ICPEAC XXX
Cairns, Australia
2017-07-25

Tutorial:
Shaping atoms and molecules with strong laser fields

Outline

- Introduction
  (what is the shape of atoms?)
- What happens to atoms in strong laser fields?
  (besides ionization and recollision)
- How to experimentally measure shape-changing atoms?
  (methods viable for neutral species)
- What do we learn?
- Where do we go with it?

Thomas Pfeifer
MPIK Heidelberg, Germany

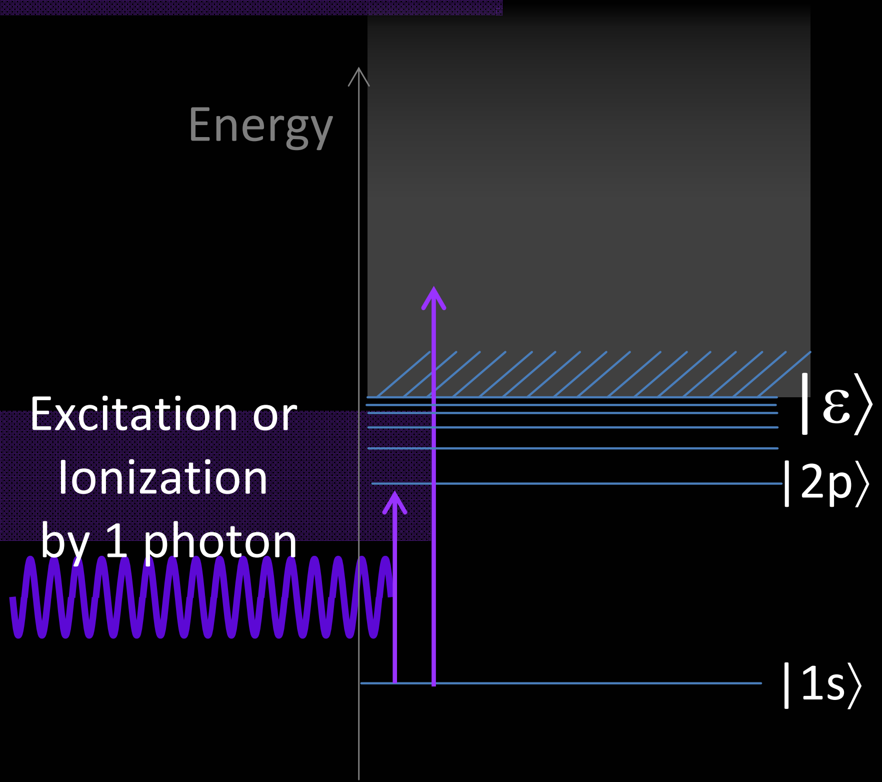
The “shape“ of atoms …

… in the Energy domain: The Spectrum

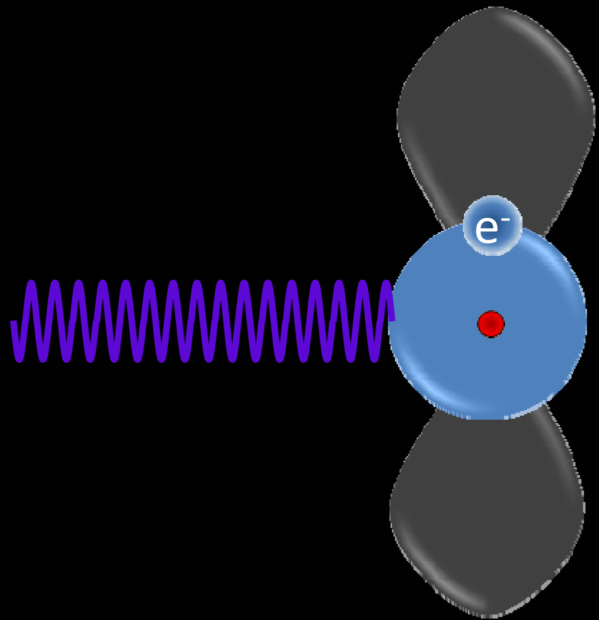
… in real space: Wavefunction/Orbital

… and their interaction with light

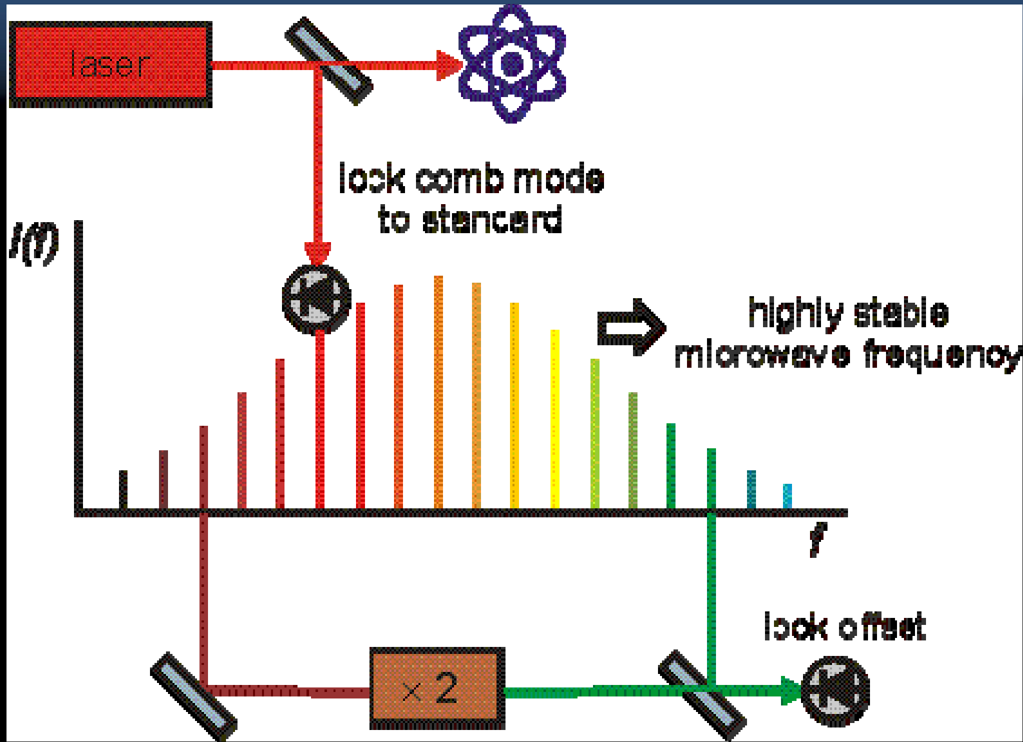
weak …



Excitation or
Ionization
by 1 photon



Atomic clocks



www.npl.co.uk/

An Atomic Clock with 10^{-18} Instability

N. Hinkley,[1,2] J. A. Sherman,[1] N. B. Phillips,[1] M. Schioppo,[1] N. D. Lemke,[1] K. Beloy,[1] M. Pizzocaro,[1,3,4] C. W. Oates,[1] A. D. Ludlow[1]*

Yb atoms

Science 341, 1215 (2013)

Sr atoms

An optical lattice clock with accuracy and stability at the 10^{-18} level

Nature 506, 71 (2014)

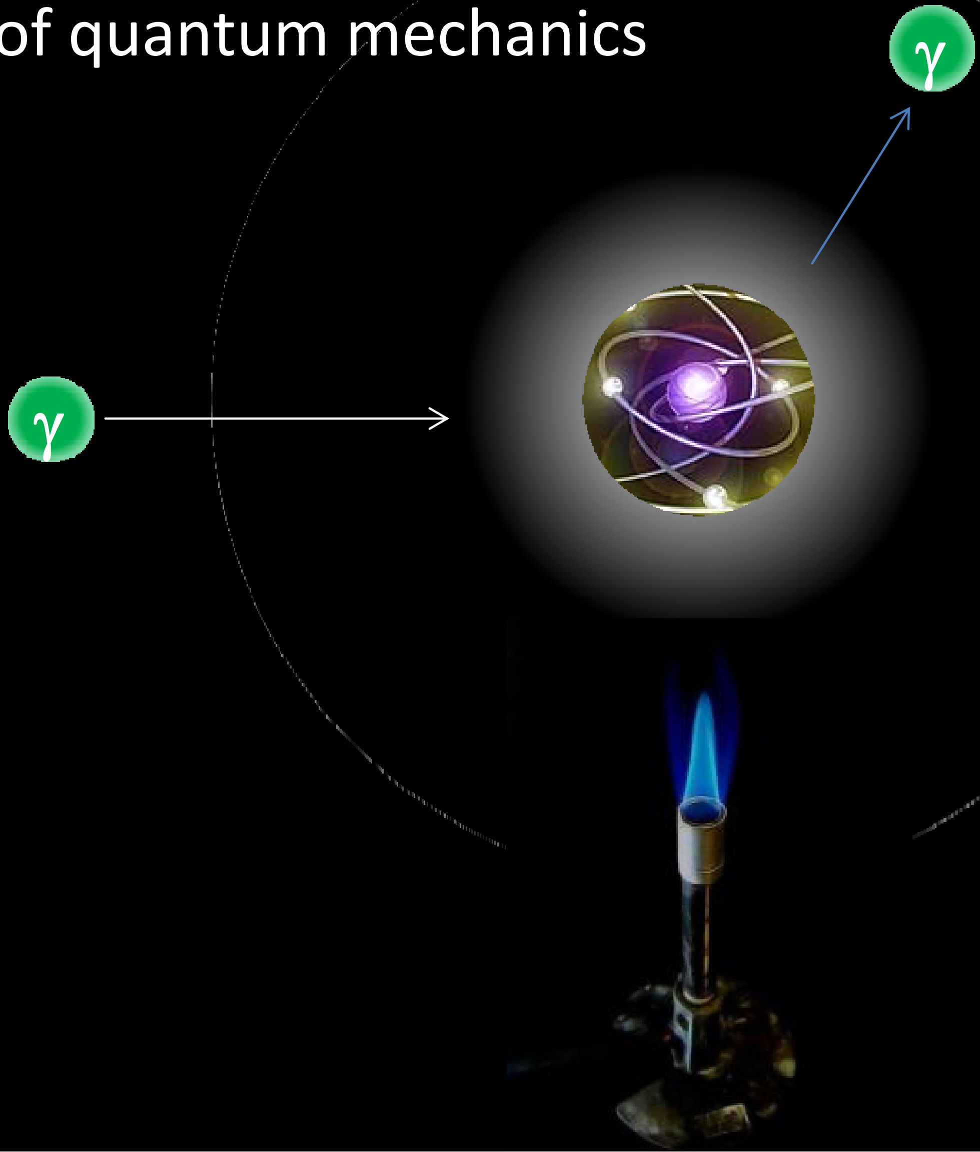
B. J. Bloom[1,2]*, T. L. Nicholson[1,2]*, J. R. Williams[1,2]†, S. L. Campbell[1,2], M. Bishof[1,2], X. Zhang[1,2], W. Zhang[1,2], S. L. Bromley[1,2] & J. Ye[1,2]

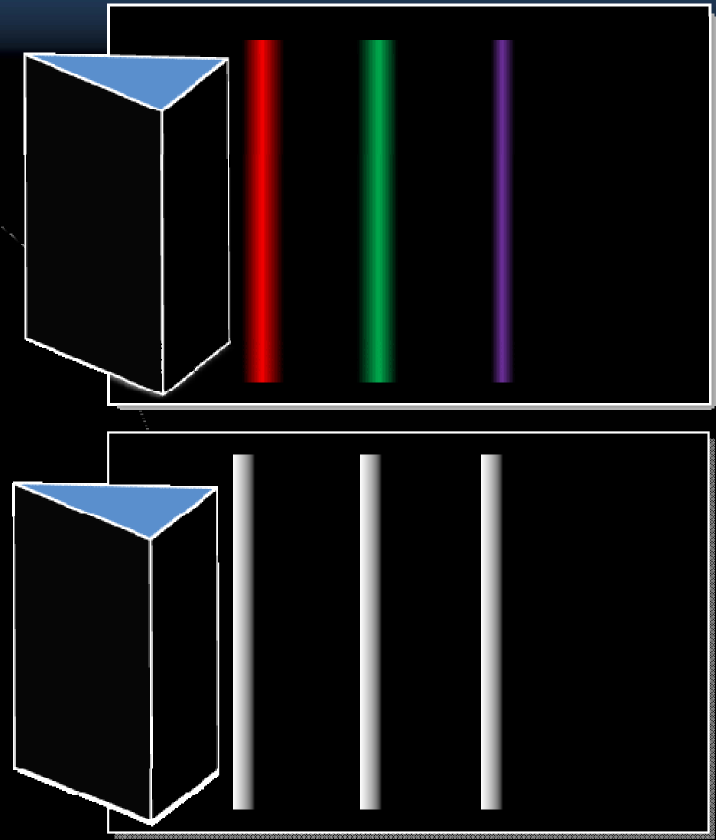
traditional spectroscopy

(Kirchhoff, Bunsen, *et al. @Heidelberg ~1860*)

the *spectral structure*:

-> key to the discovery
of quantum mechanics



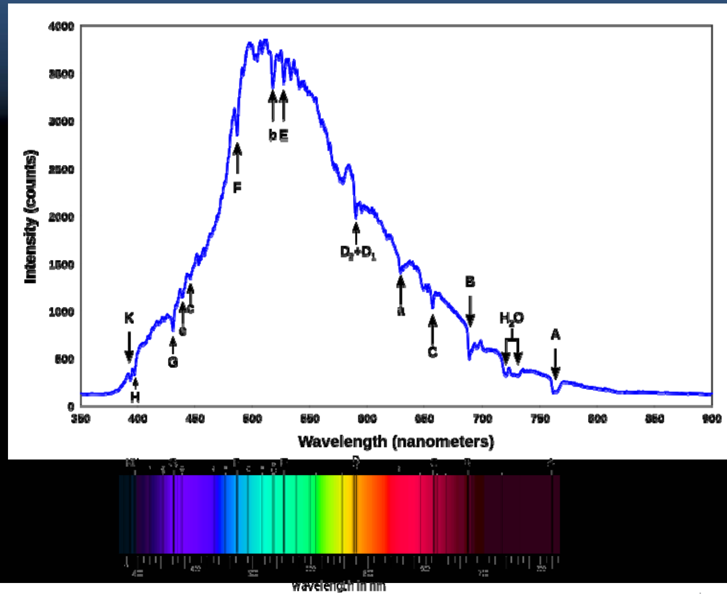


e.g. Fraunhofer *et al.*

atom image from: http://startswithabang.com/?p=1795
bunsen burner image from: http://www.west-michigan-family.com/holiday-archives-march.html
flame image from: http://commons.wikimedia.org/wiki/File:Bunsen_burner_flame_types_.jpg

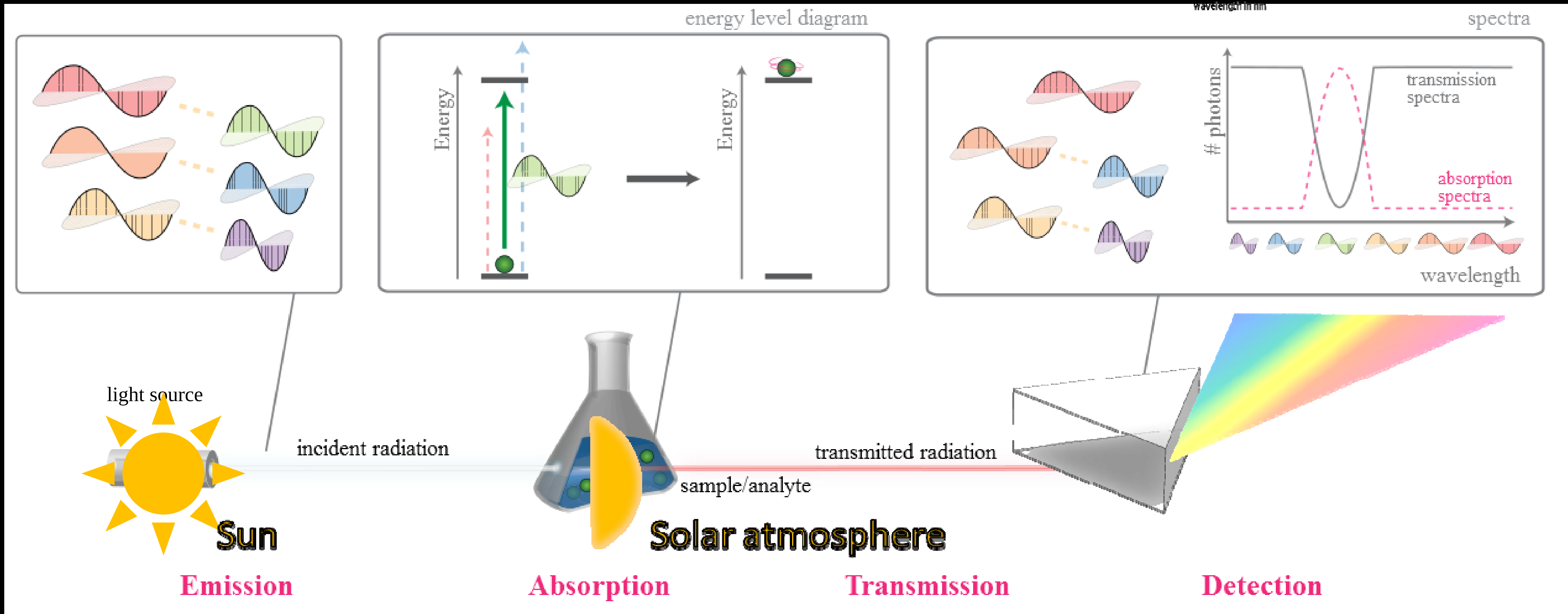
Absorption Spectroscopy

as explained on Wikipedia The Free Encyclopedia



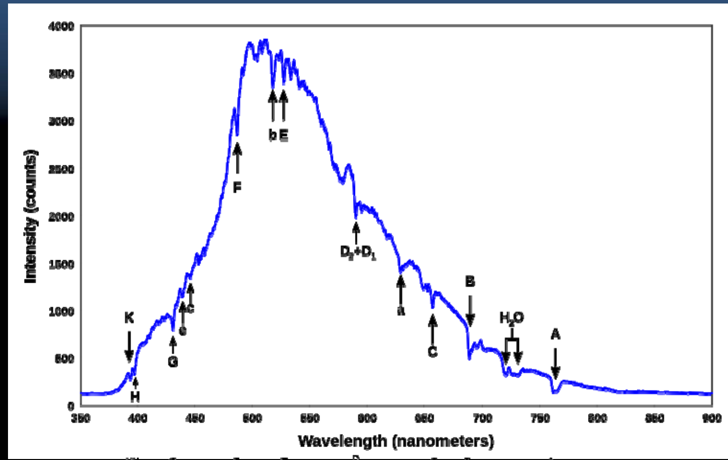
Wollaston (1802), Fraunhofer (1814):
Solar spectrum
assigned to characteristic emission lines
Kirchhoff, Bunsen (1859)

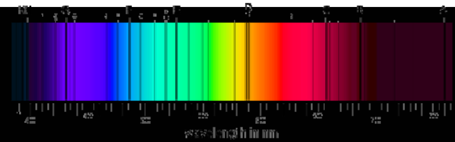
Spectral line positions



Absorption Spectroscopy

as explained on Wikipedia The Free Encyclopedia



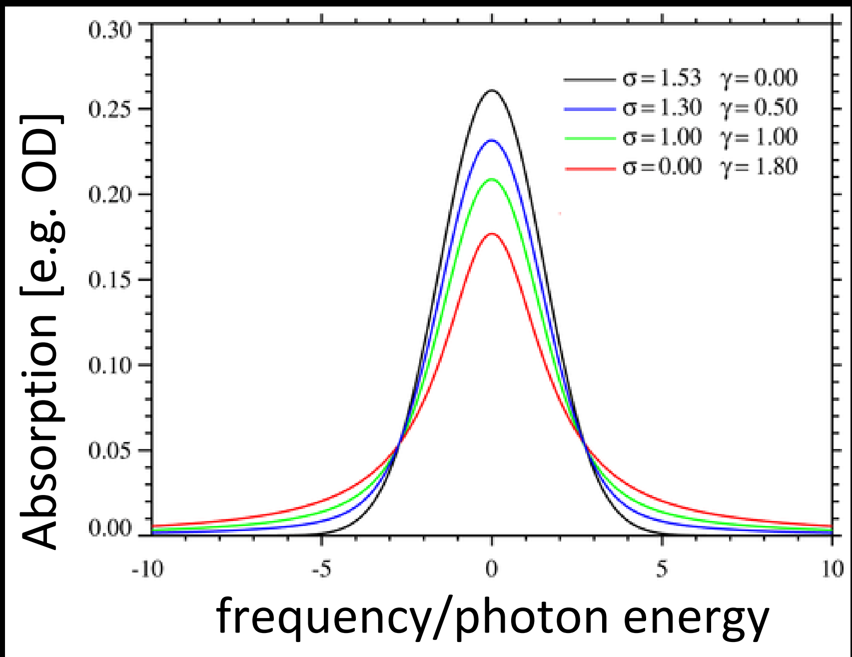


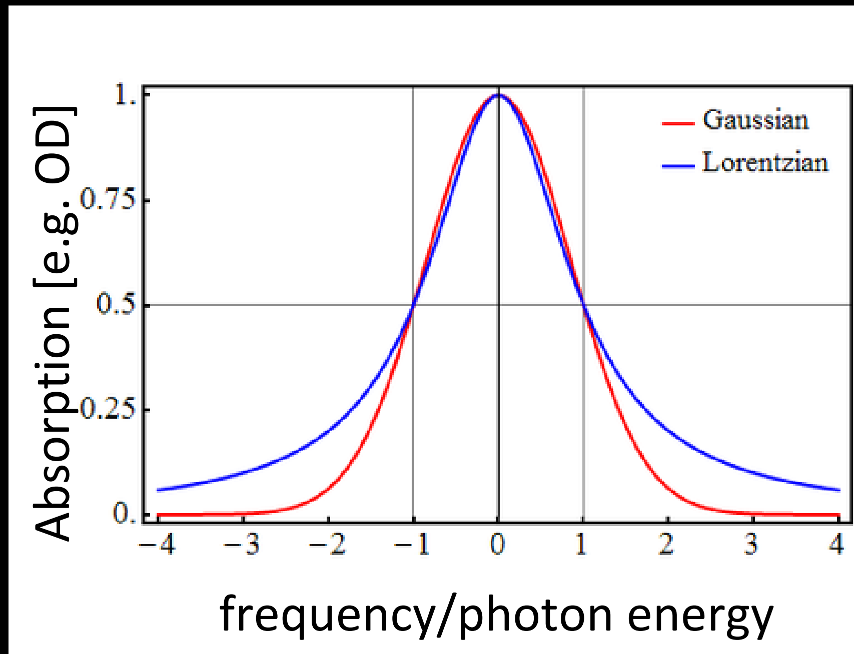
Wollaston (1802), Fraunhofer (1814):
Solar spectrum
assigned to characteristic emission lines
Kirchhoff, Bunsen (1859)

Spectral line positions

Spectral line widths

Spectral line shapes



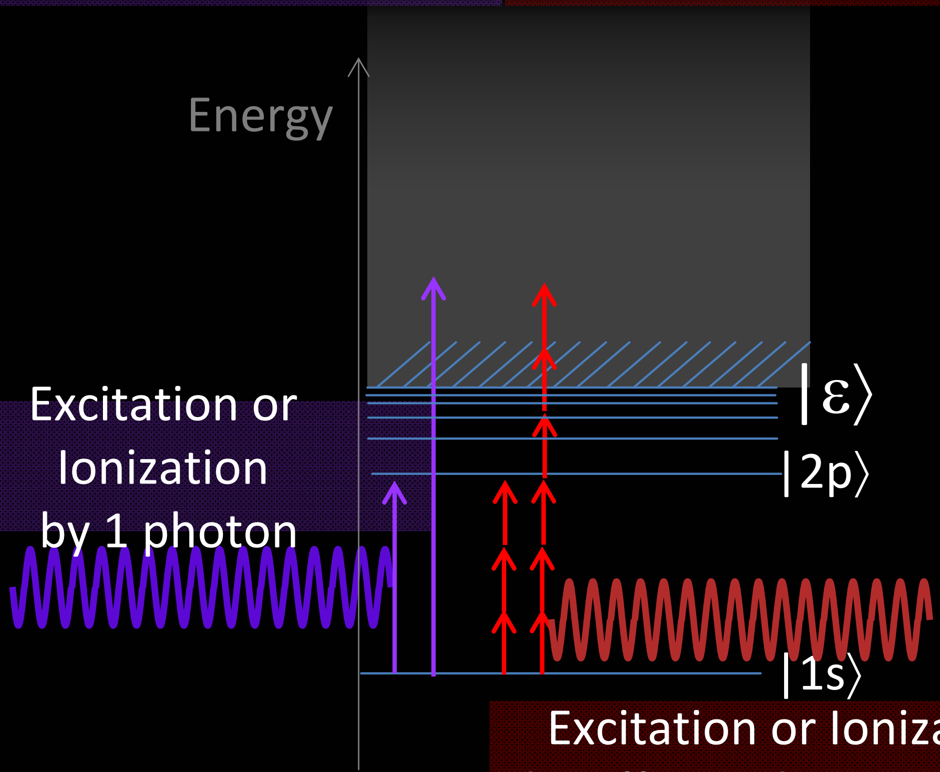


The „shape“ of atoms…

… gets ***changed*** by intense laser fields

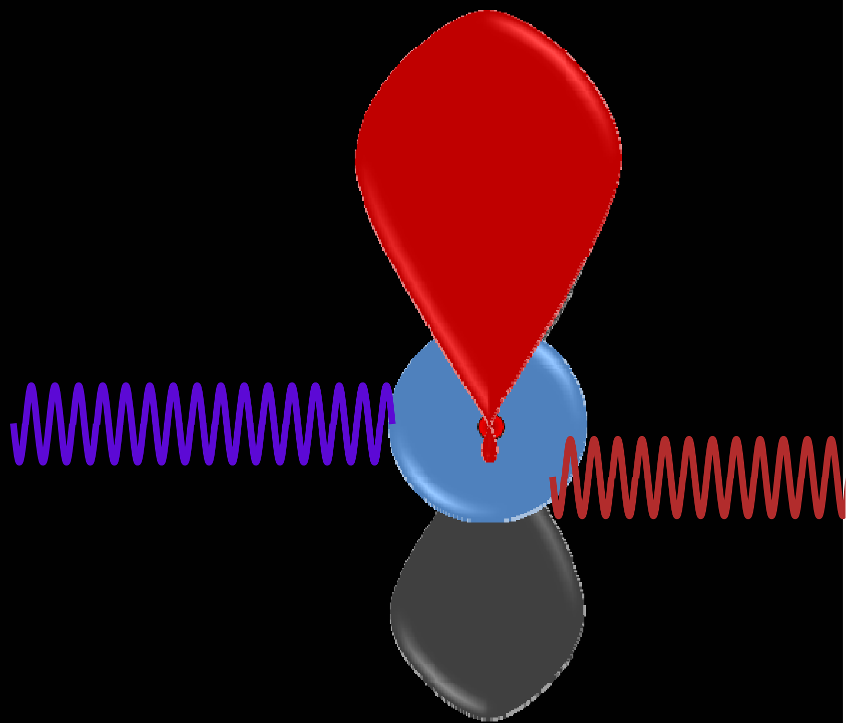
… and their interaction with light

weak …	… and strong fields

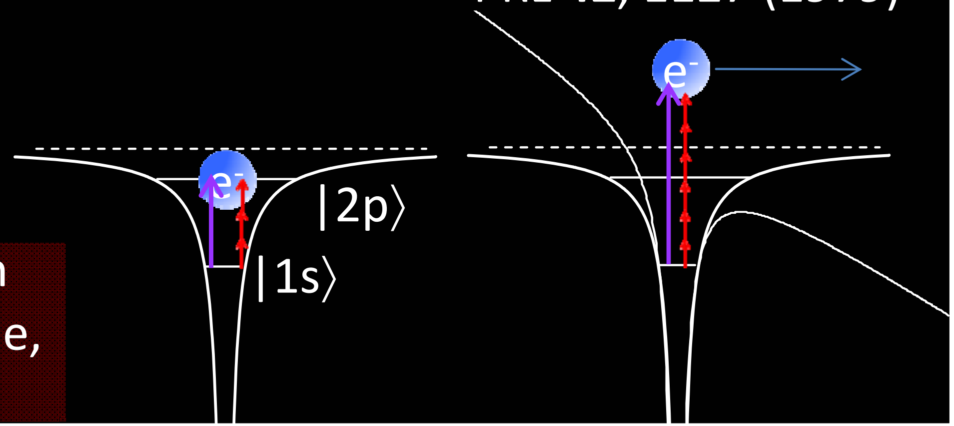


Excitation or Ionization by 1 photon

Excitation or Ionization by effectively N, multiple, photons, tunneling

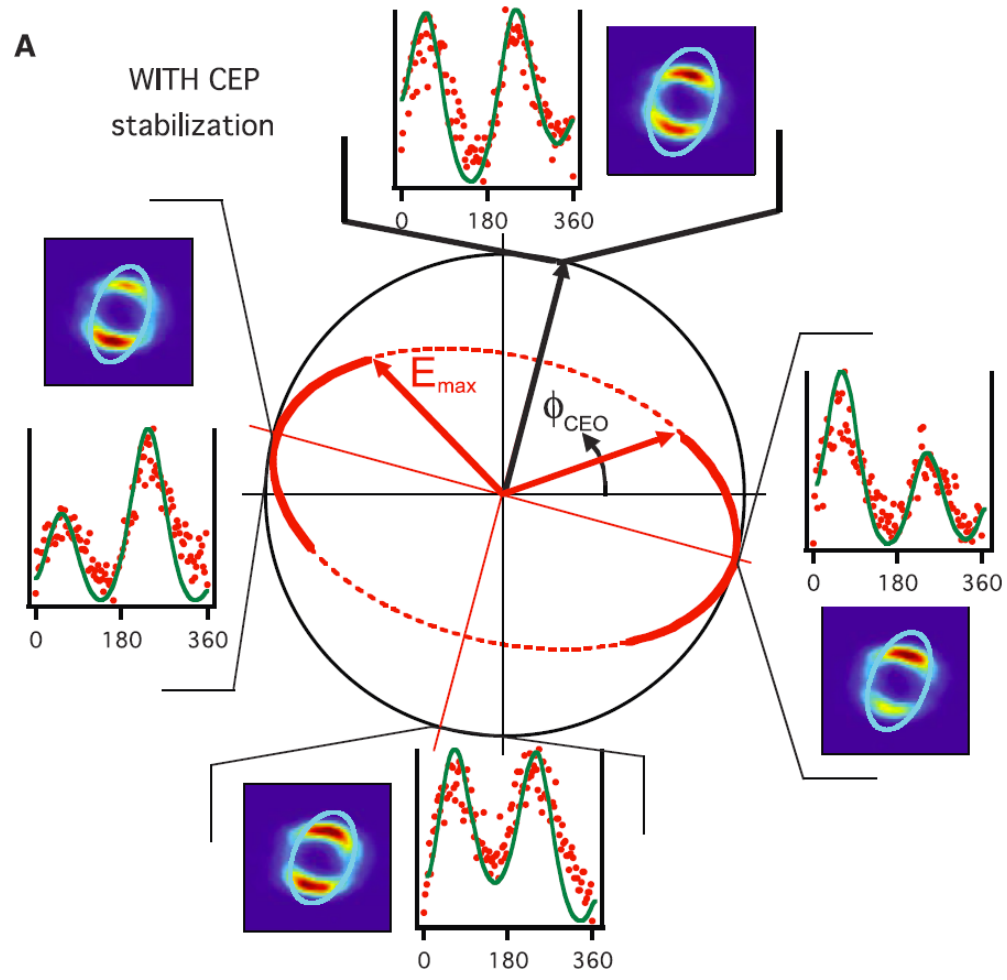


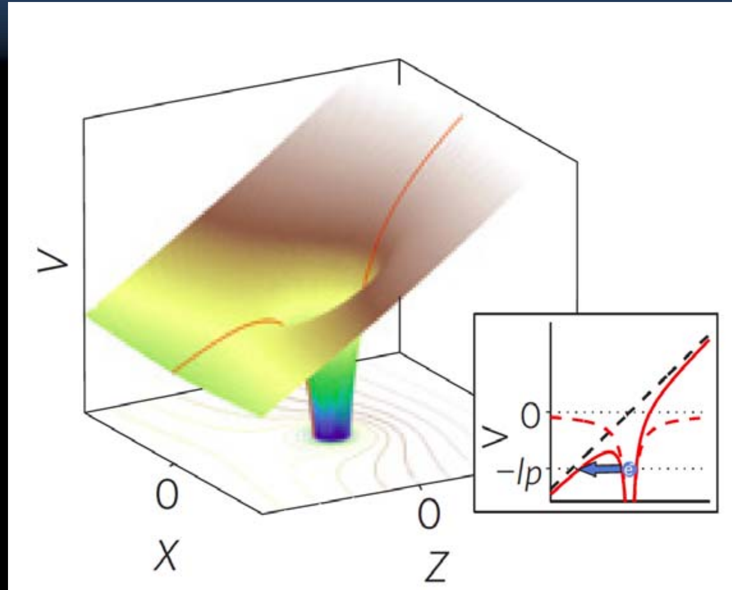
Pierre Agostini *et al.* PRL **42**, 1127 (1979)

