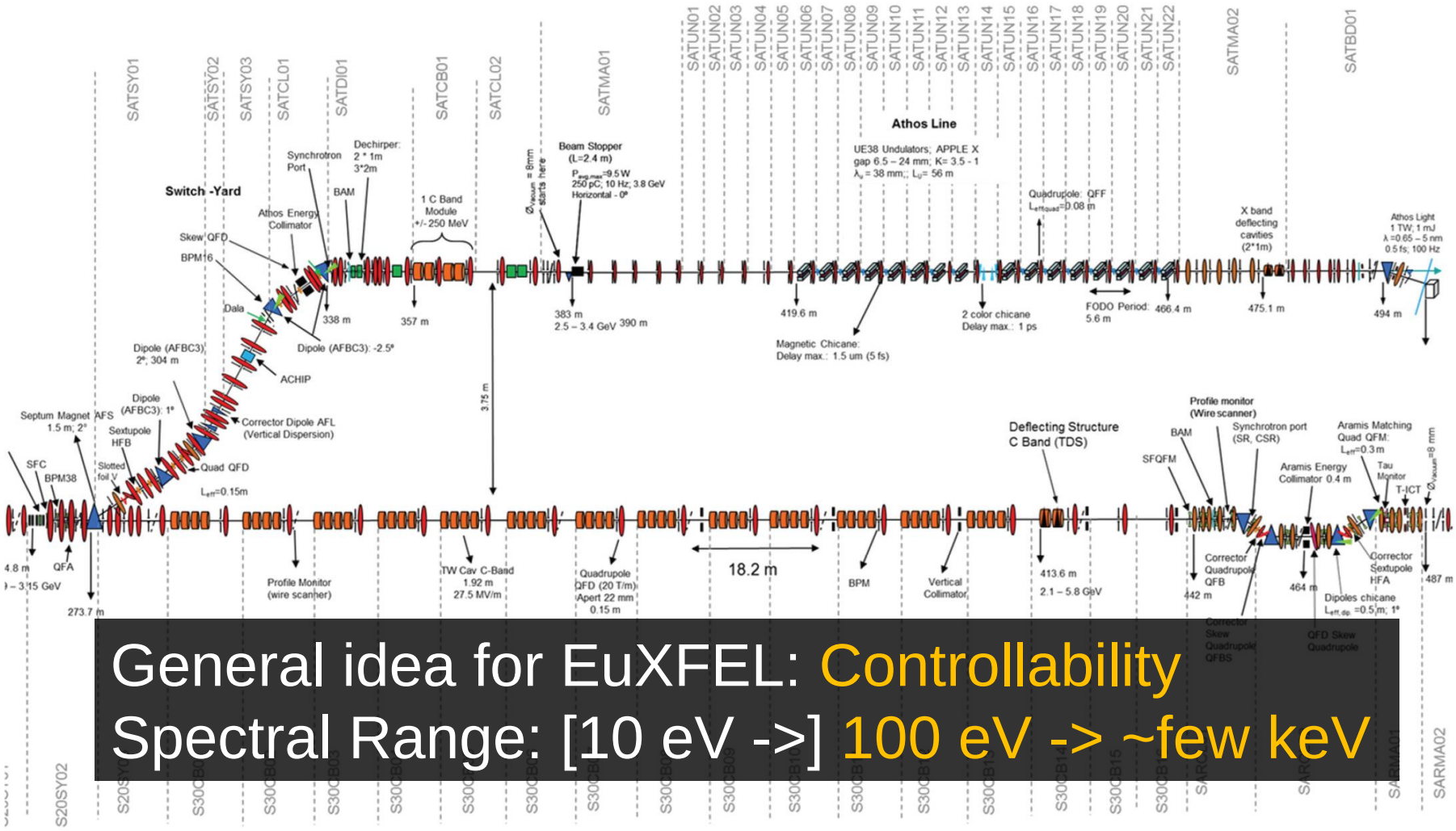
7. Superradiance for X-ray Production

Scope

Use chicanes, integrated in the undulator structure in combination with a manipulation of the electron beam, to generate high power, very short pulses comparable to the coherence time. The peak power that can be achieved this way is an order of magnitude higher than normal. Simulations for the PSI case show sub-femtosecond pulses with TW of power can be achieved in the nanometer range, but the method is in principle scalable to the European XFEL case and relative photon energies.

Advanced undulator design also @SwissFEL

38th International Free Electron Laser Conference FEL2017, Santa Fe, NM, USA JACoW Publishing
ISBN: 978-3-95450-179-3 doi:10.18429/JACoW-FEL2017-MOP038



General idea for EuXFEL: Controllability
Spectral Range: [10 eV ->] 100 eV -> ~few keV

Figure 2: Layout of the Athos FEL line with the main electron beam components going from z ~270m to z~500m (electron source photocathode is at z~0m). Athos is parallel to the linac 3 of SwissFEL.