

The sound of the phase:
Atoms&molecules
respond to intense
laser flashes

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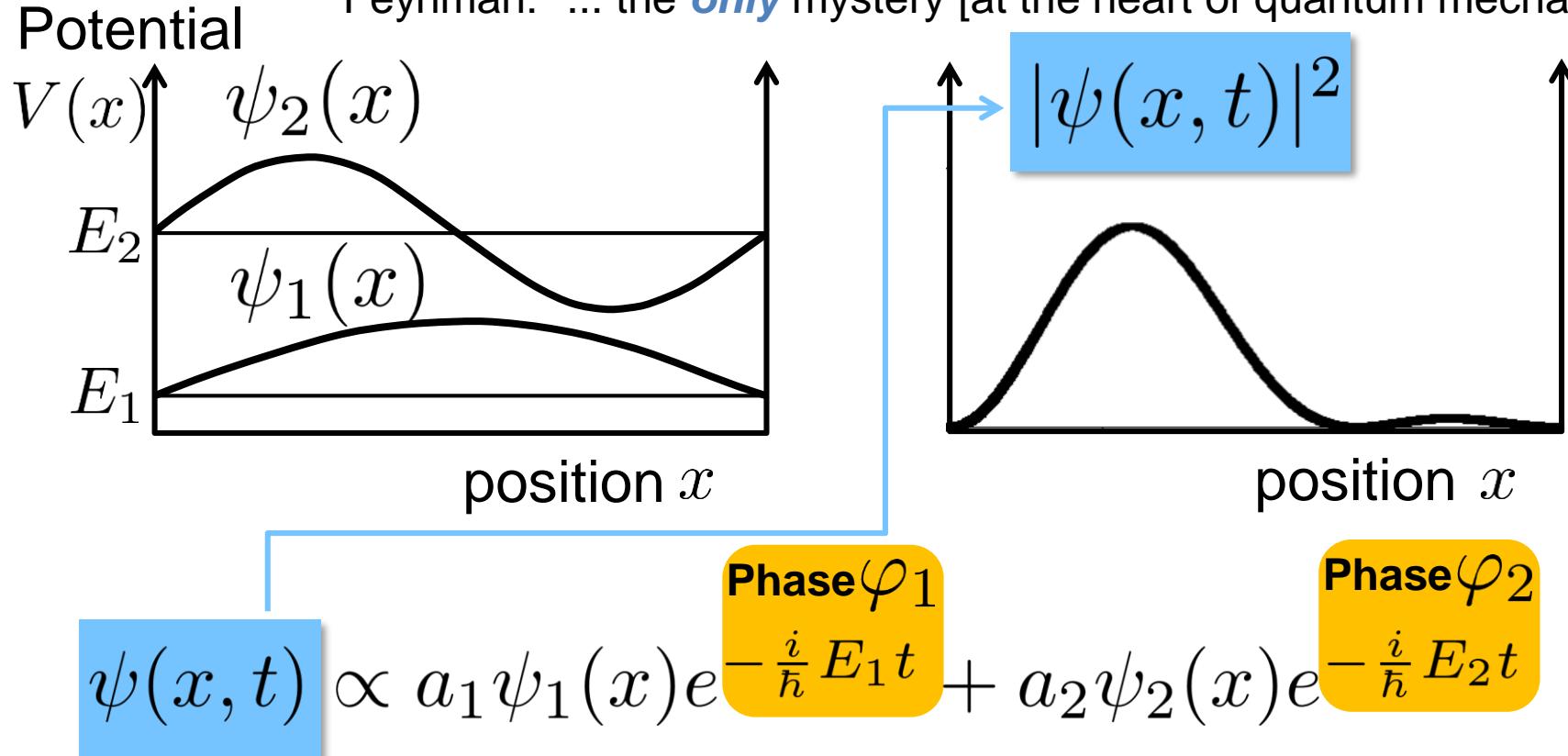


Fundamental Quantum Dynamics

$$i\hbar \frac{\partial}{\partial t} \psi(x, t) = \mathcal{H}\psi(x, t)$$

Interference of quantum states

Feynman: "... the **only** mystery [at the heart of quantum mechanics]."



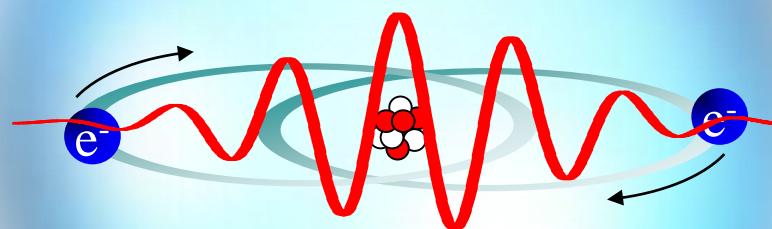
few-body quantum dynamics

a fundamental scientific question:

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

spatial scale
 $R \sim$ sub/few Å

temporal scale
 $T \sim$ sub/few fs



The
“**quantum**
few-body
problem”
in strong fields

Science goal:
measure / understand / control
*the **quantum dynamics** of*
small systems
in strong fields

(x-ray) movies of
single molecules

Laser control of
chemical reactions

Petahertz-coded
computing

x-ray precision
spectroscopy

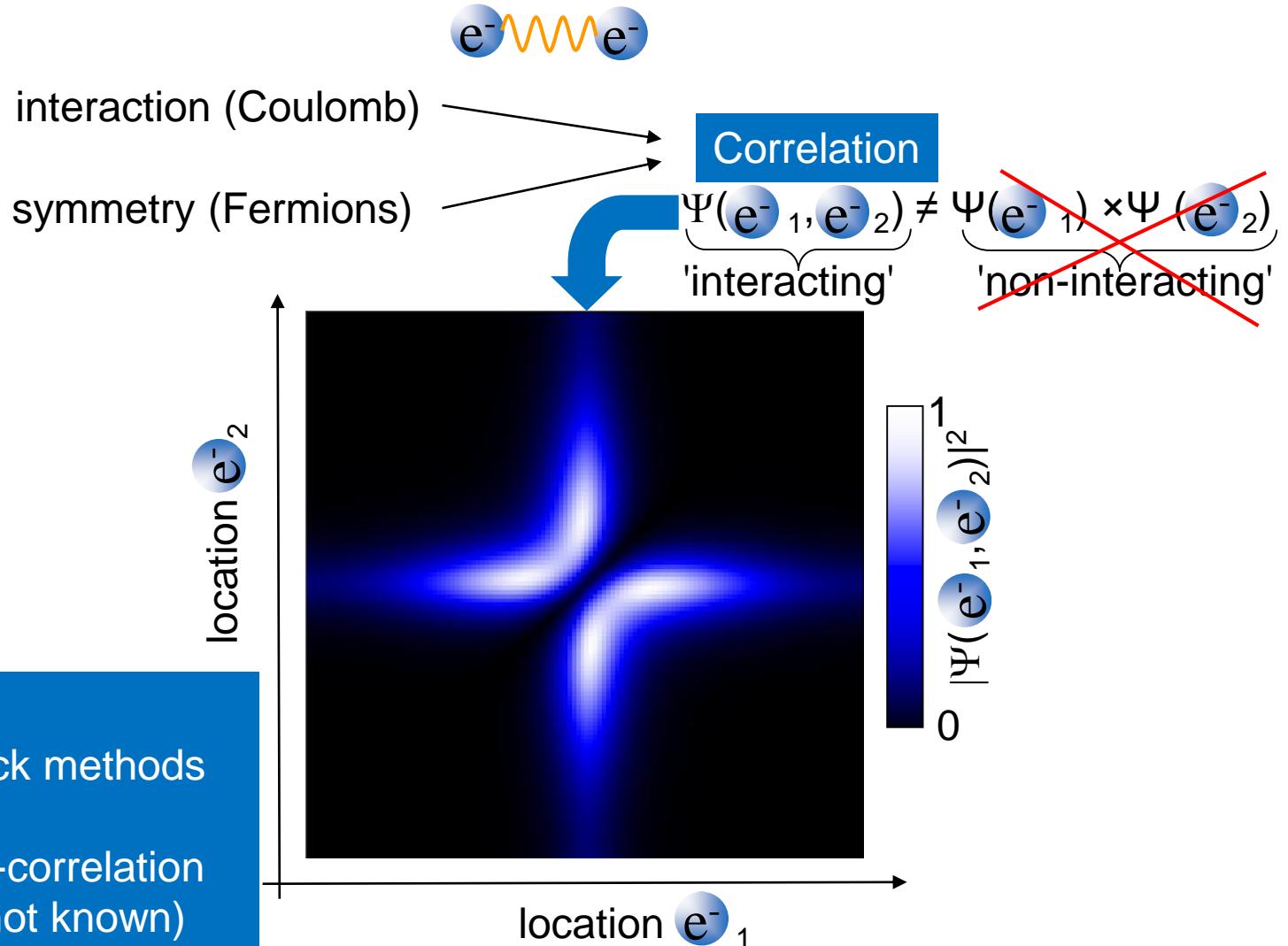
Electron Facts

- lightest charged elementary particle 
- 'chemical glue', holding molecules together
 - > making and breaking of bonds in chemical reactions
- carriers of electric current
 - > electricity generation/conversion/transport
- carriers of information
 - > electronics, computing, data storage
- mediating interactions of matter with light
 - > role in photosynthesis, photovoltaics, vision, ...

...typically more than one electron involved...

What is the problem? (with two or more electrons)

Correlated e^- lectron dynamics



Why is it important?

Time-dependent correlated electron dynamics



interaction (Coulomb)

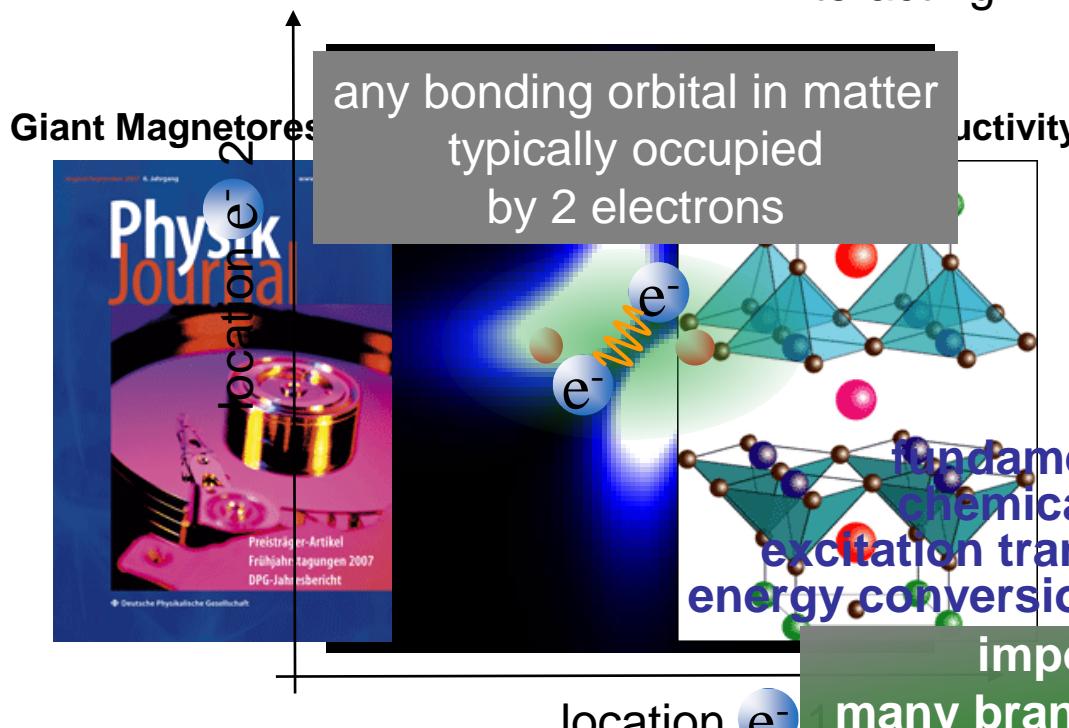
symmetry (Fermions)

Correlation

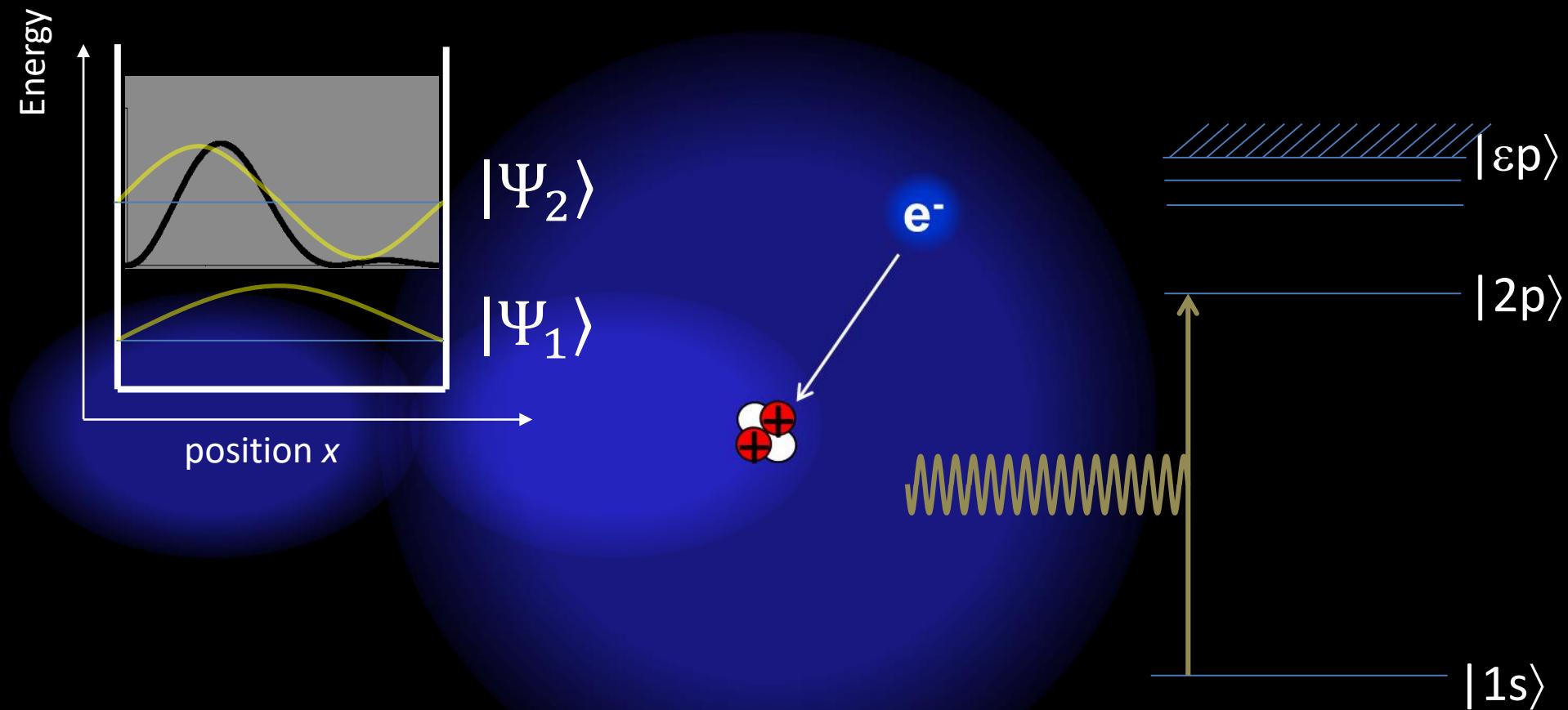
$$\Psi(e^-_1, e^-_2)$$

'interacting'

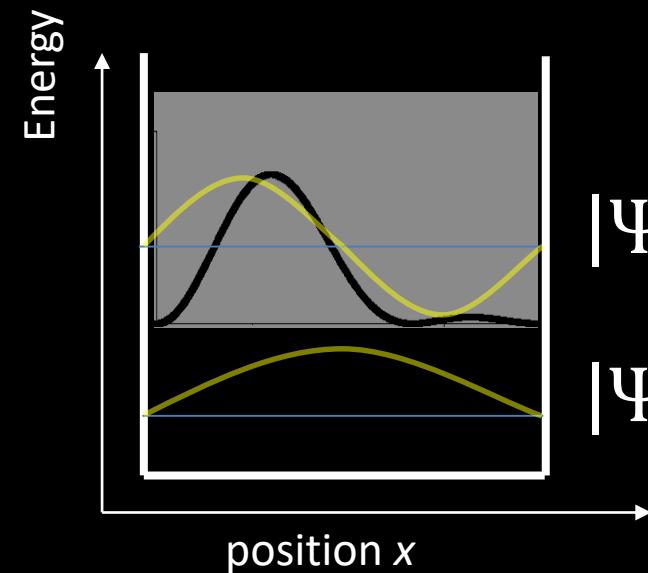
$$\underbrace{\Psi(e^-_1) \times \Psi(e^-_2)}_{\text{'non-interacting'}}$$



the language electrons speak and why is it so fast?



Wavepacket dynamics and observation



Quantum beat period:
 $\Delta T = \frac{\hbar}{\Delta E} \approx 4.1 \text{ fs} / \Delta E [\text{eV}]$

$|\Psi_2\rangle$

$|\Psi_1\rangle$

$$\Psi(t) \sim \Psi_1 e^{\frac{-i}{\hbar} E_1 t} + \Psi_2 e^{\frac{-i}{\hbar} E_2 t}$$

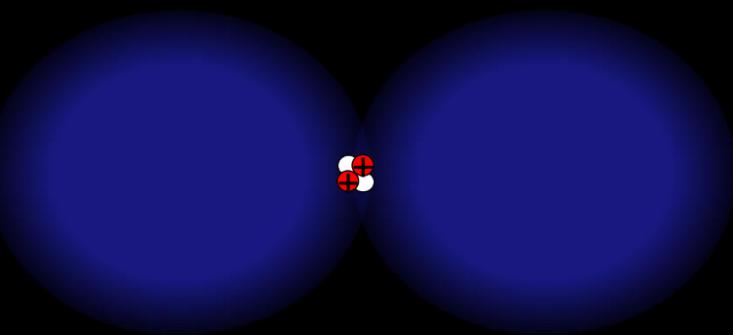
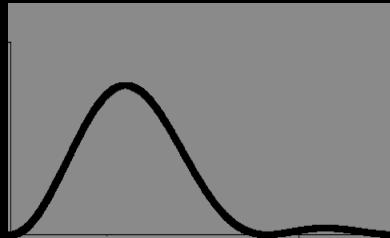
e.g. time-dependent position/dipole:

$$x(t) = \langle \Psi(t) | \hat{x} | \Psi(t) \rangle$$

$$= \frac{1}{2} \left\langle \Psi_1 e^{\frac{-i}{\hbar} E_1 t} + \Psi_2 e^{\frac{-i}{\hbar} E_2 t} \middle| \hat{x} \middle| \Psi_1 e^{\frac{-i}{\hbar} E_1 t} + \Psi_2 e^{\frac{-i}{\hbar} E_2 t} \right\rangle$$

$$= |\langle \Psi_1 | \hat{x} | \Psi_2 \rangle| \cos \left[\frac{(E_1 - E_2)}{\hbar} t + \varphi_0 \right] \rightarrow \varphi(t)$$

the language electrons talk and why is it so fast?



and the way
we "listen"....

Spectroscopy

Quantum beat period:

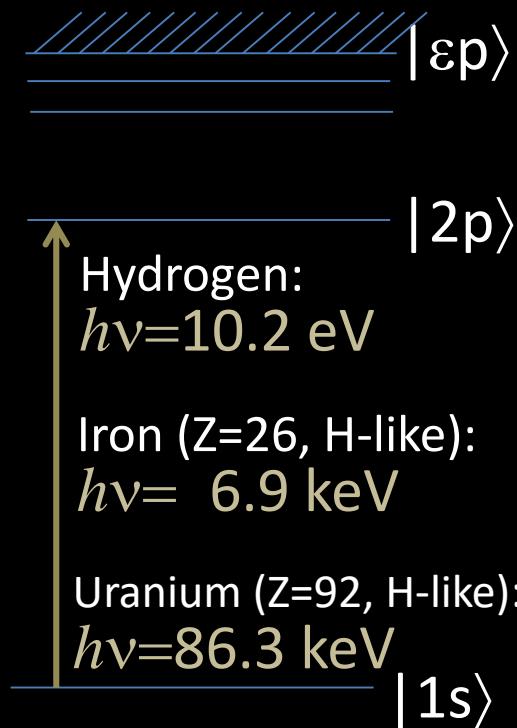
$$\Delta T = \frac{\hbar}{\Delta E} \approx 4.1 \text{ fs} / \Delta E [\text{eV}]$$

Oscillation period:

Hydrogen:
 $T = 0.4 \text{ fs} (10^{-18} \text{ s})$

Iron (H-like):
 $T = 0.6 \text{ as} (10^{-18} \text{ s})$

Uranium (H-like):
 $T = 48.0 \text{ zs} (10^{-21} \text{ s})$



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

Free Electron Lasers



SACLA

specifications demonstrated thus far:

~1 Å

~5 mJ

LCLS
@USA
~0.5 fs
partially
coherent

lowest wavelength

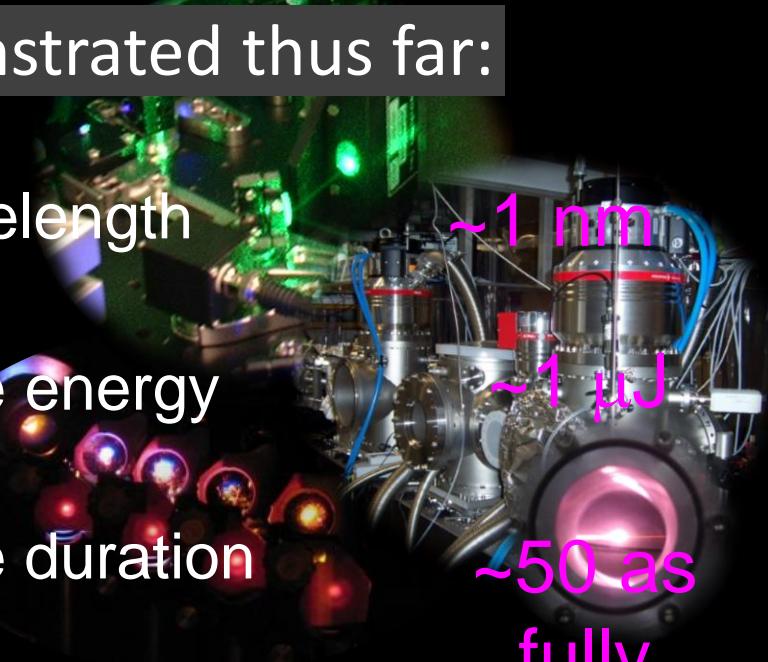
largest pulse energy

shortest pulse duration

~1 nm

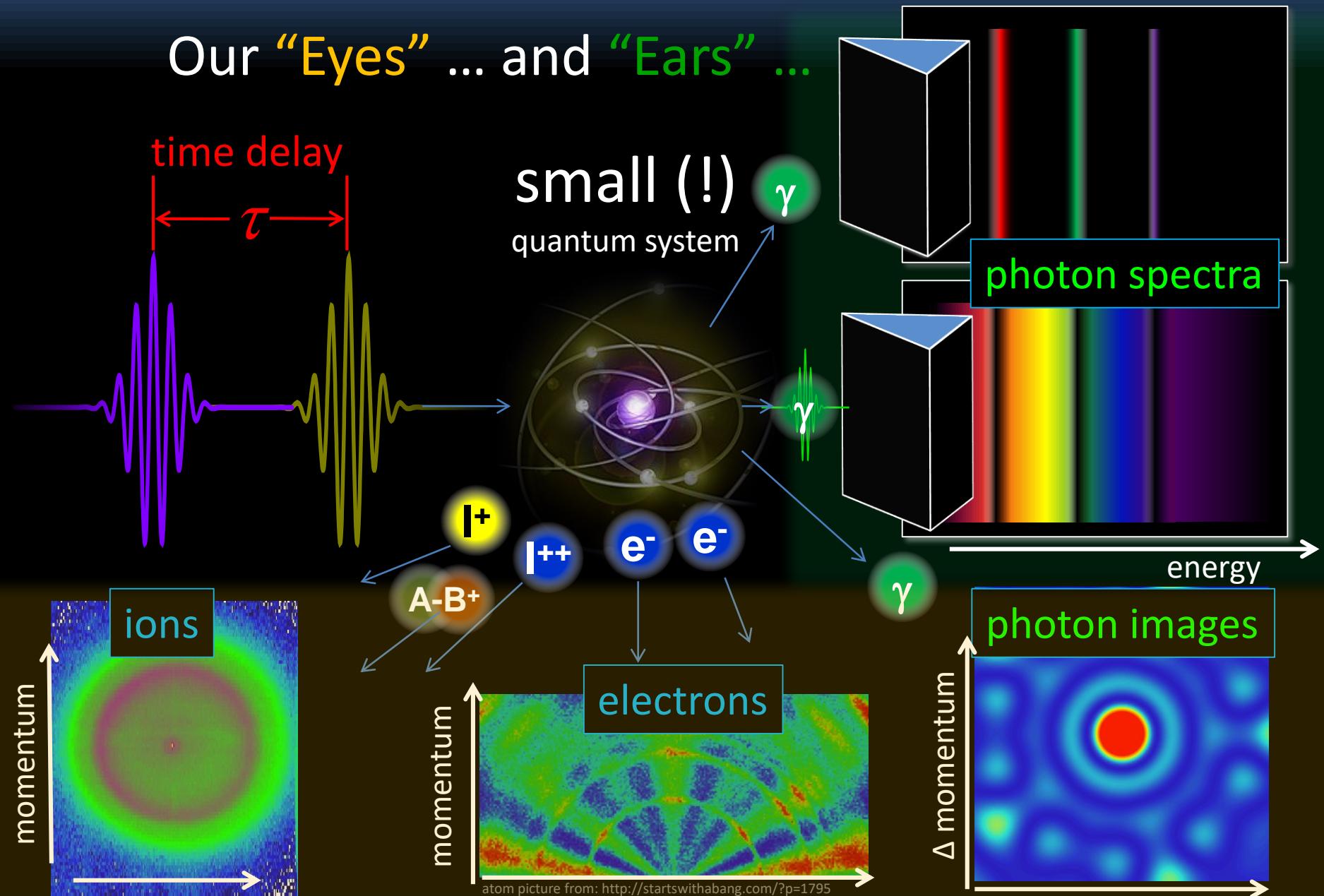
~1 μJ

~50 as
fully
coherent

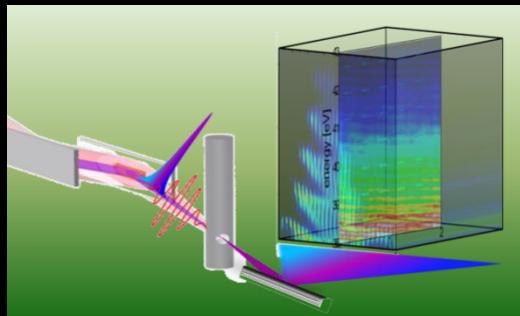
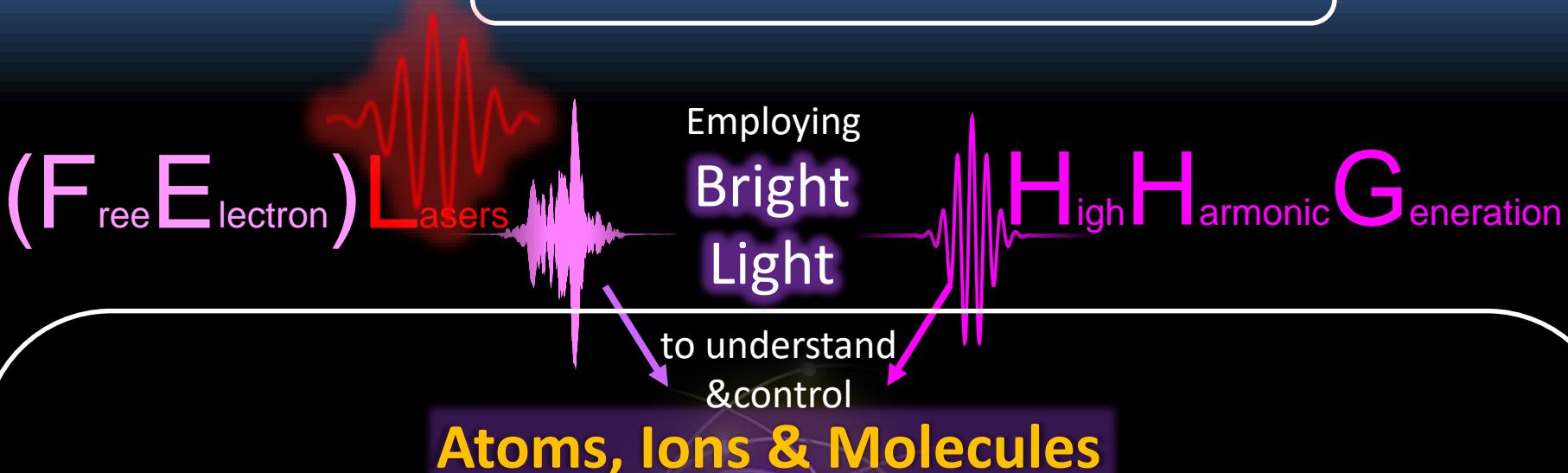


Quantum dynamics imaging and spectroscopy

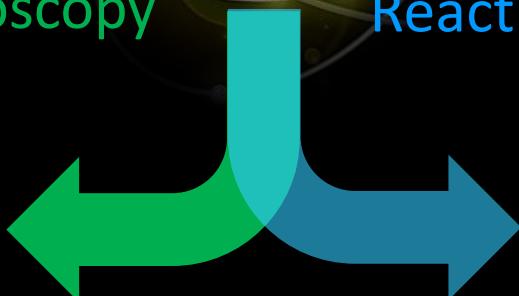
Our “Eyes” ... and “Ears” ...



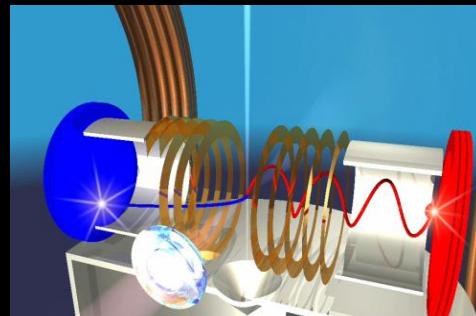
Our Experimental Focus



time&energy-resolved
detection of photons

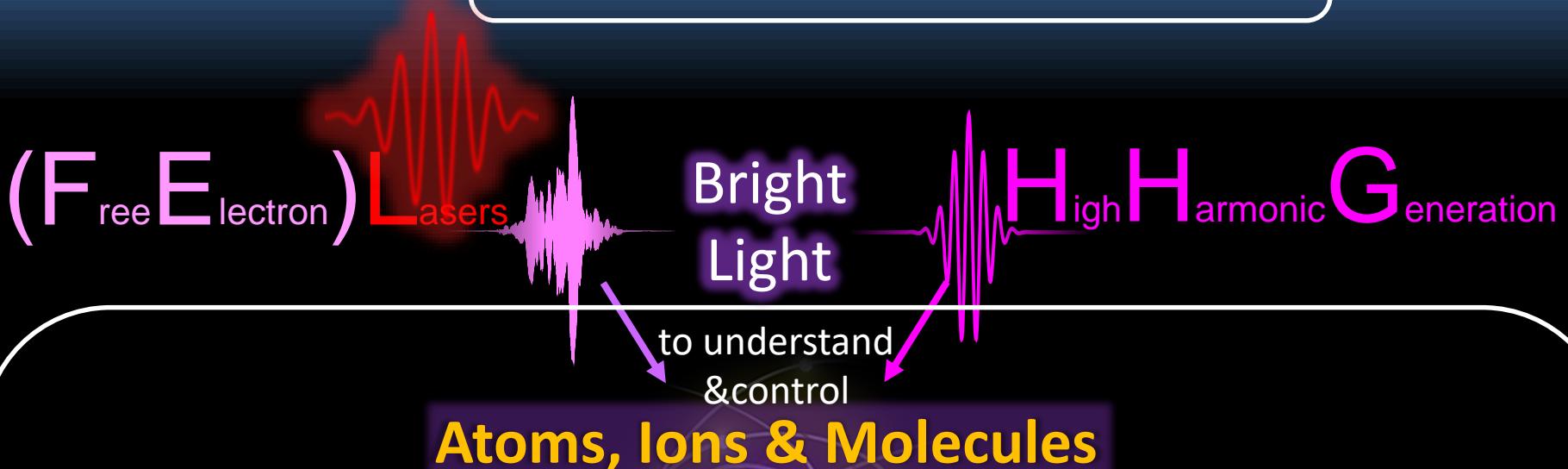


Goal: extract
complete
information



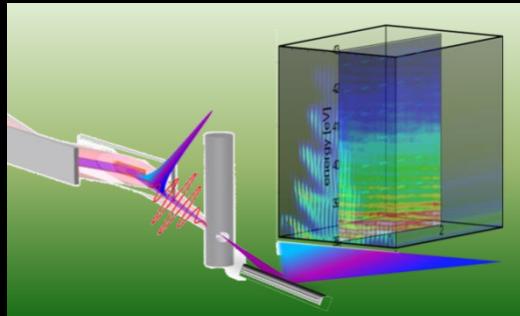
coincidence
detection of electrons/ions

Our Experimental Focus



"listening"

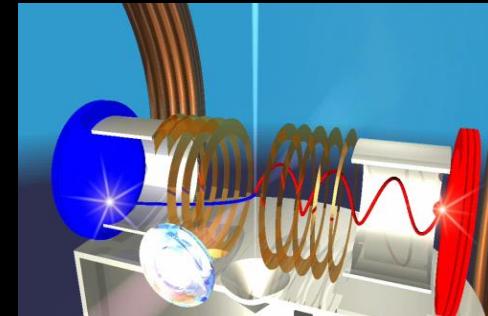
Multidim. optical spectroscopy



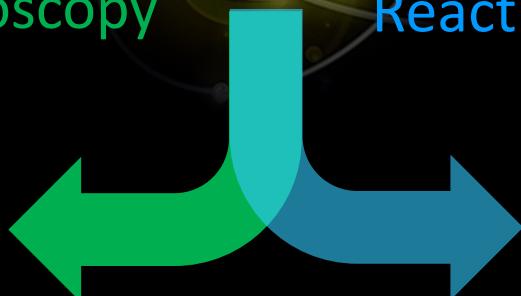
time&energy-resolved
detection of photons

"imaging"

Reaction microscope/COLTRIMS



coincidence
detection of electrons/ions

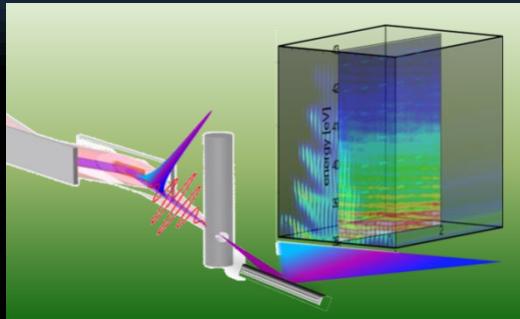


Goal: extract
complete
information

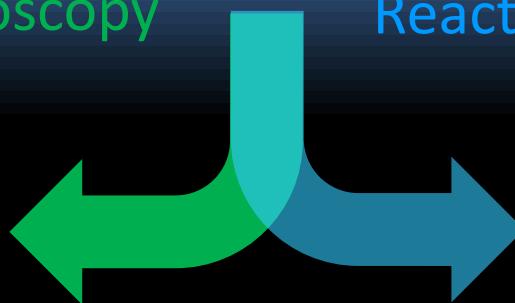
Experimental observation of few-body dynamics

Multidim. optical spectroscopy

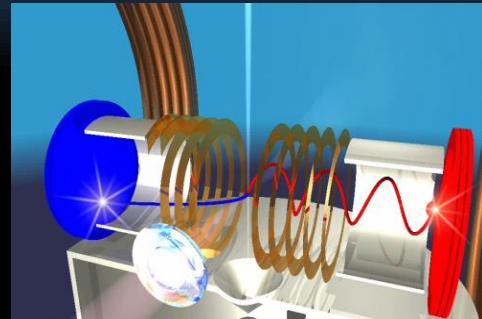
Reaction microscope/COLTRIMS



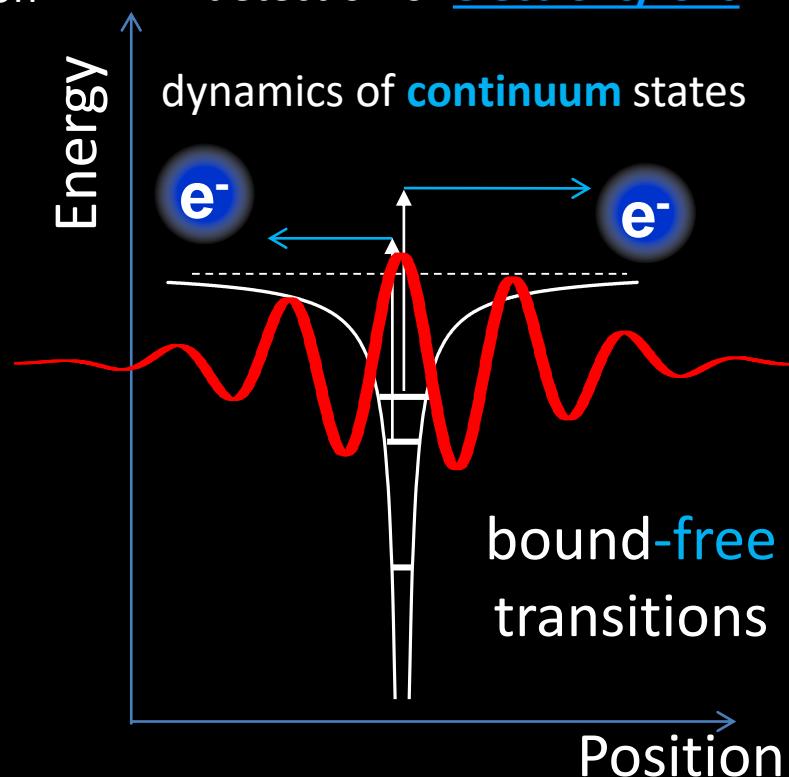
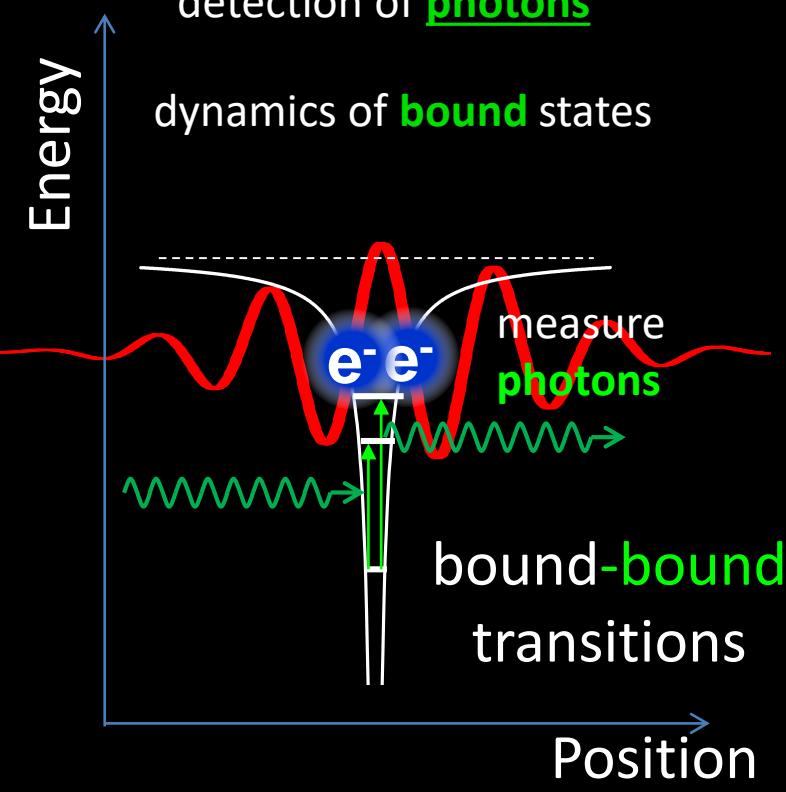
time&energy-resolved
detection of **photons**



Goal: extract
complete
information

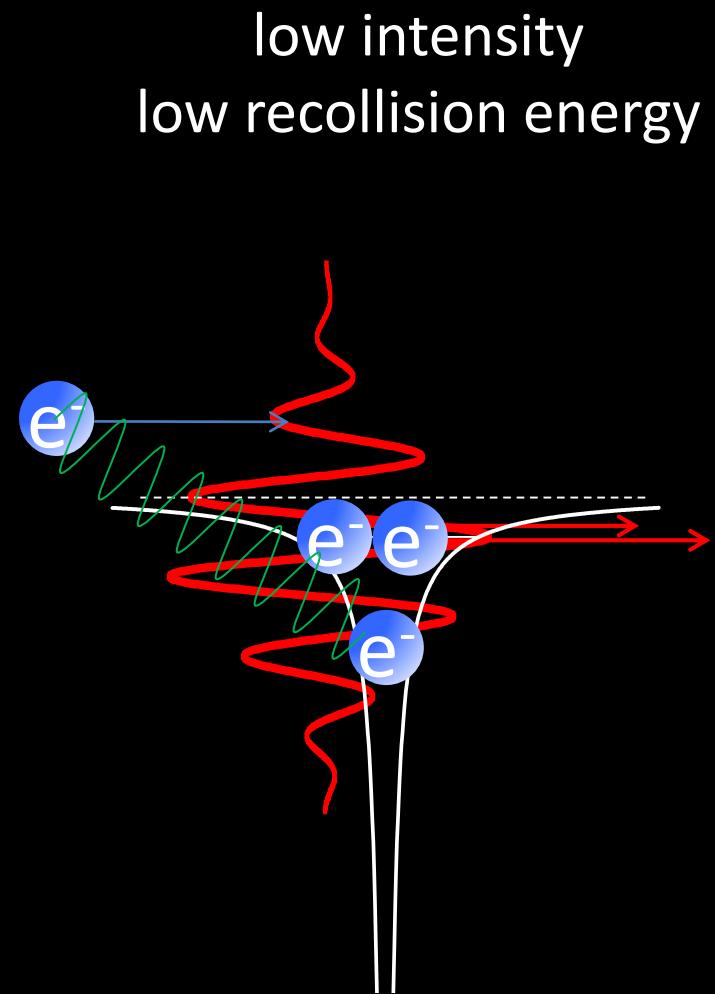
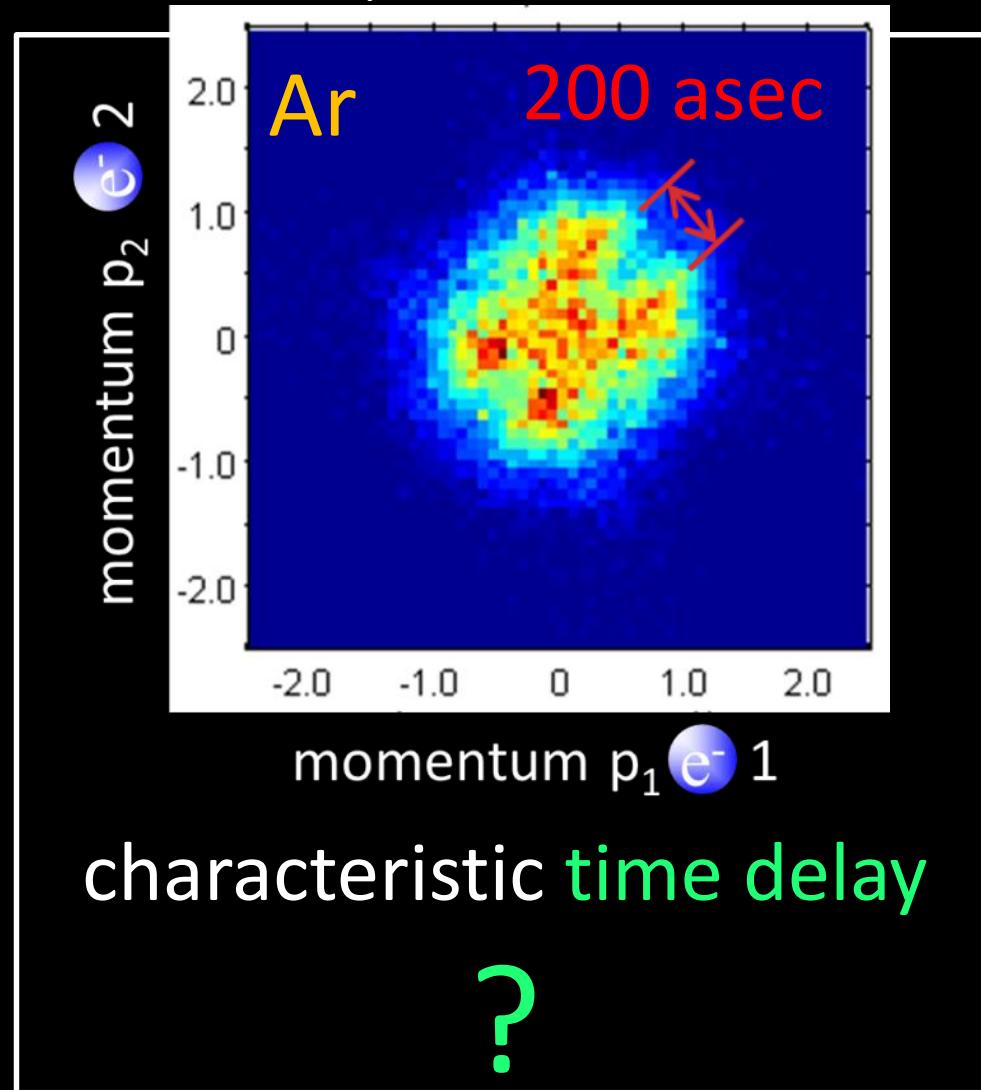


coincidence
detection of **electrons/ions**



Strong-field Recollision Physics in “nonsequential” double ionization

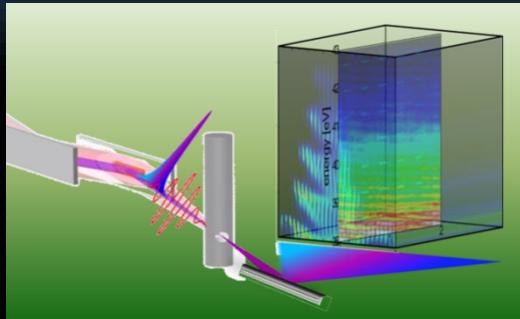
N. Camus *et al.* Phys. Rev. Lett. **108**, 073003