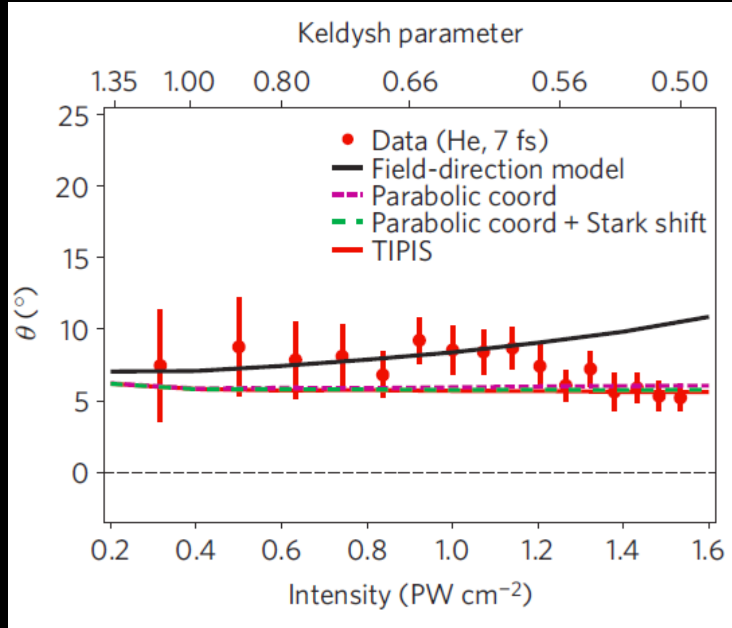


Strong-field ionization of Atoms

Keller group @ ETH
Attoclock, 10^{-18} sec precision



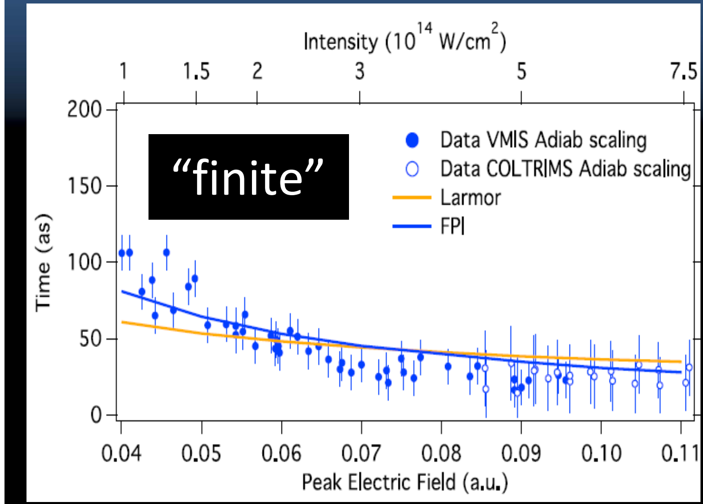




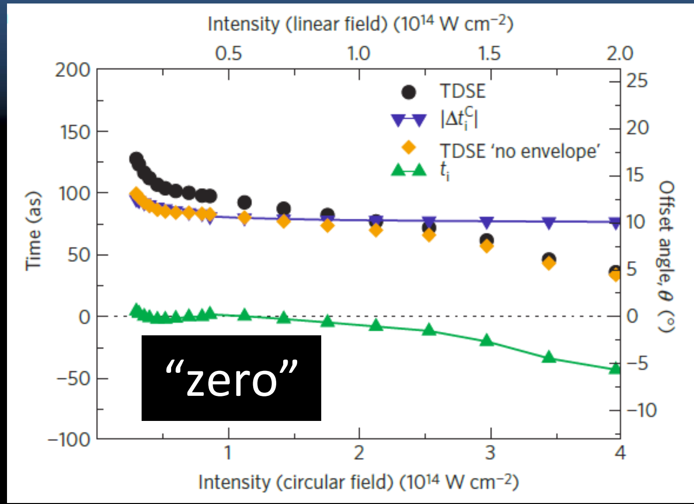
P. Eckle *et al.* Science **322**, 1525 (2008)

A. Pfeiffer *et al.* Nat. Physics (2012)

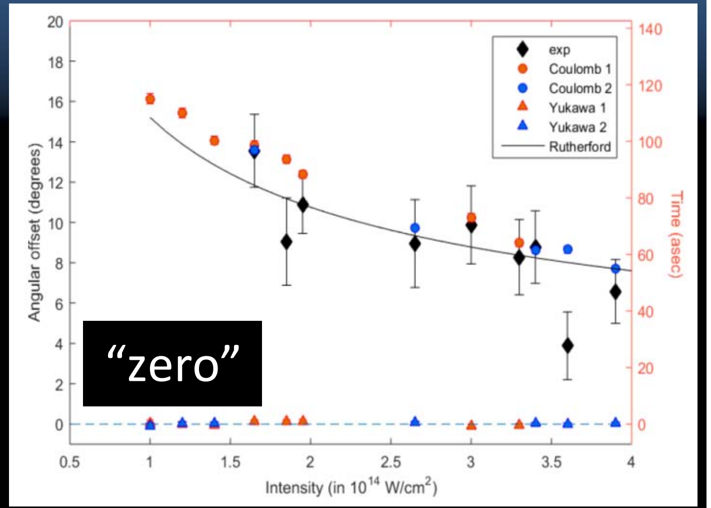
A never-ending controversy: “tunneling-time measurement”



Landsman *et al.* (Keller group)
Optica (2014)



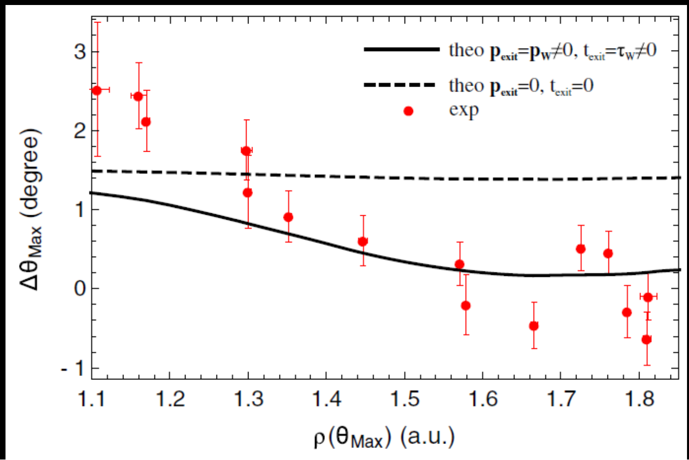
Torlina *et al.* (Smirnova group)
Nat. Physics (2015)



Sainadh *et al.* (Litvinyuk group)
arXiv:1707.05445 (2017)

<u>Where is the problem?</u>
- No time operator, hence: time not directly observable
- need to define a “time standard” (“controversy”)
- here in particular: mapping quantum dynamics (tunneling) into classical dynamics (trajectory)

Camus *et al.* PRL (2017)



Definition: Wigner time (Keitel group)
->Key: allow both **tunneling** <u>time</u> and <u>momentum</u>
Experimental test (Moshammer group)
->Key: Compare two species (Ar, Kr)
by coincidence measurements
under otherwise identical conditions
with high angular precision

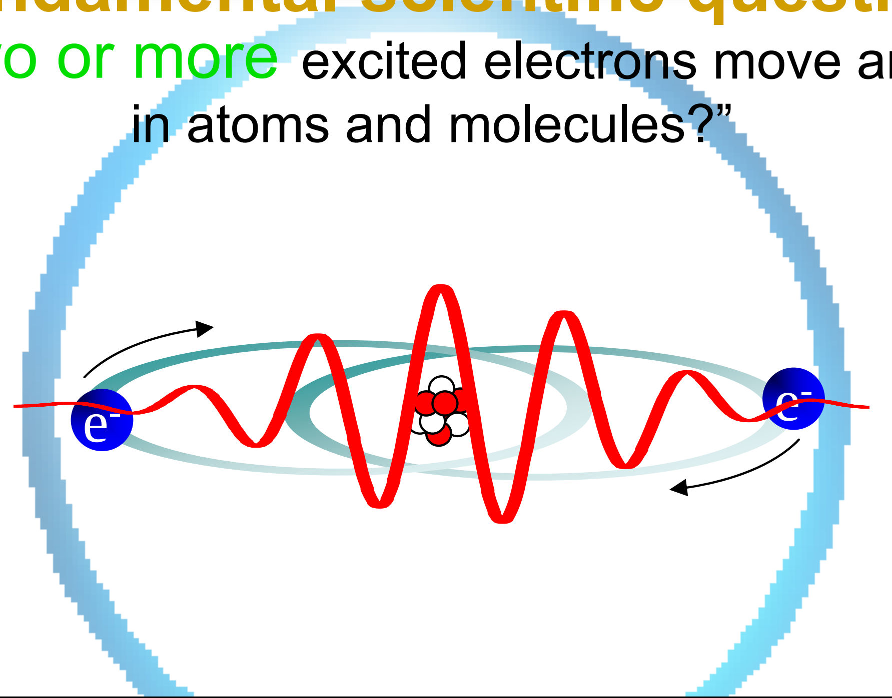
few-body quantum dynamics

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***

Sciencific goal:

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

0 as

(x-ray) movies of single molecules

Petahertz-clocked computing

x-ray precision spectroscopy

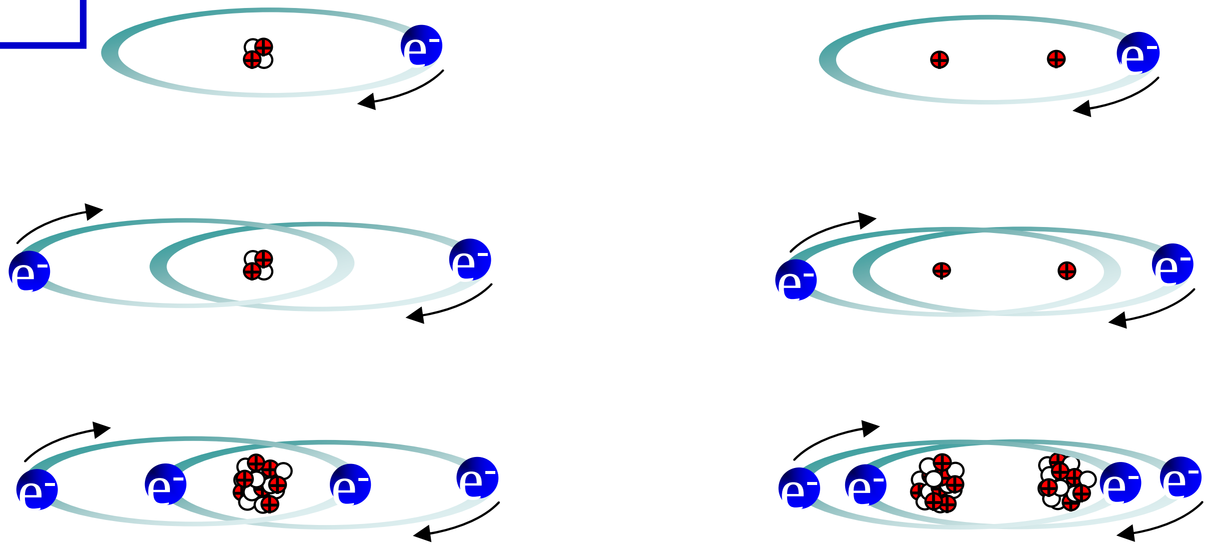
Laser control of chemical reactions

Fundamental Quantum Dynamics

Fermionic condensates in a nanotrap!

more fundamental

in atoms and small molecules:

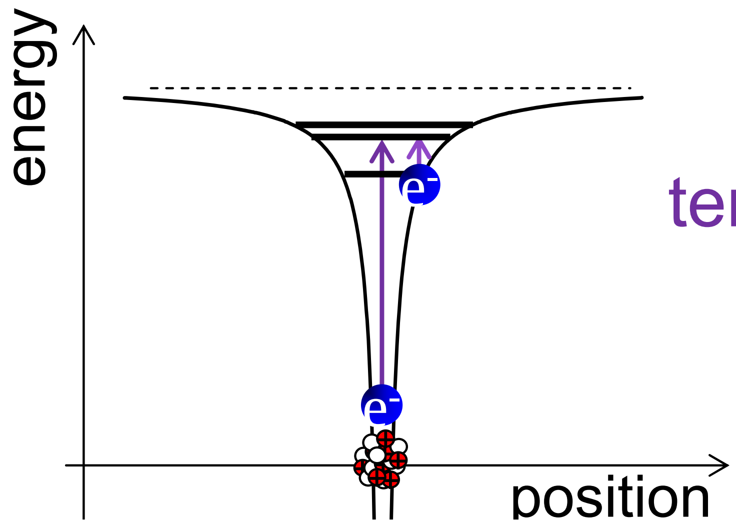


The **"quantum few-body problem"**

more complex

naturally well-defined, isolated, Å-sized 'labs'

What are the tools?



typical ΔE for transitions:

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

XUV to **x-ray**

light required for measurement and control

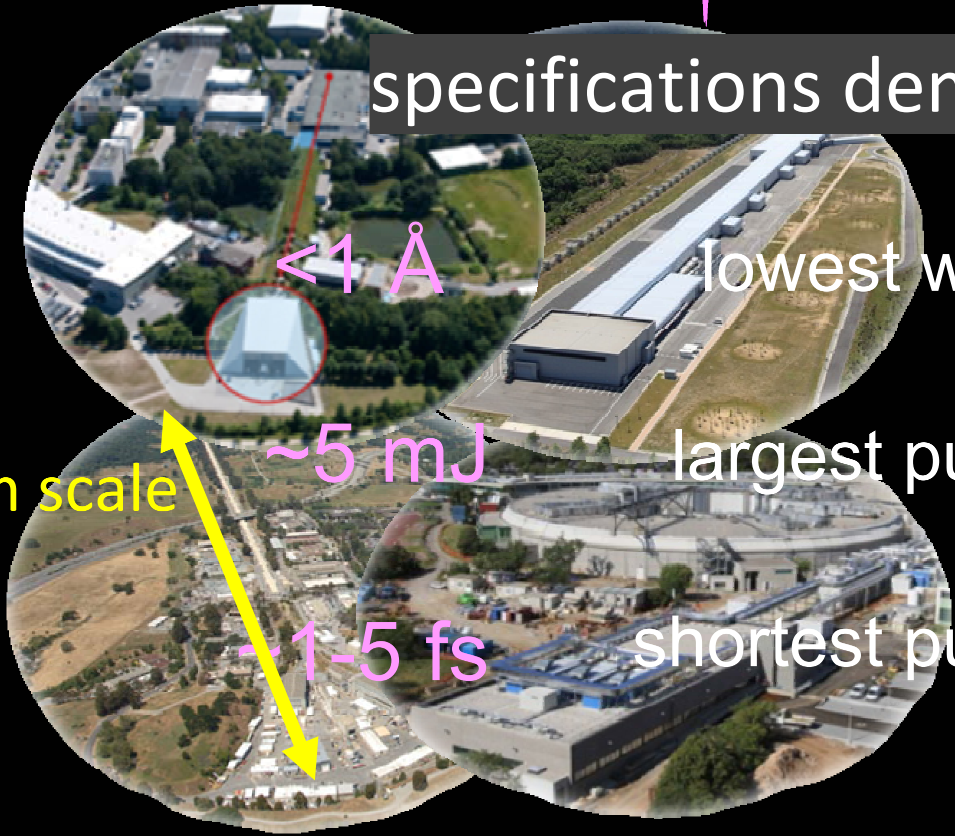
Two parallel revolutions in ultrashort x-ray/XUV laser science

Free **E**lectron **L**asers

High **H**armonic **G**eneration

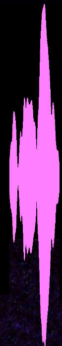
specifications demonstrated thus far:

Free Electron Lasers		High Harmonic Generation
<1 Å	lowest wavelength	~1 nm
~5 mJ	largest pulse energy	~few µJ
~1-5 fs	shortest pulse duration	~50 as

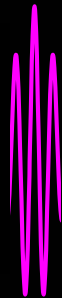


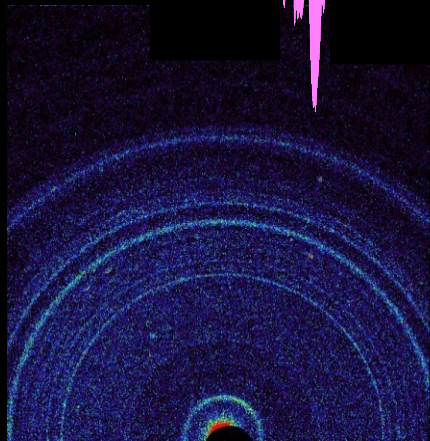
Time-resolved Science with novel x-ray/XUV laser sources

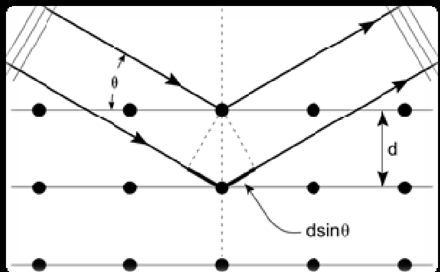
Free Electron Lasers



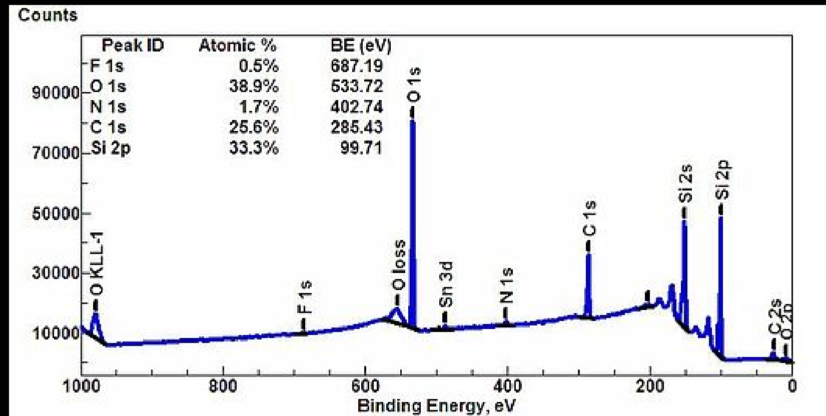
High Harmonic Generation

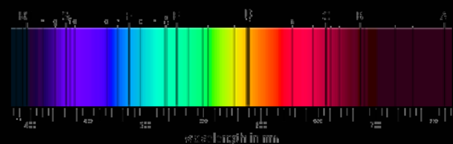






X-ray diffraction

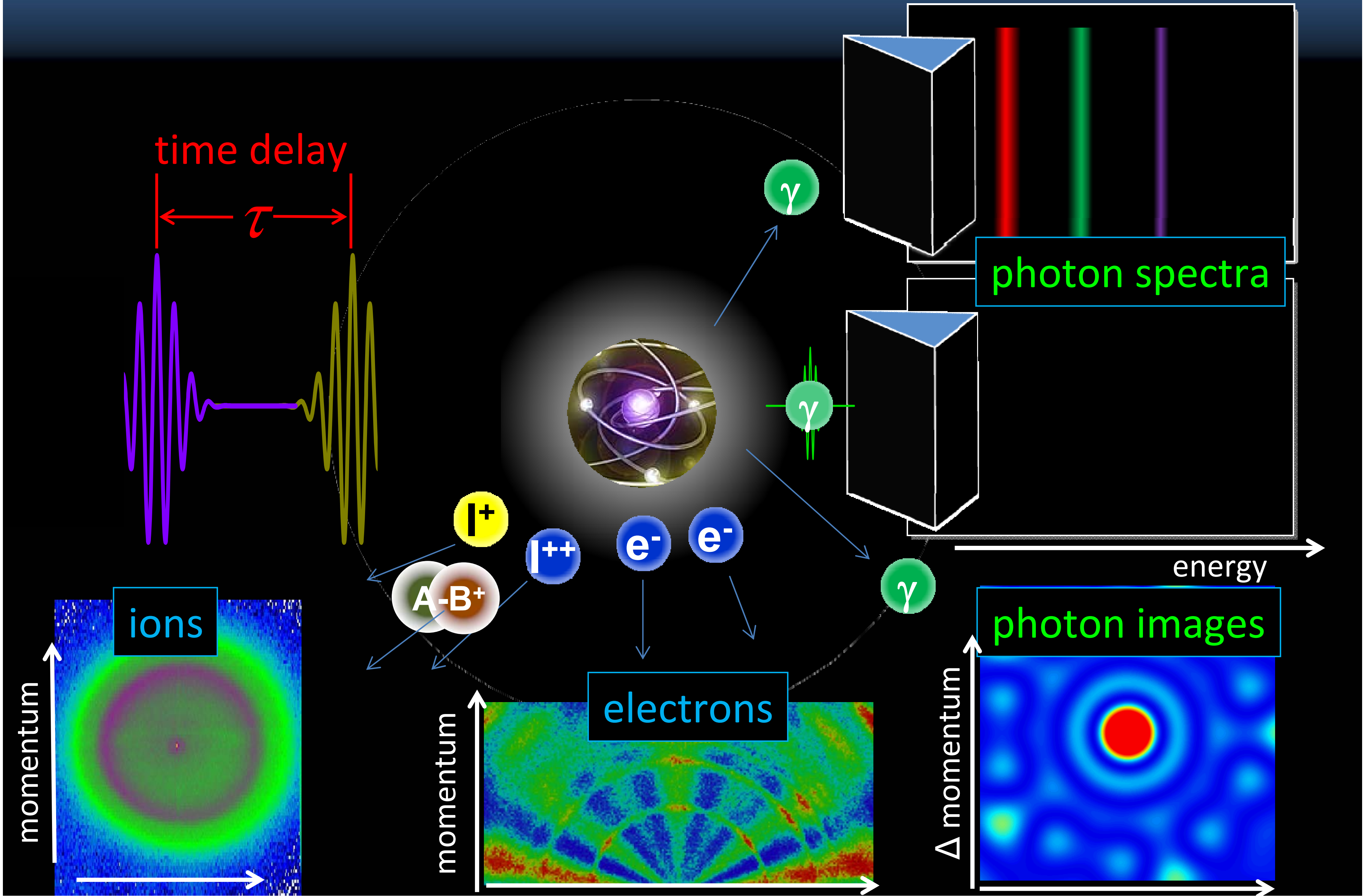




XUV spectroscopy

Figs.: Wikipedia

Time-resolved imaging and spectroscopy

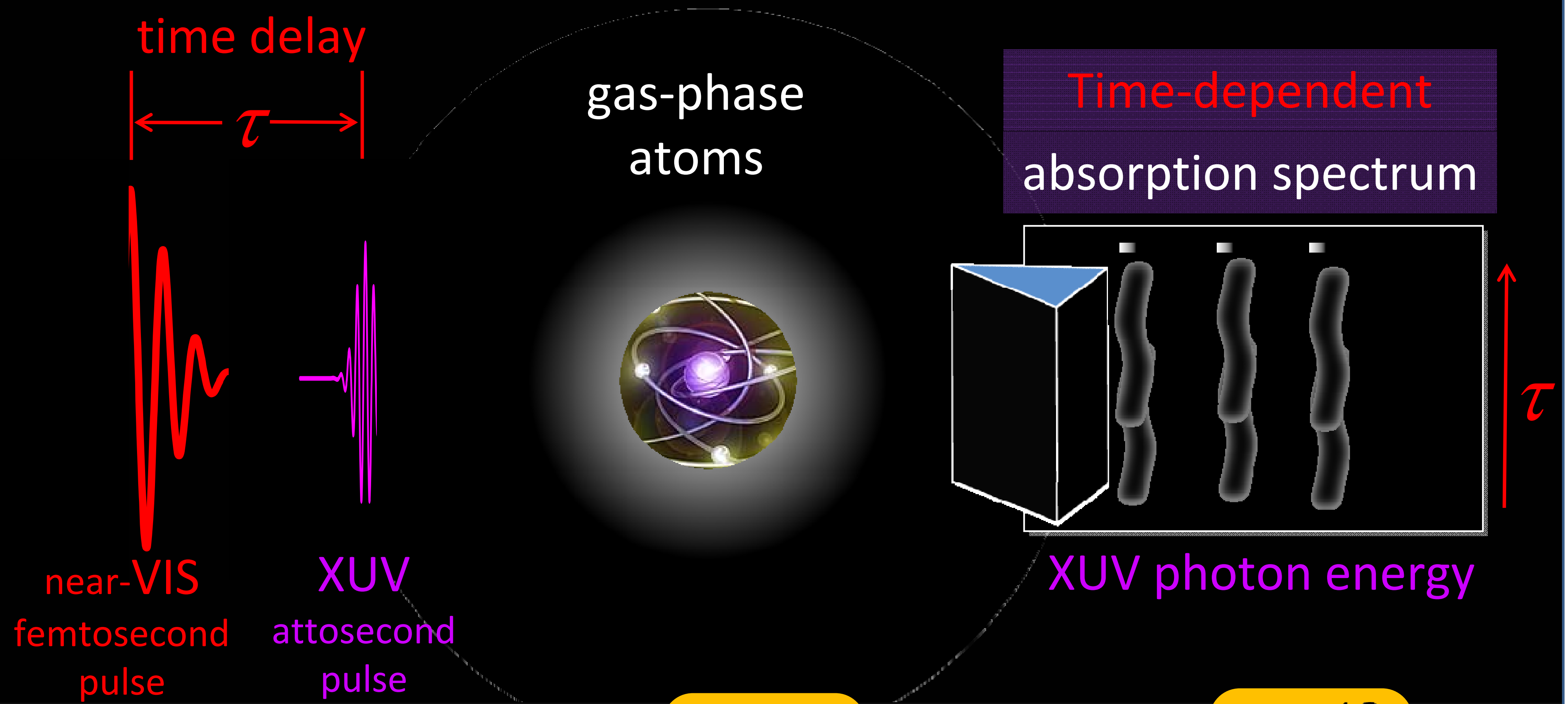


time-dependent XUV absorption spectroscopy

HHG-based applications, active groups:

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

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



Phase φ_1 Phase φ_2

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

time-dependent XUV absorption spectroscopy

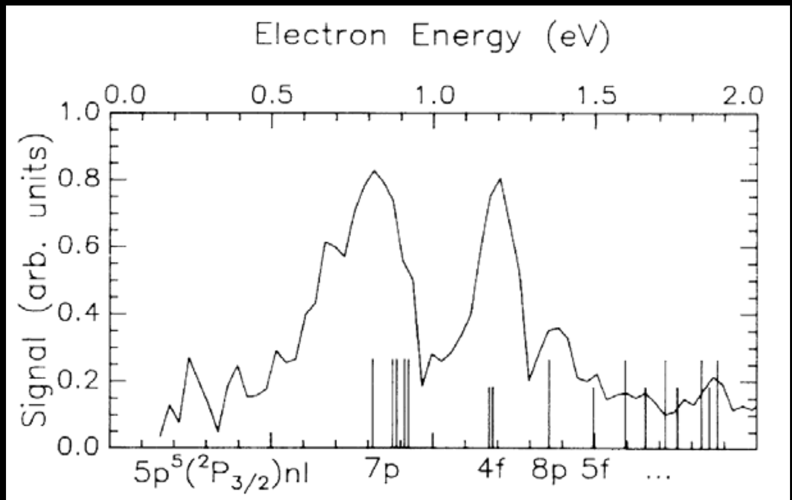
What happens to bound states and resonances in *short* and **strong** fields ?

Phase φ_1 Phase φ_2

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

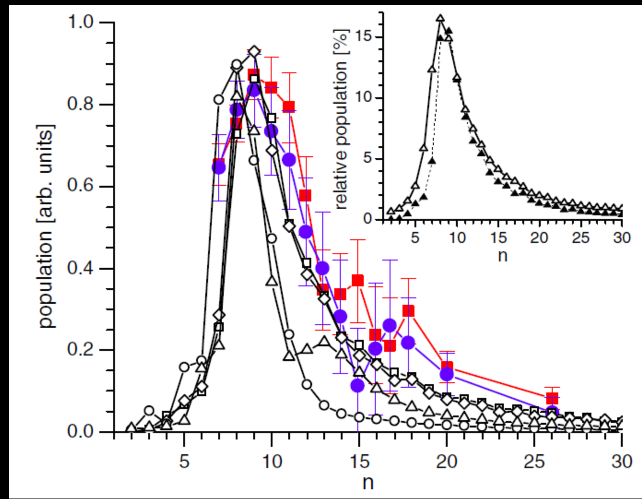
Role of bound (excited) states and resonances in (short) strong fields

Freeman resonances in strong-field ionization



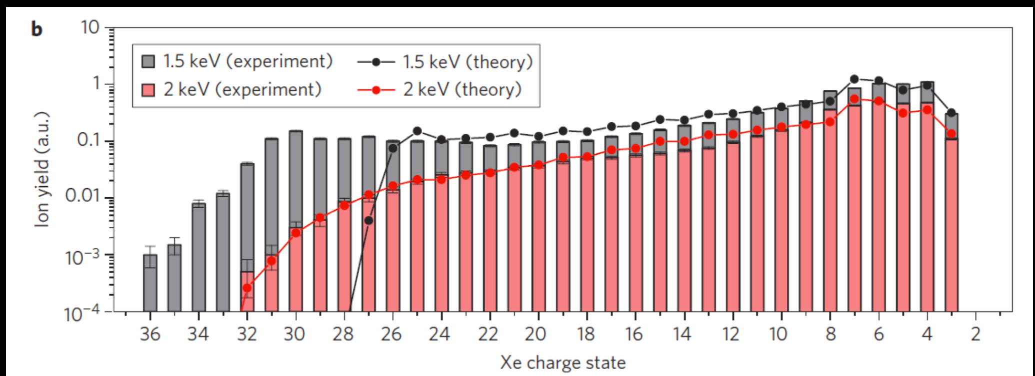
Freeman, Bucksbaum *et al.* PRL **59**, 1092 (1987)

frustrated tunnel ionization



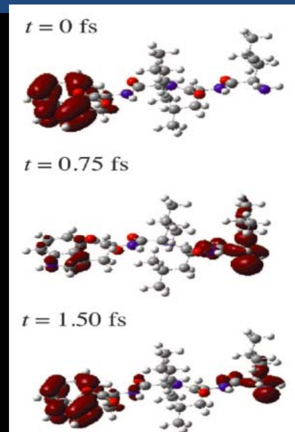
Zimmermann, Buller, Eilzer, Eichmann PRL **114**, 123003 (2015)

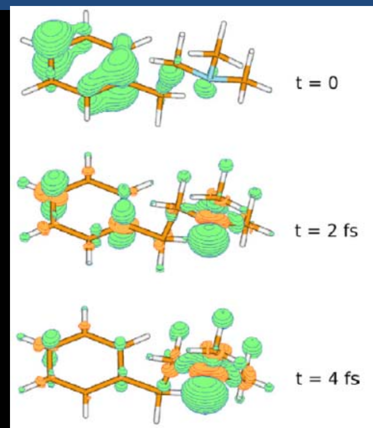
x-ray FEL multiple ionization



Rudek *et al.* Nat. Photonics **6**, 858 (2012)

molecular charge migration





Remacle, Levine PNAS 103, 6793 (2006)

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

General Physics: coupling of states

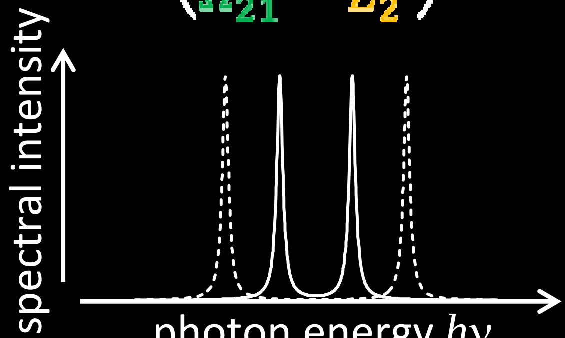
coupling of **one** to **one other** state	coupling of **one** to **multiple** states	coupling of **one** to a **continuum** of states

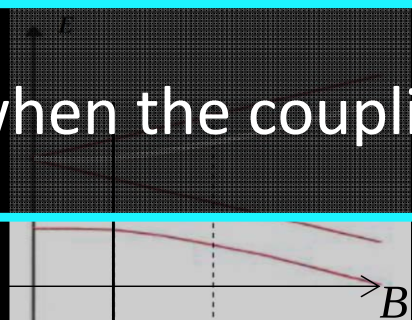
Quite well understood for cases of time-independent (or adiabatic) couplings

What happens when the coupling is **ultrashort** ?

Rabi oscillation in strong coupling

$$\begin{pmatrix} E_1 & \Omega_{12} \\ \Omega_{21} & E_2 \end{pmatrix}$$

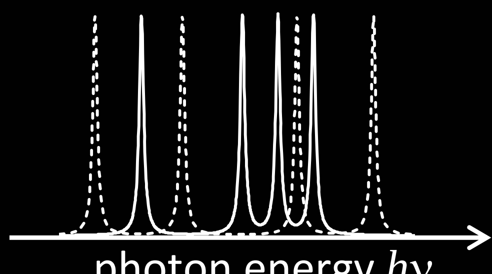


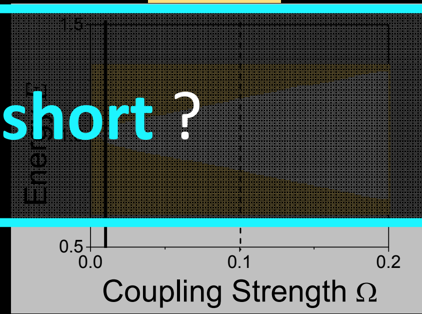


Breit-Rabi

e.g. Paschen-Back regime

$$\begin{pmatrix} E_1 & \Omega_{12} & \Omega_{13} & \Omega_{14} \\ \Omega_{21} & E_2 & \Omega_{23} & \Omega_{24} \\ \Omega_{31} & \Omega_{32} & E_3 & \Omega_{34} \\ \Omega_{41} & \Omega_{42} & \Omega_{43} & E_4 \end{pmatrix}$$





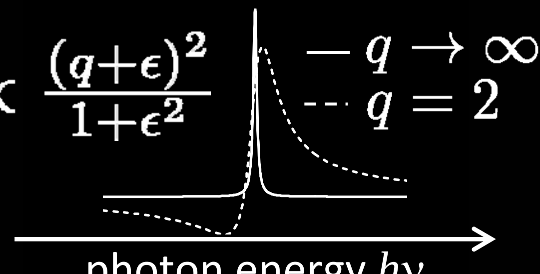
U. Fano (1935)

Phys. Rev. **124**, 1866 (1961)

Nuovo Cim. 12, 154 (1935)

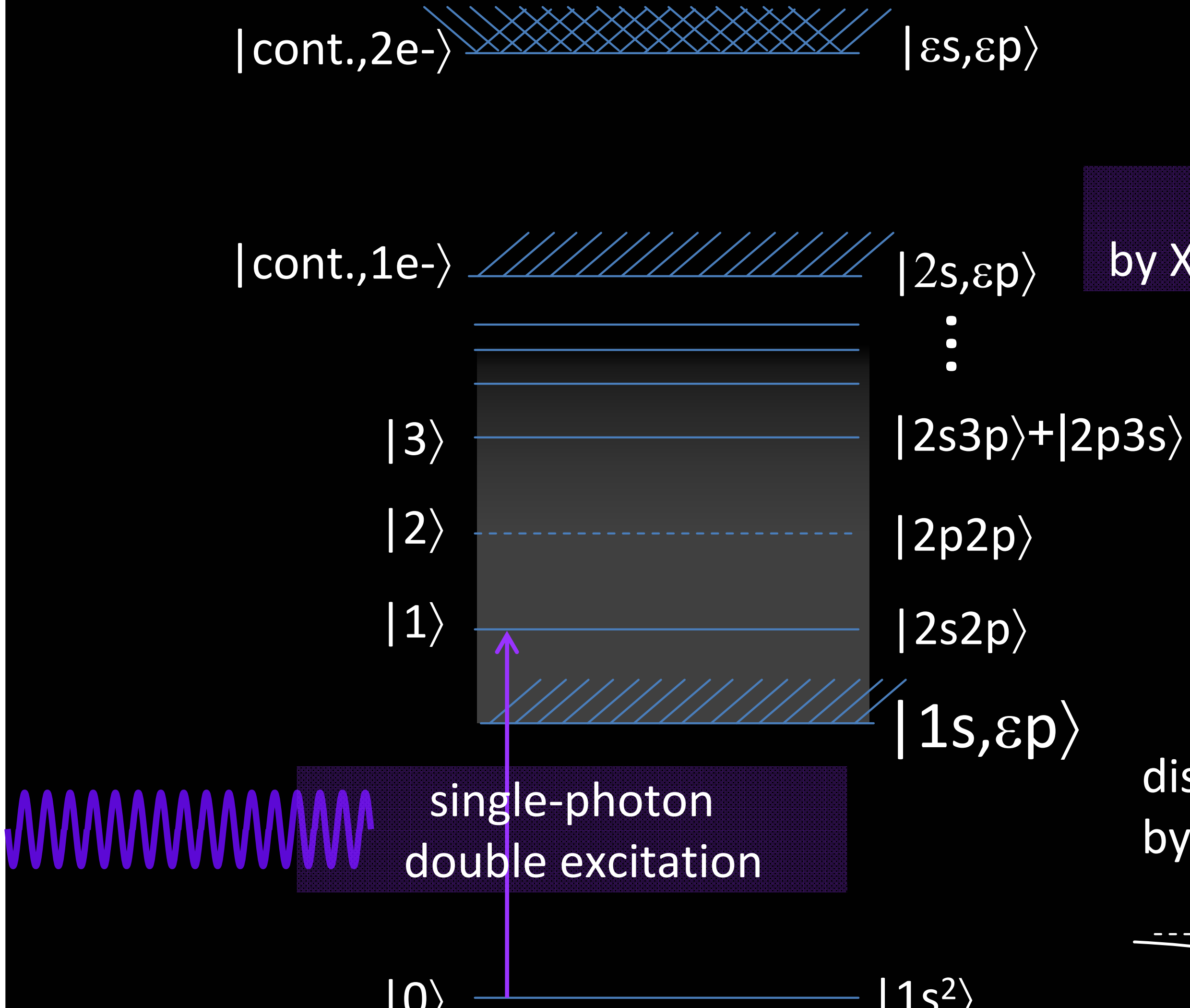
$$\begin{pmatrix} E & \Omega(\varepsilon) \\ \Omega(\varepsilon) & \varepsilon \end{pmatrix}$$

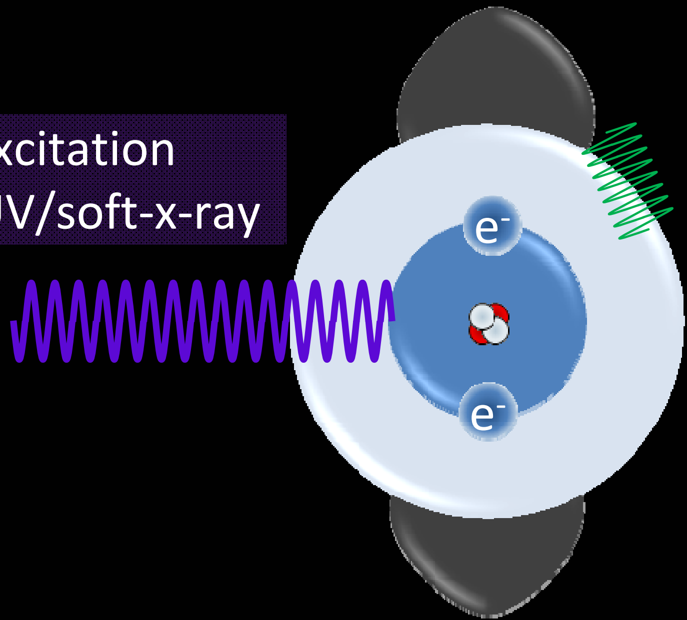
$$\sigma \propto \frac{(q+\epsilon)^2}{1+\epsilon^2}$$



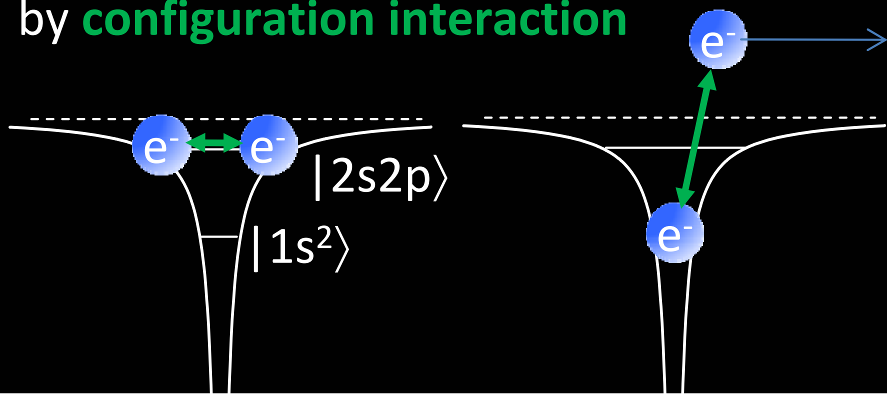
correlated bound-state dynamics:
Doubly excited helium

a prototype system
for electron correlation



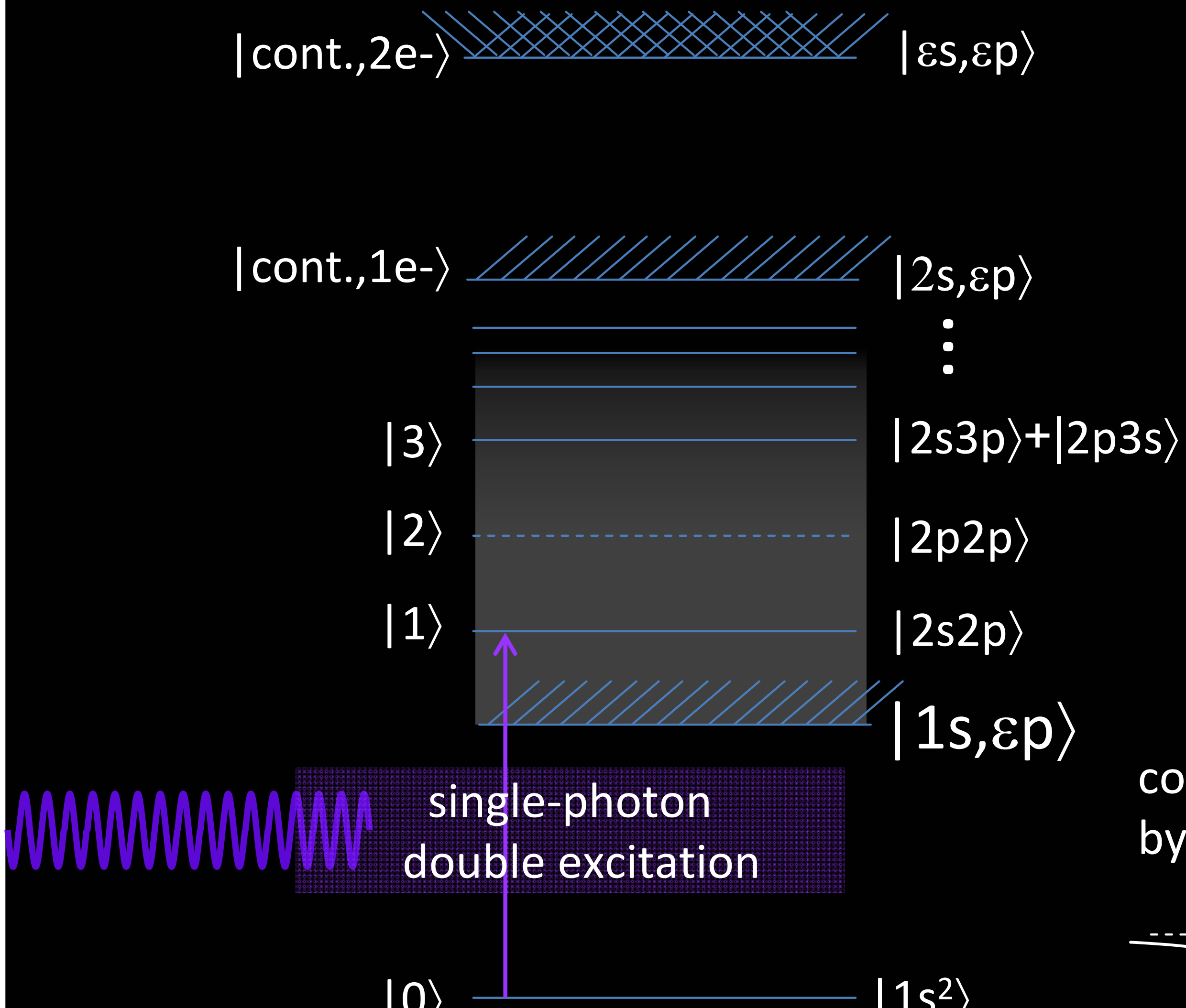


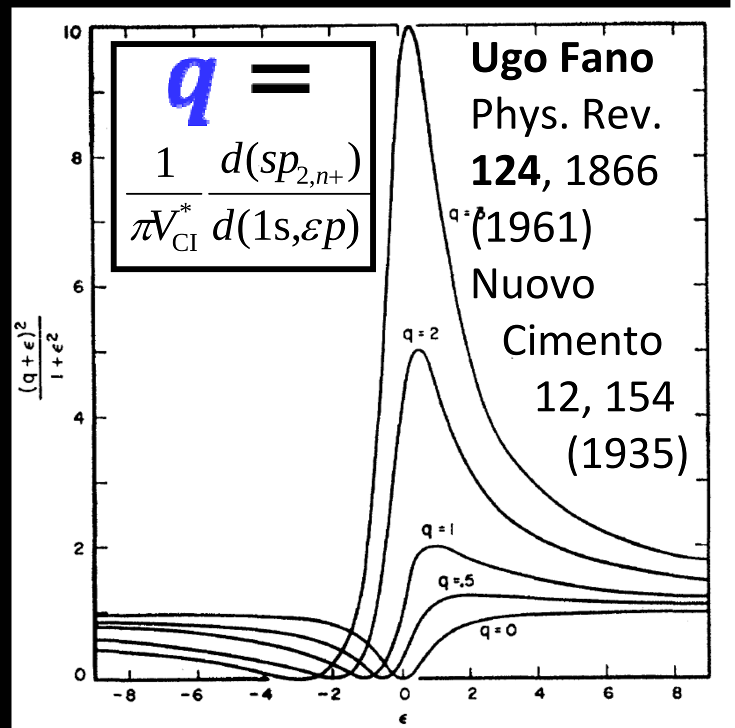
discrete states coupled to continuum
by **configuration interaction**



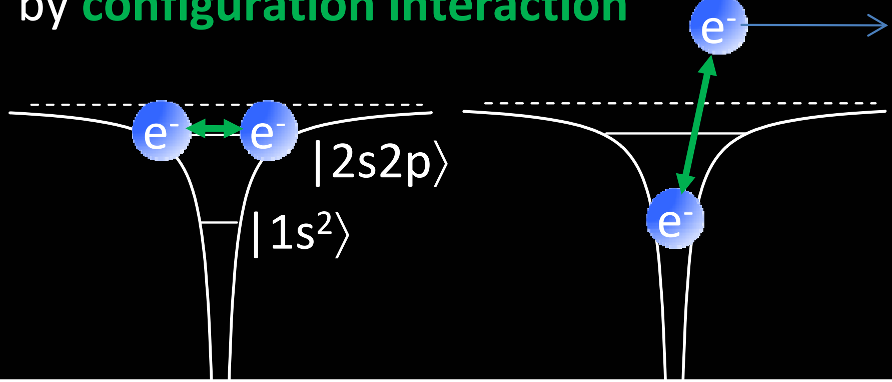
doubly excited helium: Fano resonance

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



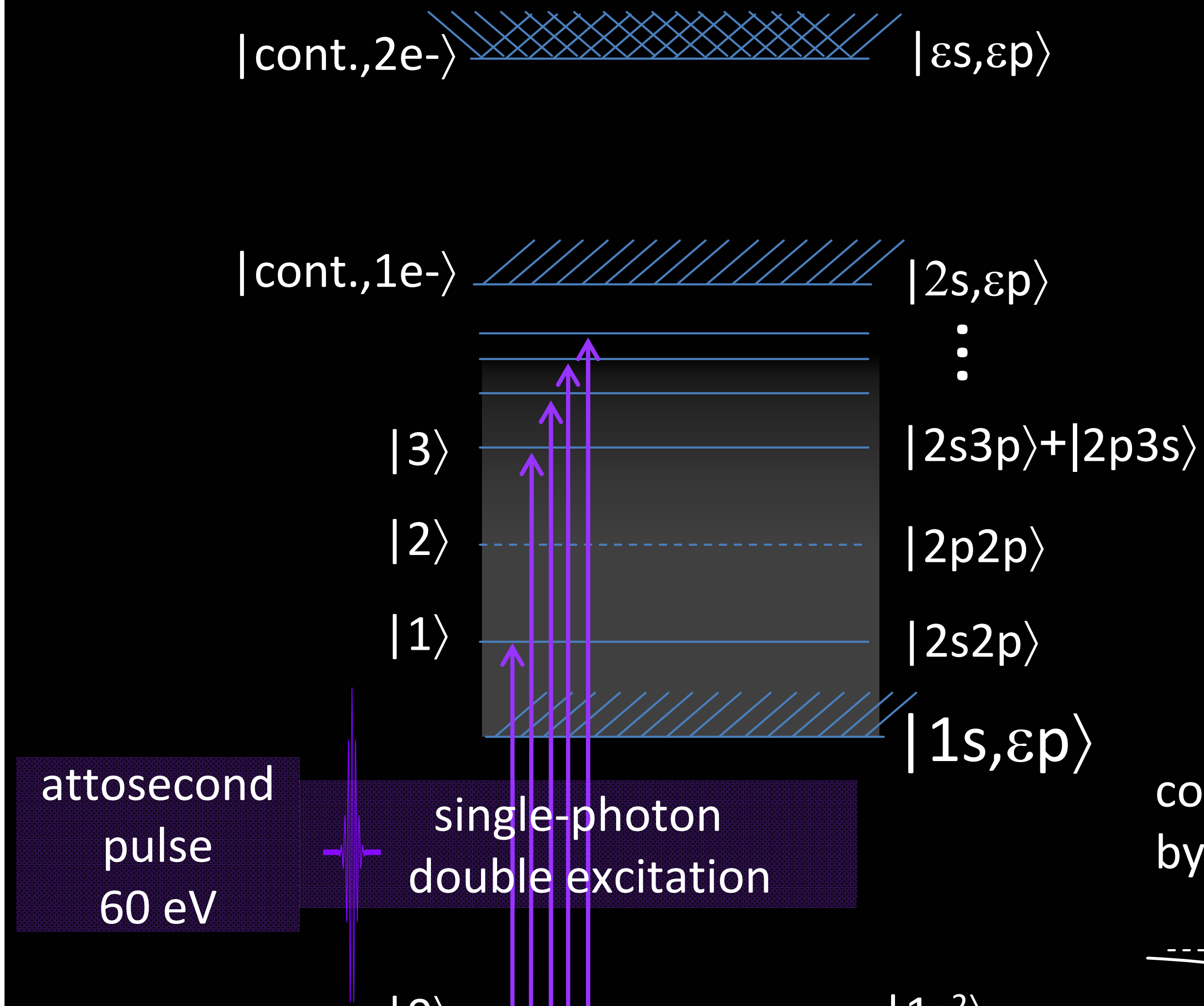


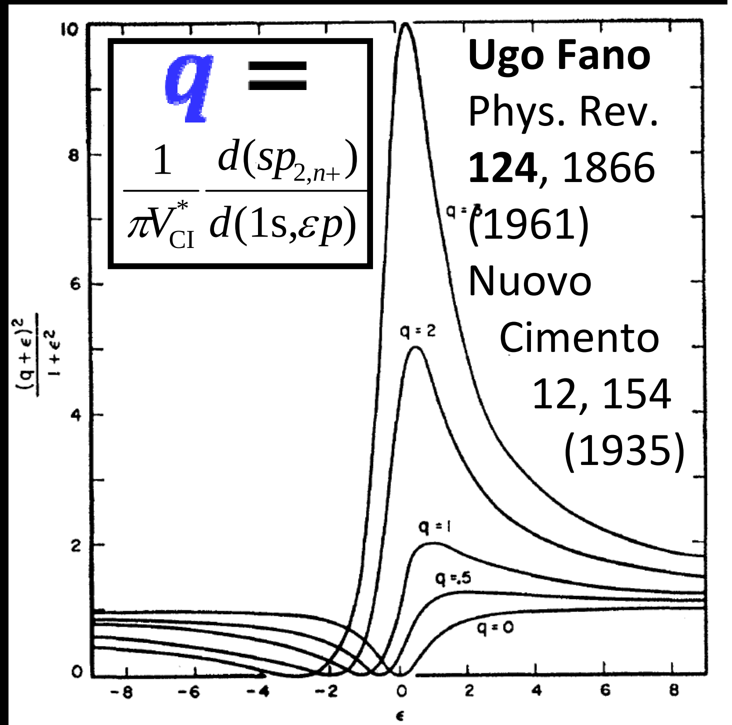
coupled
by **configuration interaction**



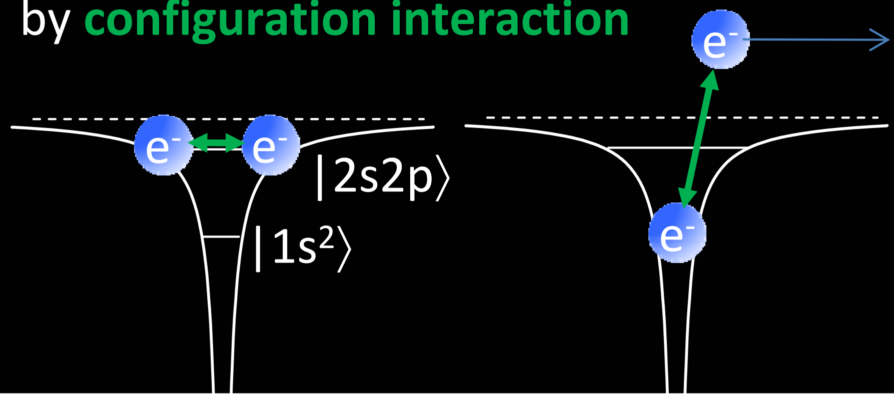
doubly excited helium: Fano resonance

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



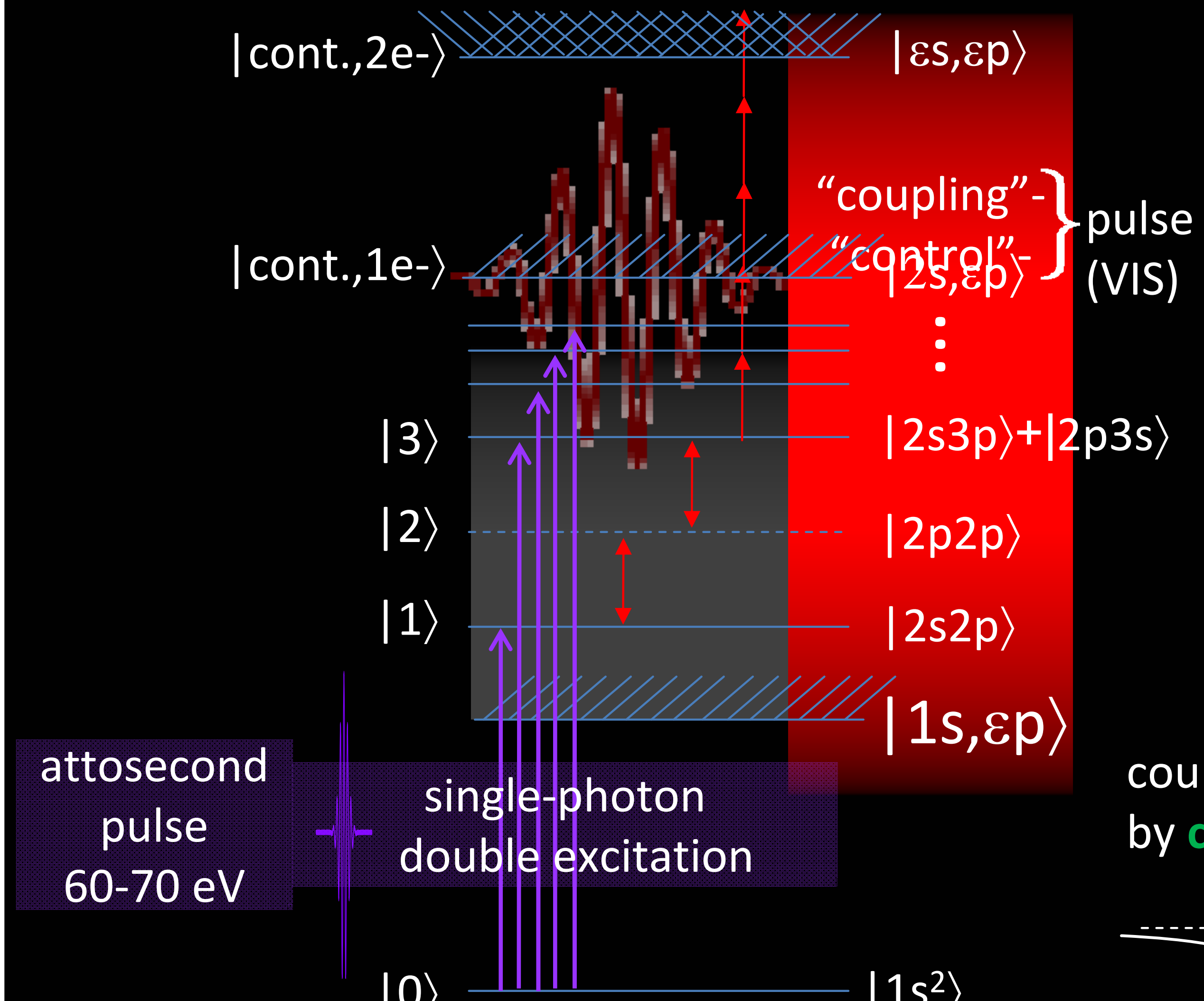


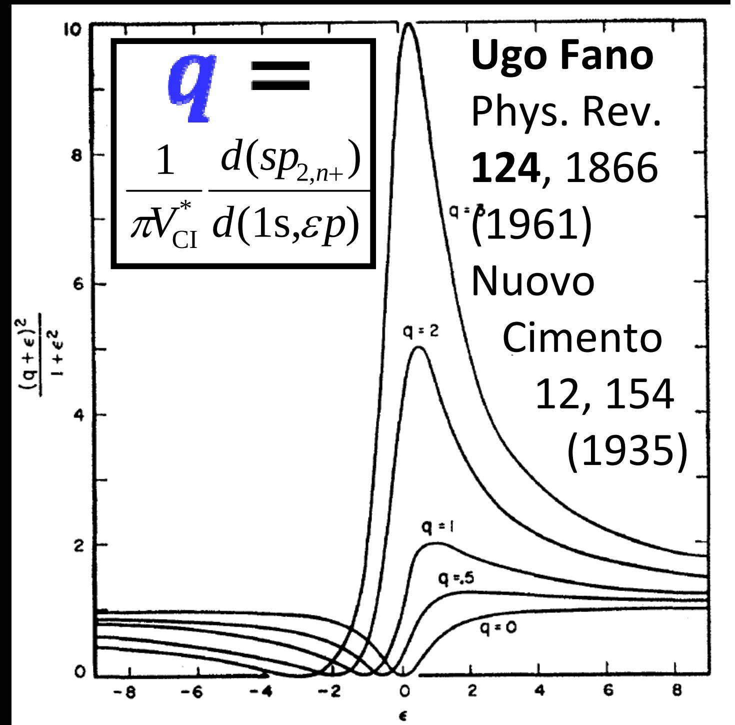
coupled by **configuration interaction**



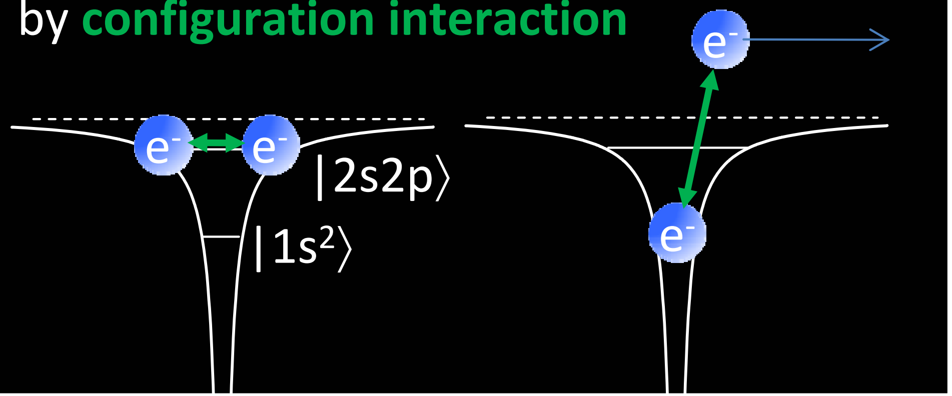
doubly-excited helium, in a strong laser field

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

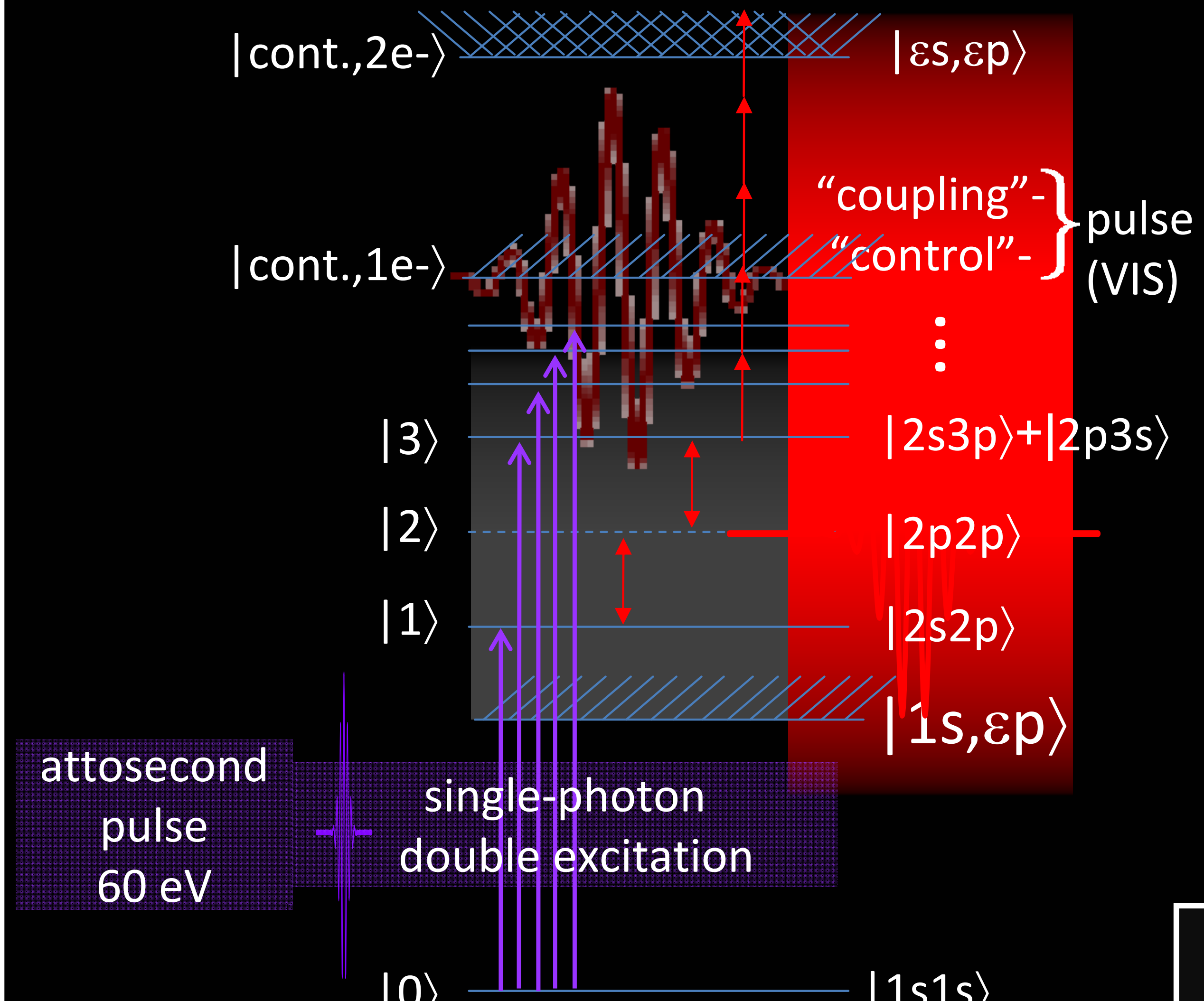




coupled
by **configuration interaction**



doubly-excited helium, coupled to a laser field



Previous work
(on laser coupling of doubly-excited helium):

Theory:
- **Madsen, Themelis, Lambropoulos**
- **Zhao, Chu, Lin et al.**

...