Experimental observation of few-body dynamics

Multidim. optical spectroscopy

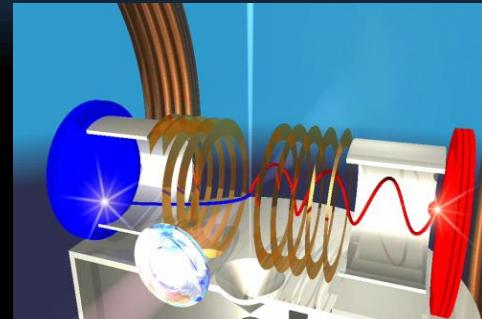
Reaction microscope/COLTRIMS



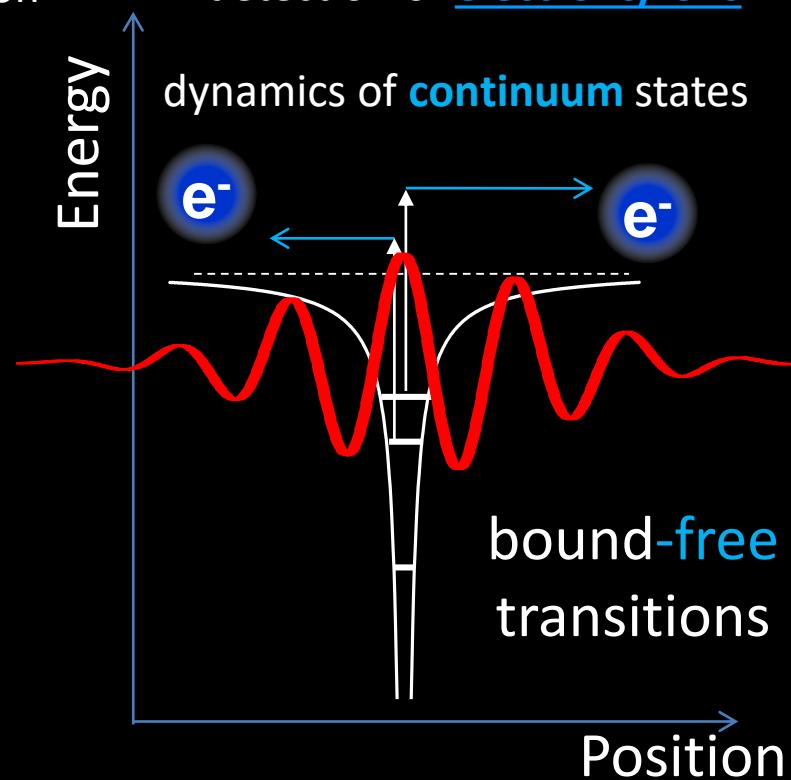
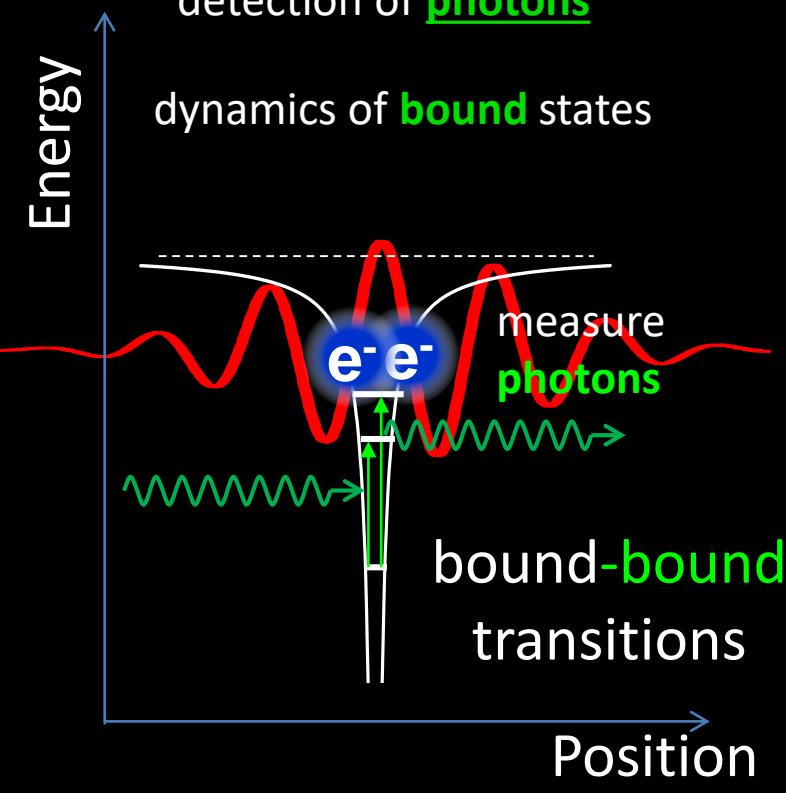
time&energy-resolved
detection of **photons**



Goal: extract
complete
information



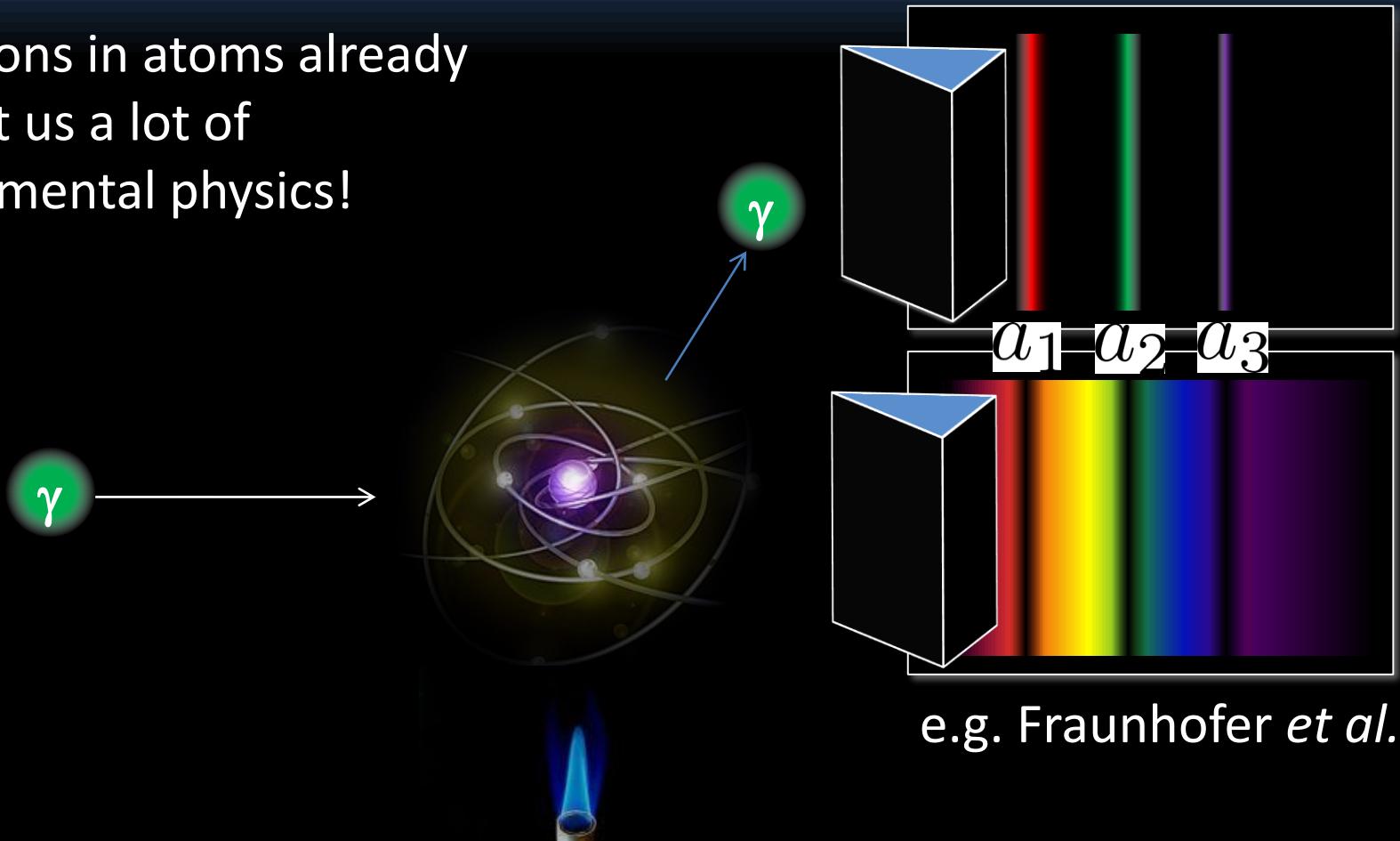
coincidence
detection of **electrons/ions**



traditional spectroscopy

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

Electrons in atoms already taught us a lot of fundamental physics!



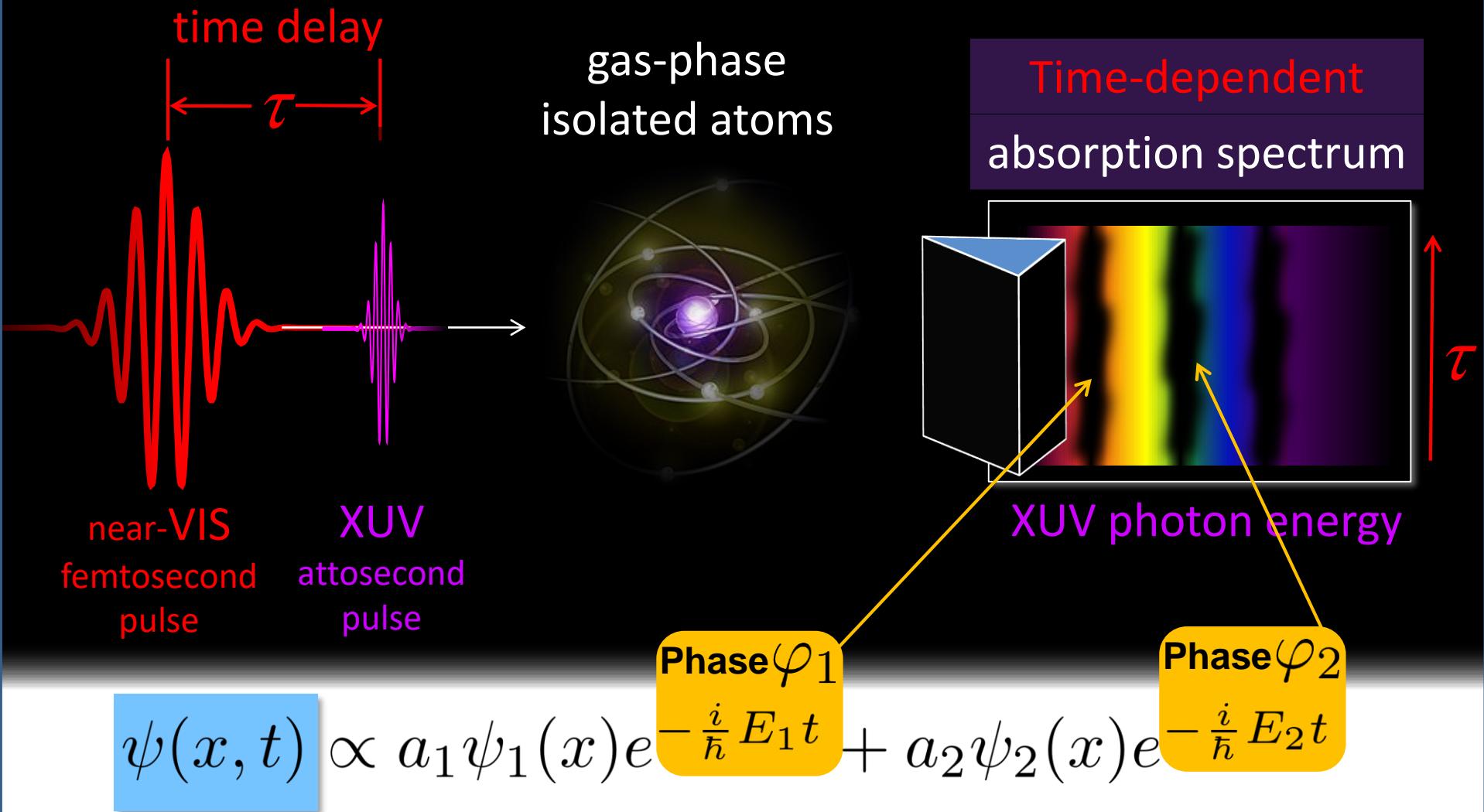
e.g. Fraunhofer *et al.*

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

Phase φ_1

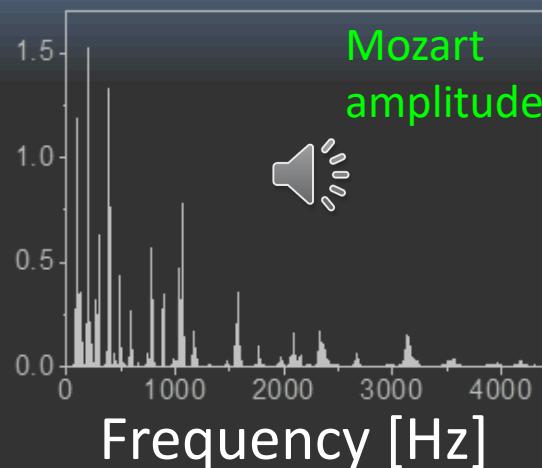
Phase φ_2

time-dependent absorption spectroscopy

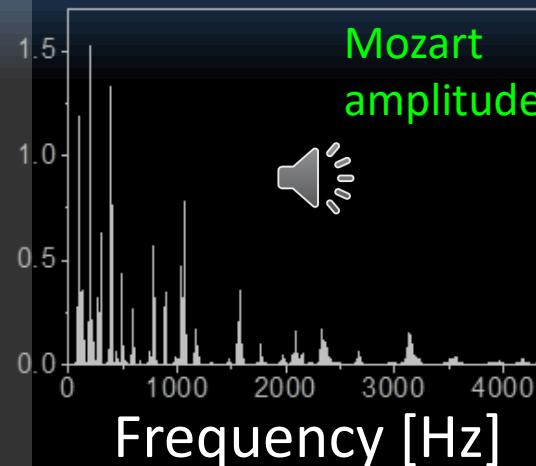


Listening to phases

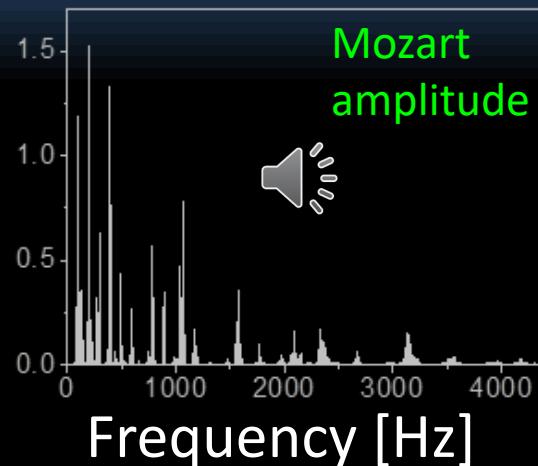
spectral intensity



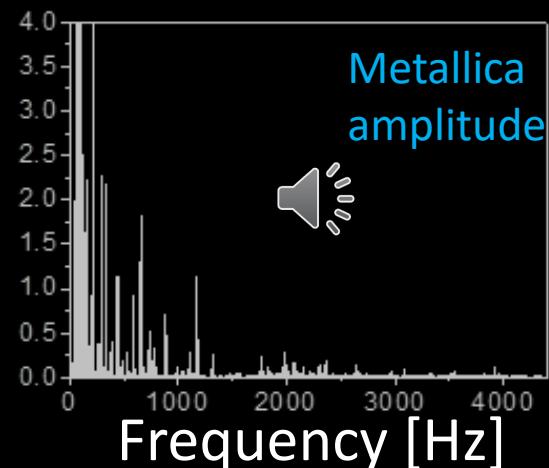
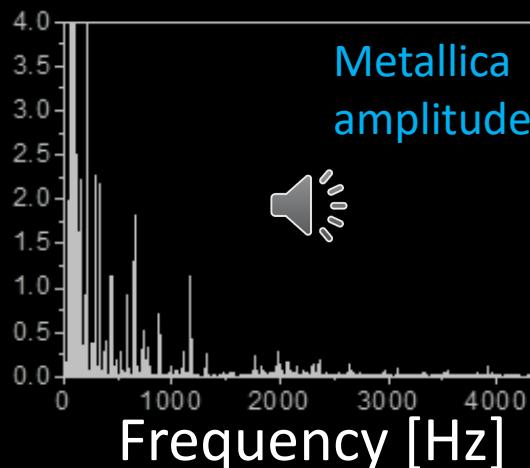
spectral phase by
by W. A. Mozart (1787)
(Eine kleine Nachtmusik)



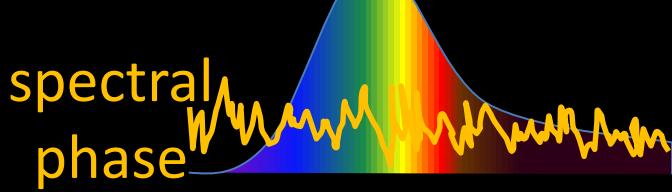
spectral phase
by Chance
(randomized)



spectral phase
by Metallica (1992)
(Nothing else matters)



Properties of different light sources

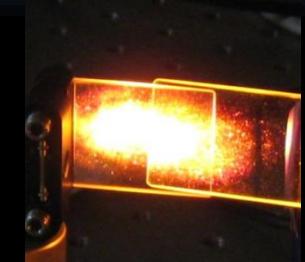


solar spectrum

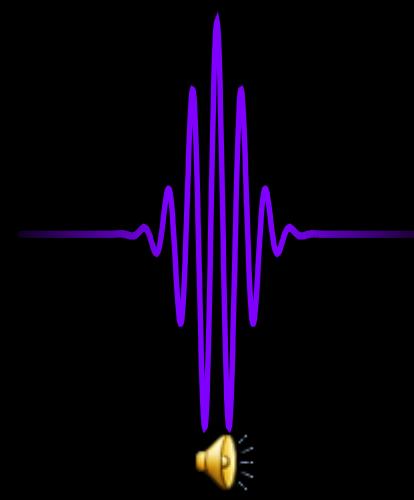
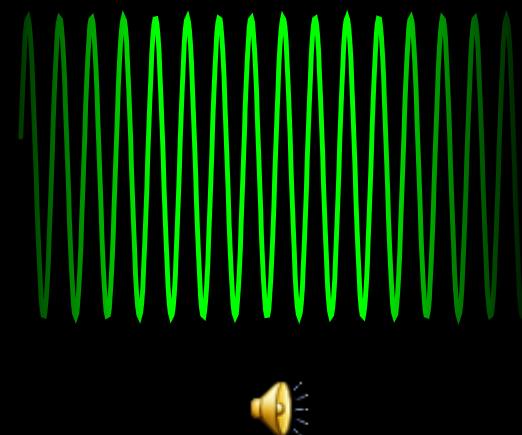
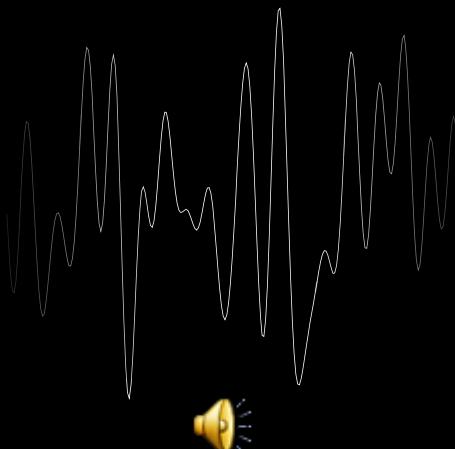


.

spectral
phase

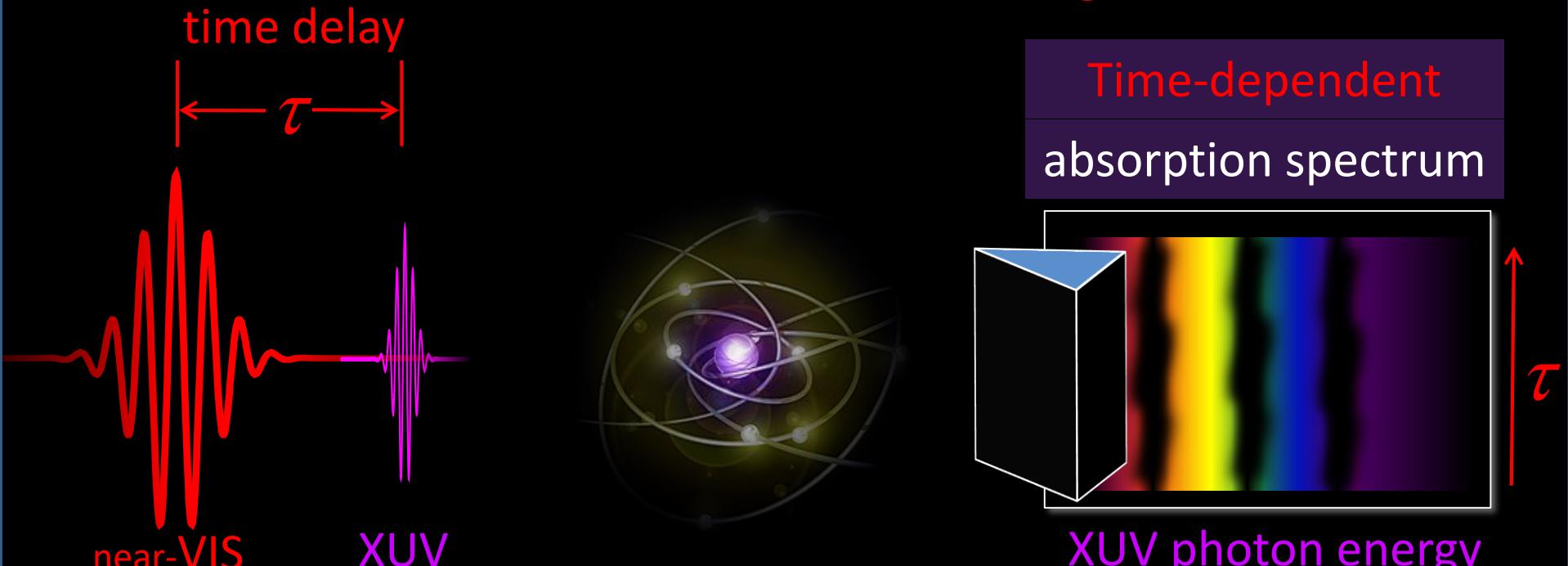


fs pulse laser spectrum



time-dependent absorption spectroscopy

What's the dynamics of bound states and resonances
in **short** and **strong** fields?