Experiment:
- **Loh, Greene, Leone, et al.**
- **Gilbertson, Chang et al.**

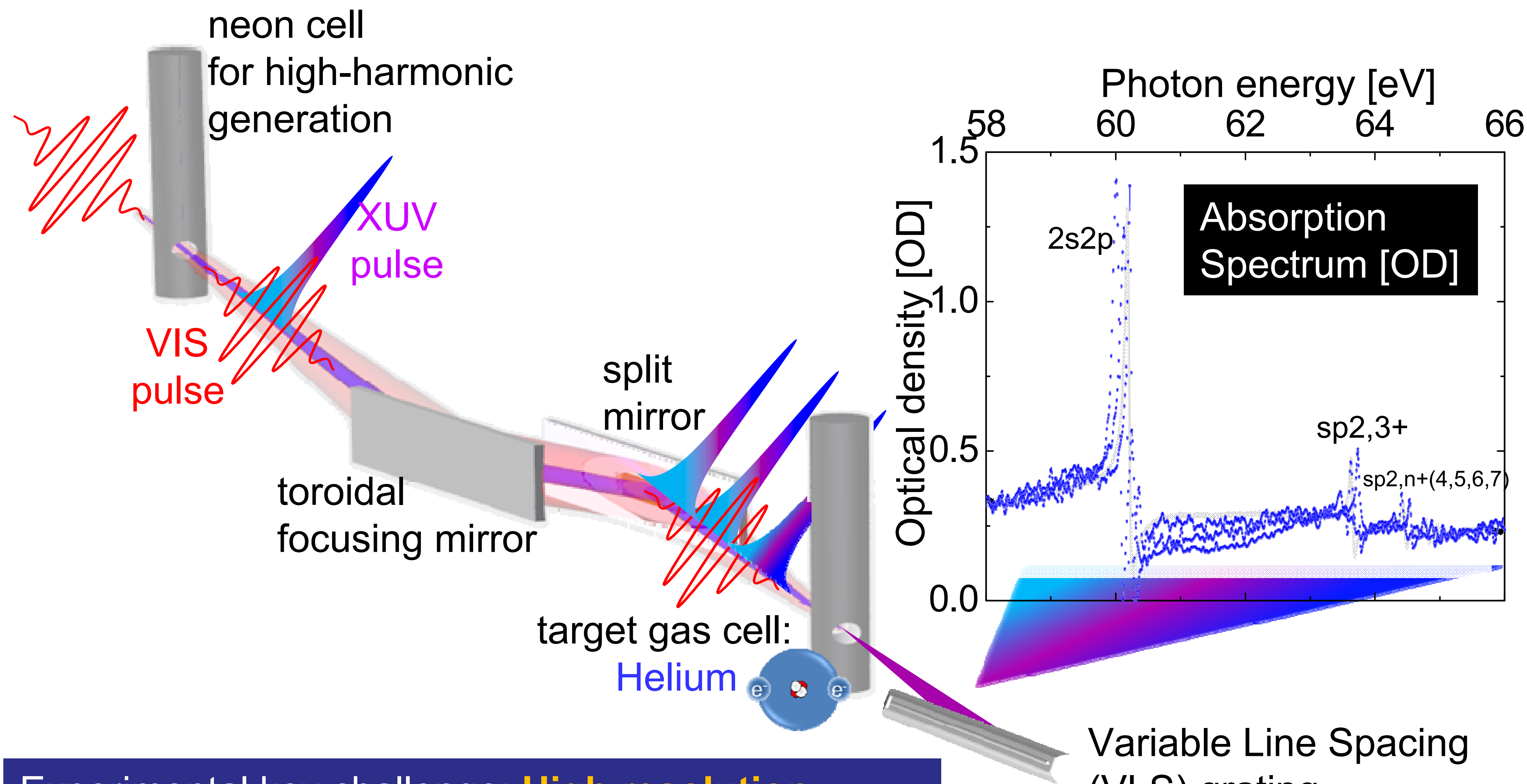
...

Experimental challenge:
- **high (asec) temporal and**
- **high (meV) spectral resolution**

required at the same time

Experimental setup

for time-resolved XUV absorption spectroscopy

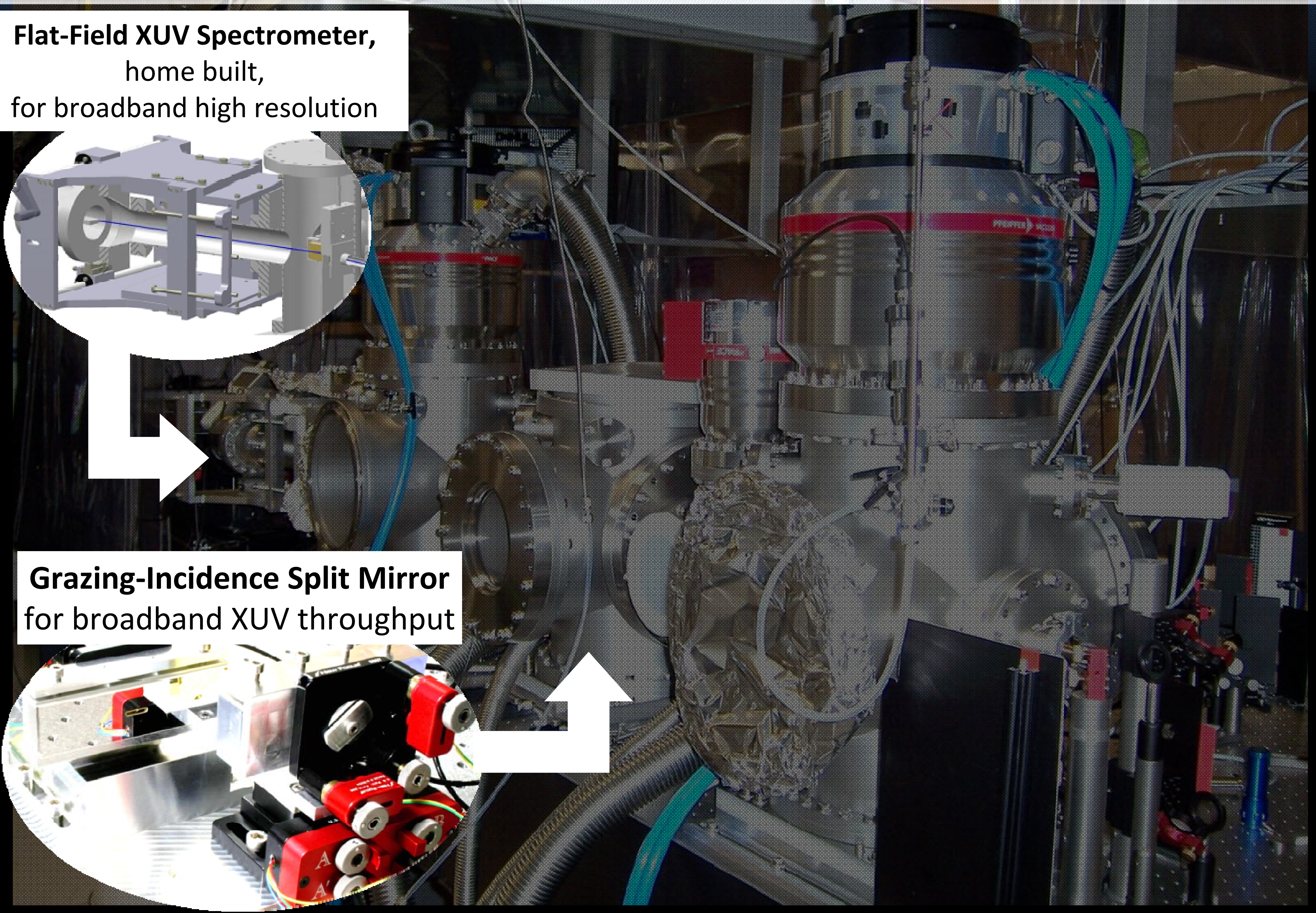


Experimental key challenge: **High resolution**

In photon energy ... : **ΔE = 20 meV (@60 eV)**

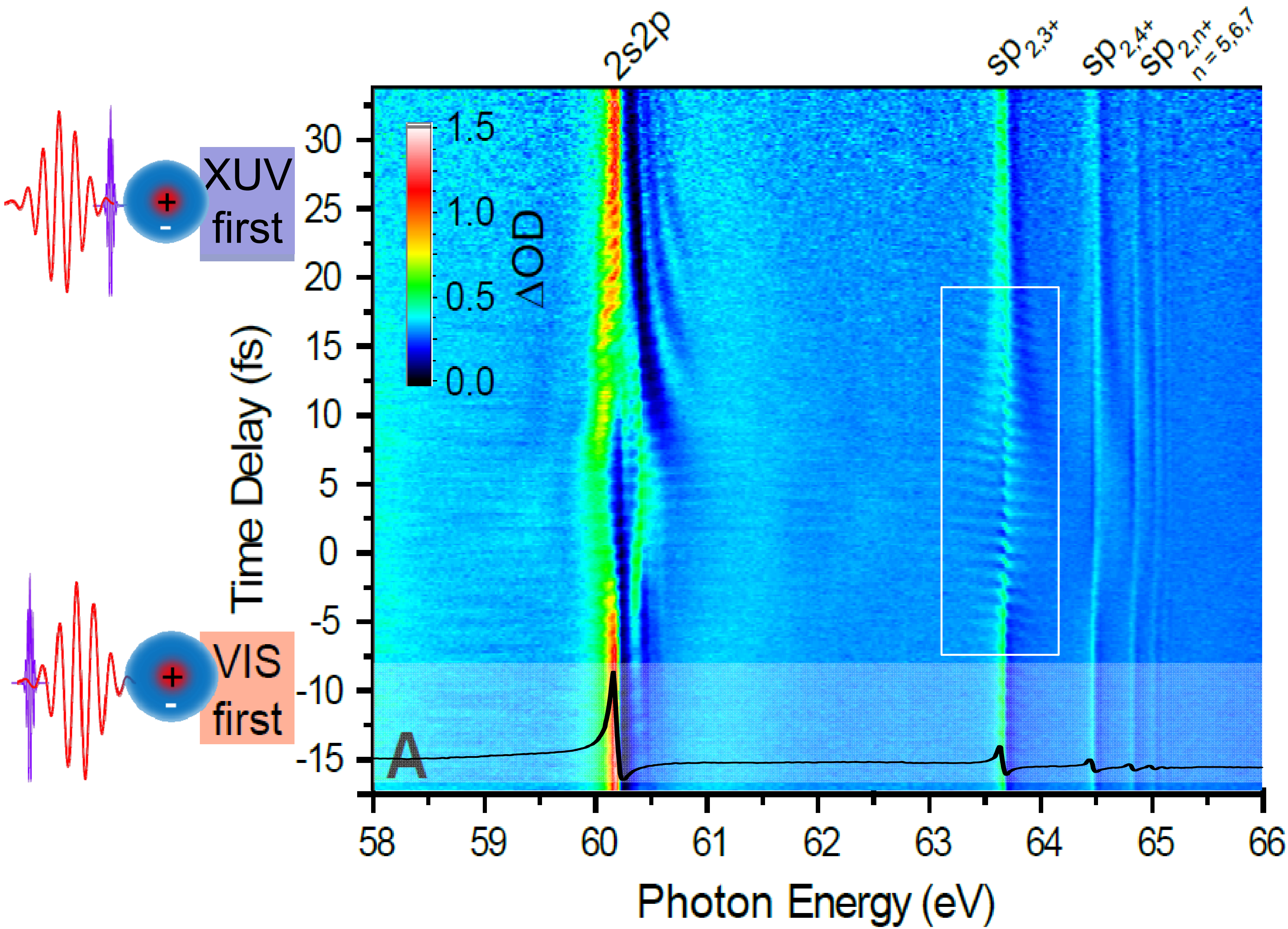
... and time delay : **Δτ = 10 as**

Experimental Setup in the Lab

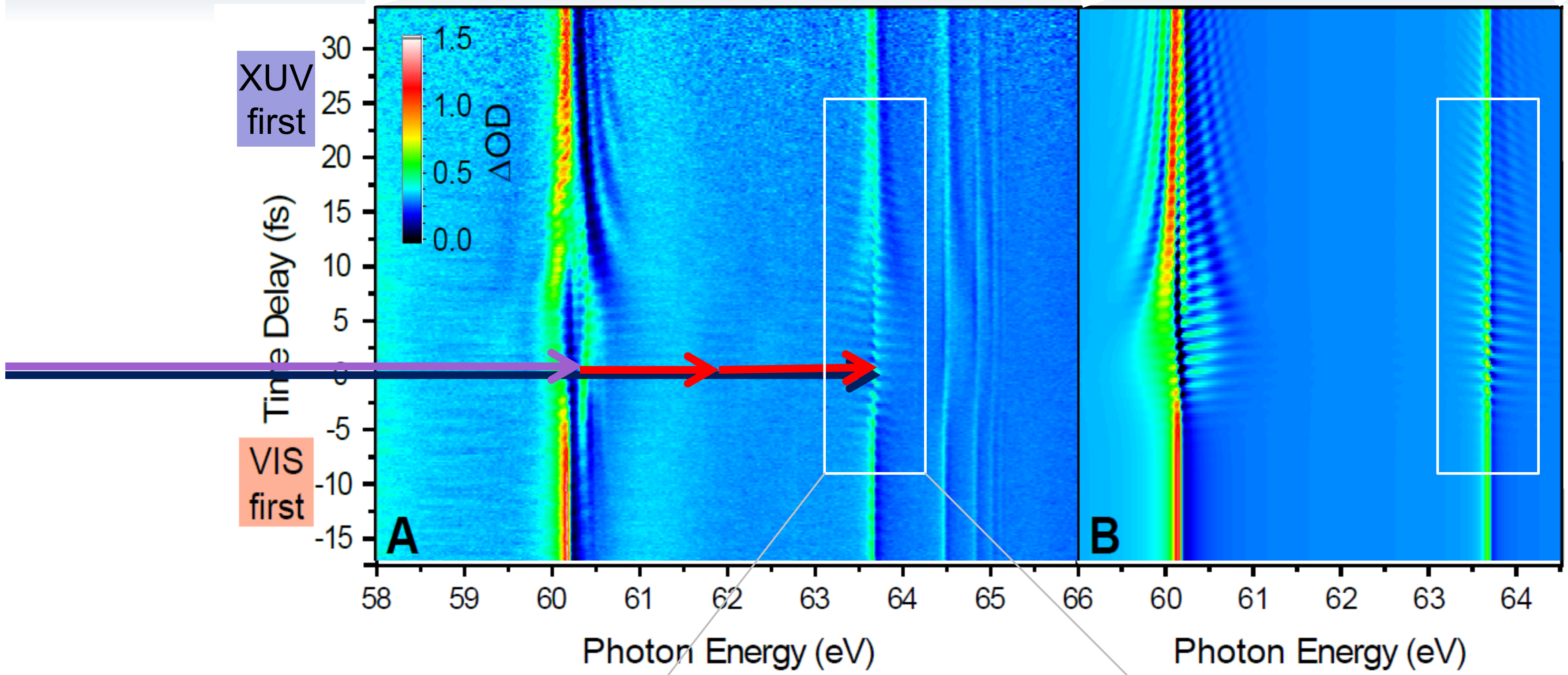


Time-resolved doubly-excited $2e^-$ dynamics in He

Experimental data



comparison experiment and theory

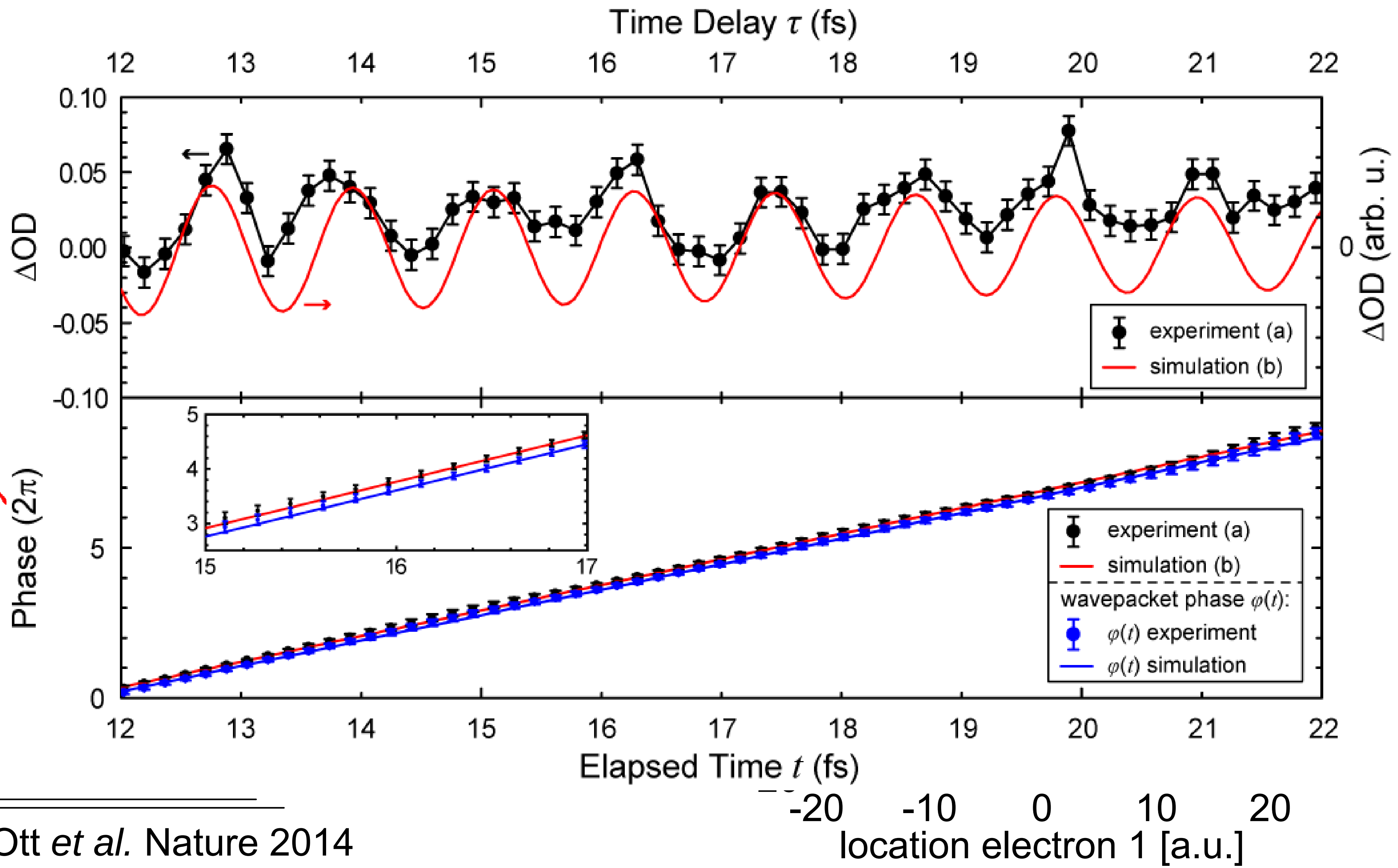


quantum-state interference
$\Rightarrow$ phase $\varphi(t)$

Probing a time-dependent superposition state:
$\Rightarrow$ Wavepacket

Measuring the time-dependent phase difference of 2s2p and sp_{23+} autoionization states

cooperation with Javier Madroñero (Theory, TU München)
Luca Argenti, Fernando Martín (Theory, UAM Madrid)

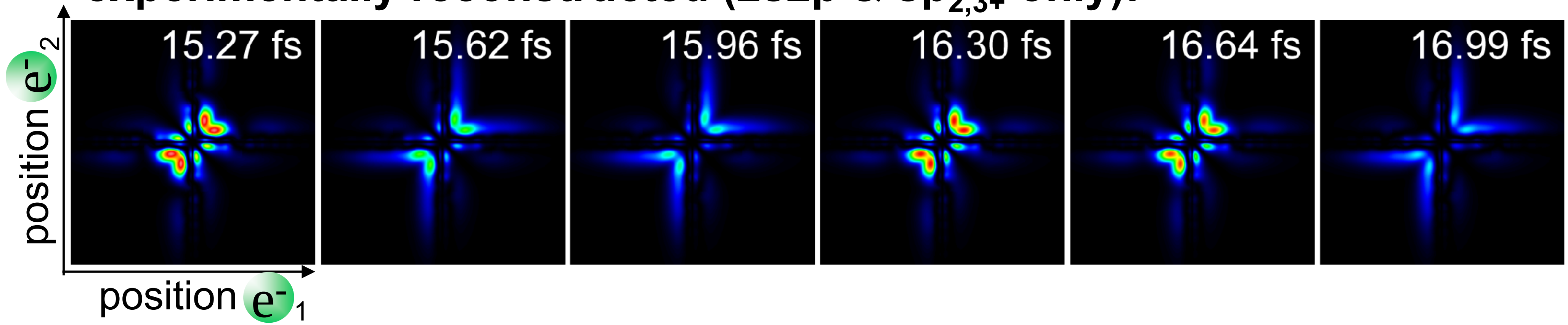


C. Ott *et al.* Nature 2014

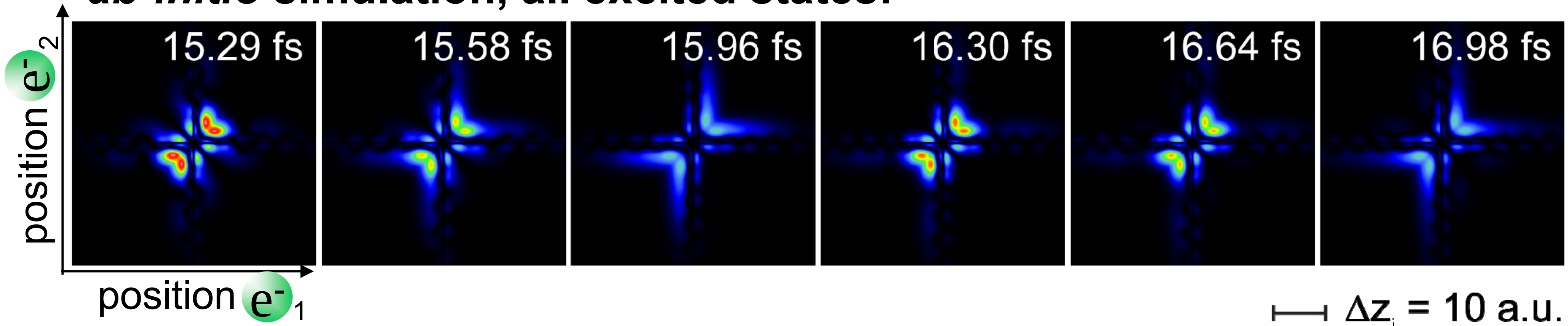
Testing *ab-initio* theory of e^- correlation dynamics

cooperation: Luca Argenti & Fernando Martín (UAM Madrid, Spain)
Javier Madroñero (TU Munich)

experimentally reconstructed (2s2p & $sp_{2,3+}$ only):



ab-initio **simulation, all excited states:**

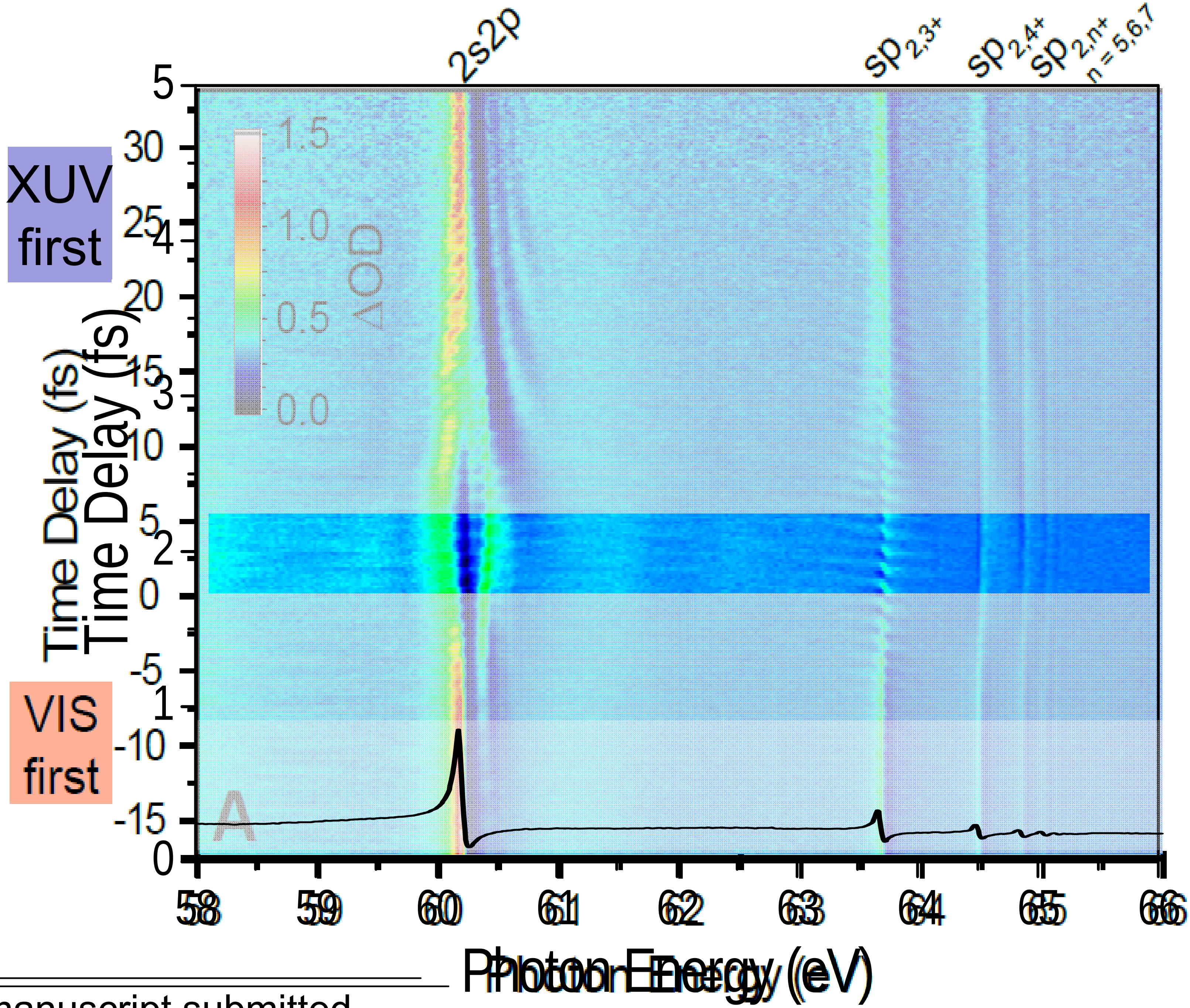


Δz_i = 10 a.u.