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

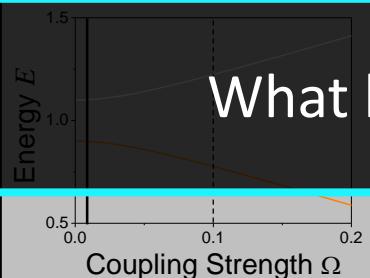
General: coupling of states

coupling of one to
one other state

coupling of one to
multiple states

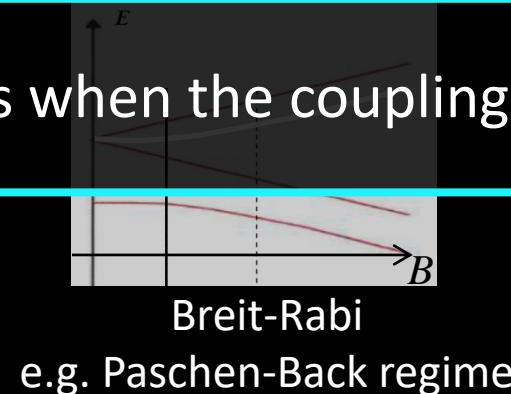
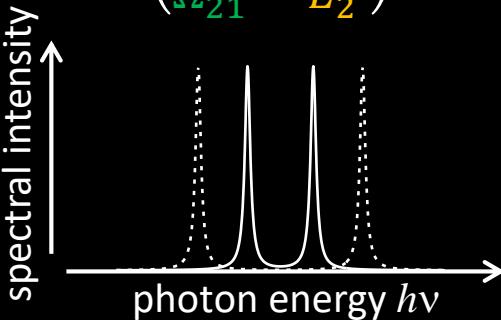
coupling of one to a
continuum of states

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



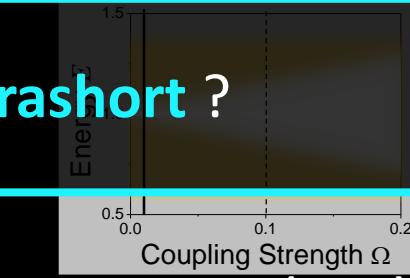
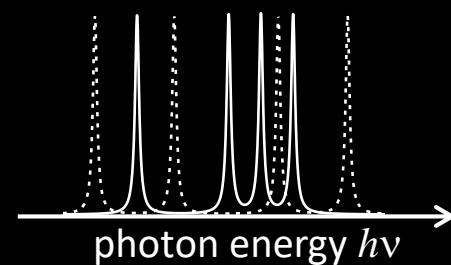
Rabi oscillation
in strong coupling

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



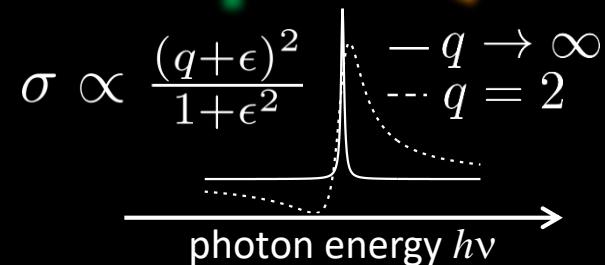
Breit-Rabi
e.g. Paschen-Back regime

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



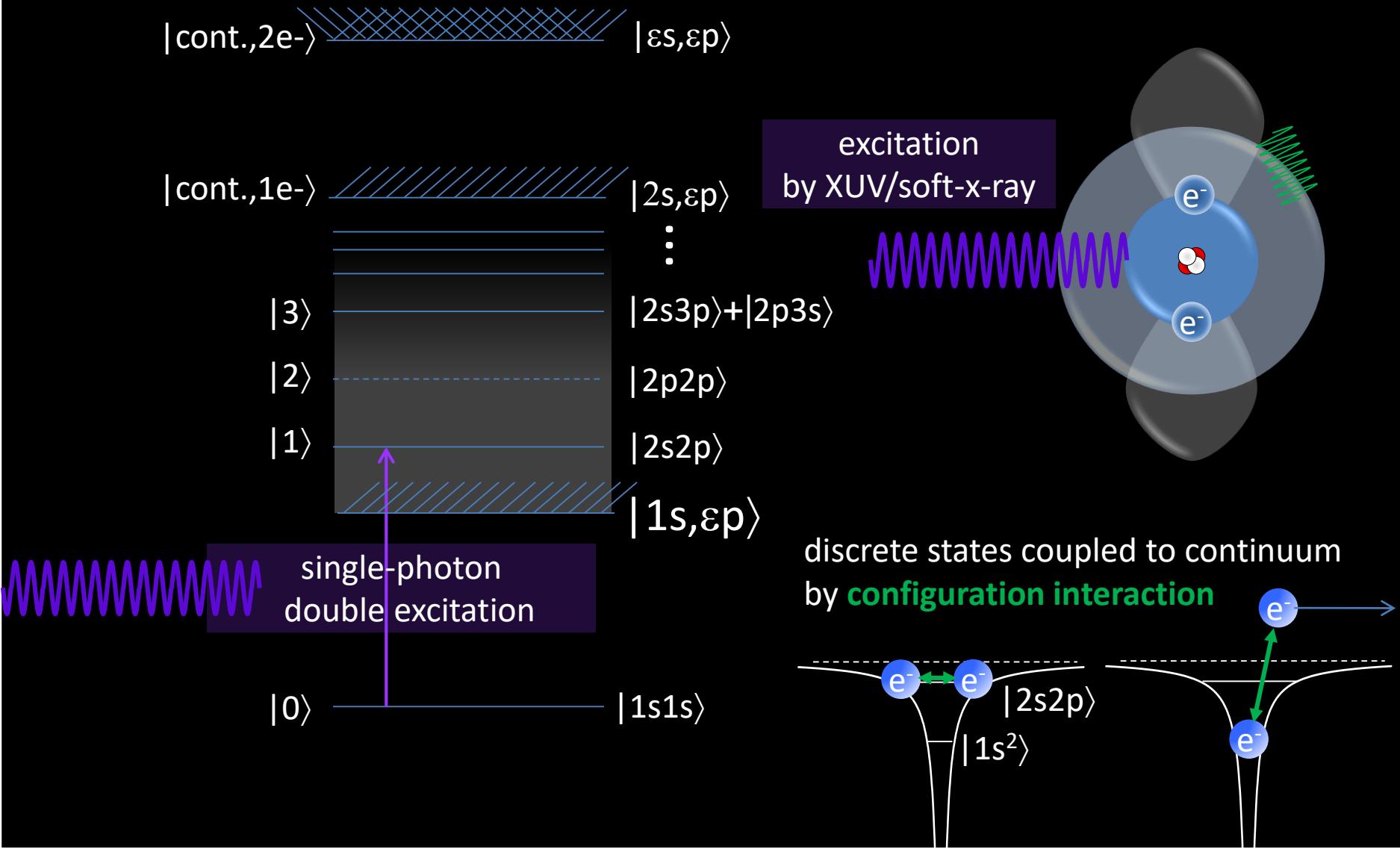
U. Fano (1935)
Phys. Rev. **124**, 1866 (1961)
Nuovo Cim. **12**, 154 (1935)

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

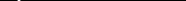


fundamental e^- - e^- correlation in atoms

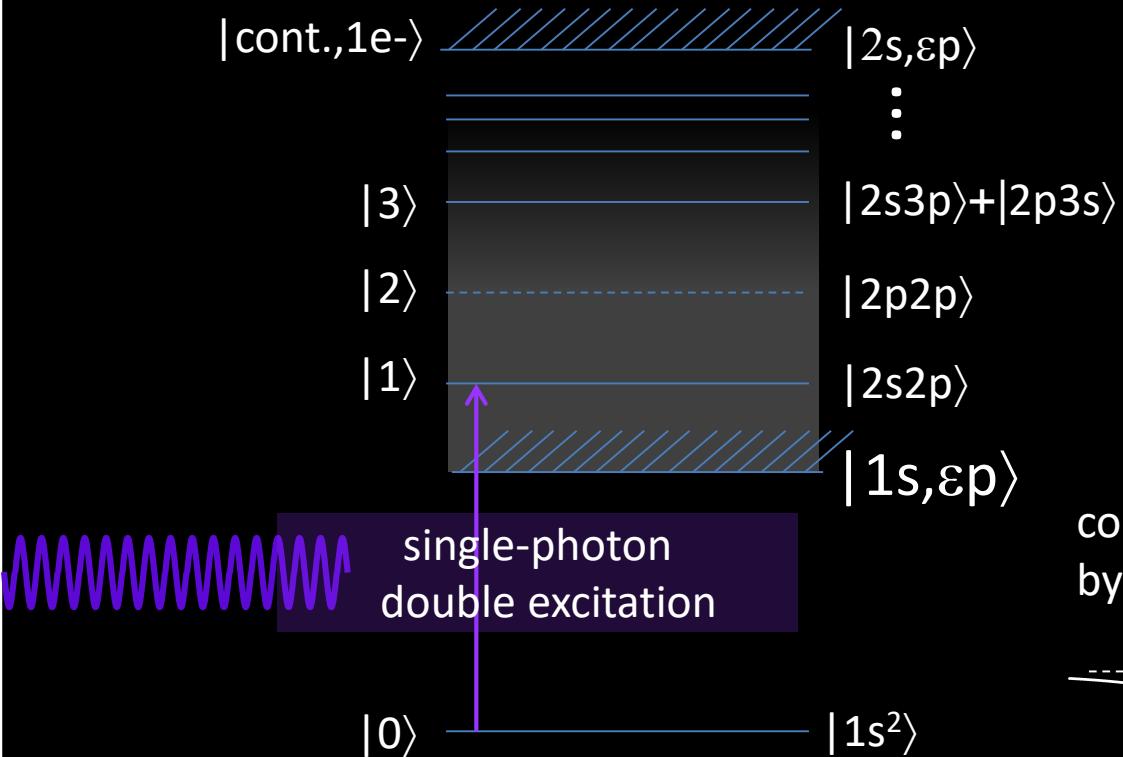
prototypical target: He atom



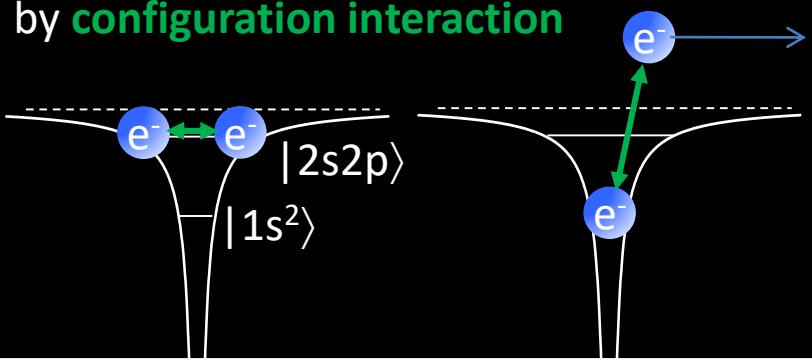
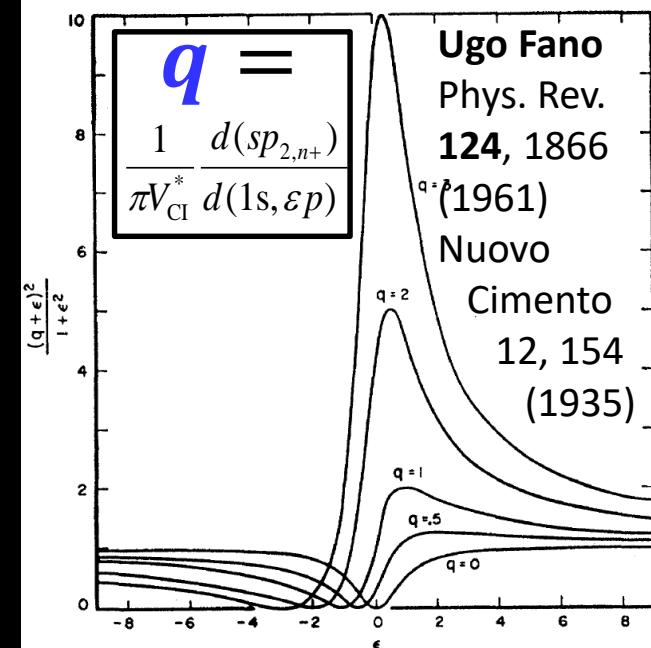
doubly excited helium: Fano resonance

$|\text{cont.,}2e-\rangle$  $|\varepsilon s,\varepsilon p\rangle$

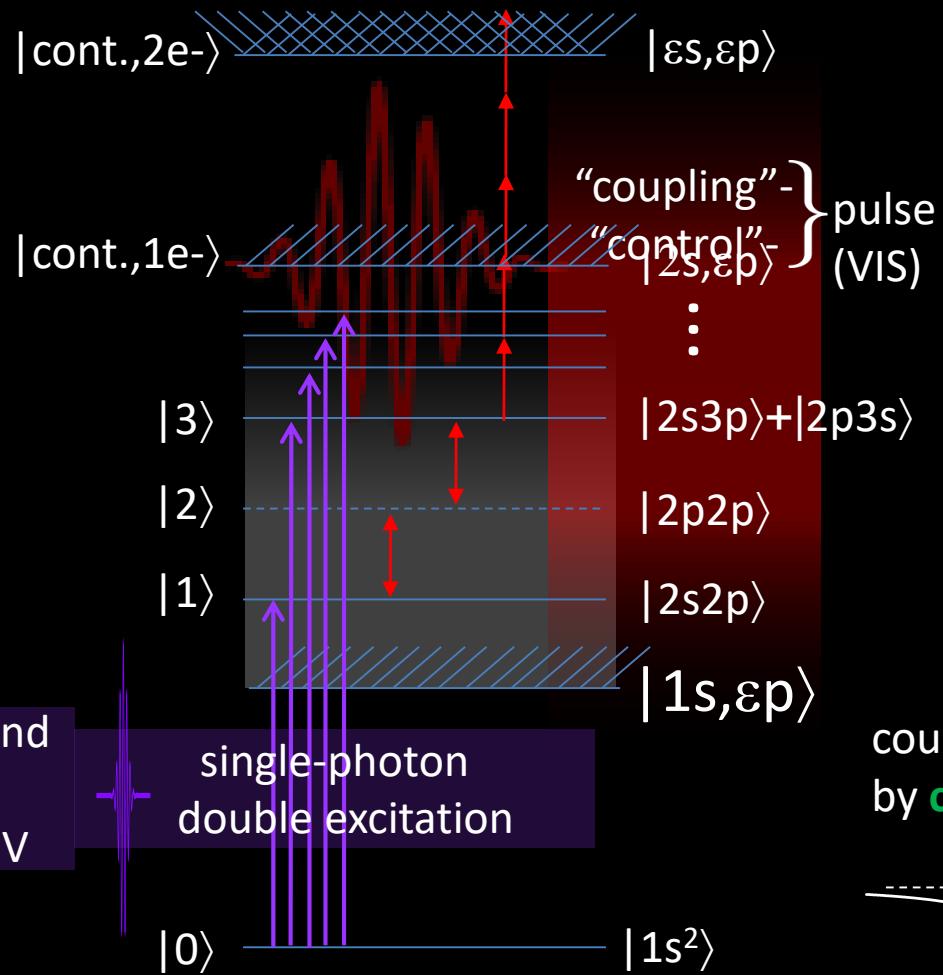
$$\sigma_{Fano} \sim \frac{(q + \varepsilon)^2}{1 + \varepsilon^2}$$



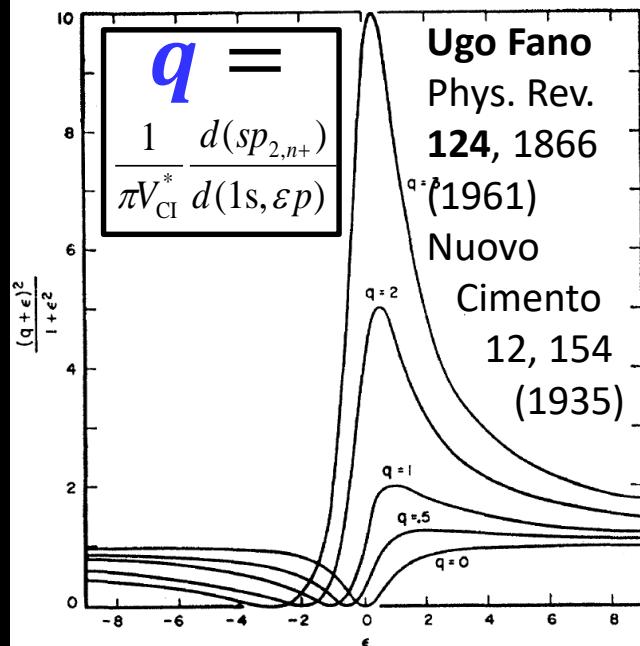
coupled by configuration interaction



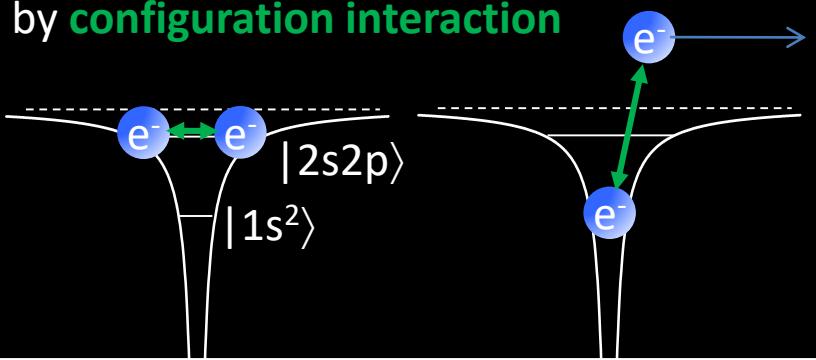
doubly-excited helium, in a strong laser field



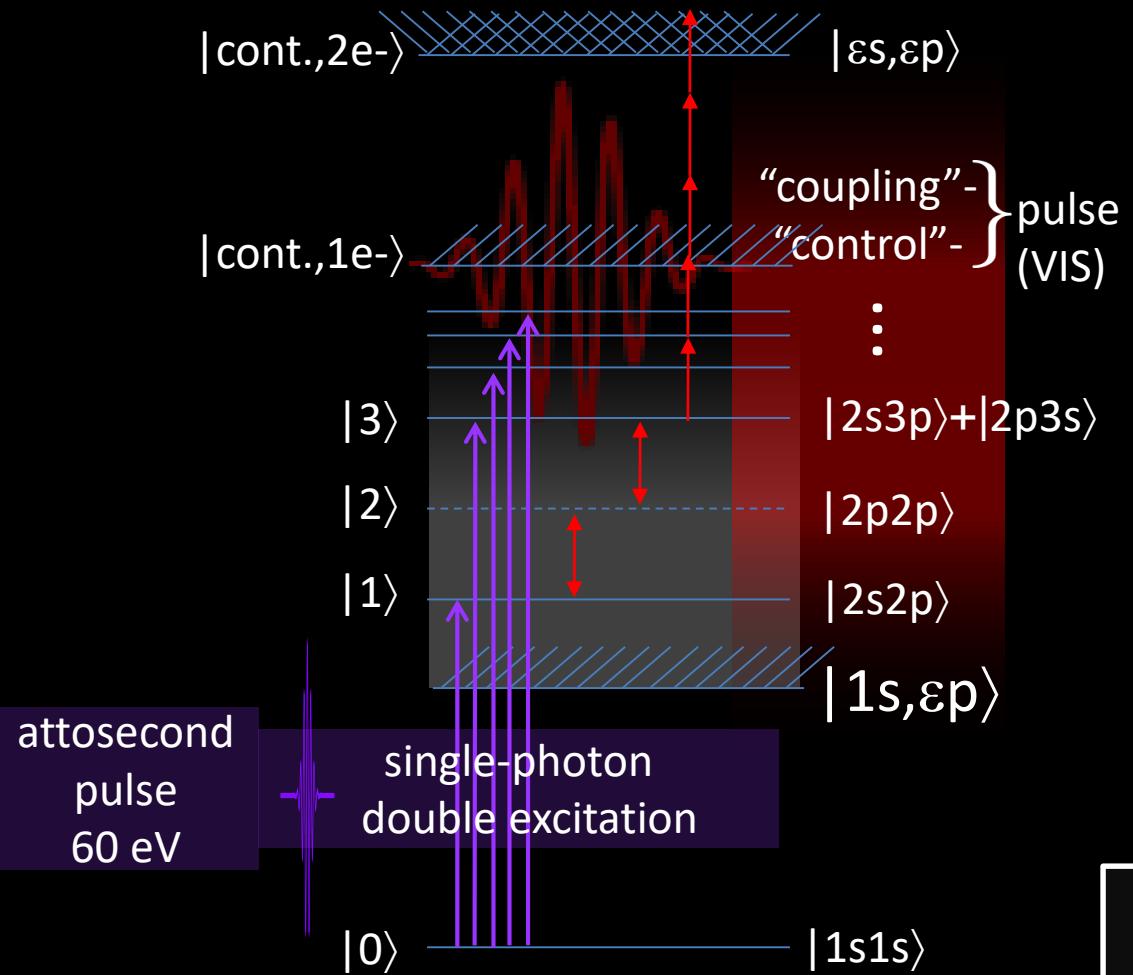
$$\sigma_{Fano} \sim \frac{(\mathbf{q} + \boldsymbol{\varepsilon})^2}{1 + \boldsymbol{\varepsilon}^2}$$



coupled by **configuration interaction**



doubly-excited helium, in a strong laser field



Previous work

(on laser coupling of doubly-excited helium):

Theory:

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

e.g. Phys. Rev. A 24, 379 (1981)

e.g. Phys. Rev. A 87, 013415 (2013)

Experiment:

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

Chem. Phys. 350, 7 (2008)

Phys. Rev. Lett. 105, 263003 (2010)

Experimental challenge:

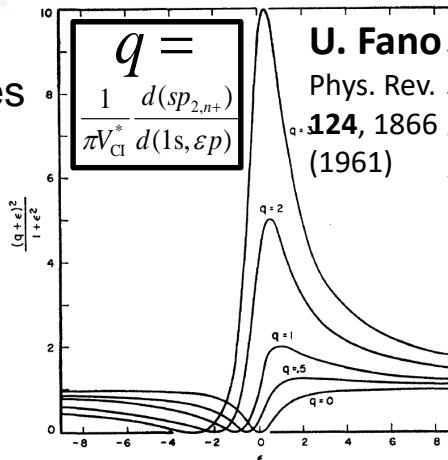
- high (asec) temporal and
- high (meV) **spectral resolution**
required at the same time

Experimental setup

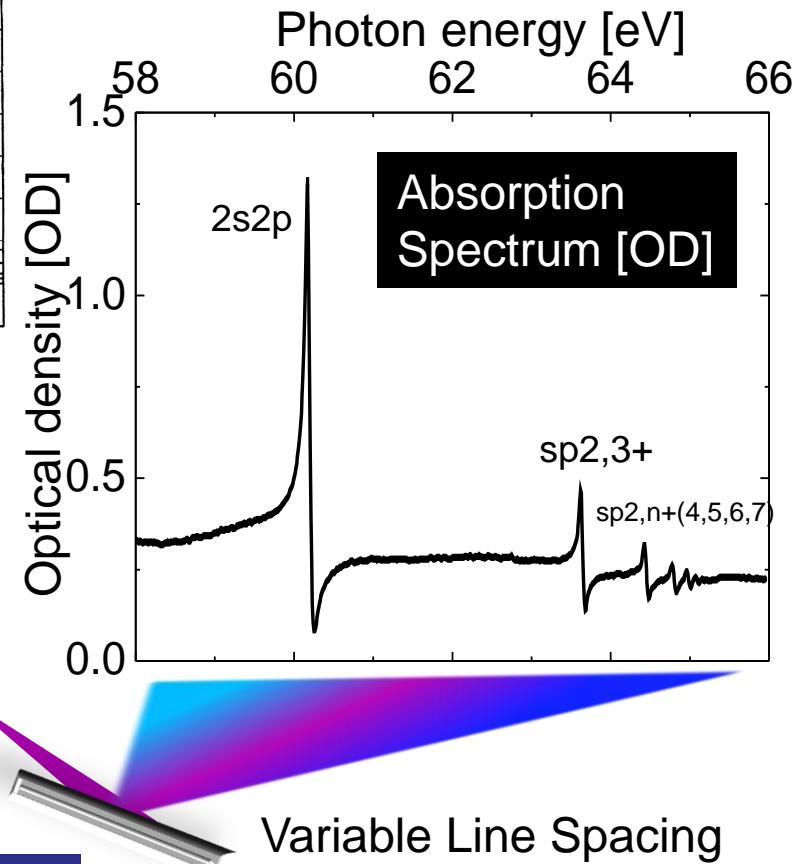
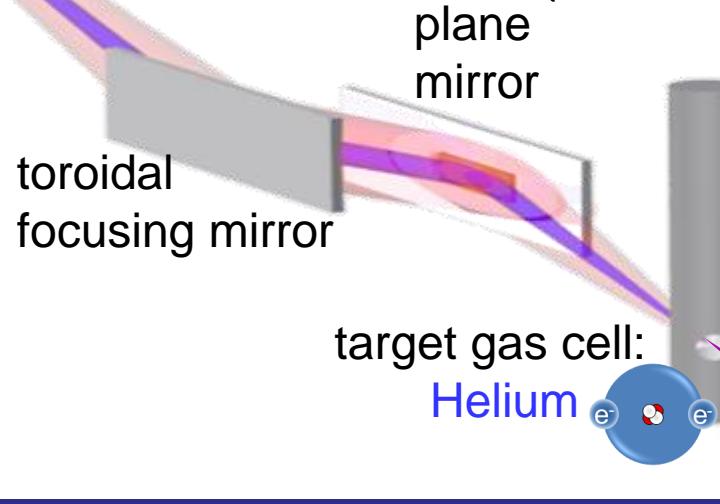
for

XUV absorption spectroscopy

Fano resonances
of autoionizing states



XUV
light

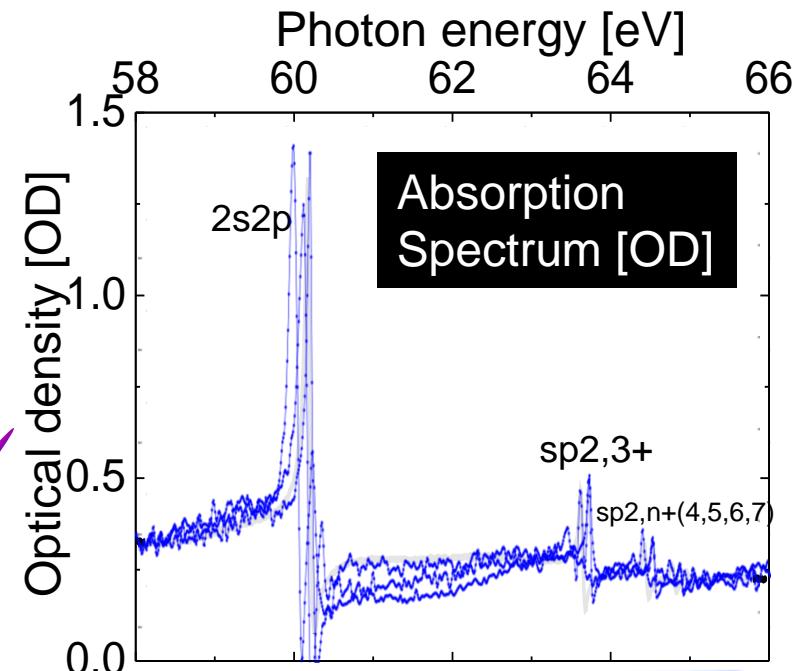
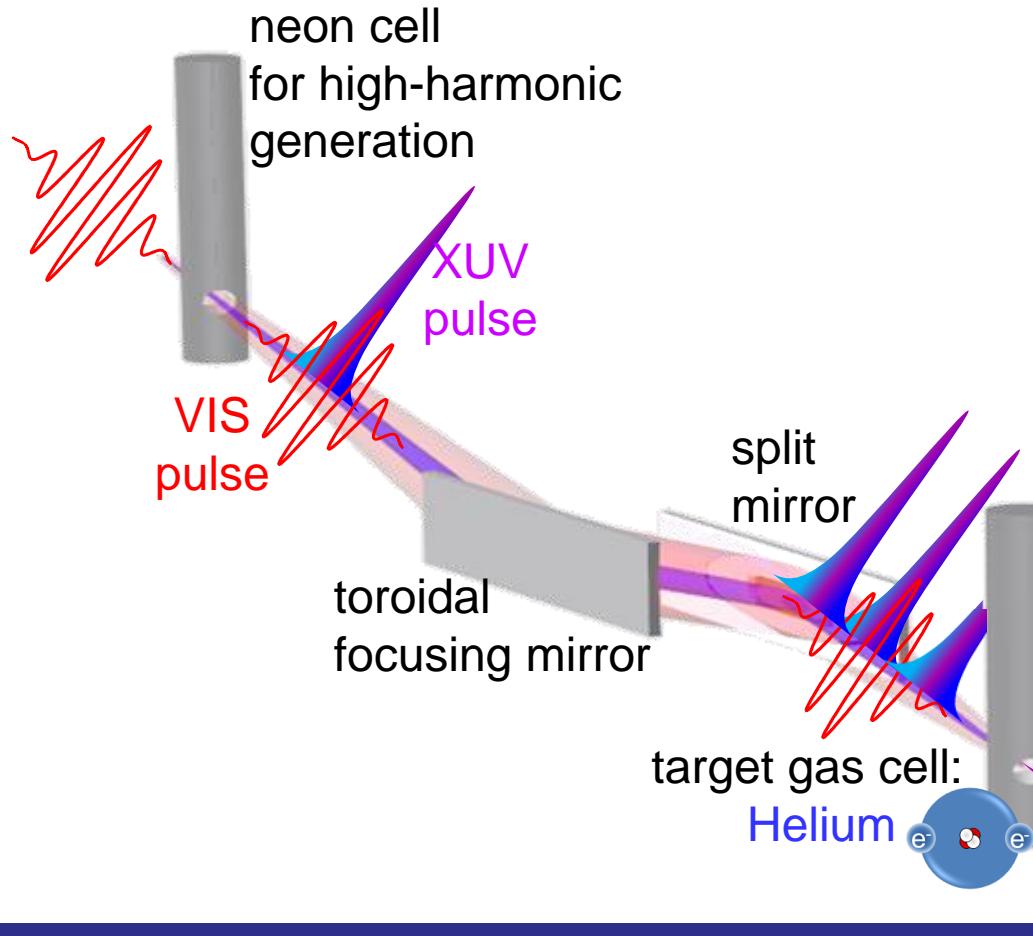


Experimental key challenge: High resolution
In photon energy ... : $\Delta E = 20 \text{ meV} (@60 \text{ eV})$

Variable Line Spacing
(VLS) grating
flat-field spectrometer

Experimental setup

for time-resolved XUV absorption spectroscopy

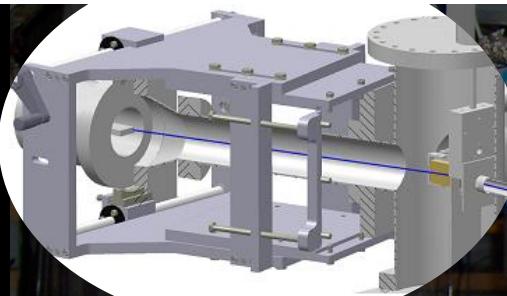


Experimental key challenge: High resolution
In photon energy ... : $\Delta E = 20 \text{ meV}$ (@60 eV)
... and time delay : $\Delta \tau = 10 \text{ as}$

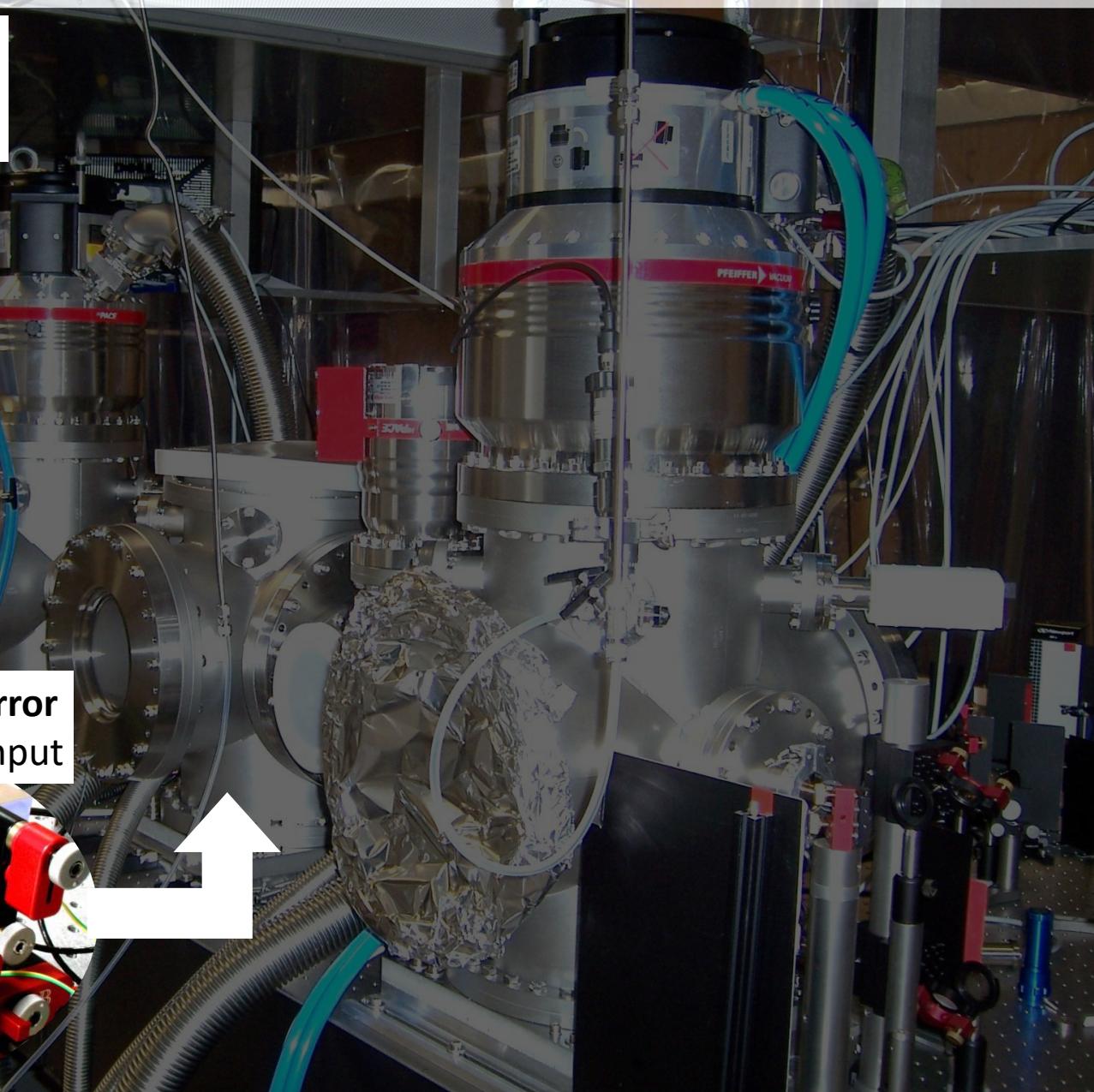
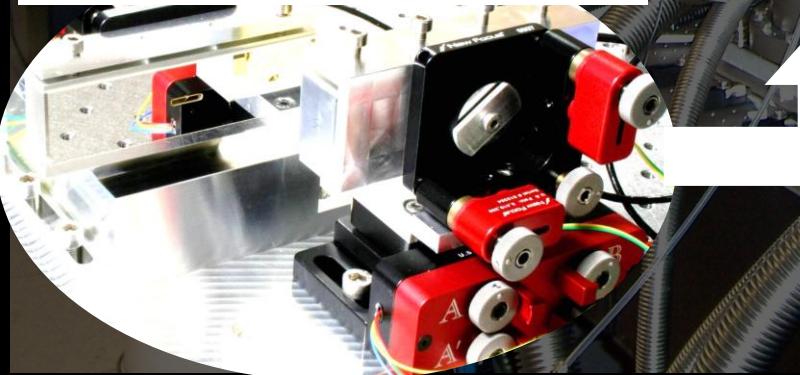
Variable Line Spacing
(VLS) grating
flat-field spectrometer

Experimental Setup in the Lab

Flat-Field XUV Spectrometer,
home built,
for broadband high resolution

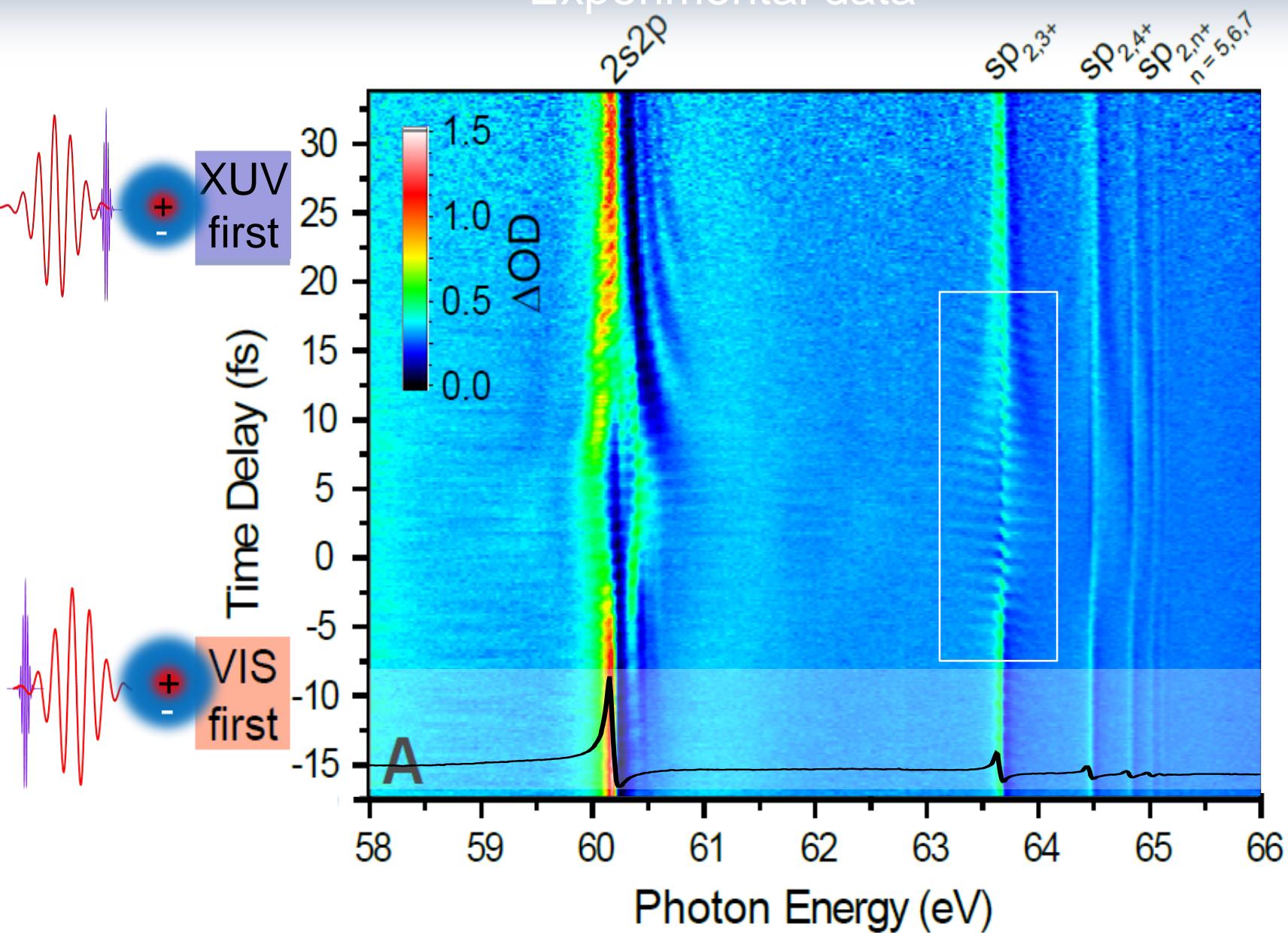


Grazing-Incidence Split Mirror
for broadband XUV throughput

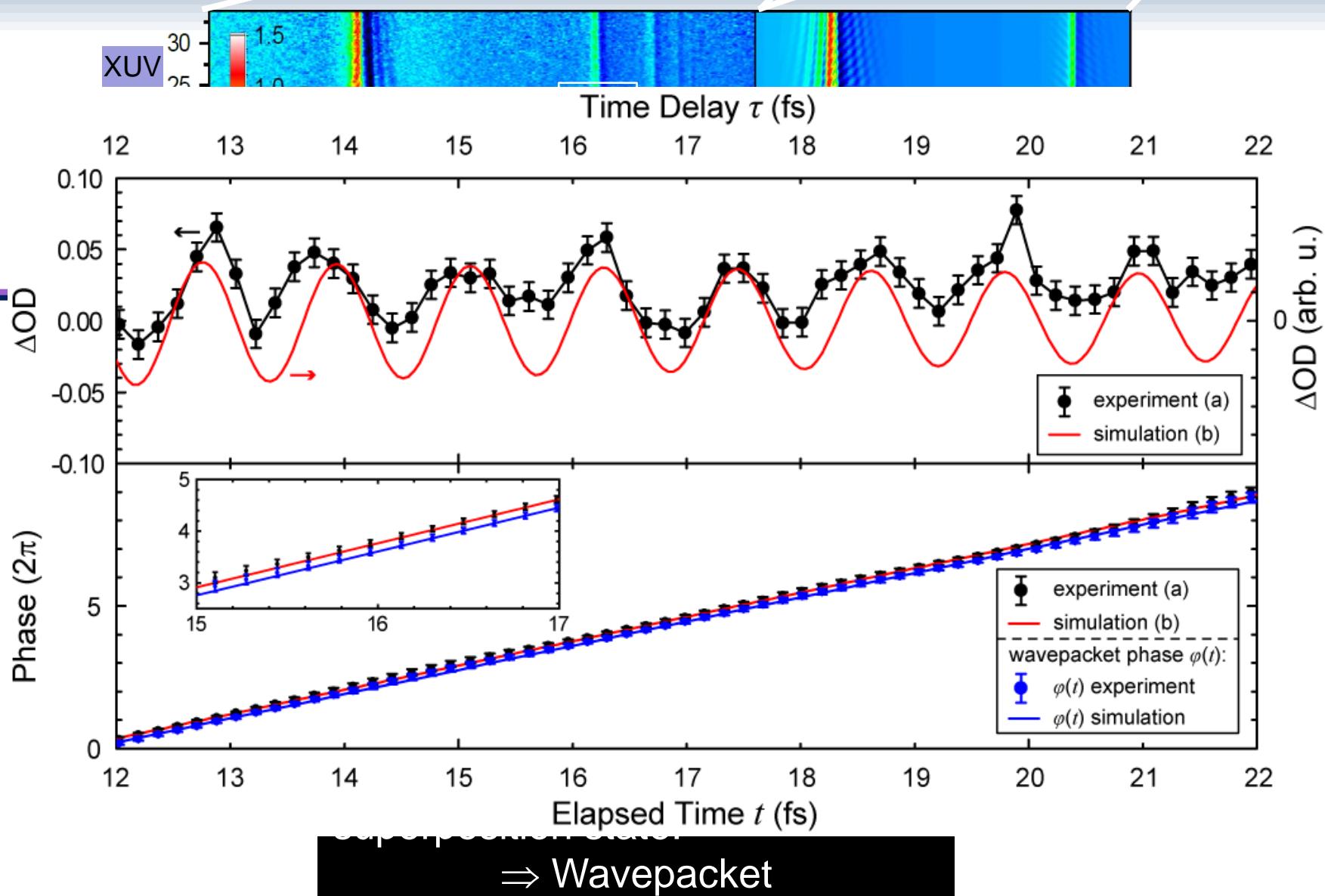


Time-resolved doubly-excited 2e⁻ dynamics in He

Experimental data

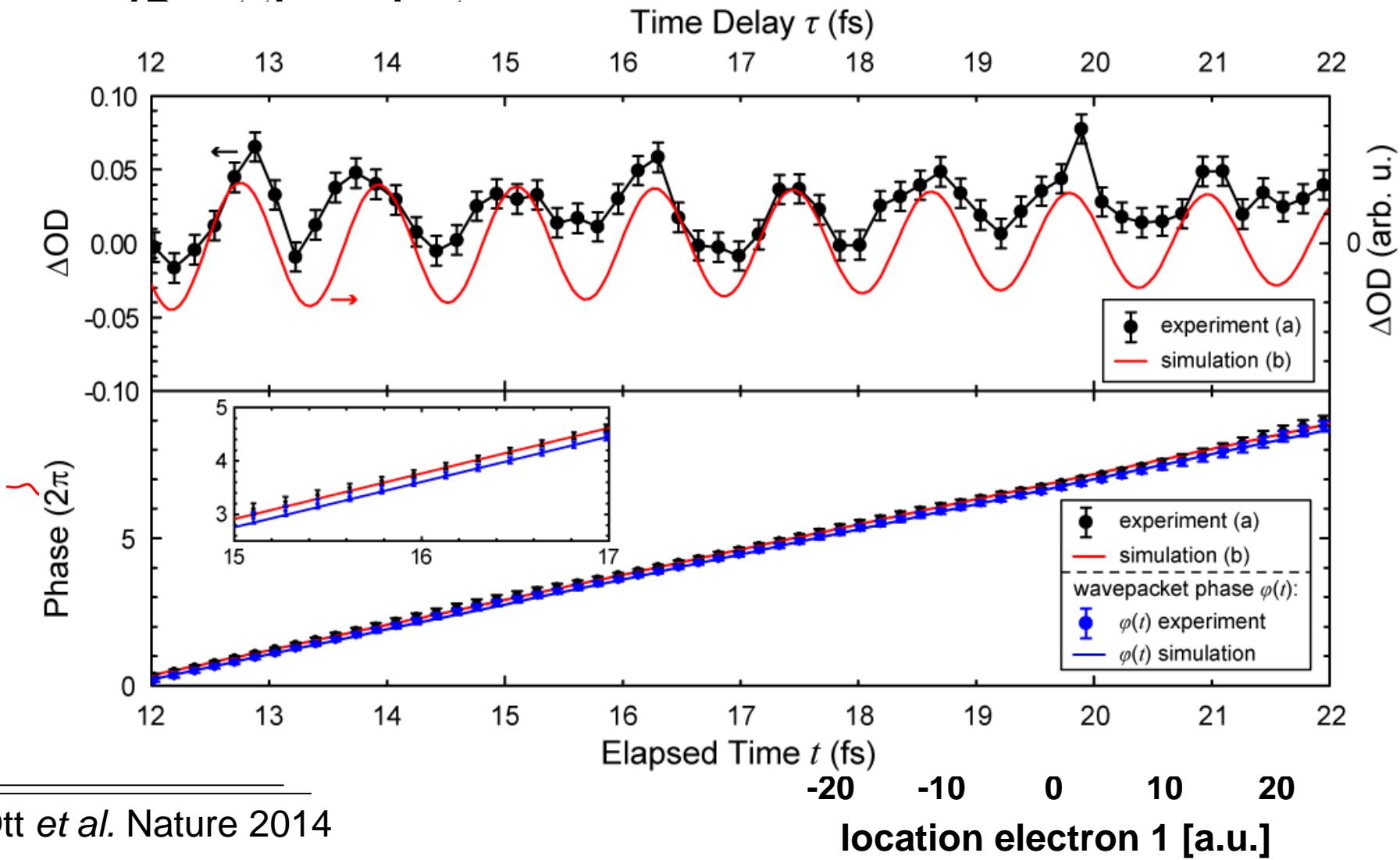


comparison experiment and theory



Measuring the time-dependent phase difference of 2s2p and sp₂₃₊ autoionization states