first experimental observation
of **two-electron wavepacket** motion

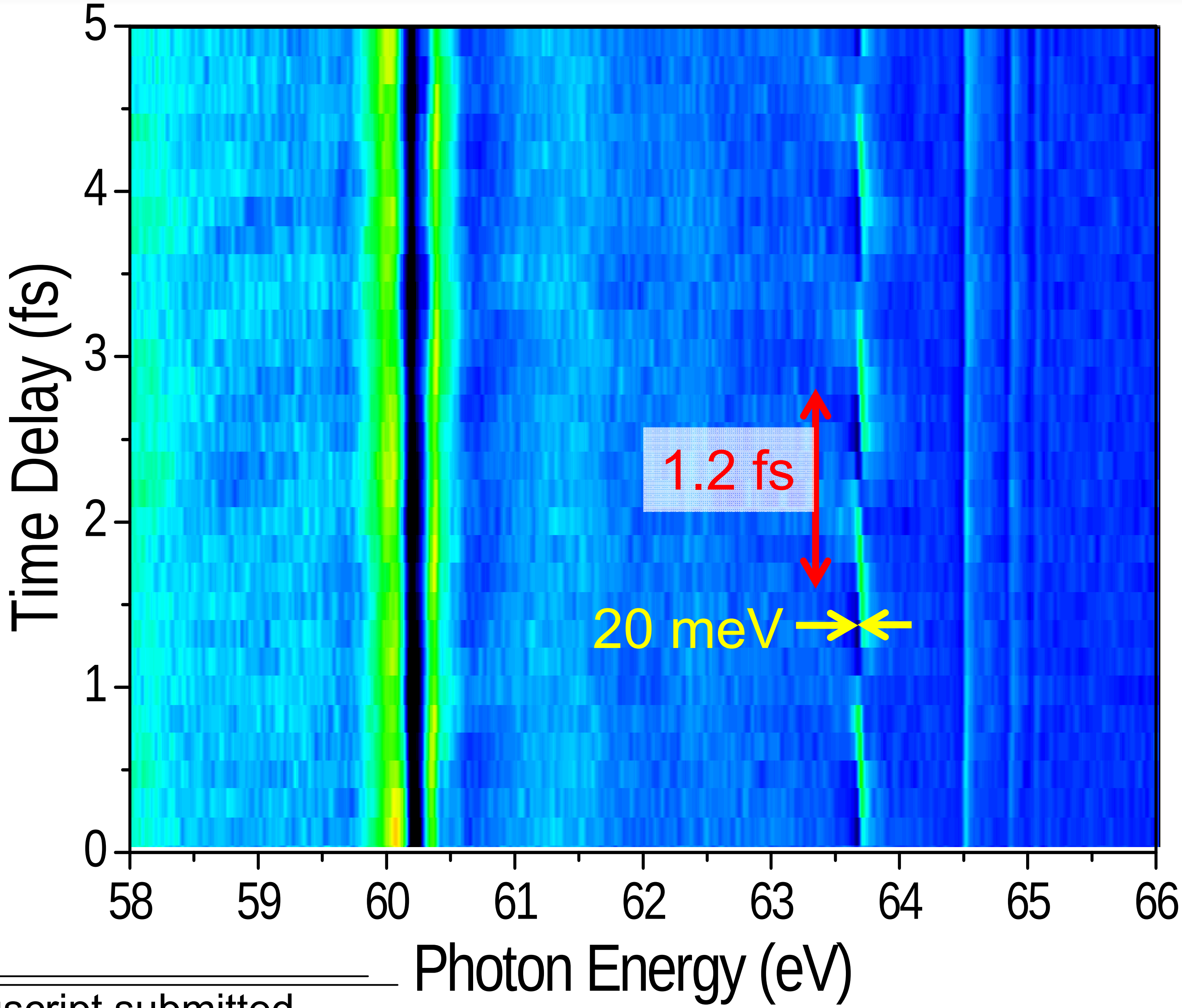
C. Ott *et al.* Nature 2014

Time-resolved doubly-excited 2e⁻ dynamics in He



manuscript submitted

Time-resolved doubly-excited 2e⁻ dynamics in He

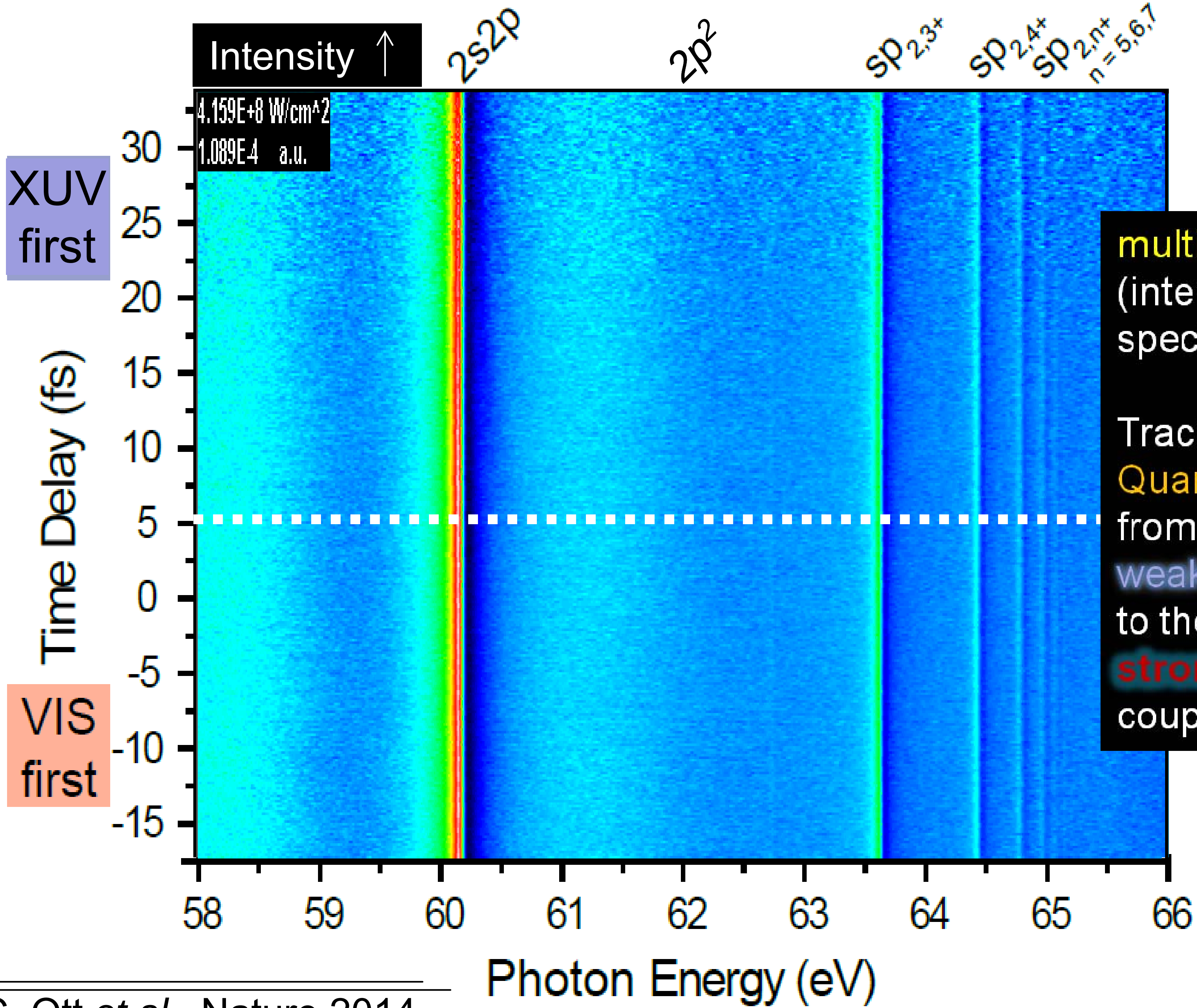


high temporal
and
high spectral
resolution
are
required
simultaneously

manuscript submitted

Intensity dependence of 2-e⁻ quantum dynamics

Experimental data



multi-dimensional
(intensity, time, energy)
spectroscopy

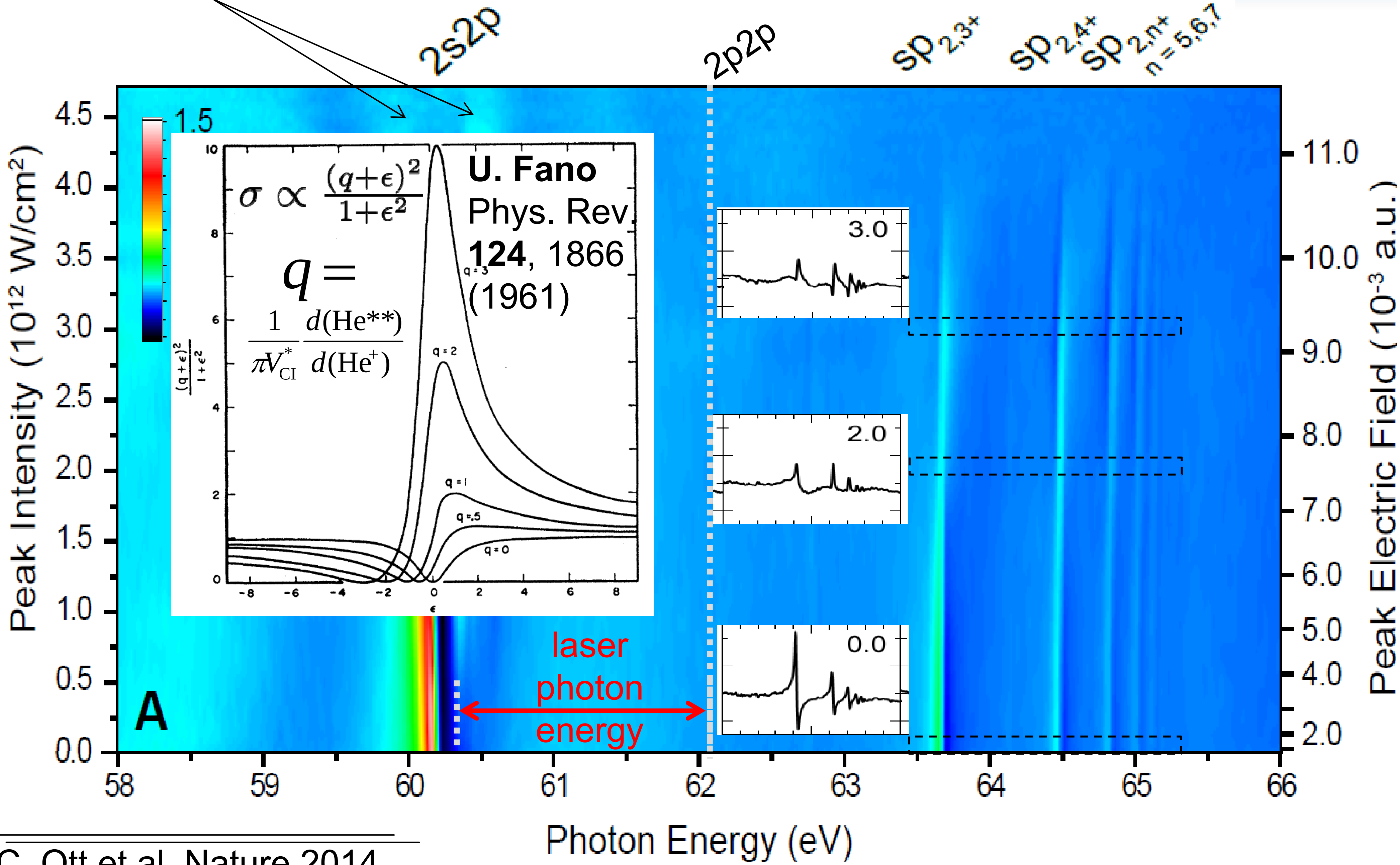
Tracking
Quantum Dynamics
from the
weak perturbative
to the
strong
coupling regime

C. Ott *et al.*, Nature 2014

Intensity, a key parameter (the coupling strength)

Rabi cycling of autoionizing states)
Autler-Townes doublet

tuning from weak perturbative to strong fields

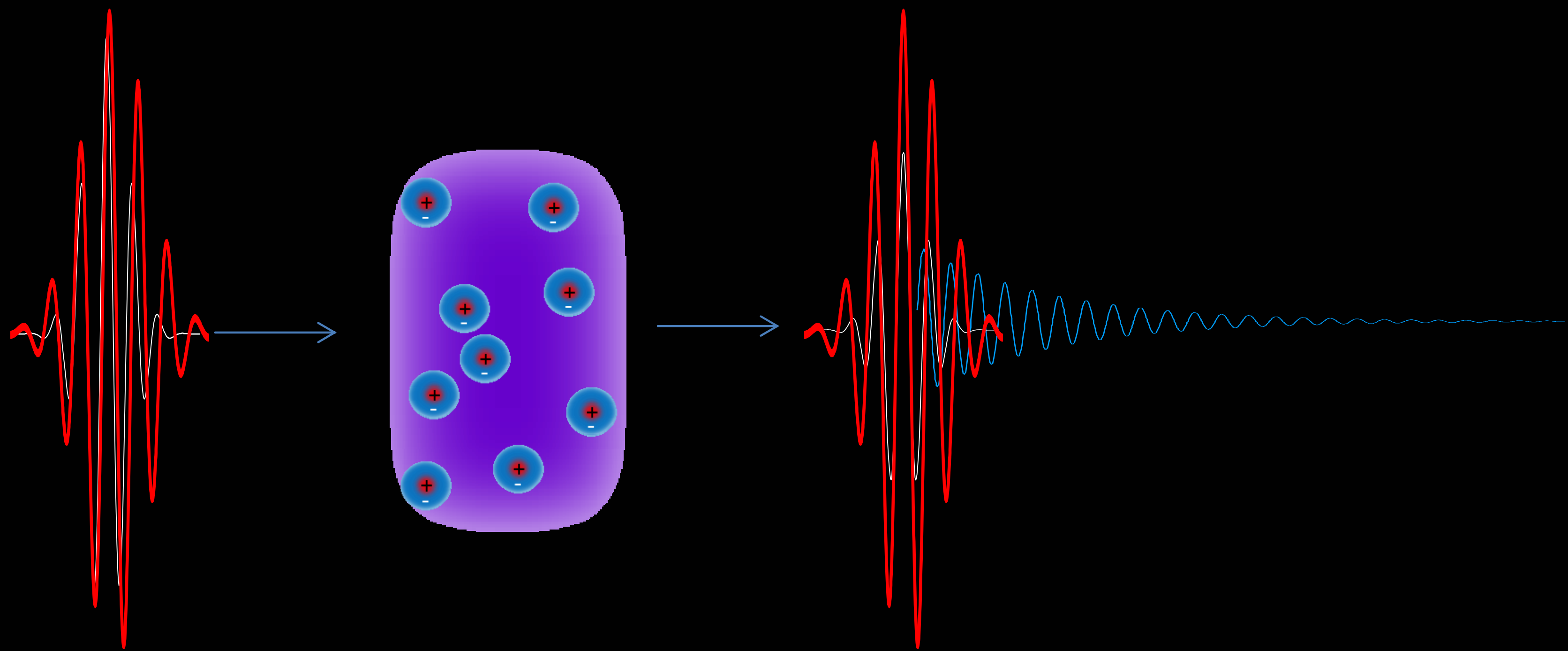


C. Ott et al. Nature 2014

asking two electrons in He
some quick questions ...

What is absorption?

And how does it respond to intense pulsed light?

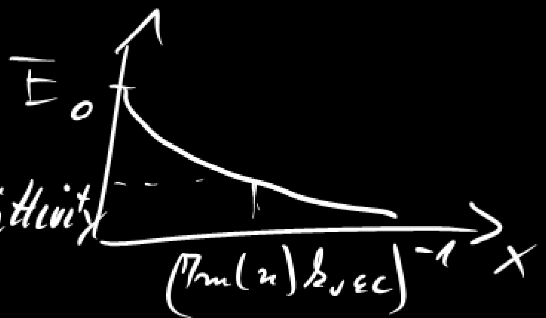


The refractive index n

$$E(x,t) = E_0 \cdot e^{i(k_{med}x - \omega t)} \qquad k_{med} = \frac{\omega}{c_{med}} = n \cdot \frac{\omega}{c_{vac.}} = n k_{vac}$$

$$\hookrightarrow = E_0 \cdot e^{i(n k_{vac} x - \omega t)} \qquad n = Re(n) + i\, Im(n)$$

$$\hookrightarrow = E_0 \cdot e^{i(Re(n) k_{vac} x - \omega t)} \cdot e^{-Im(n) k_{vac} x}$$



macr. polarization

$$P = \chi \cdot E$$

χ: susceptibility (linear)

rel. permittivity: $\chi = \varepsilon_r - 1$

$$\left[D = \varepsilon_0 \varepsilon_r E = P + \varepsilon_0 E \right]$$

$$P = \rho d$$

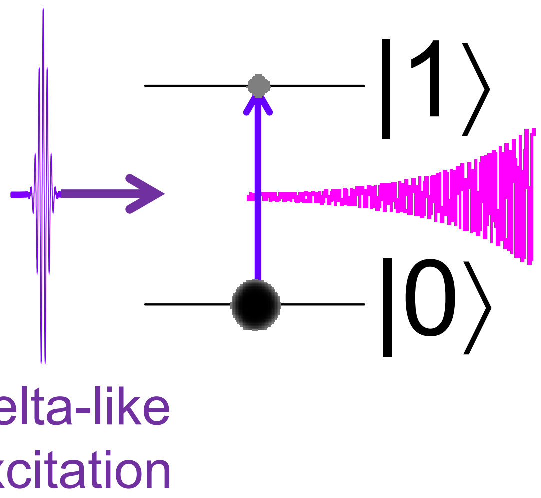
d: dipole moment

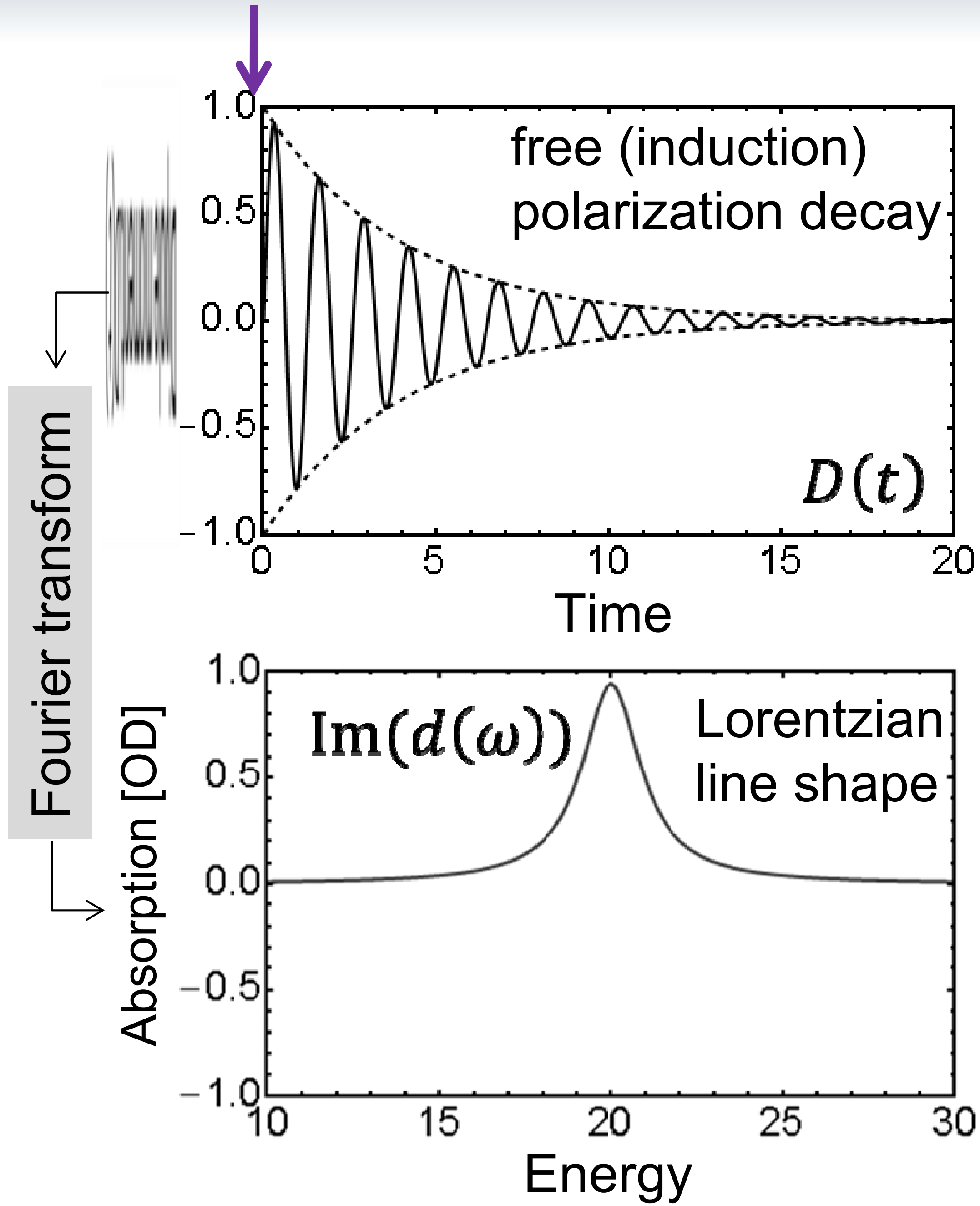
ρ: density of dipoles (e.g. atoms)

$$n^2 = \varepsilon_r = 1 + \chi$$

$$n = \sqrt{1+\chi} \sim 1 + \frac{\chi}{2} = 1 + \frac{\rho d}{E} \Rightarrow \Delta n(\omega) = \rho \frac{d(\omega)}{E(\omega)}$$

Optical response and absorption

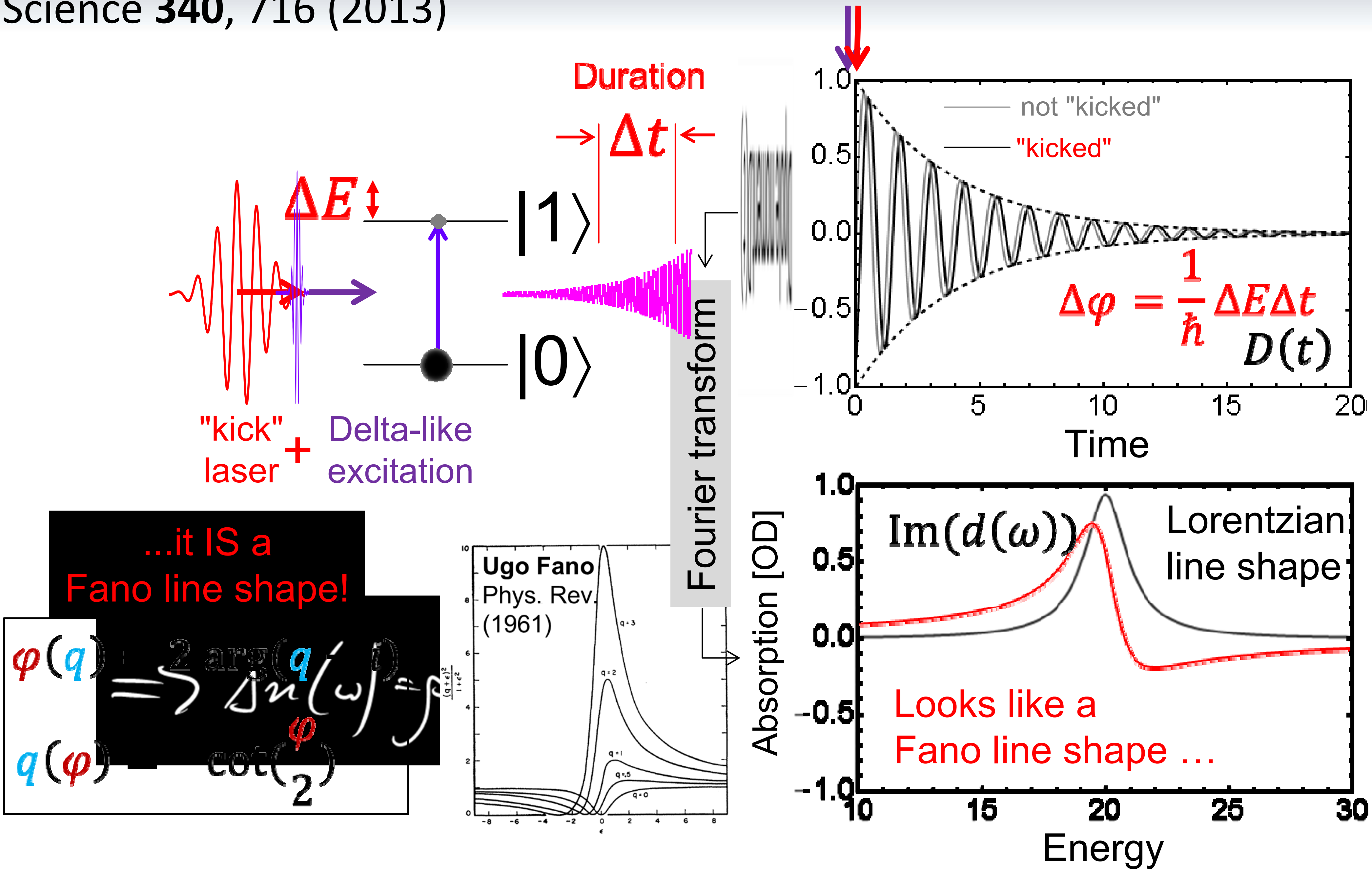




$$\Rightarrow \Delta n(\omega) \propto \frac{d(\omega)}{E(\omega)}$$

Resonance absorption in the Time Domain

Science **340**, 716 (2013)



The Fano dipole phase

Exact mapping from Fano q parameter to temporal phase shift φ

cooperation with C. Greene (Purdue), J. Evers and C. Keitel (MPIK)

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

$$=$$

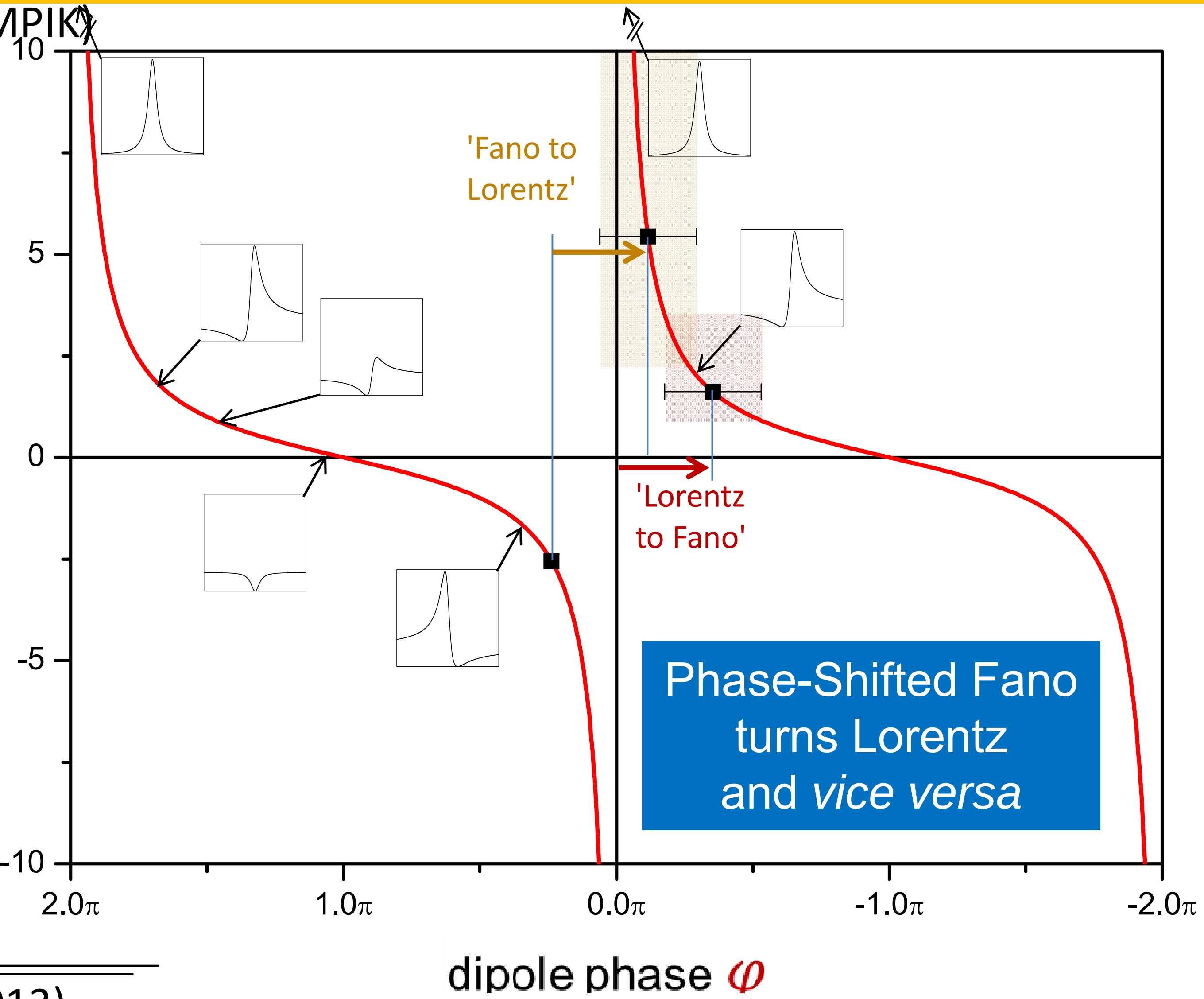
$$\mathrm{Im}\left(\frac{-1}{i+\varepsilon}\exp(i\varphi)\right) + const.$$

Phase-shifted Lorentzian

...it IS a Fano line shape!

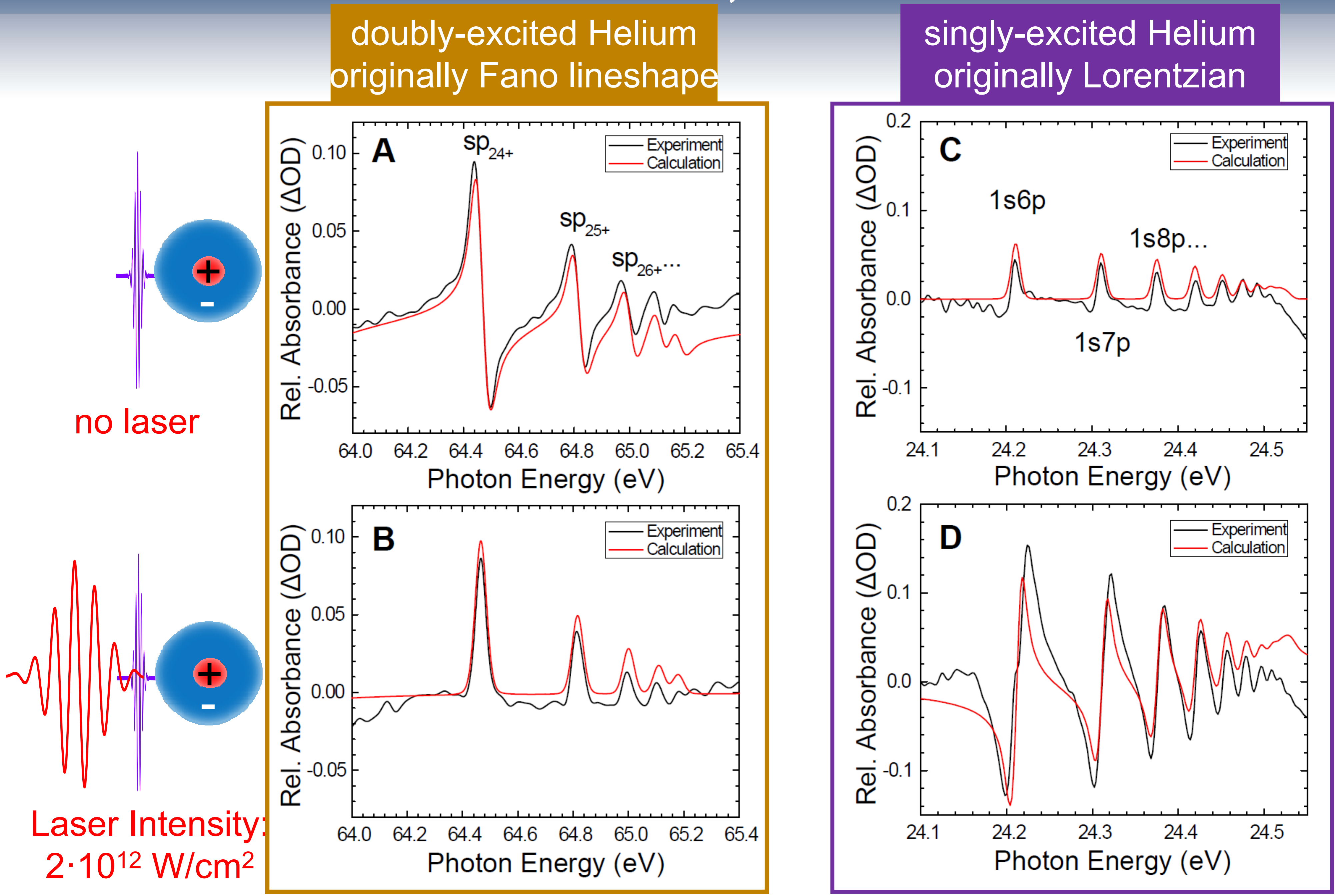
$$\varphi(q) = 2\arg(q - i)$$

$$q(\varphi) = -\cot\left(\frac{\varphi}{2}\right)$$



C. Ott *et al.* Science **340**, 716 (2013)

Fano to Lorentz, and Lorentz to Fano

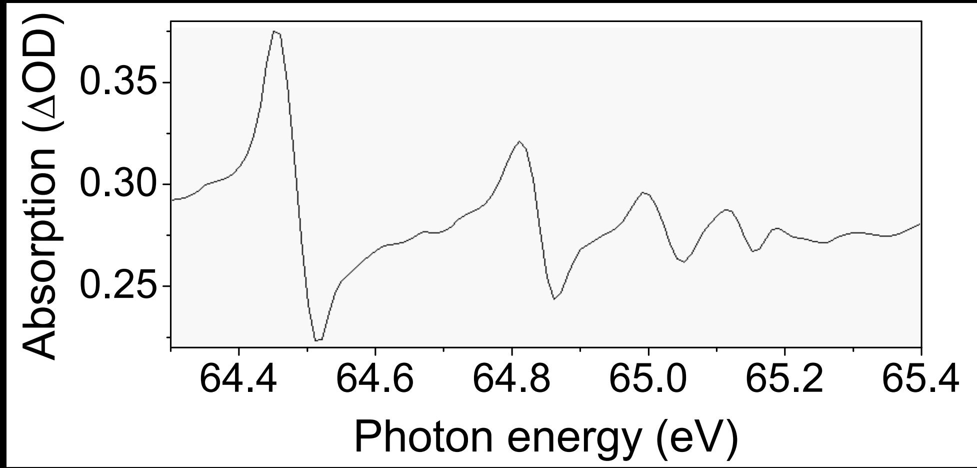


Science **340**, 716 (2013)

Changing the (spectral) shape of atoms

Helium **doubly** excited (**above** the continuum threshold)

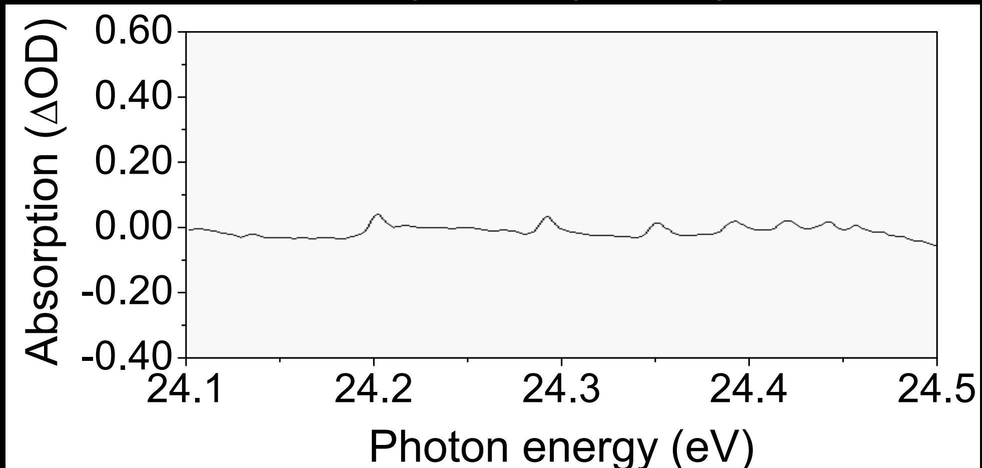
'2s4p' '2s5p' '2s6p' ...



turning original 'Fano' into 'Lorentz'

Helium **singly** excited (**below** the continuum threshold)

1s6p 1s7p 1s8p ...



turning original 'Lorentz' into 'Fano'

$$q = -\cot\left(\frac{\varphi}{2}\right)$$

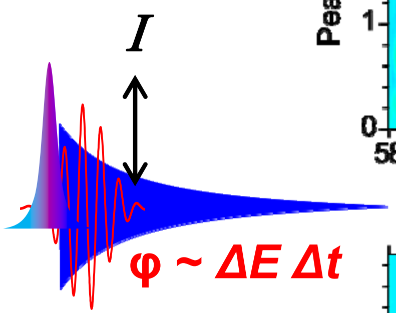
$q = -2.55 \Rightarrow \varphi = 0.24\,\pi$

Science **340**, 716 (2013)

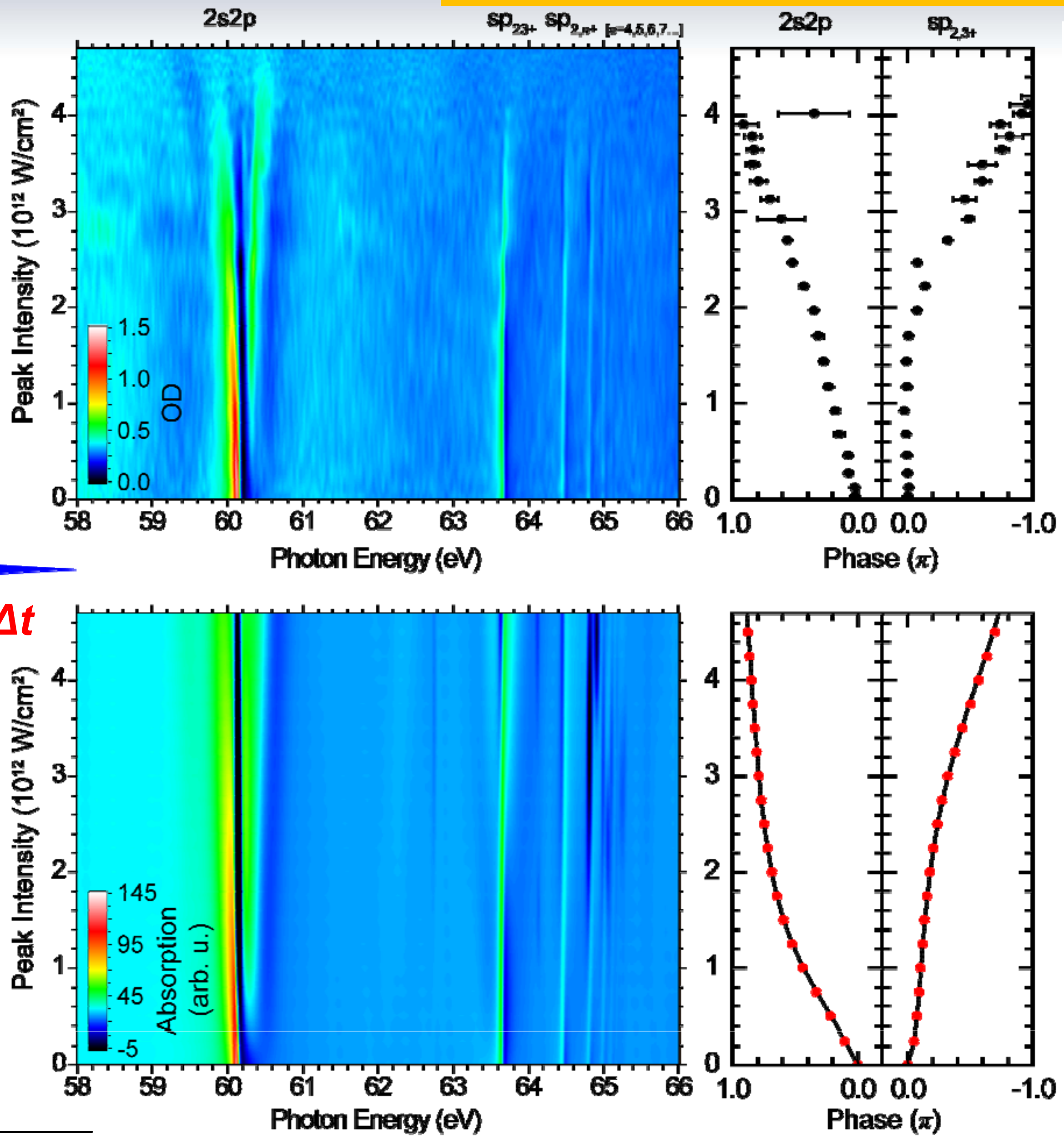
Extracting the laser-induced phase shift

Cooperation with J. Madronero, L. Argenti, F. Martín

experiment

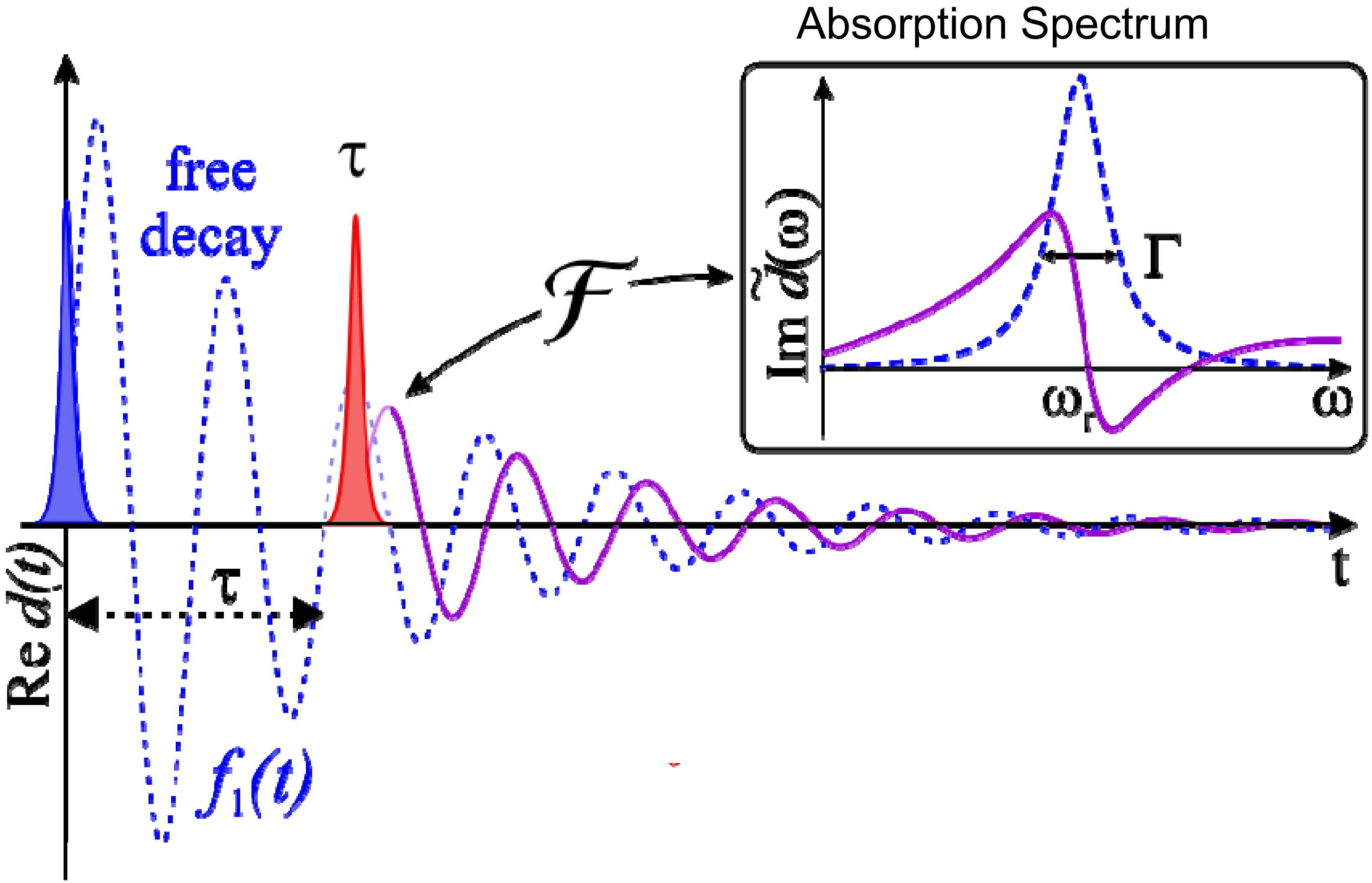


***ab-initio* theory**
(Argenti/Martin)



C. Ott *et al.* Nature 2014

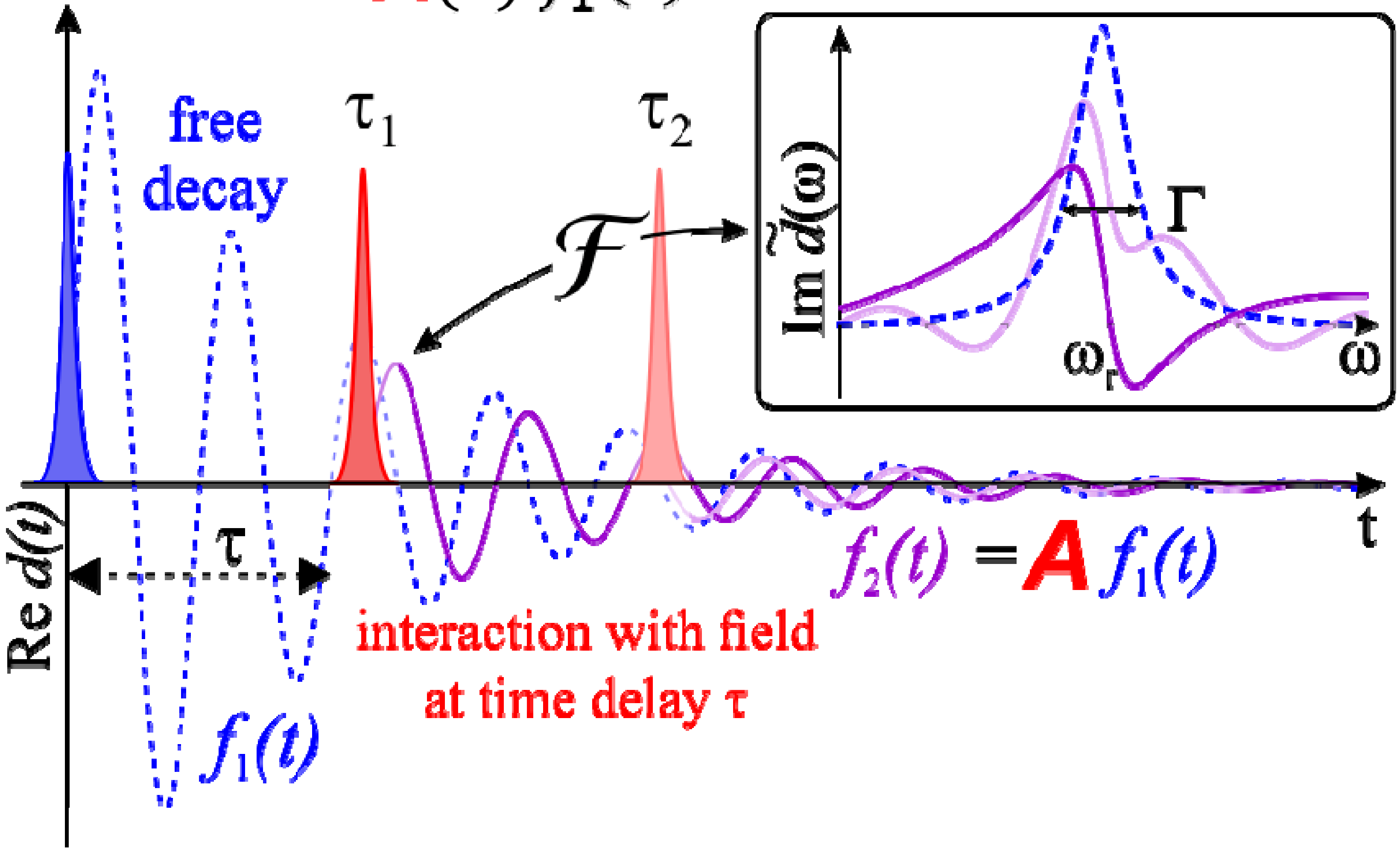
The Fano Phase Shift



The dipole-control model

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

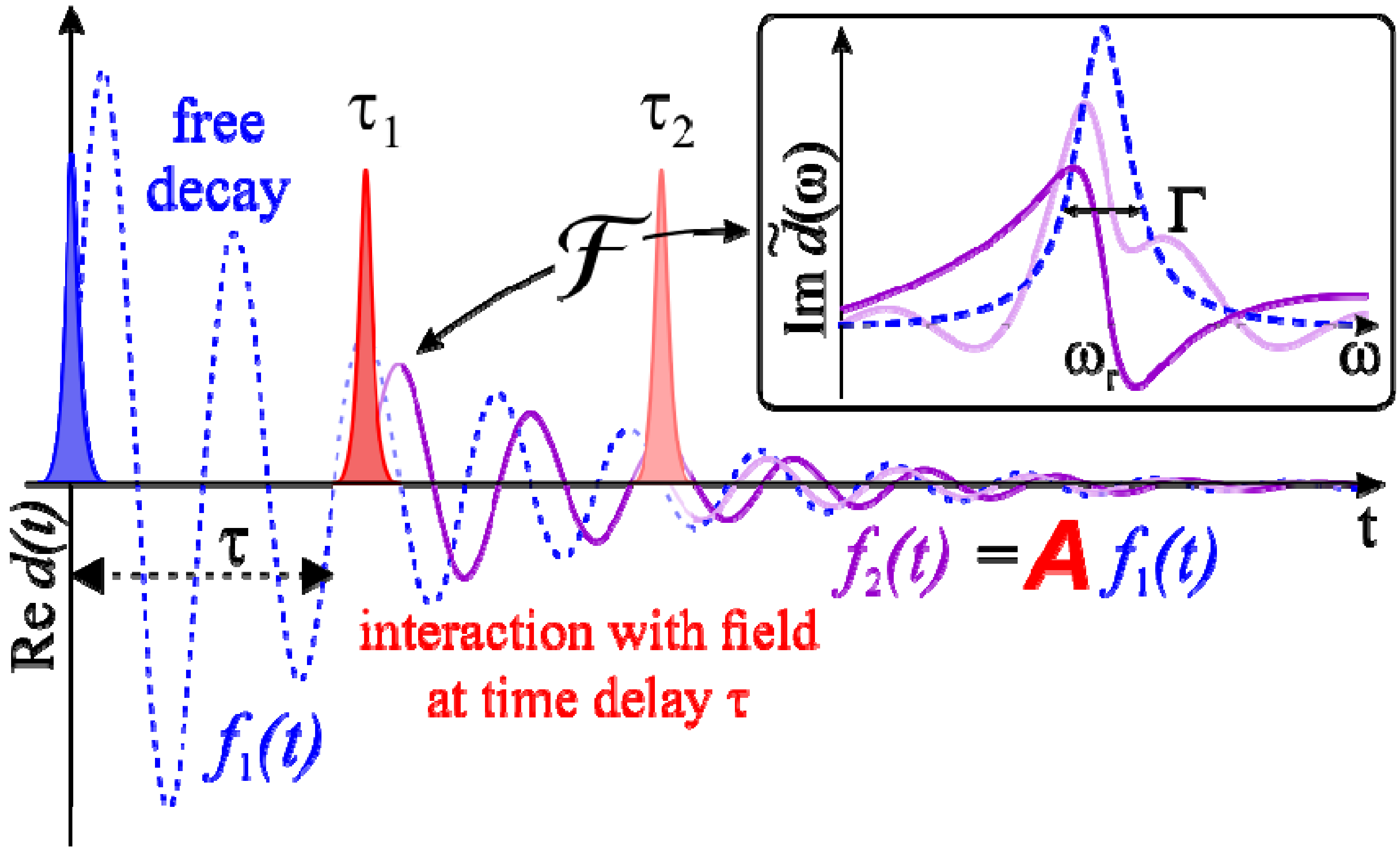
$$d_\tau(t,\tau) \propto \begin{cases} 0 & t < 0 \\ f_1(t) & 0 < t < \tau \\ A(\tau)\, f_1(t) & t > \tau \end{cases}$$



The dipole-control model

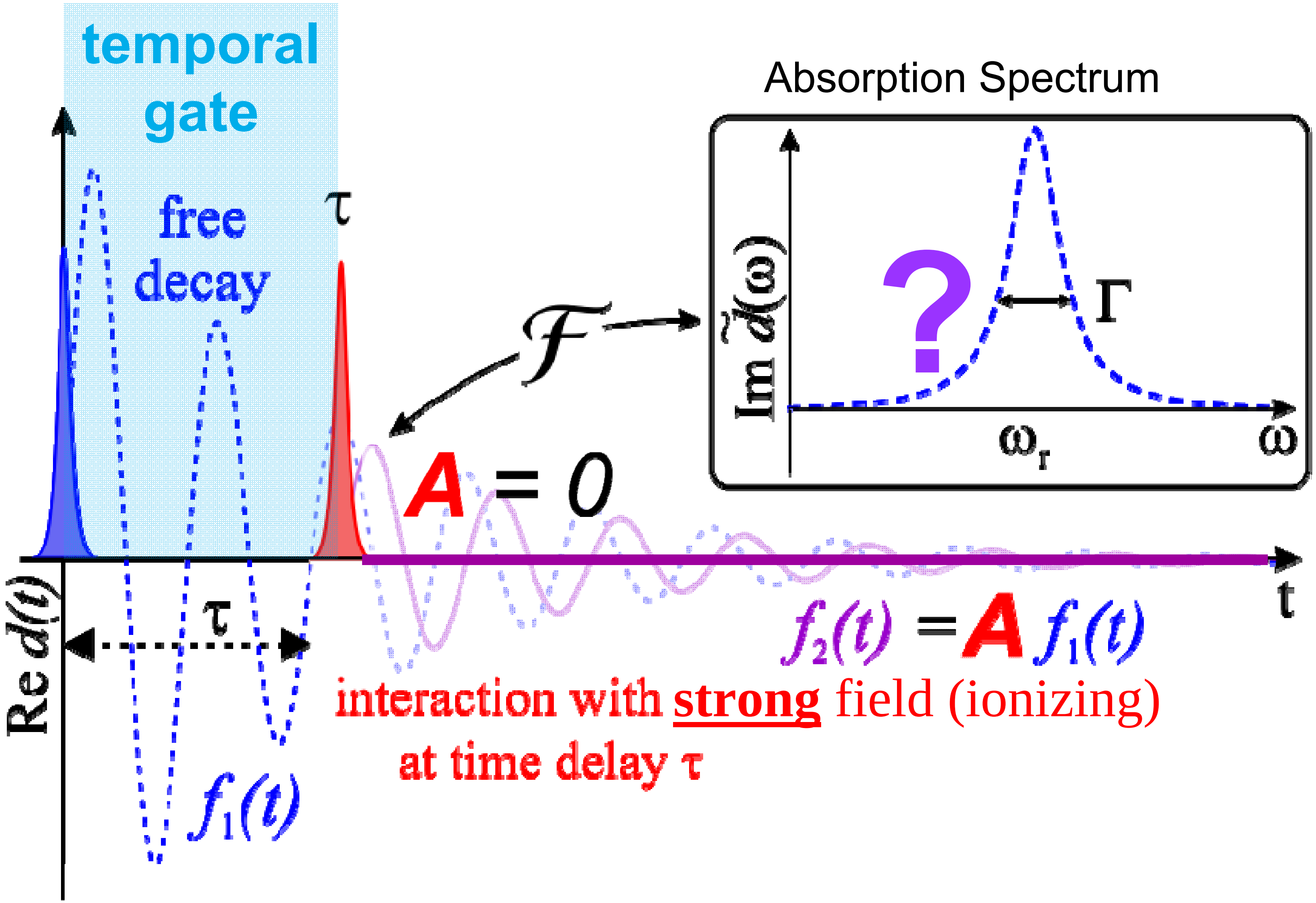
Analytical result: Blättermann *et al.* J. Phys. B: At. Mol. Opt. Phys. **47** 124008 (2014)

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



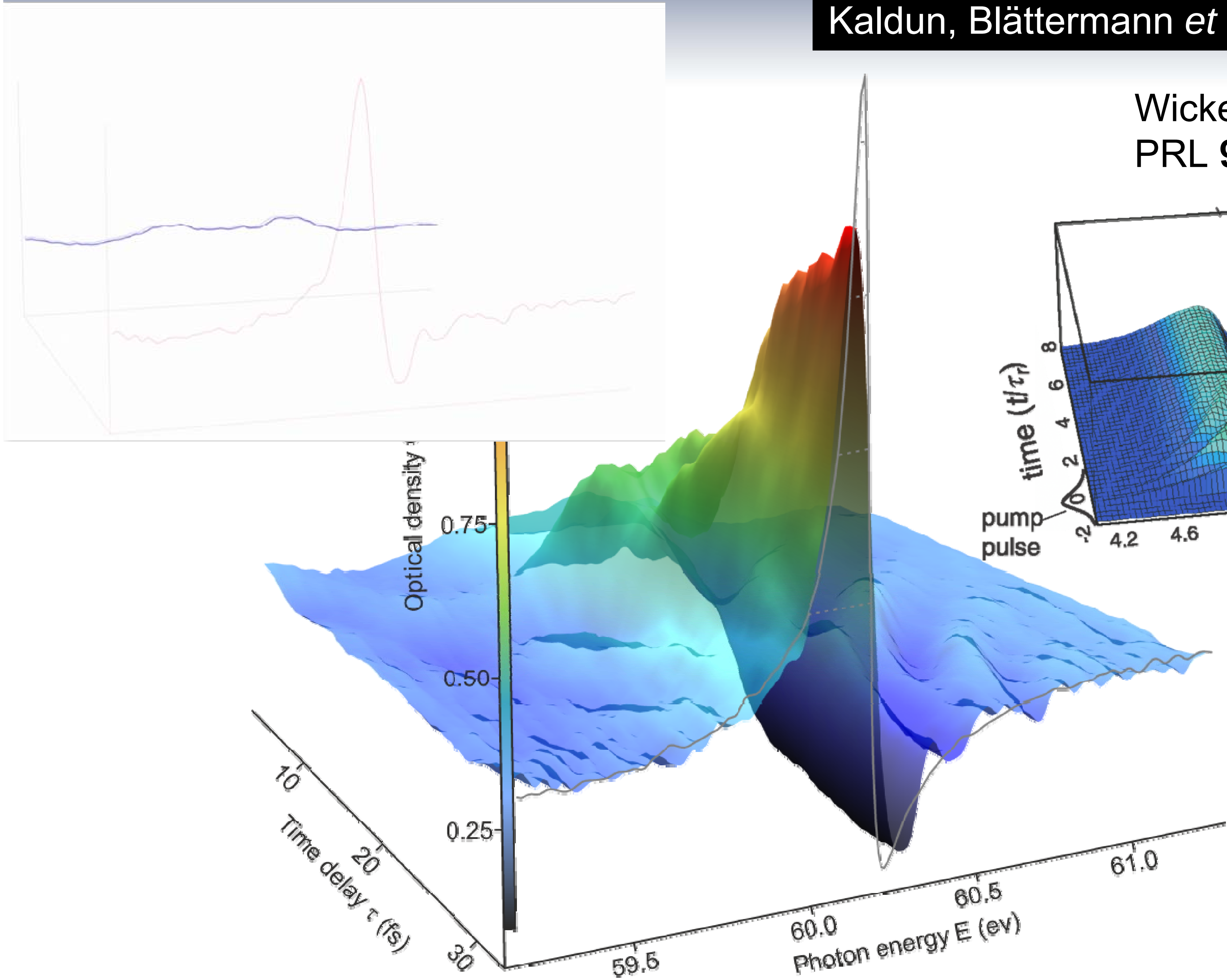
The Dipole-Amplitude Gate

Kaldun, Blättermann *et al.* Science 2016

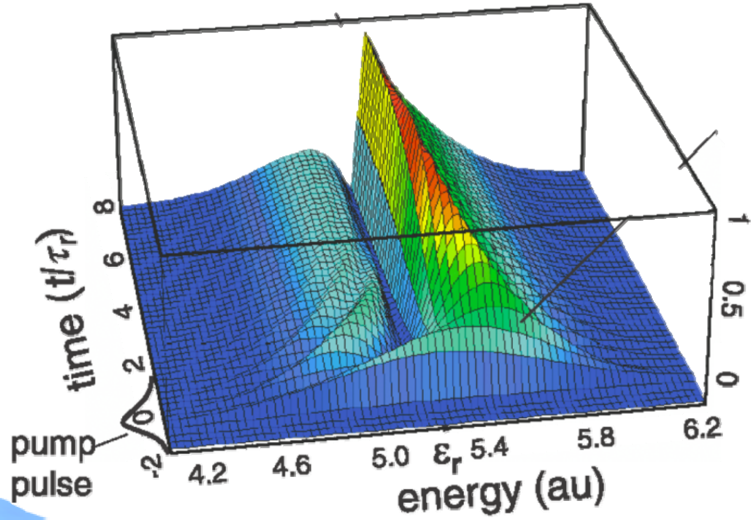


The birth of a Fano resonance

Kaldun, Blättermann *et al.* Science 2016



Wickenhauser *et al.*
PRL **94,** 023002 (2005)



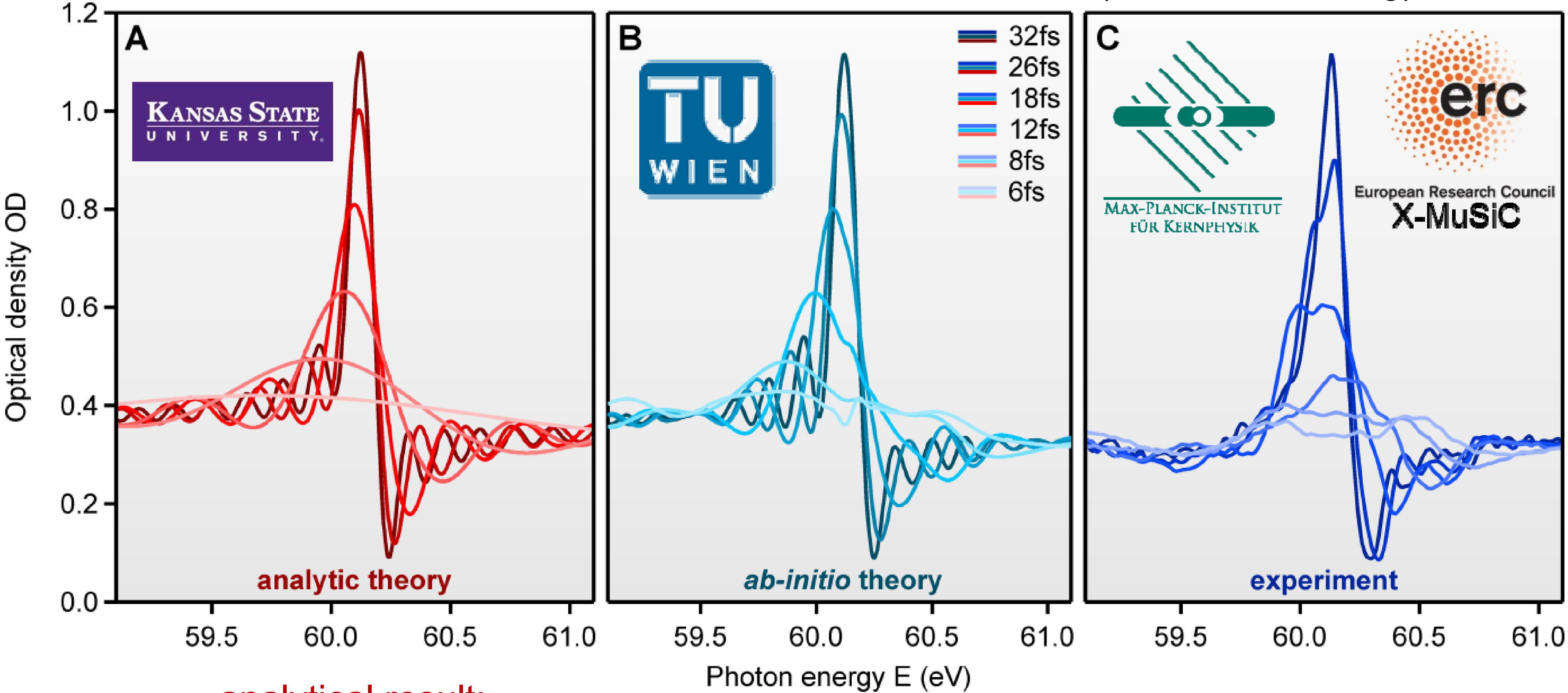
From theory to experiment

Kaldun, Blättermann *et al.* Science 2016

by Hui Wei and Chii-Dong Lin (KSU, USA)

by R. Pazourek, S. Donsa J. Burgdörfer (TU Vienna, Austria)

Experiment (MPIK, Heidelberg)



analytical result:

$$\sigma(\epsilon,\tau) \propto \mathrm{Re}\left\{1+\frac{(q-i)^2}{1-i\epsilon}\left[1-e^{-\frac{\Gamma}{2}(1-i\epsilon)\tau}\right]\right\}$$

ideal case approximation

TDSE (TDCC)

time-gated absorption spectroscopy

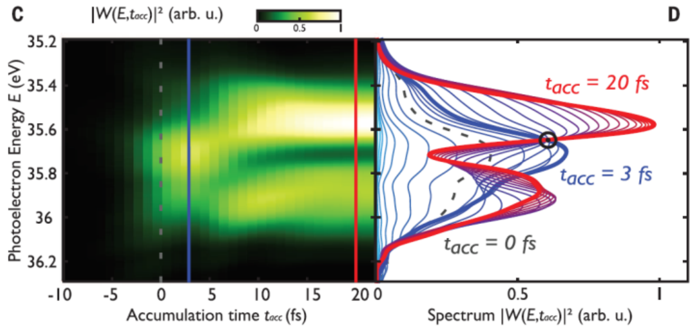
Two complementary studies

CHEMICAL PHYSICS

Attosecond dynamics through a Fano resonance: Monitoring the birth of a photoelectron

V. Gruson,[1*] L. Barreau,[1*] Á. Jiménez-Galan,[2] F. Risoud,[3] J. Caillat,[3] A. Maquet,[3] B. Carré,[1] F. Lepetit,[1] J.-F. Hergott,[1] T. Ruchon,[1] L. Argenti,[2]† R. Taïeb,[3] F. Martín,[2,4,5]‡ P. Salières[1]‡

Photoelectron spectroscopy



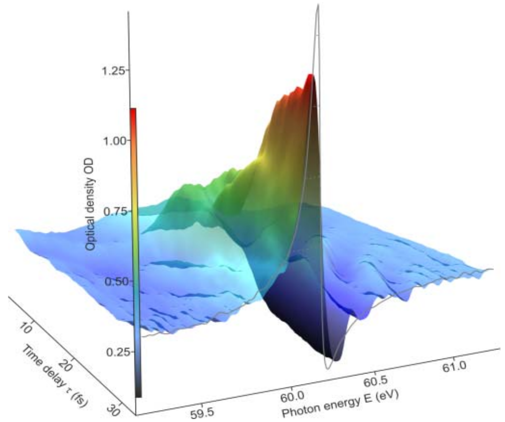
Science **354** 734 (2016)

CHEMICAL PHYSICS

Observing the ultrafast buildup of a Fano resonance in the time domain

A. Kaldun,[1*]† A. Blättermann,[1]† V. Stooß,[1] S. Donsa,[2] H. Wei,[3] R. Pazourek,[2] S. Nagele,[2] C. Ott,[1] C. D. Lin,[3] J. Burgdörfer,[2] T. Pfeifer[1,4]‡

Absorption spectroscopy



Science **354** 738 (2016)

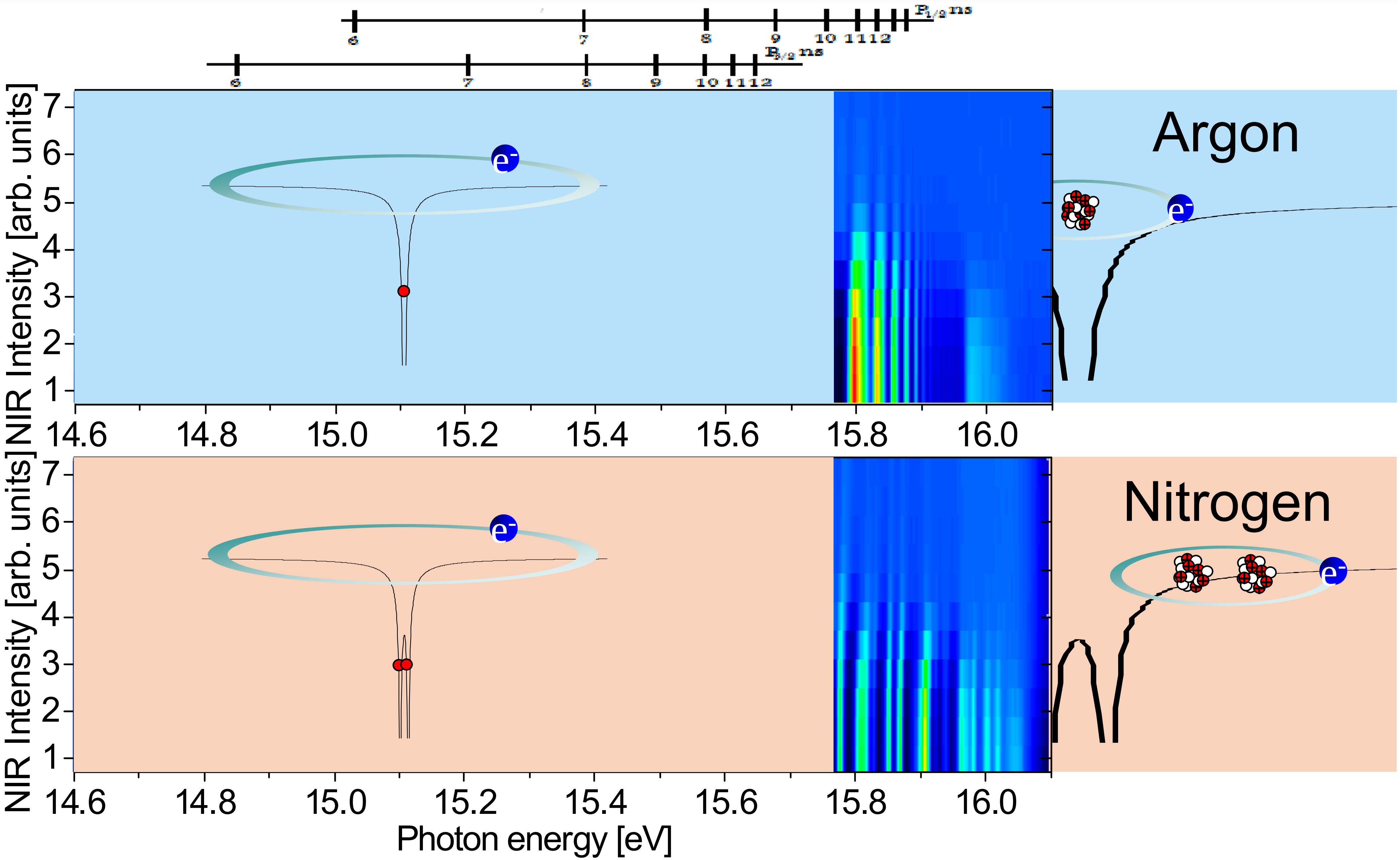
From atoms to molecules

Can we change the shape of molecular states?