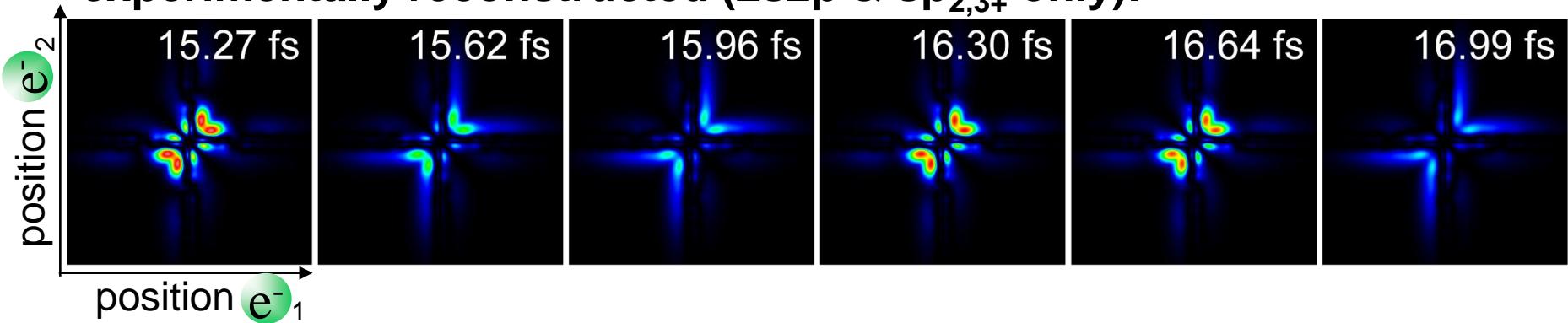
cooperation with Javier Madroñero (Theory, TU München)



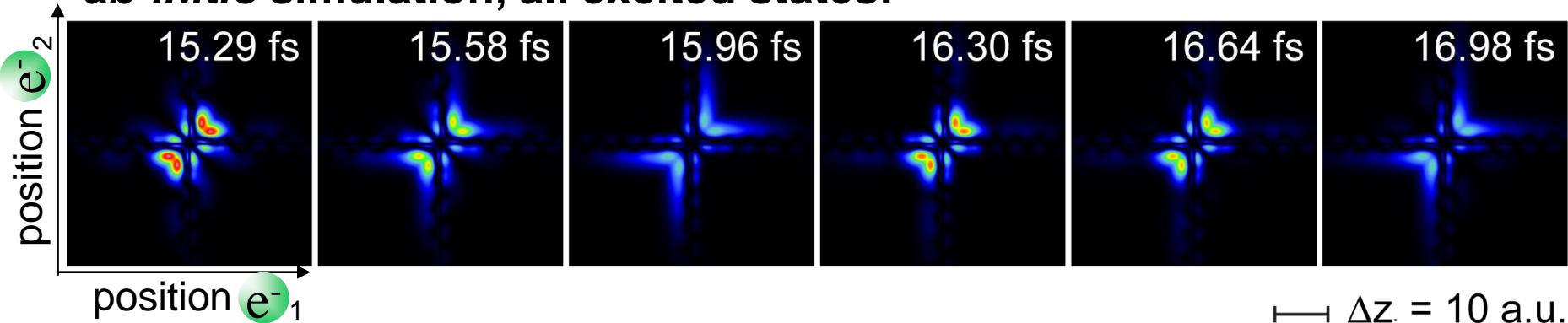
Testing *ab-initio* theory of e⁻ correlation dynamics

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

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

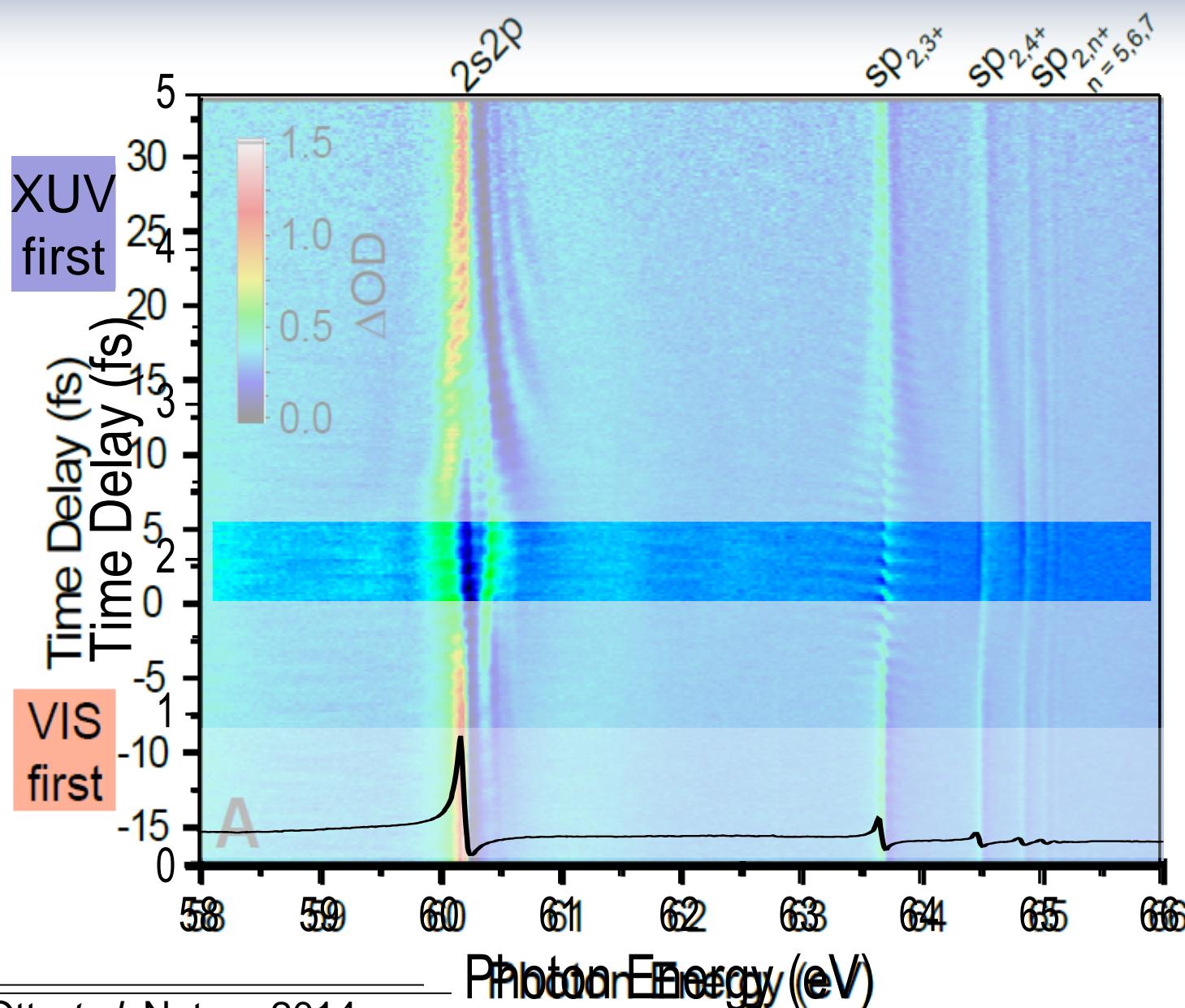


***ab-initio* simulation, all excited states:**

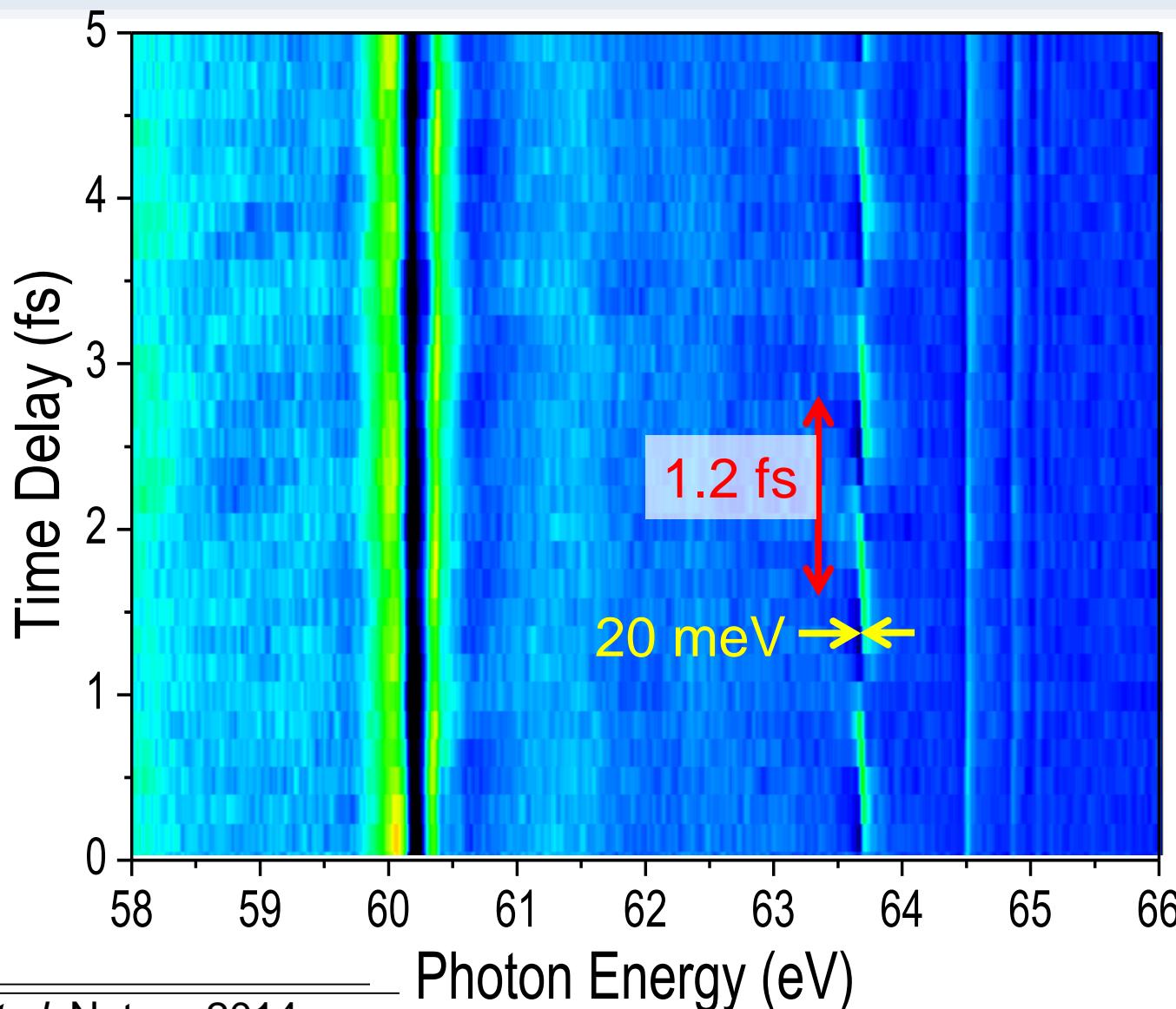


first experimental observation
of **two-electron wavepacket** motion

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



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



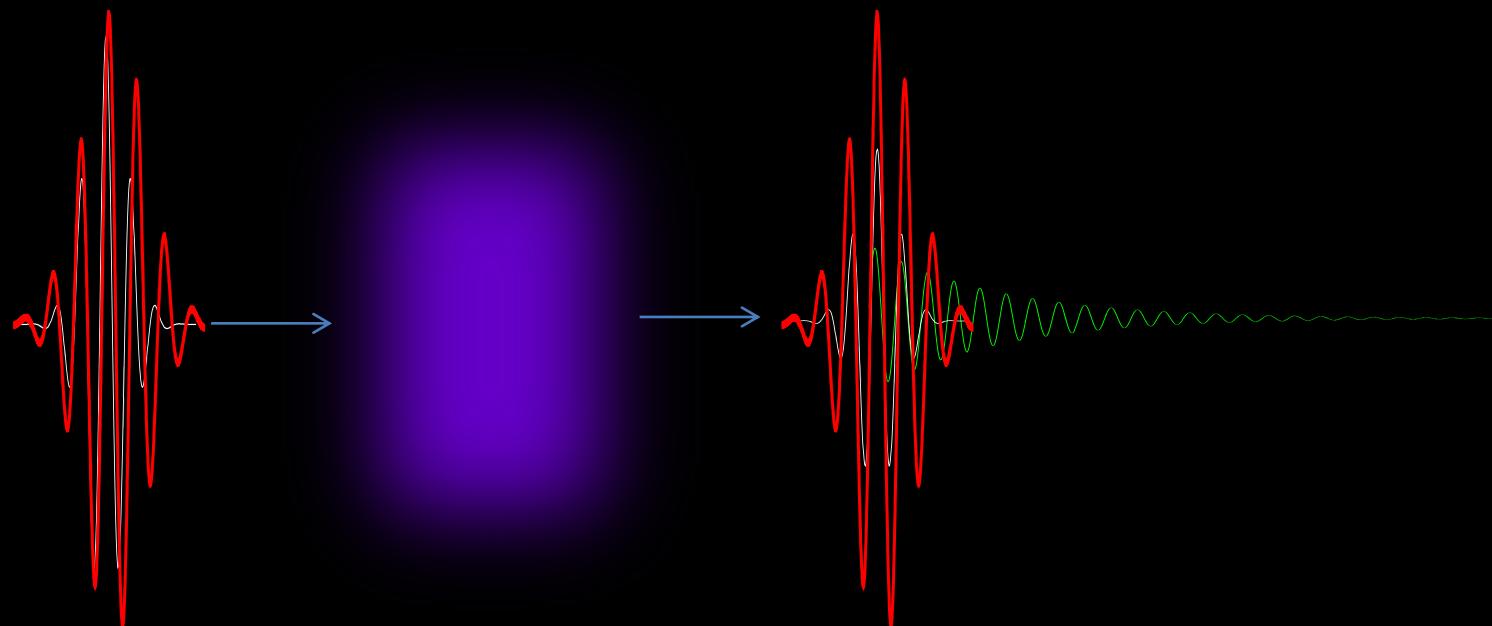
high **temporal**
and
high **spectral**
resolution
are
required
simultaneously

Hold on, can I ask you a few questions?



What is absorption?

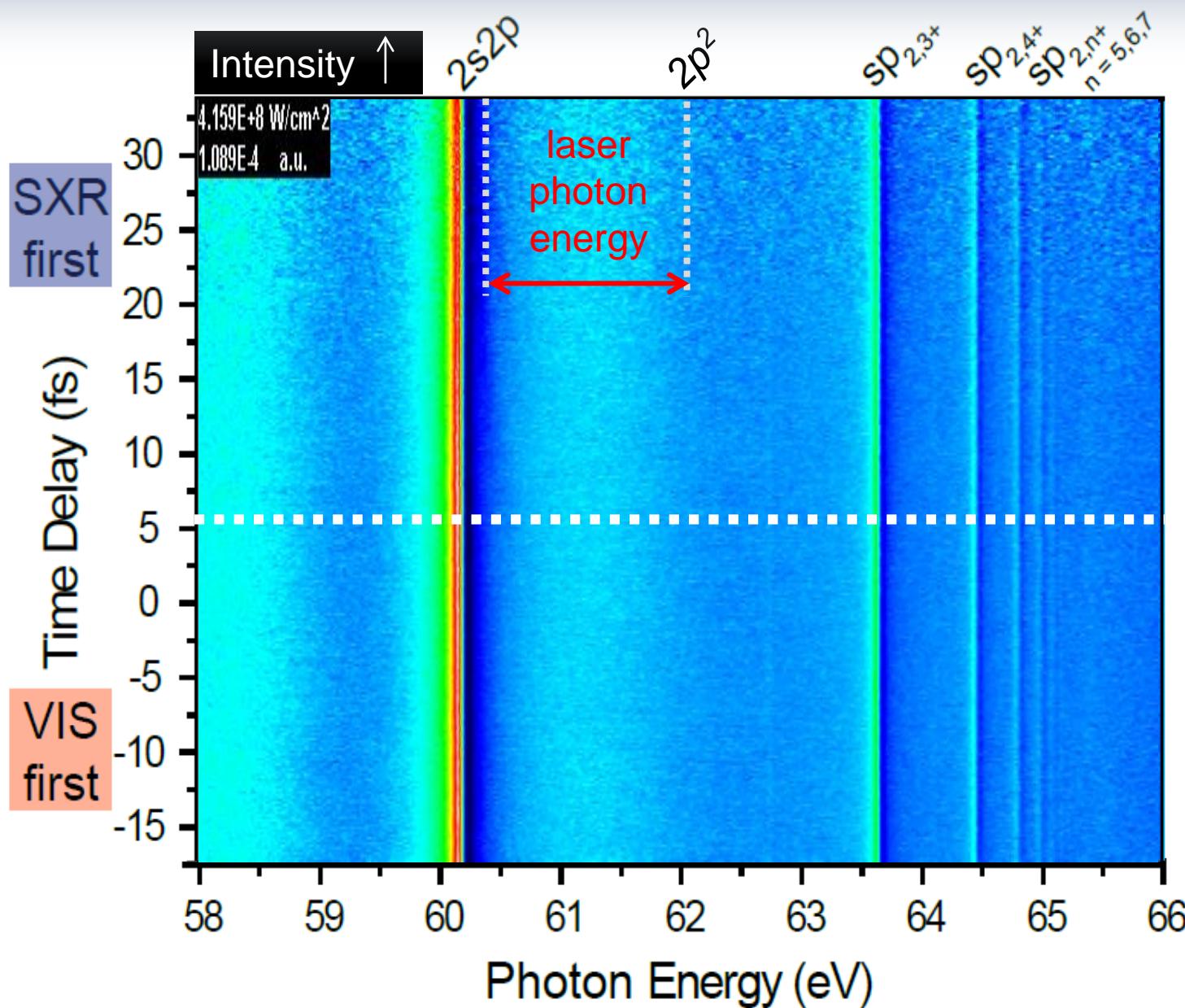
And how does it respond to **intense pulsed light**?



Control the electrons, and measure their response

"Ask"

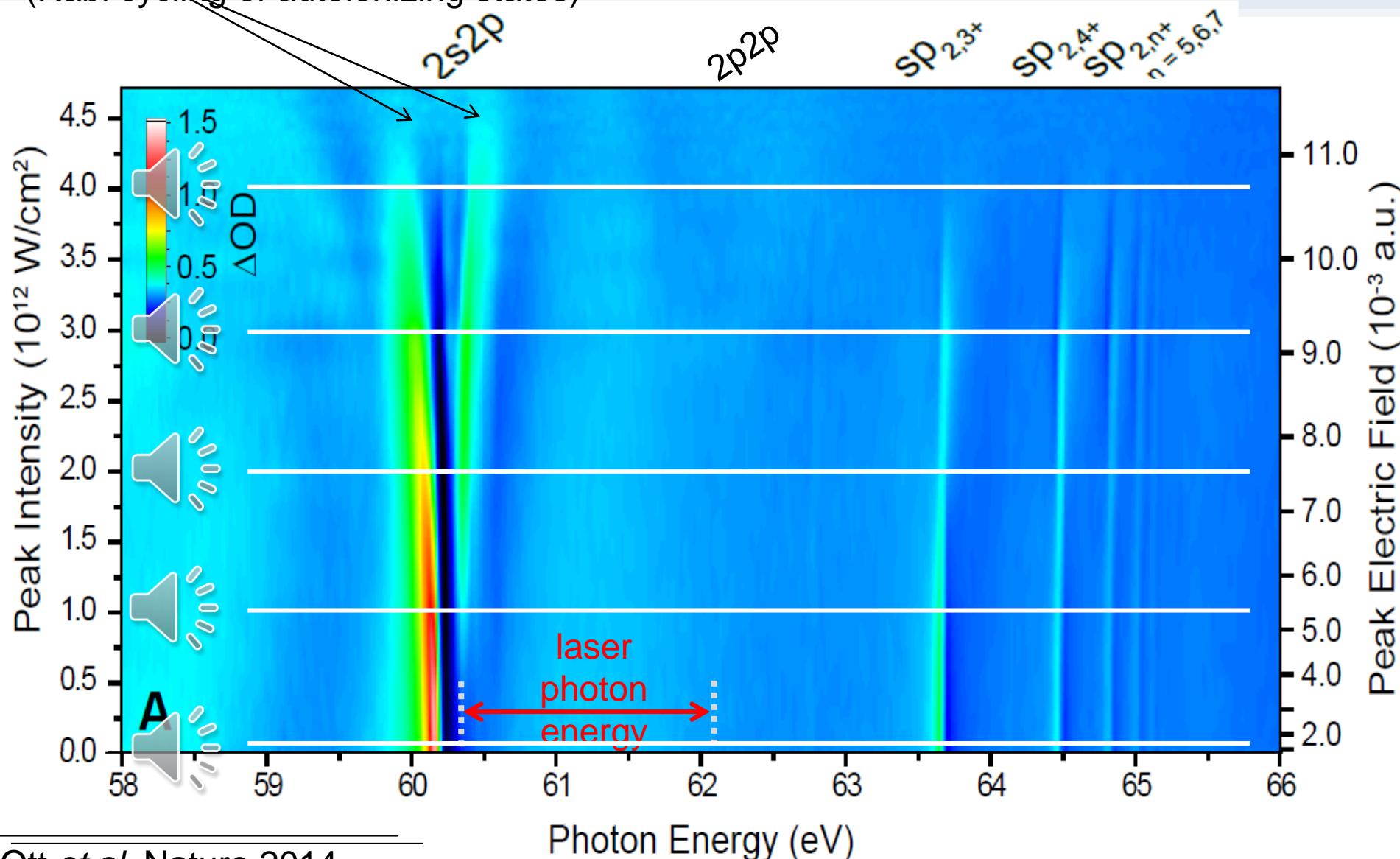
"listen"



Intensity, the control knob

Autler-Townes doublet
(Rabi cycling of autoionizing states)

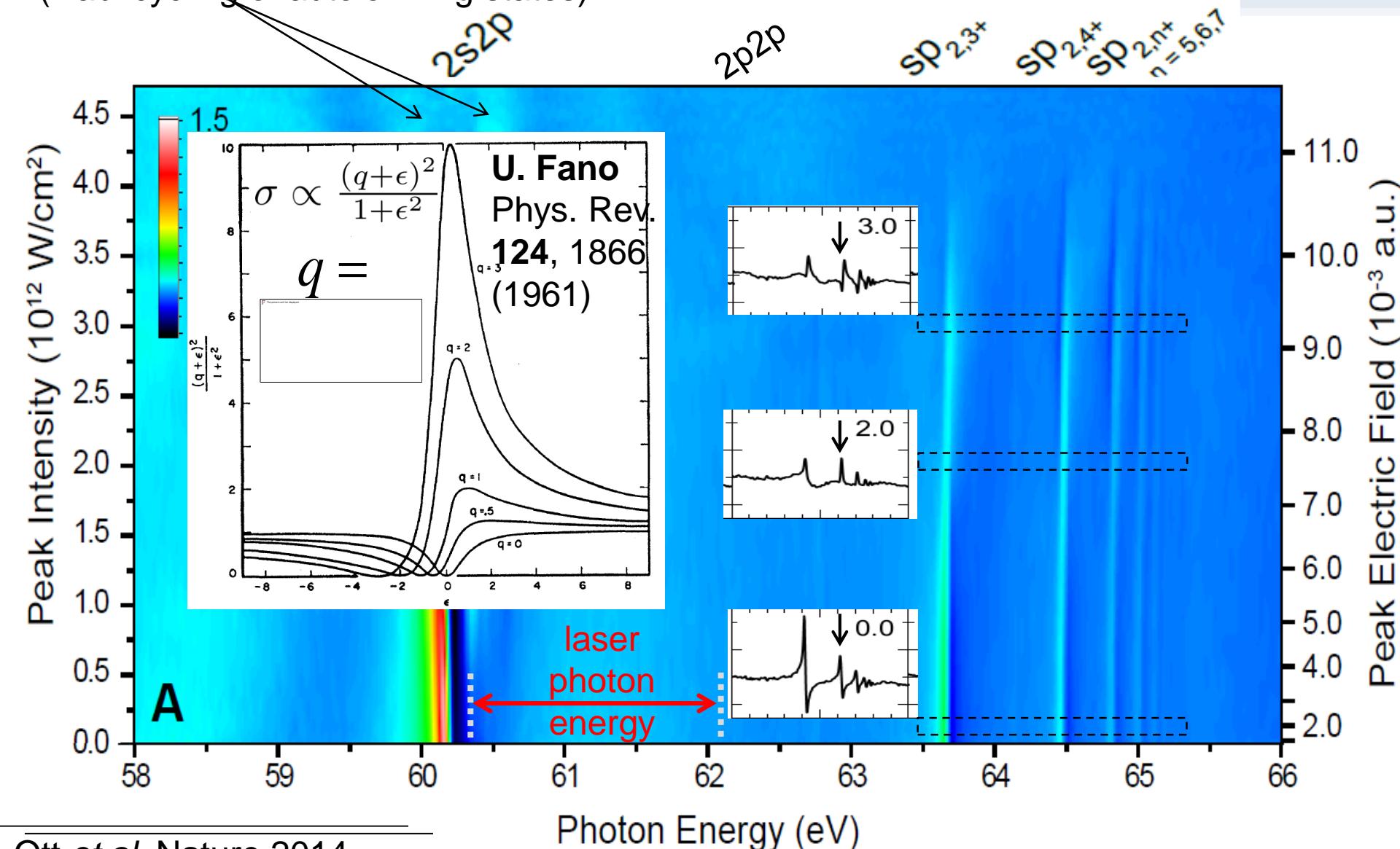
tuning from weak perturbative to strong fields



Intensity, the control knob

Autler-Townes doublet
(Rabi cycling of autoionizing states)

tuning from weak perturbative to strong fields



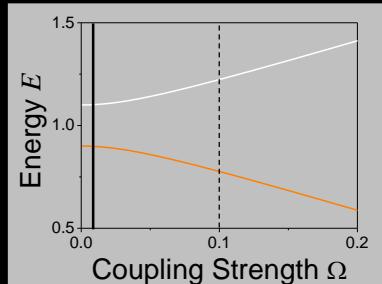
General: coupling of states

coupling of one to
one other state

coupling of one to
multiple states

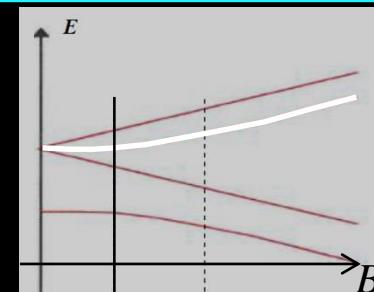
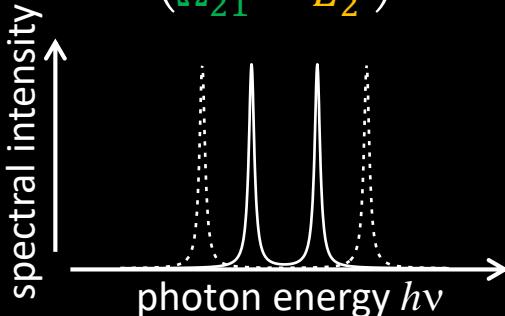
coupling of one to a
continuum of states

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



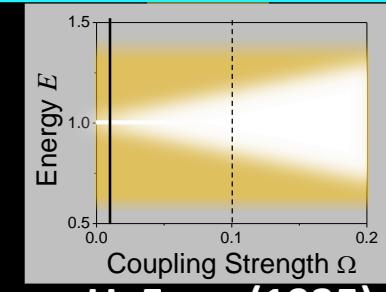
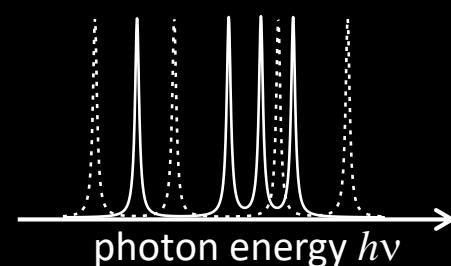
Rabi oscillation
in strong coupling

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



Breit-Rabi
e.g. Paschen-Back regime

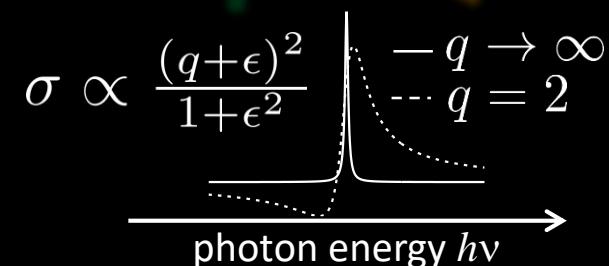
$$\begin{pmatrix} E_1 & \Omega_{12} & \Omega_{13} & \Omega_{14} \\ \Omega_{21} & E_2 & \Omega_{23} & \Omega_{24} \\ \Omega_{31} & \Omega_{32} & E_3 & \Omega_{34} \\ \Omega_{41} & \Omega_{42} & \Omega_{43} & E_4 \end{pmatrix}$$



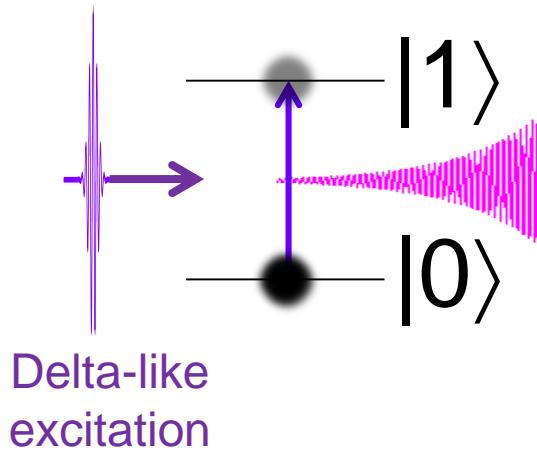
U. Fano (1935)

Phys. Rev. **124**, 1866 (1961)
Nuovo Cim. **12**, 154 (1935)

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

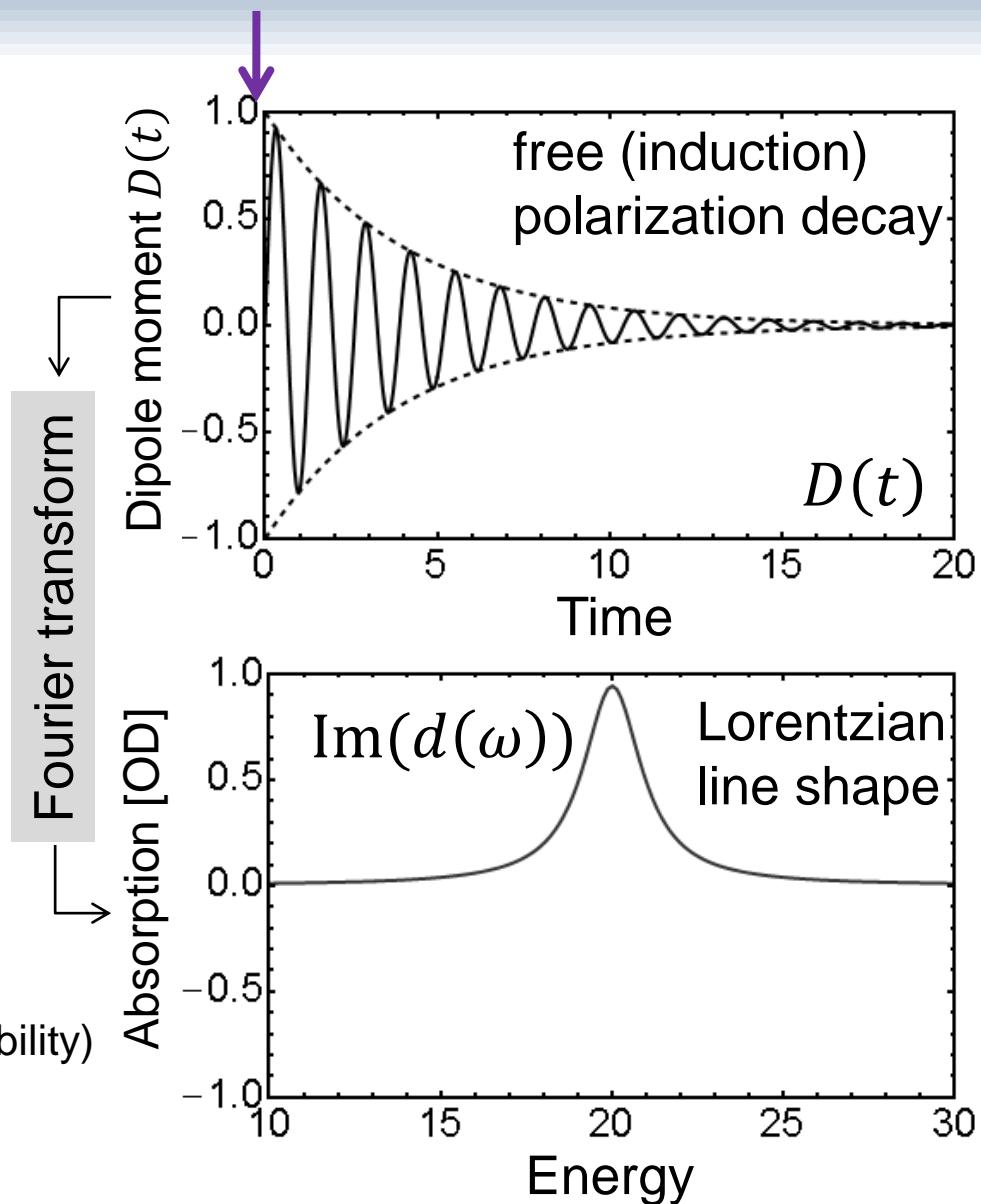


Resonance absorption in the Time Domain



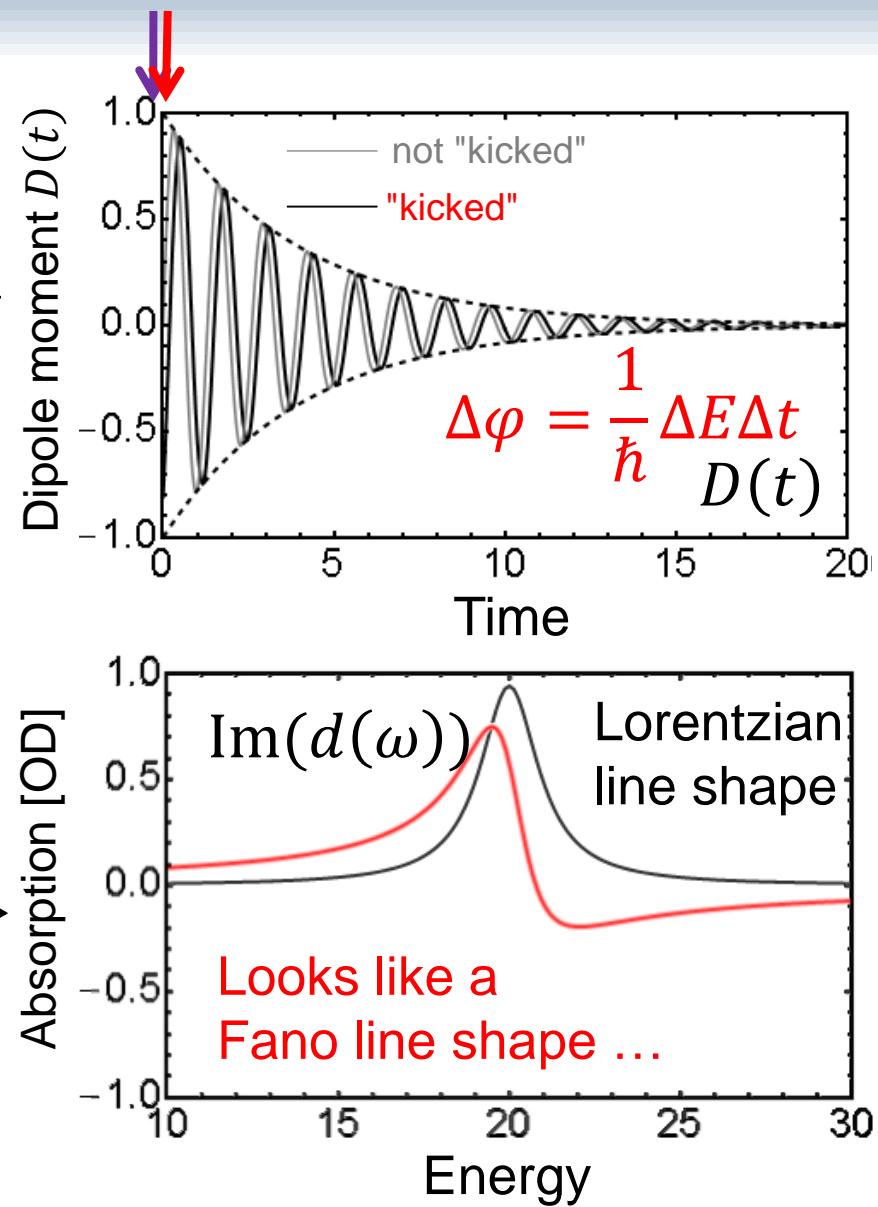
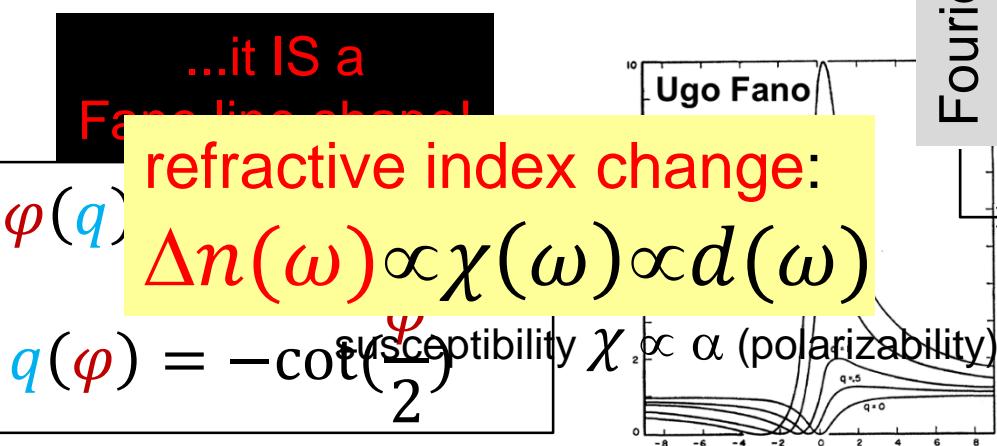
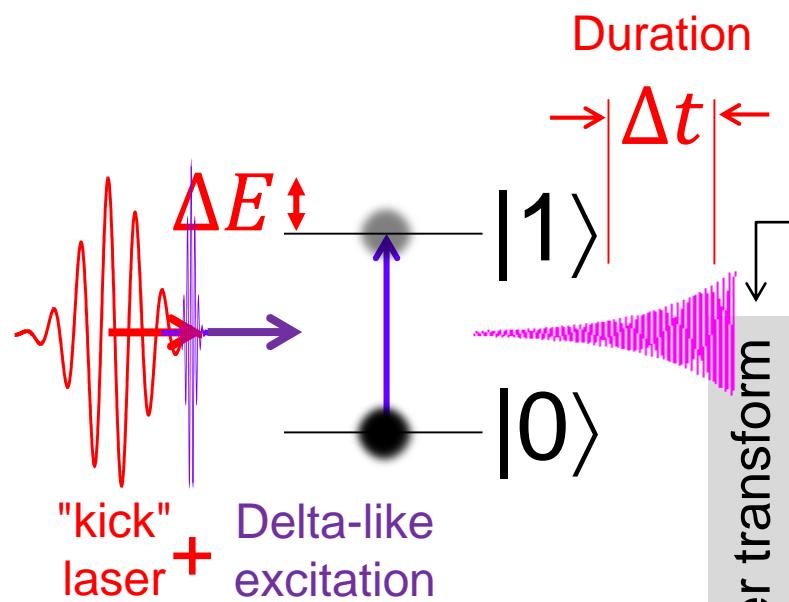
refractive index change:
 $\Delta n(\omega) \propto \chi(\omega) \propto d(\omega)$

susceptibility $\chi \propto \alpha$ (polarizability)



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)

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

=

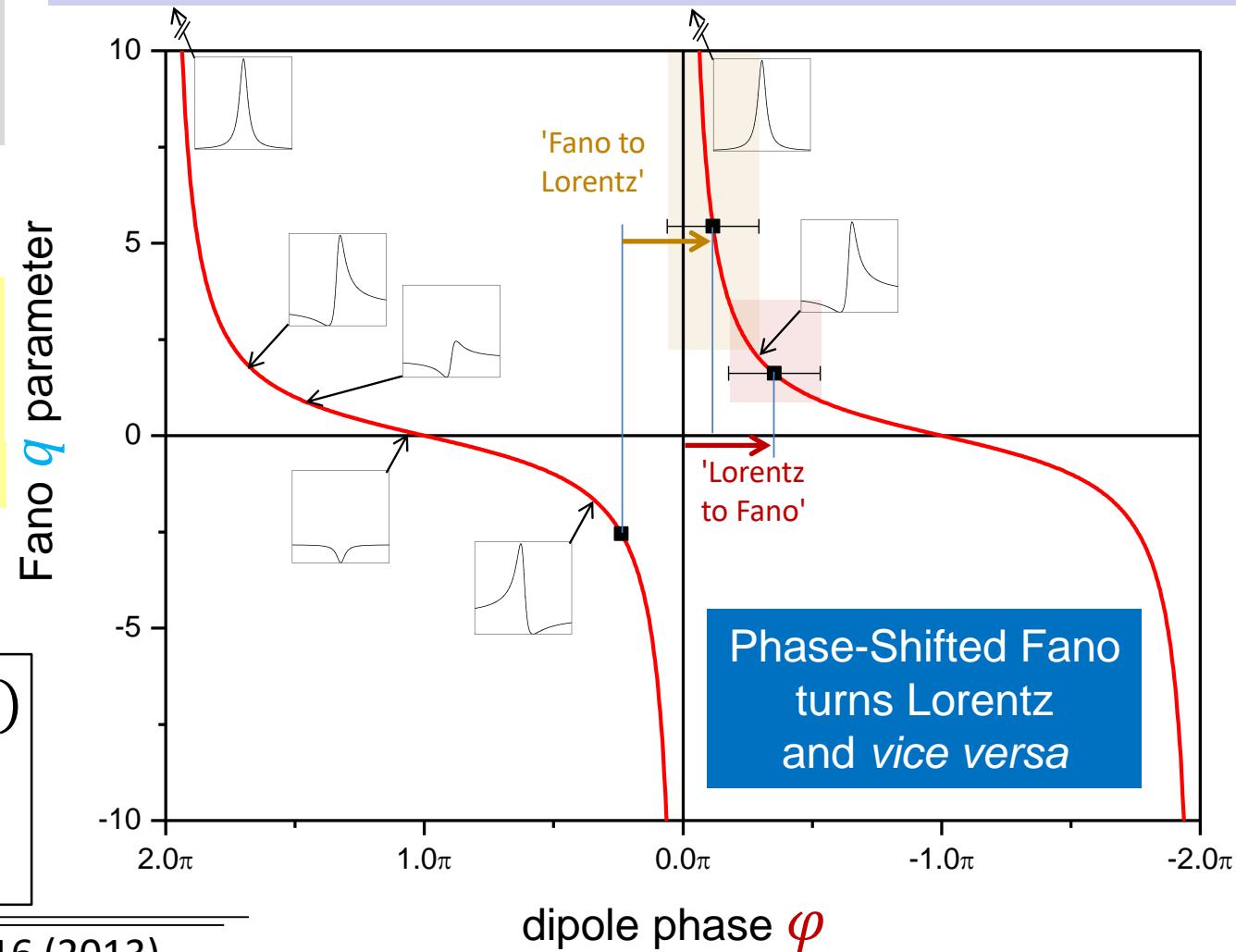
$$\text{Im}\left(\frac{-1}{i+\varepsilon} \exp(i\varphi)\right) + \text{const.}$$

Phase-shifted Lorentzian

...it IS a
Fano line shape!