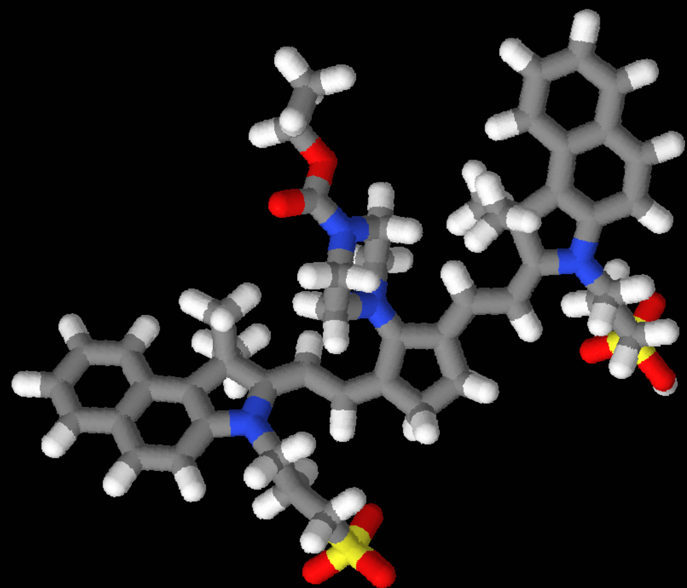
If such control works for (two) bonding electrons in molecules ...
... this would open doors to laser-directed chemistry.

Atomic and molecular resonances

interacting with weak to strong laser fields



Can we change the shape of complex molecules?

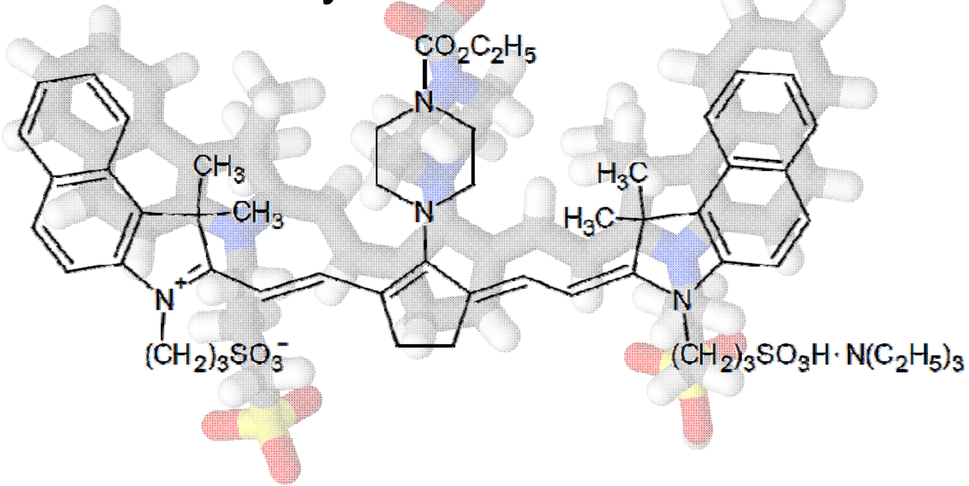


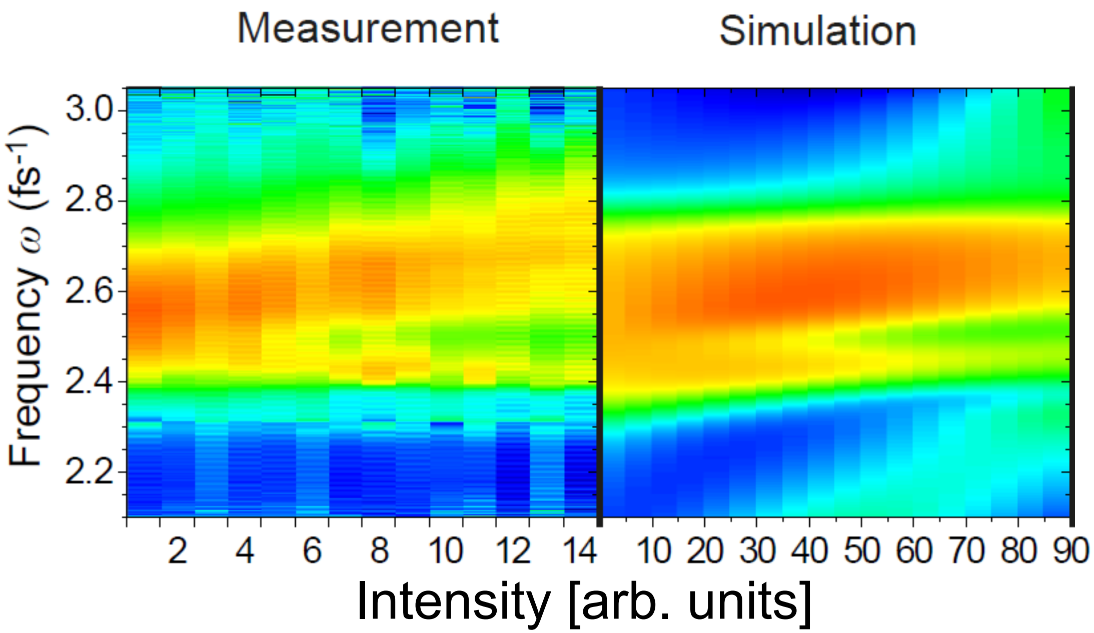
Fano control of molecules in the liquid phase

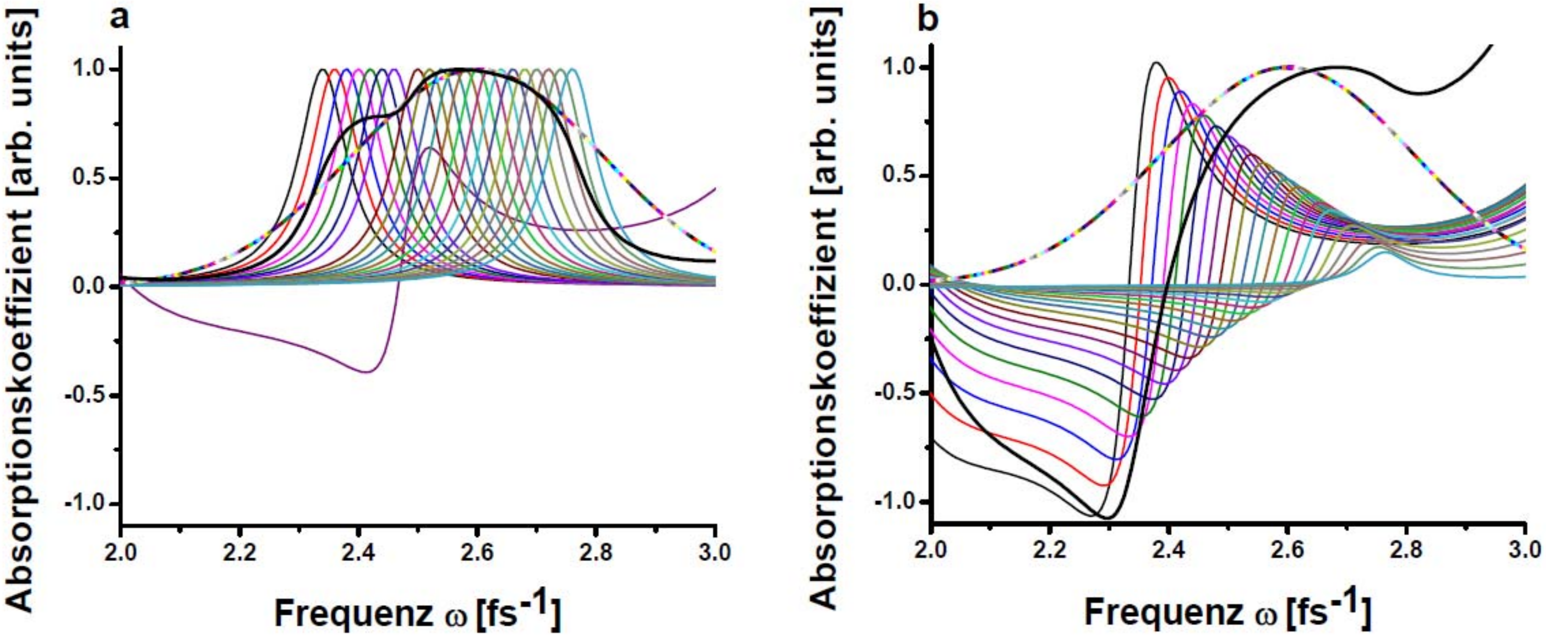
cooperation with J.-M. Mewes, A. Dreuw, Univ. Heidelberg

Kristina Meyer *et al.*, PNAS (2015)

Laser Dye IR144 in methanol

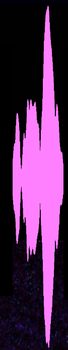




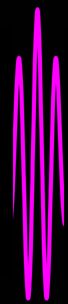


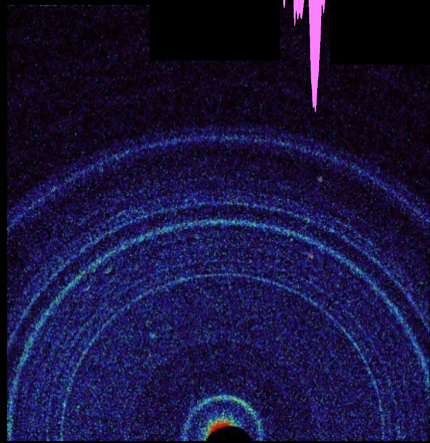
Time-resolved Science with novel x-ray/XUV laser sources

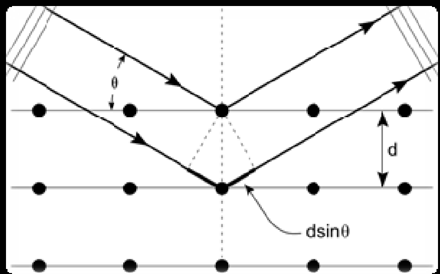
Free Electron Lasers



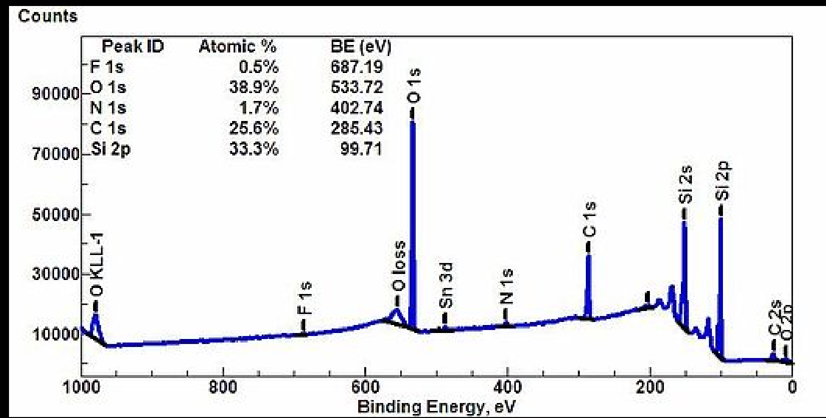
High Harmonic Generation

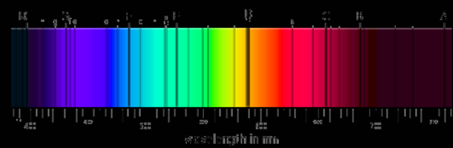






X-ray diffraction

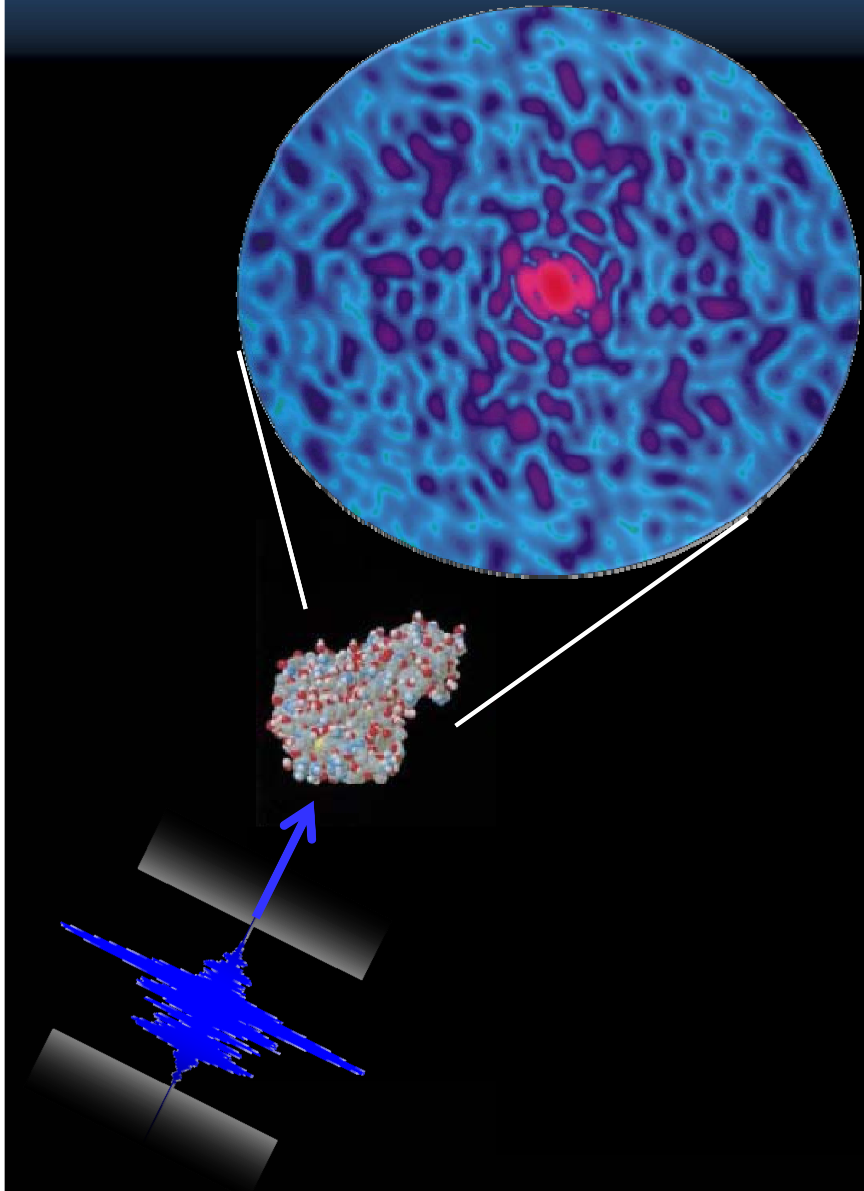




XUV spectroscopy

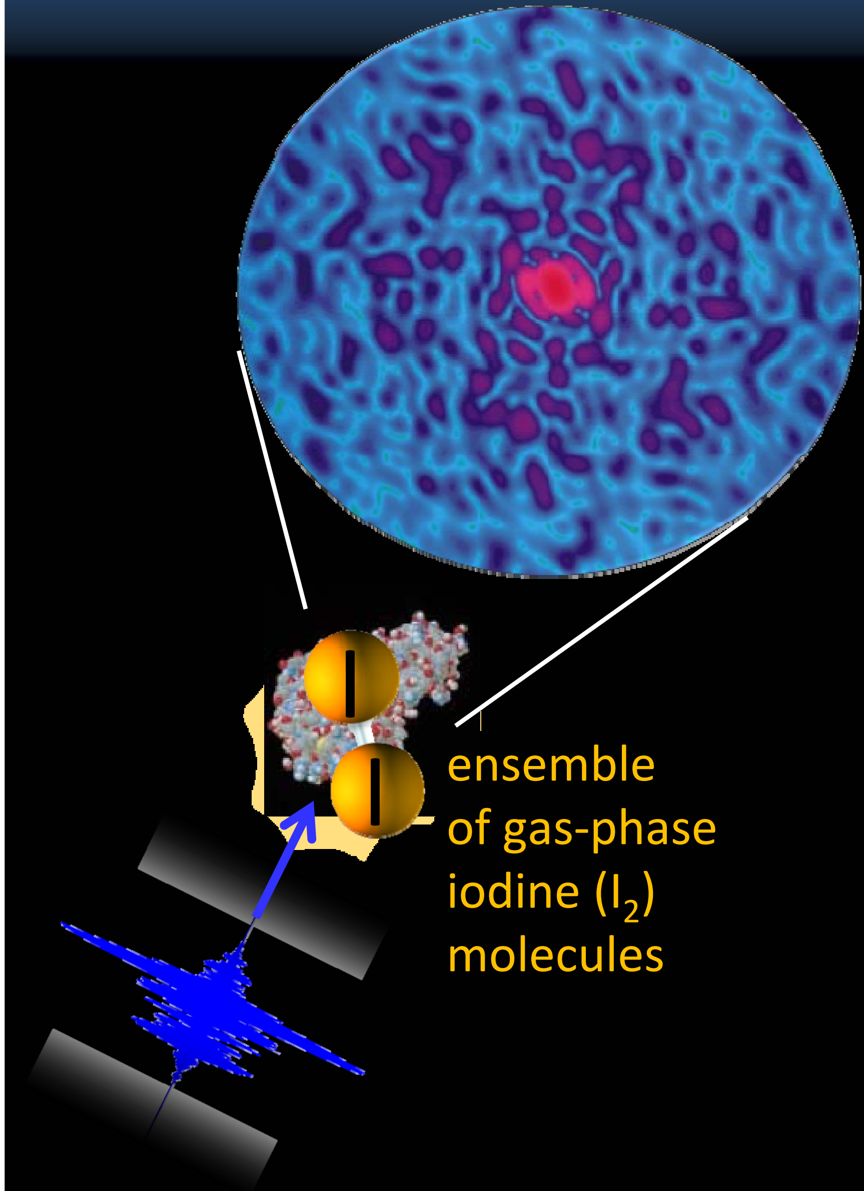
Figs.: Wikipedia

Challenge: "See" (image) molecules in x-ray Light

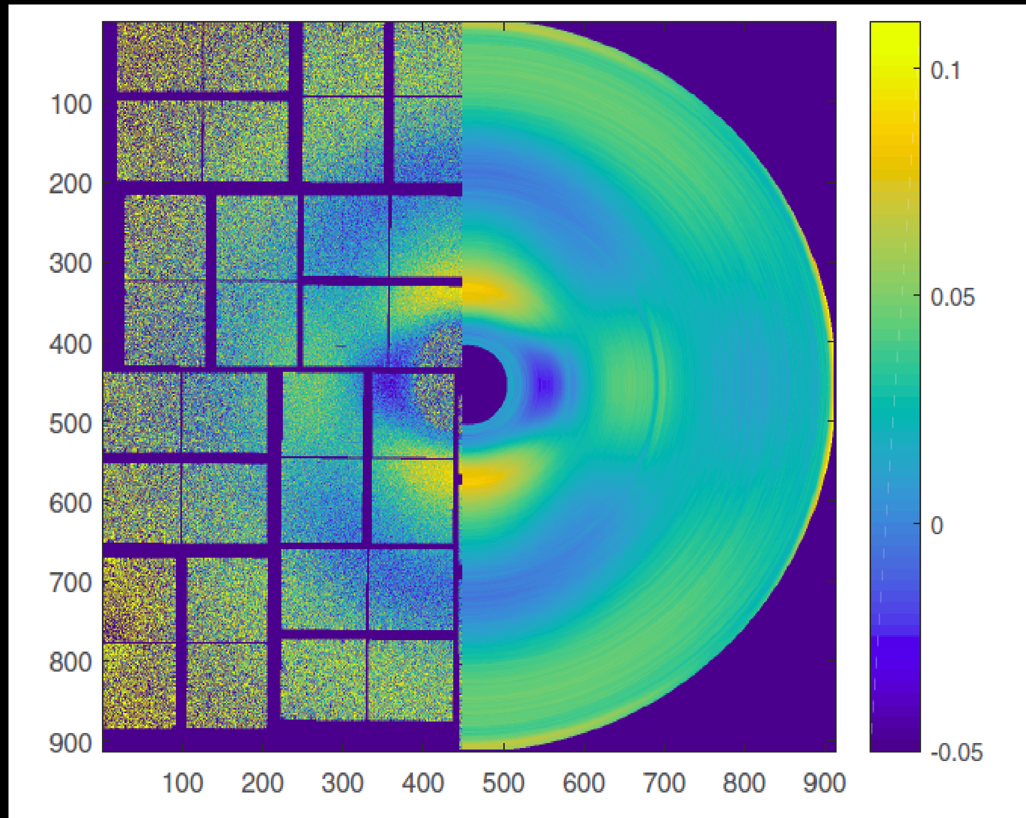


Neutze *et al.*, Nature **406**, 752 (2000)

One Goal: “See” (image) molecules in x-ray Light

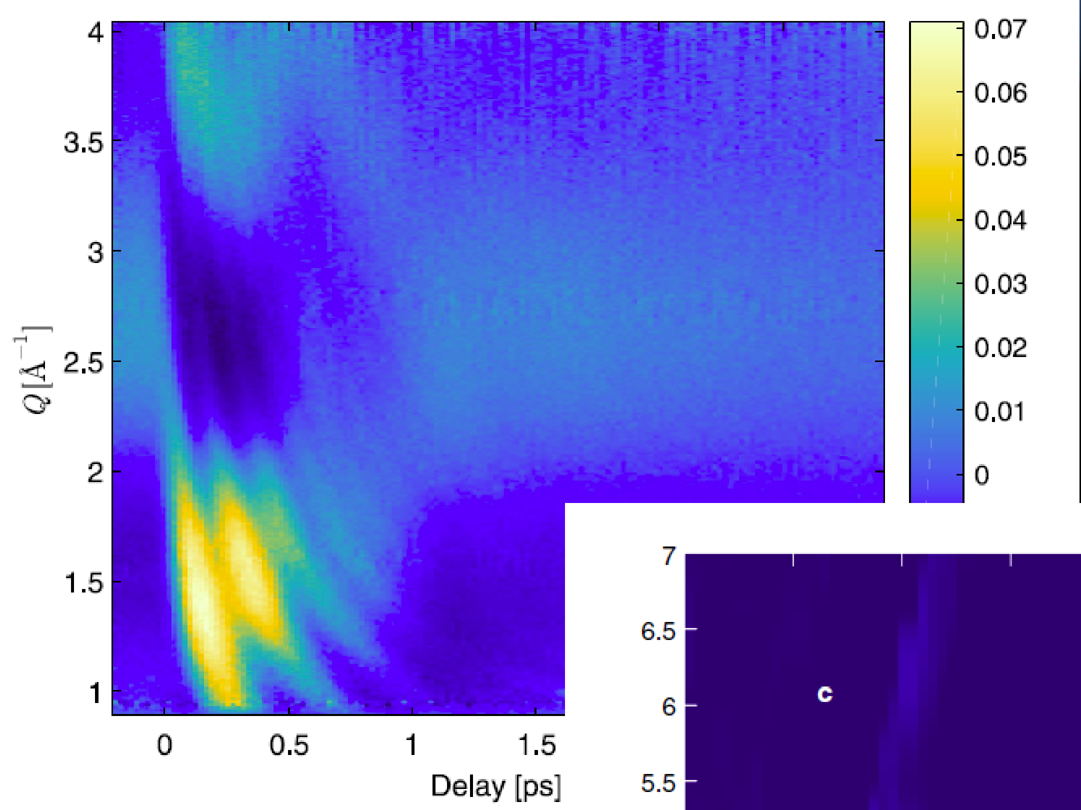


ensemble of gas-phase iodine (I_2) molecules



Glownia et al. PRL **117**, 153003 (2016)
(Bucksbaum group@SLAC)

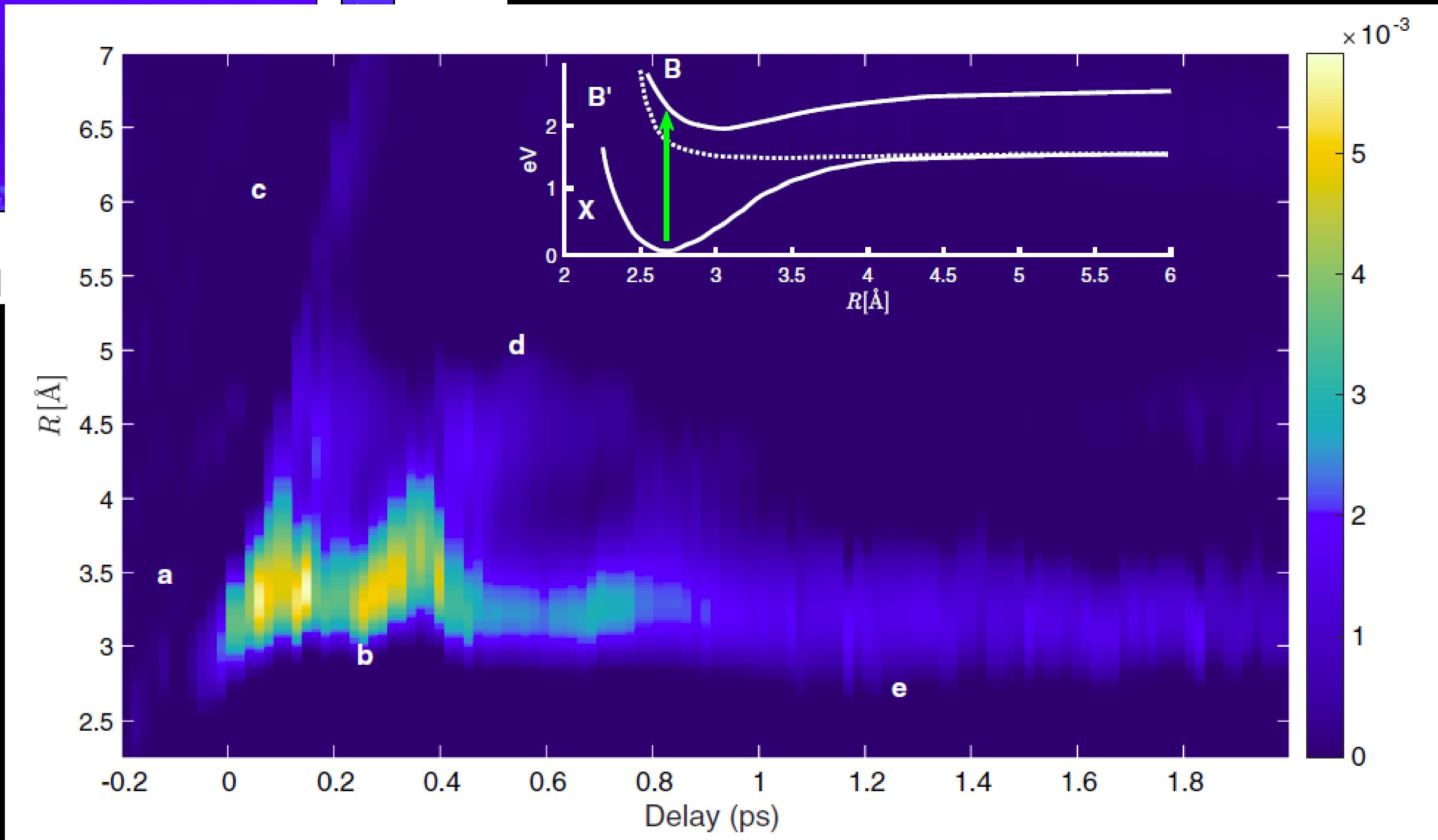
Time-resolved dynamics



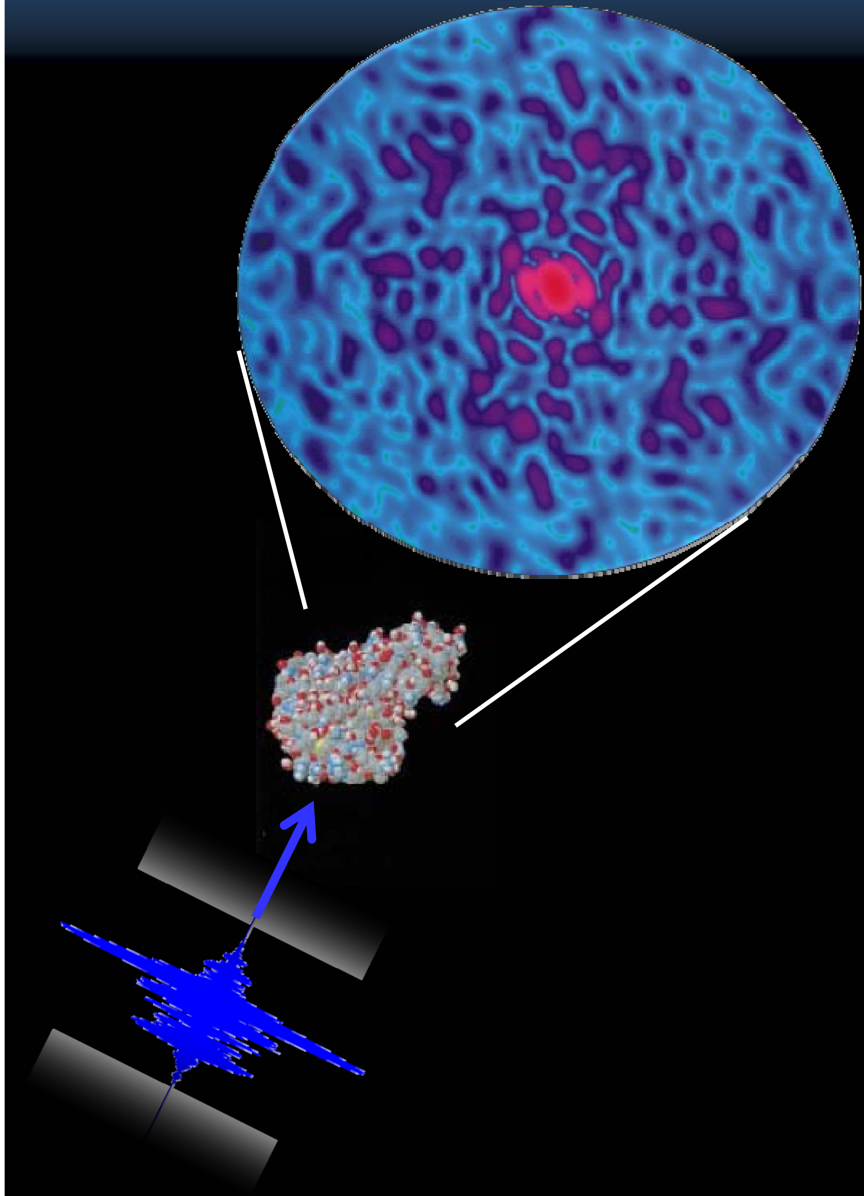
Scattering image acquired for various delay times

Glownia et al. PRL **117**, 153003 (2016)
(Bucksbaum group@SLAC)

Reconstructed molecular wavepacket after single-photon excitation

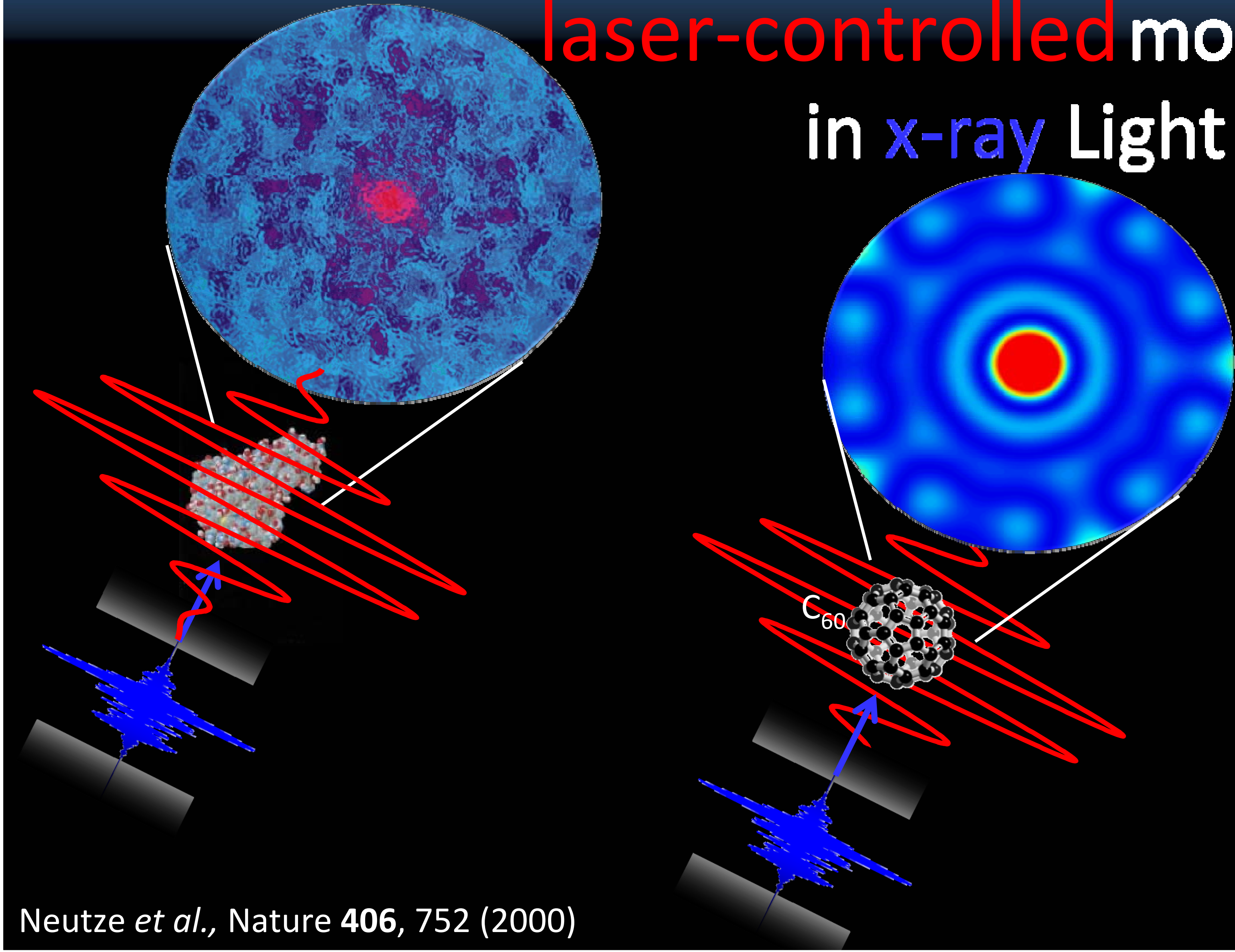


One Goal: “See” (image) molecules in x-ray Light



Neutze *et al.*, Nature **406**, 752 (2000)

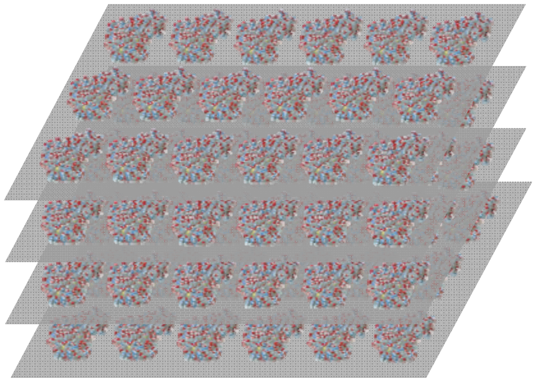
Next Step: "See" (image) laser-controlled molecules in x-ray Light



Neutze *et al.,* Nature **406**, 752 (2000)

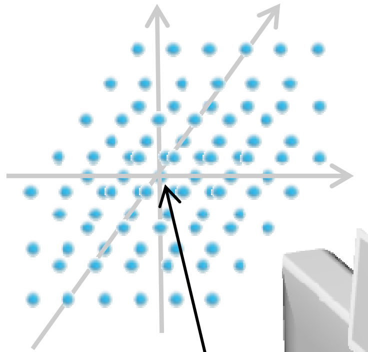
X-ray diffraction crash course/refresher

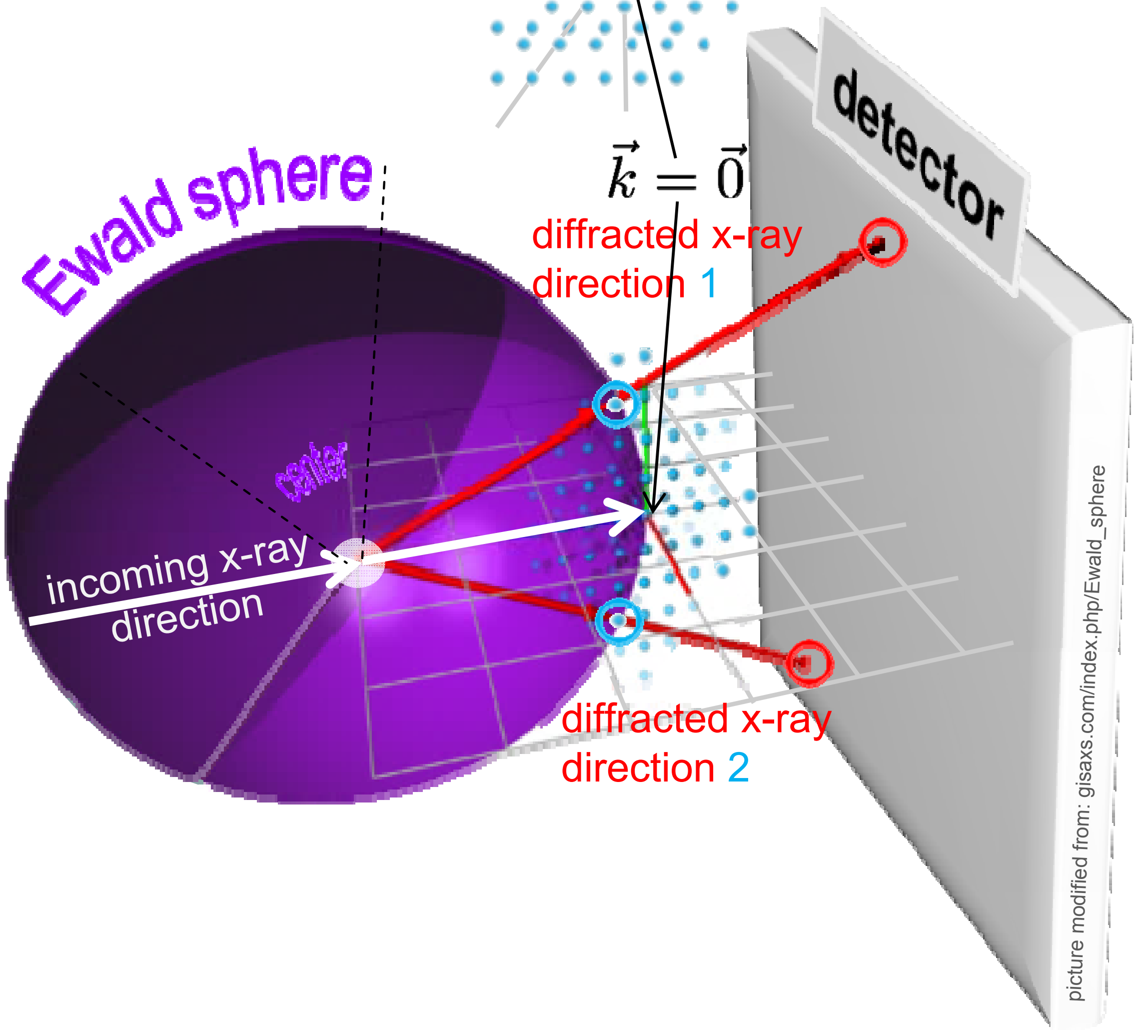
real space



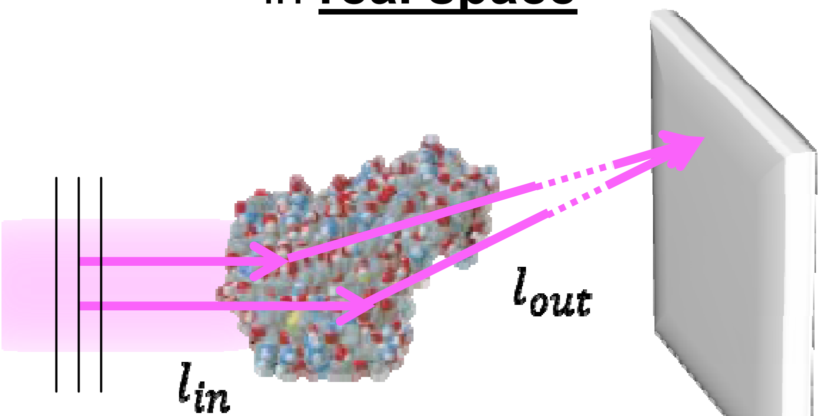
Fourier Transform

k/momentum space





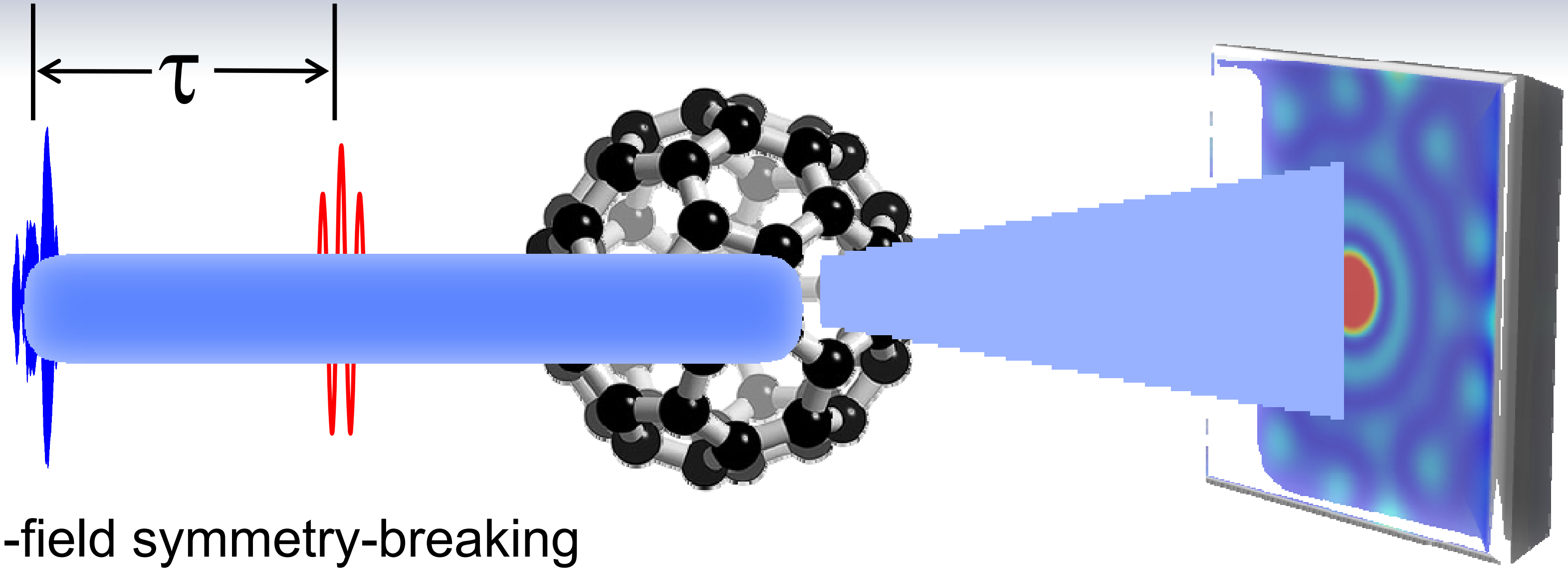
Alternative route for calculation:
-> addition of geometric paths
in **real space**



l_{out} : function of detector pixels and atomic positions x,y,z (simplified by letting $l_{out} \gg \Delta x, \Delta y, \Delta z$

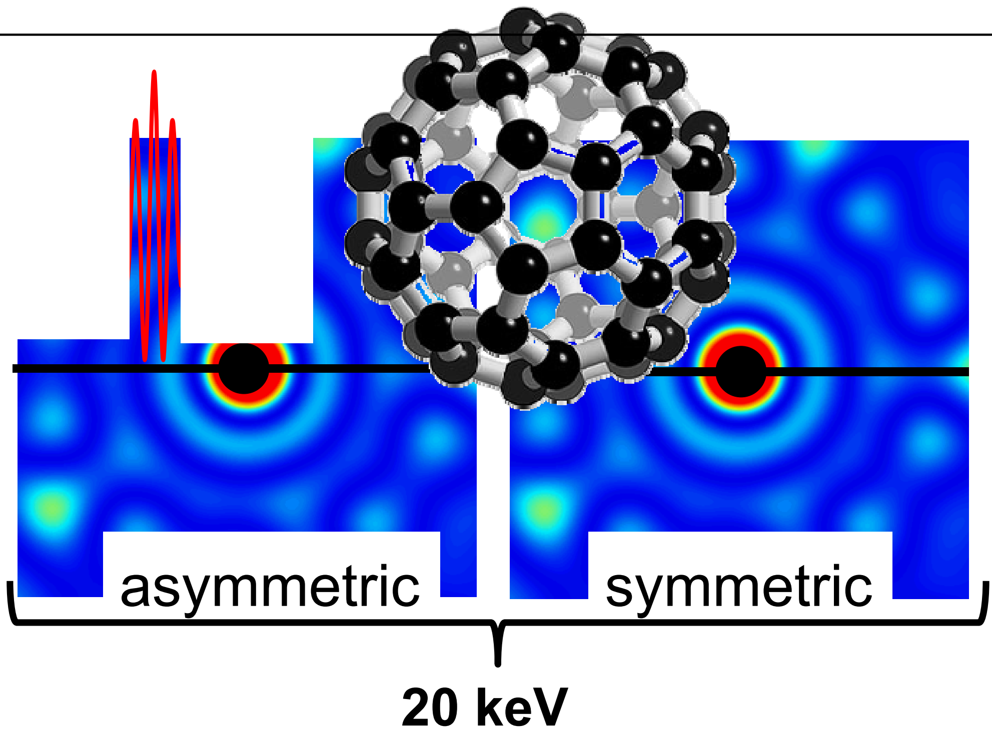
C_{60} vibrational dynamics imaging

C60 real-space image
from: 3DChem.com



study strong-field symmetry-breaking

simulation results:



LCLS Proposal
(1st submission 2012)

X-ray imaging of laser-induced coherent dynamics in C_{60} fullerenes

Claus Peter Schulz*, Ilme Schlichting, Lutz Foucar, Thomas Möller, Christoph Bostedt, Timur Osipov, Arnaud Rouzée, Marc Vrakking, Artem Rudenko, Katharina Kubicek, Simone Techert, Nora Berrah, Jochen Küpper, Ulf Saalmann, Jan Michael Rost, Rüdiger Schmidt, Kiyoshi Ueda, Louis DiMauro, Robert Moshammer, Joachim Ullrich, and Thomas Pfeifer*

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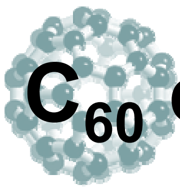
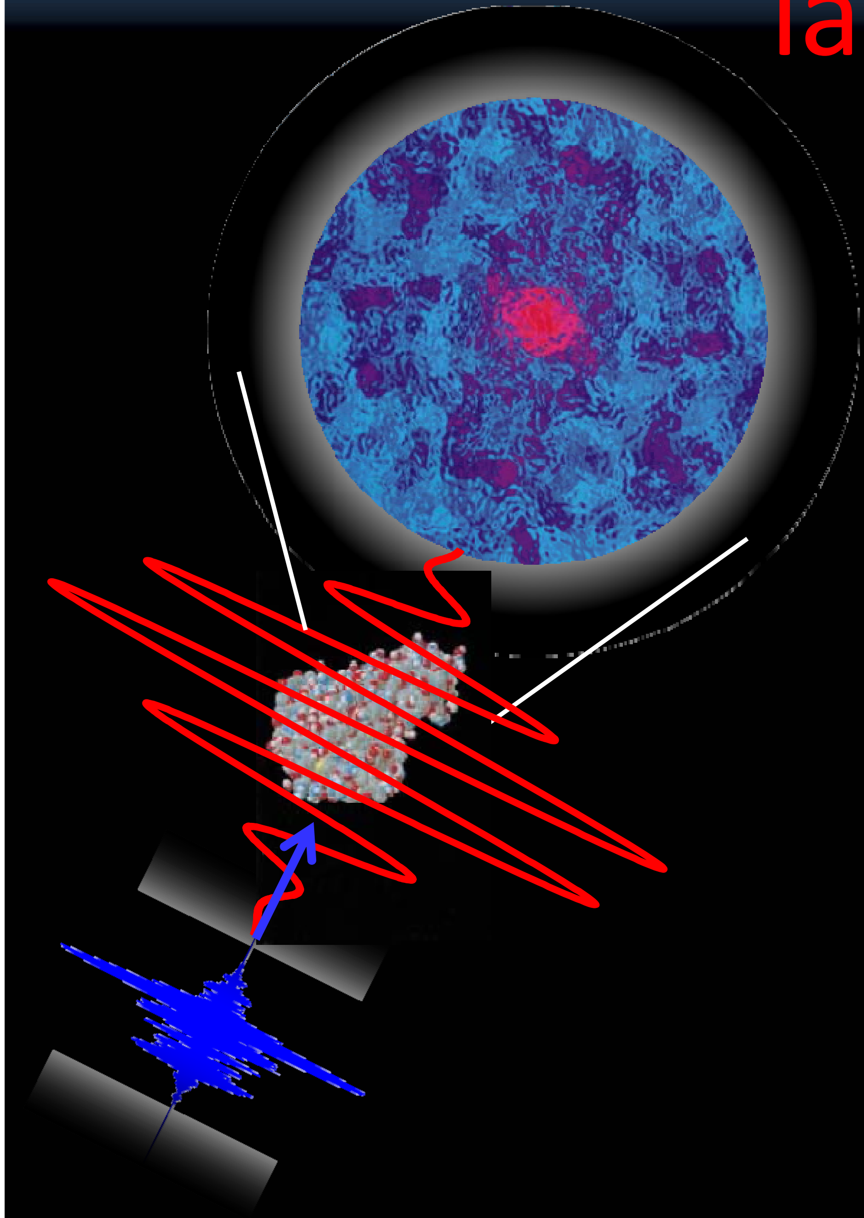
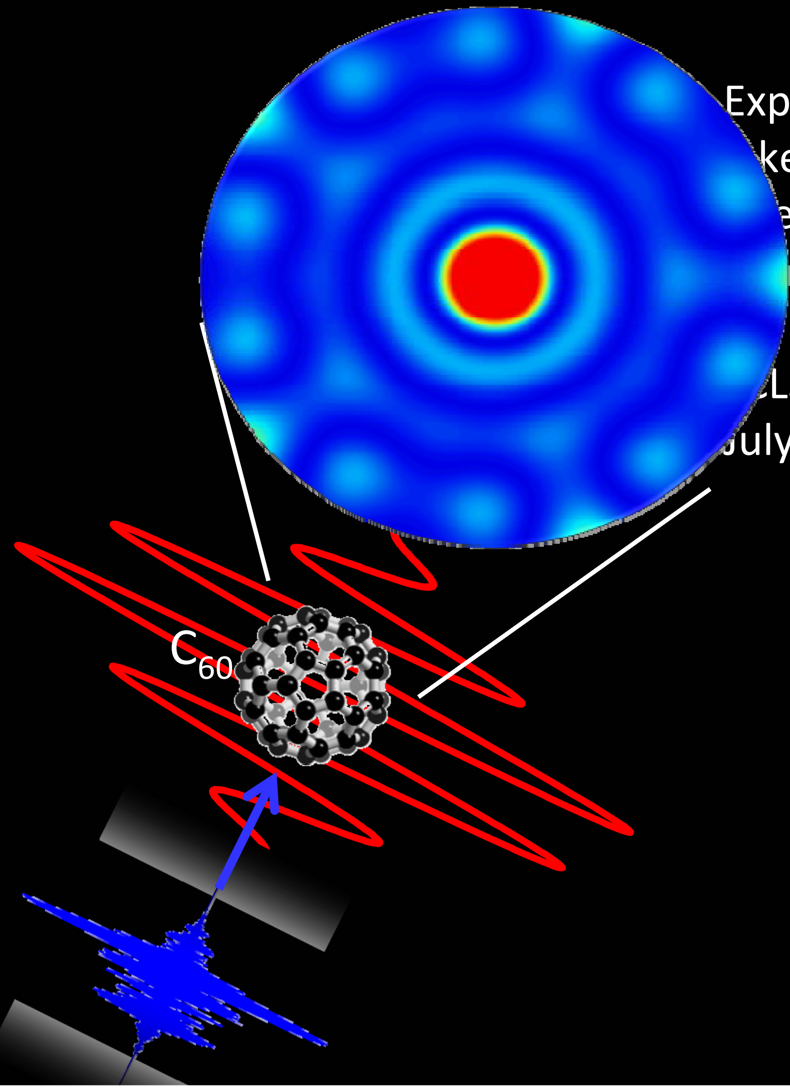
Abstract: Optical lasers interacting with C_{60} fullerene molecules induce coherent electronic and vibrational excitations, resulting in the deformation of the originally symmetric structure. The strong-field symmetry-breaking (leading to asymmetric prolate/oblate deformation vs symmetric breathing mode) and the corresponding coherence lifetimes have only been indirectly explored in experiments, while direct imaging could provide answers to these scientific questions. This experiment can only be conducted at the LCLS due to its specific attributes as explained in the experimental section. C_{60} represents an important model system at the interface between clusters and molecules, and also between inorganic and organic medium-size molecules. The results of this proposed experiment are also expected to have benchmark character for future studies in dynamical x-ray diffractive imaging of molecules.



"Can we >see< C_{60} changing shape in a strong field"

One Goal: “See” (image) laser-controlled molecules in x-ray Light





Experimental Data
keV photon
ergy
ractive imaging

LS Experiment
July 2016

C_{60} dynamical transitions

delay → IR early

delay ↓ IR early

IR pulse energy: 2.57 mJ

0.76 mJ

0.35 mJ

Imaging time-resolved laser-induced symmetry breaking?

horizontal width

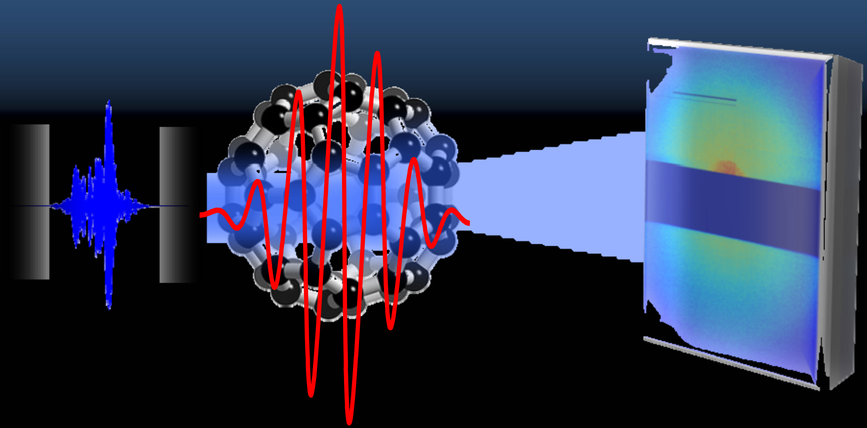
vertical width

total diffraction signal

IR-Laser polarization: horizontal

LCLS Experiment July 2016, preliminary results, analysis in progress

Opening new science opportunities



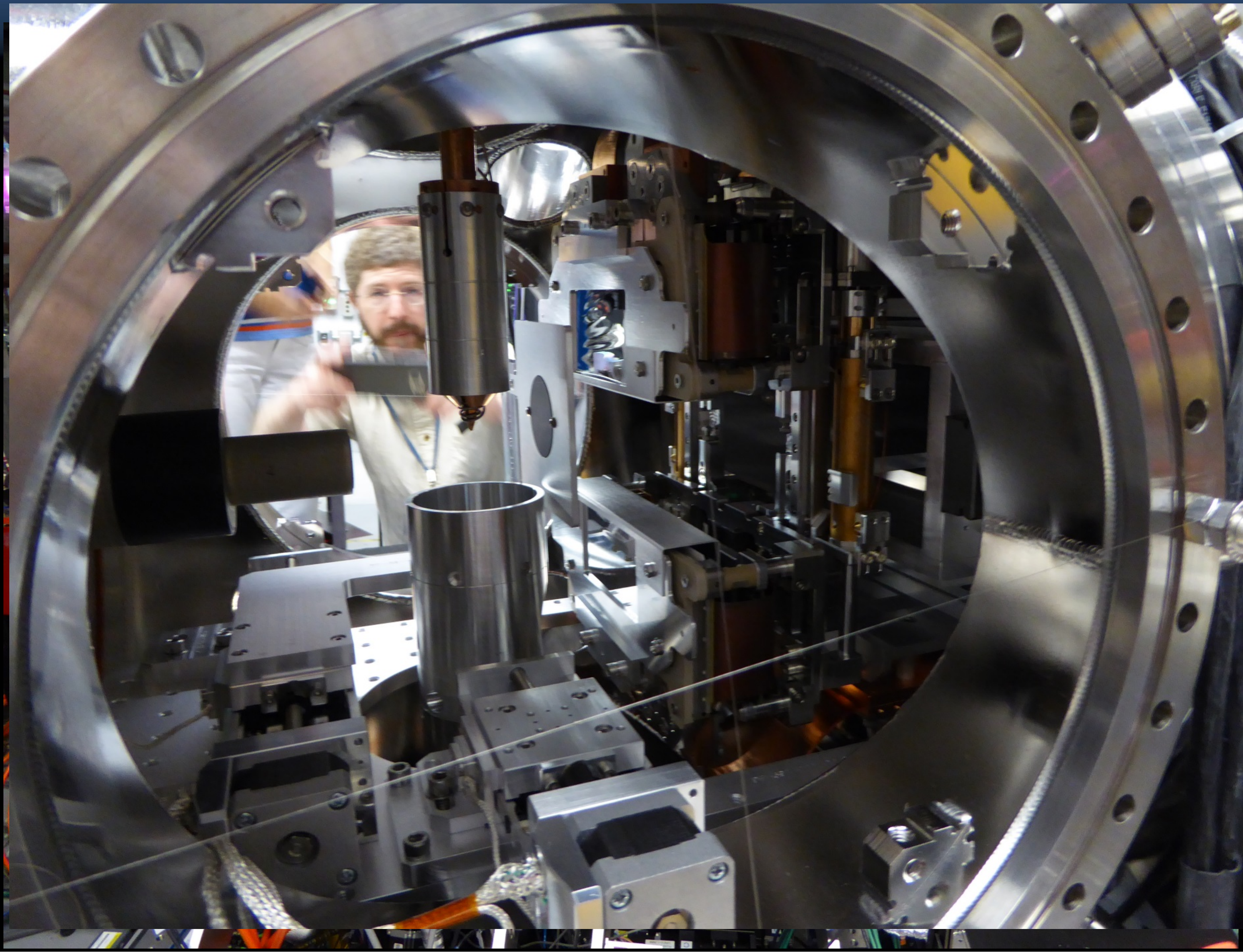
of particular interest:
- for LCLS II
- for European XFEL (higher rep rates)

- 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

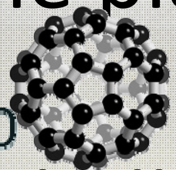
high intensity $10^{15} \frac{W}{cm^2}$ ← low intensity $10^{14} \frac{W}{cm^2}$

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

C_{60} imaging: AMO@LCLS



Thanks to the players of the

C_{60} 

quantum football/soccer team

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erc X-MuSiC

Heidelberg Center for Quantum Dynamics

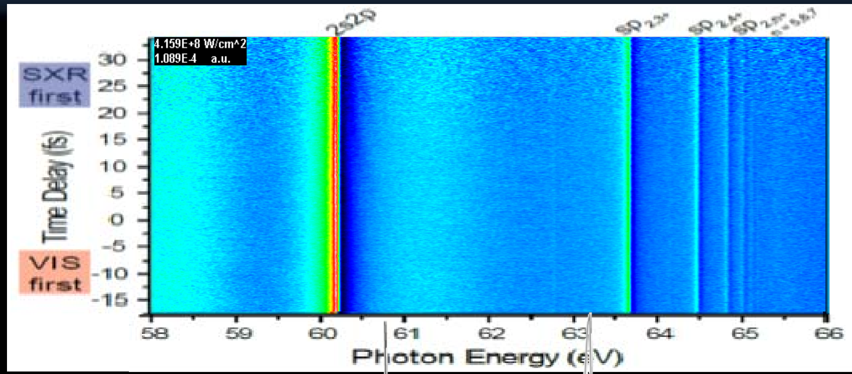
Take-home messages

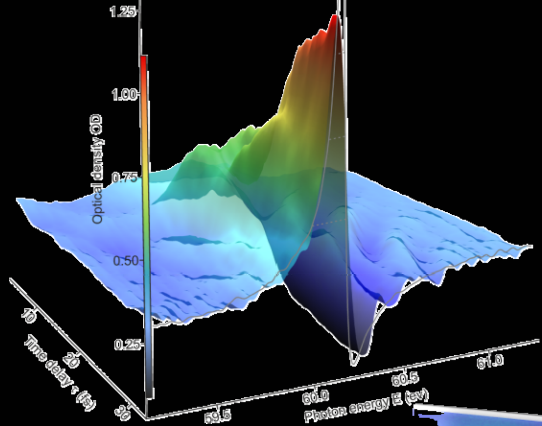
Atoms and molecules change their spectral (line) shape

as a function of laser intensity

quantum-state phase $\varphi = \Delta E \Delta t$

$$q(\varphi) = -\cot\left(\frac{\varphi}{2}\right)$$



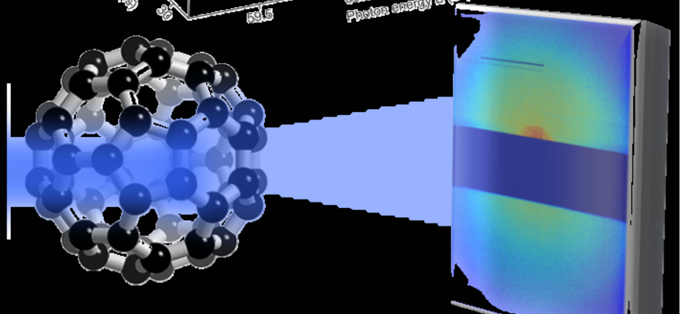


- Phase control tunes the Fano q parameter
- Amplitude gate resolves the Fano resonance

... and these mechanisms are general...

- See/image molecules changing their spatial shape

  Intensity dependence of structural dynamics in C_{60}



Understand & Control Matter in Strong laser fields