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

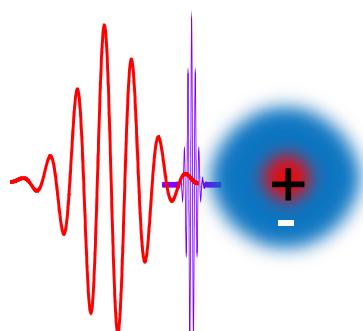
$$q(\varphi) = -\cot\left(\frac{\varphi}{2}\right)$$



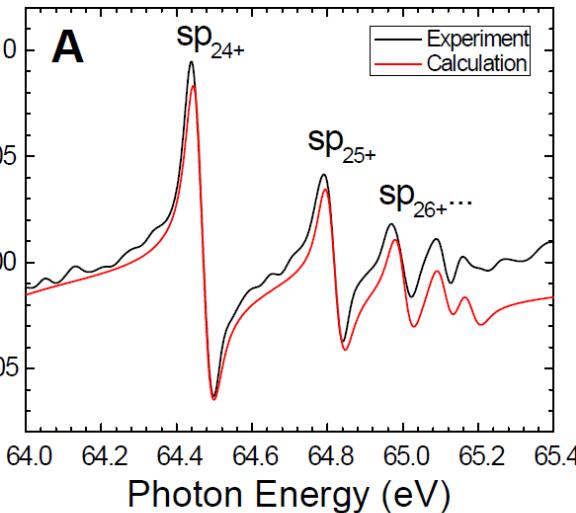
Fano to Lorentz, and Lorentz to Fano

doubly-excited Helium
originally Fano lineshape

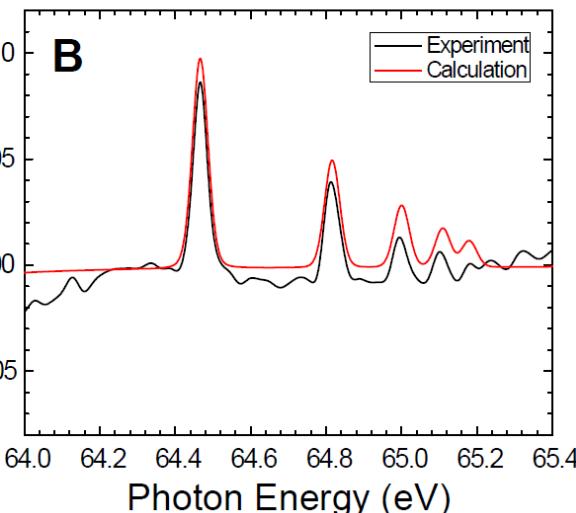
no laser



Rel. Absorbance (ΔOD)

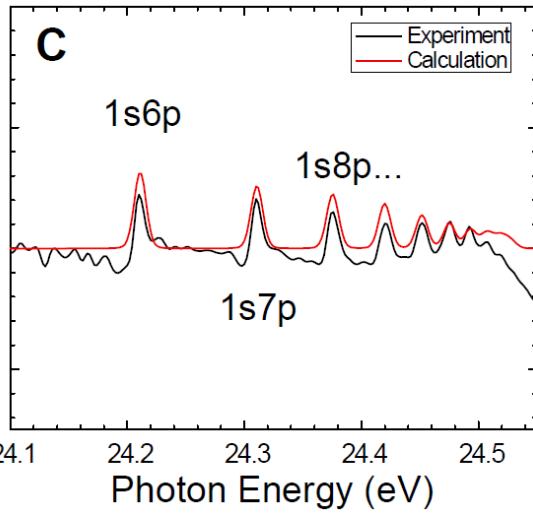


Rel. Absorbance (ΔOD)

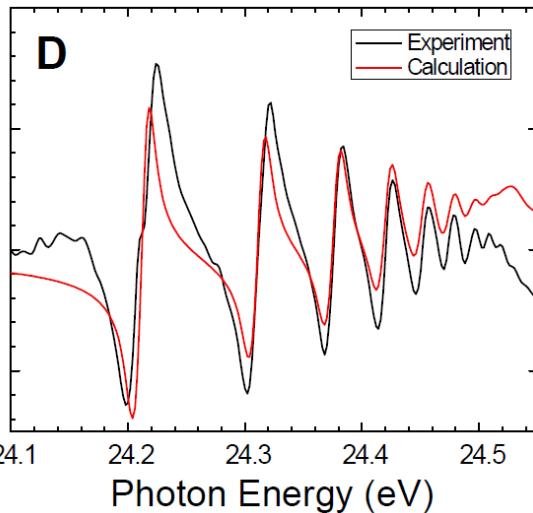


singly-excited Helium
originally Lorentzian

Rel. Absorbance (ΔOD)

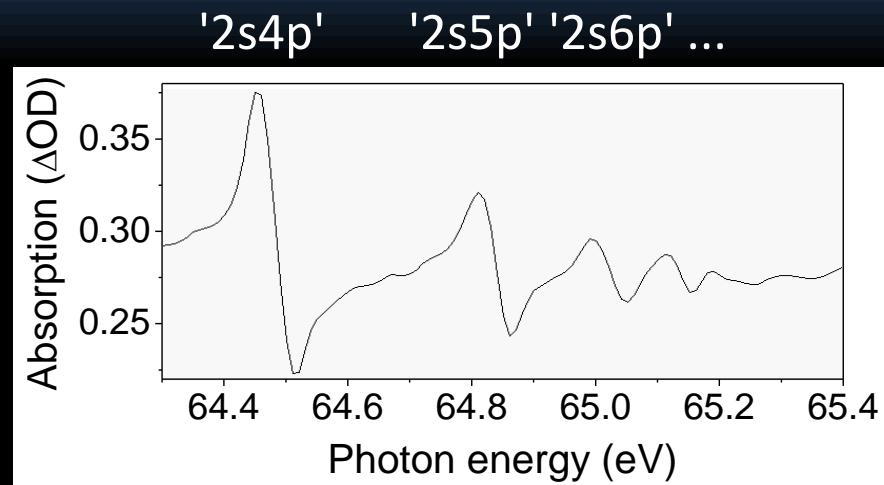


Rel. Absorbance (ΔOD)



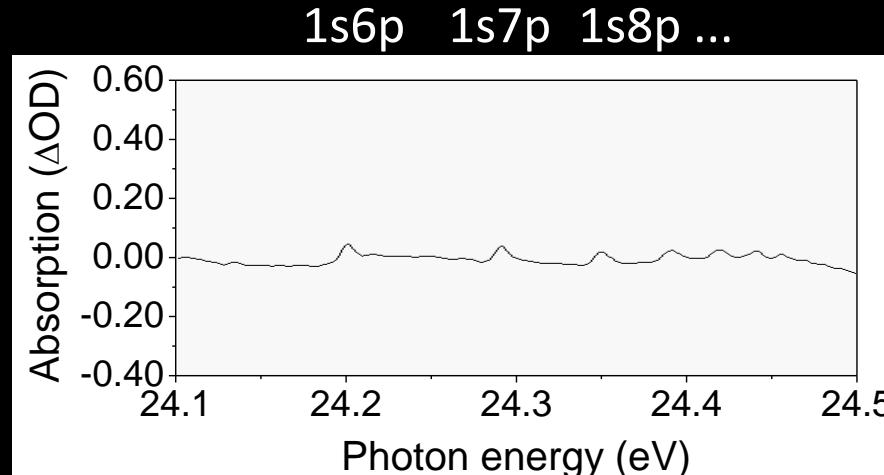
Laser Control of Fano and Lorentzian resonances

Helium
doubly excited
(above the
continuum
threshold)



turning
original 'Fano'
into 'Lorentz'

Helium
singly excited
(below the
continuum
threshold)



turning
original 'Lorentz'
into 'Fano'

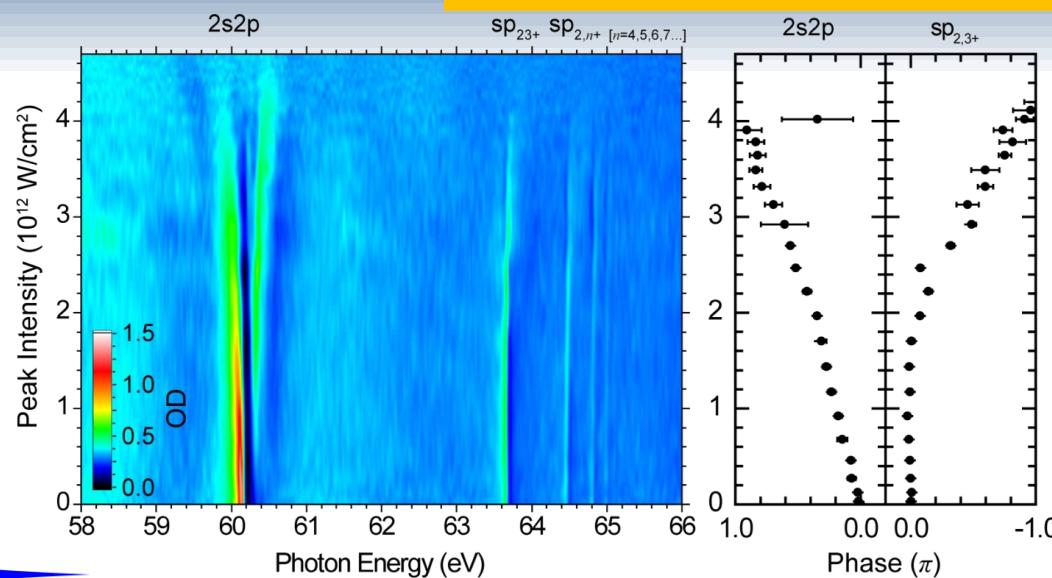
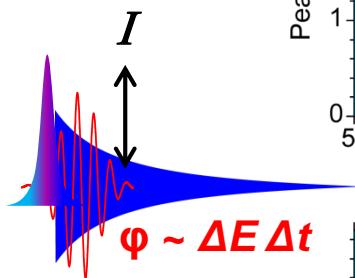
$$q = -\cot\left(\frac{\varphi}{2}\right)$$

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

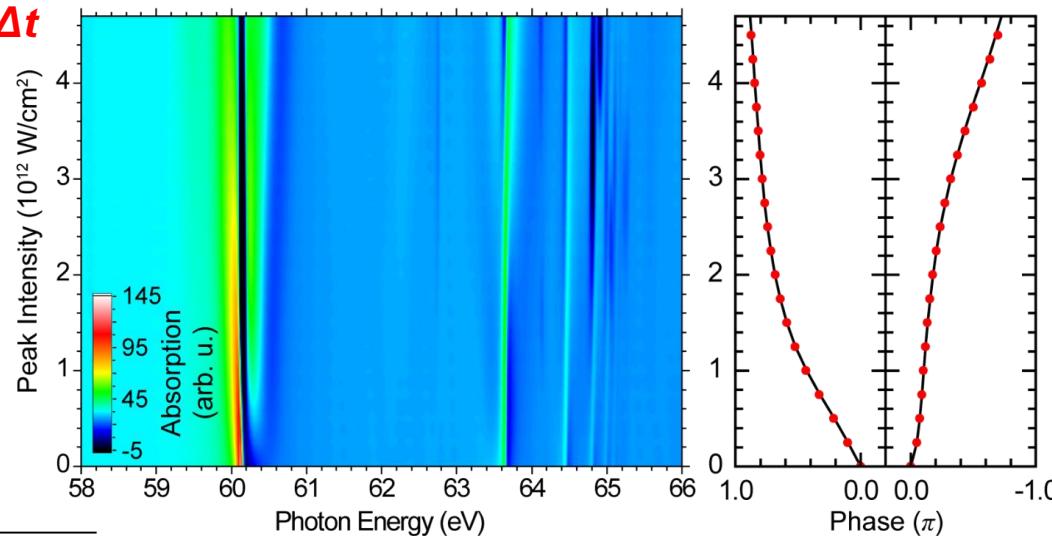
Extracting the laser-induced phase shift

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

experiment

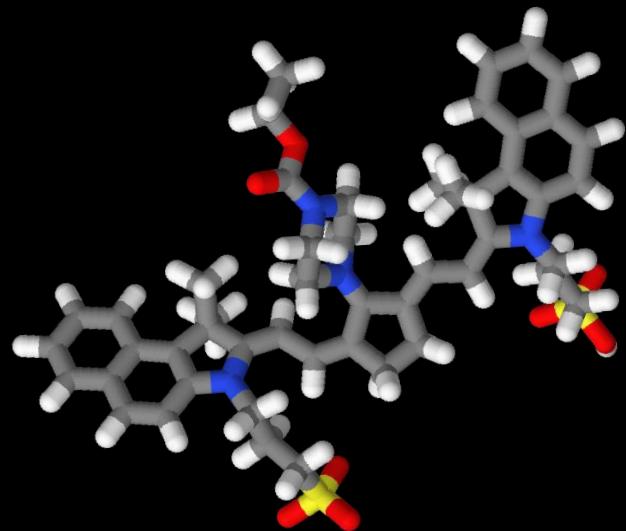


ab-initio theory
(Argenti/Martin)



Phase control of "real molecules"

Can we change the (spectral) shape
of complex molecules?

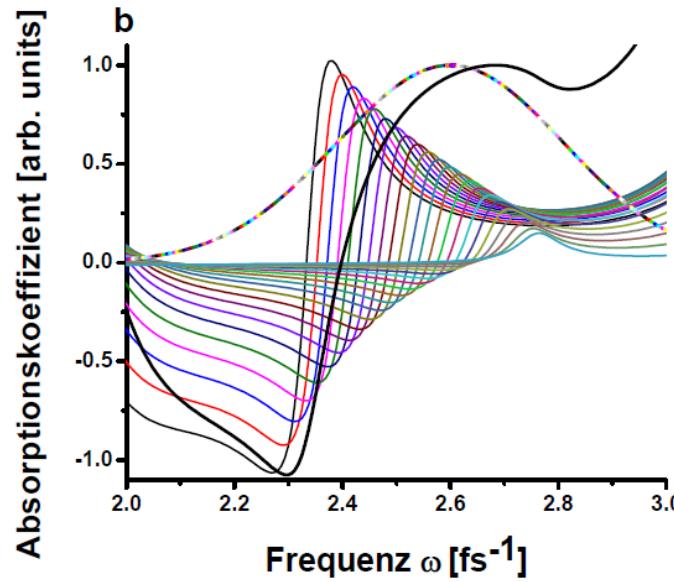
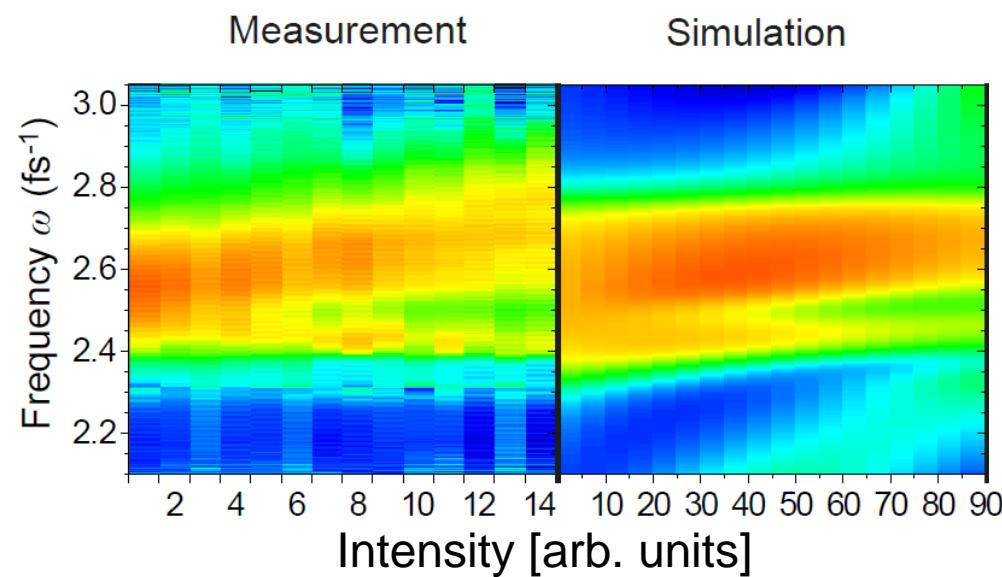
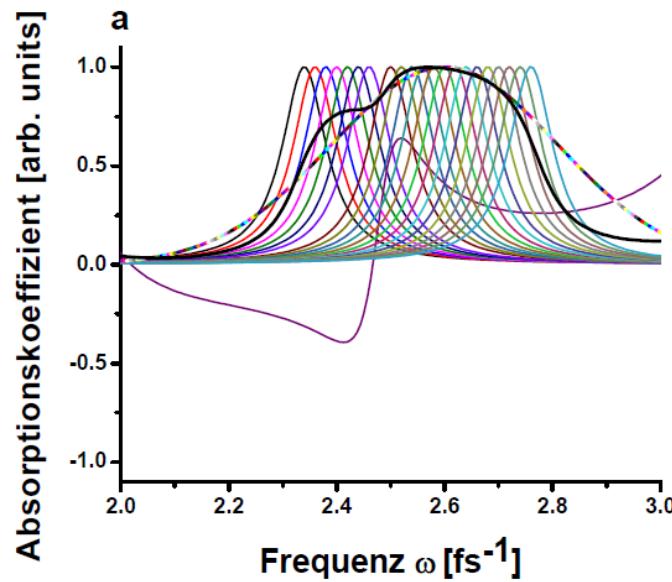
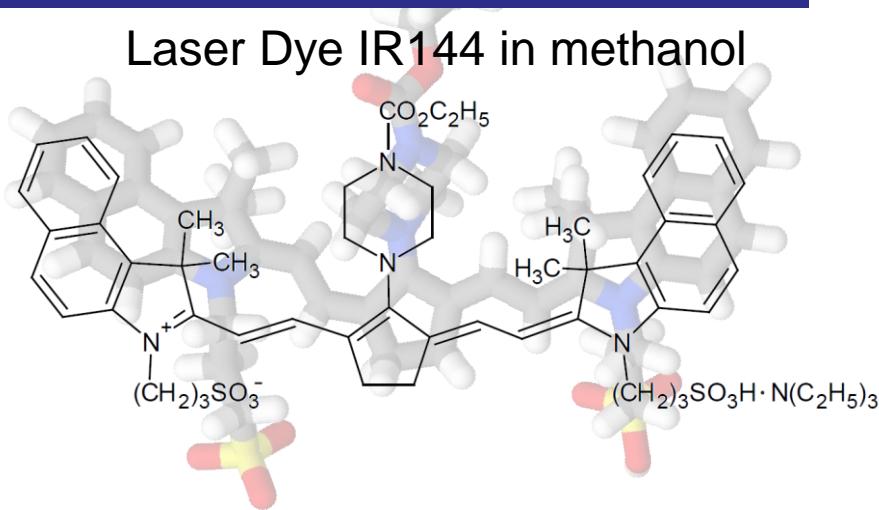


Fano control of molecules in the liquid phase

cooperation with J.-M. Mewes, A. Dreuw, T. Buckup, M. Motzkus@Univ. Heidelberg

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

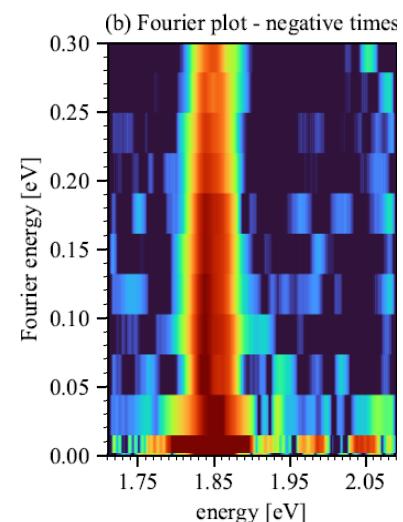
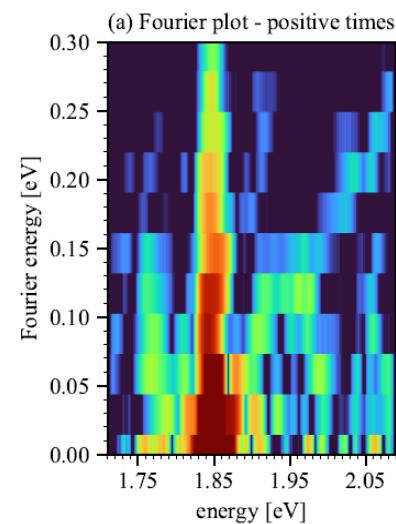
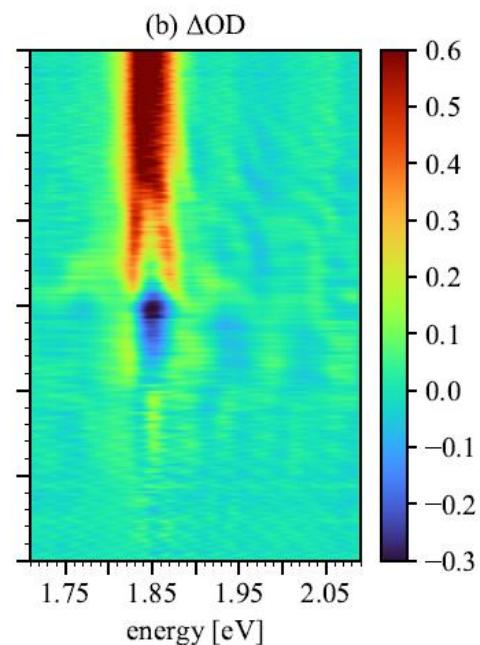
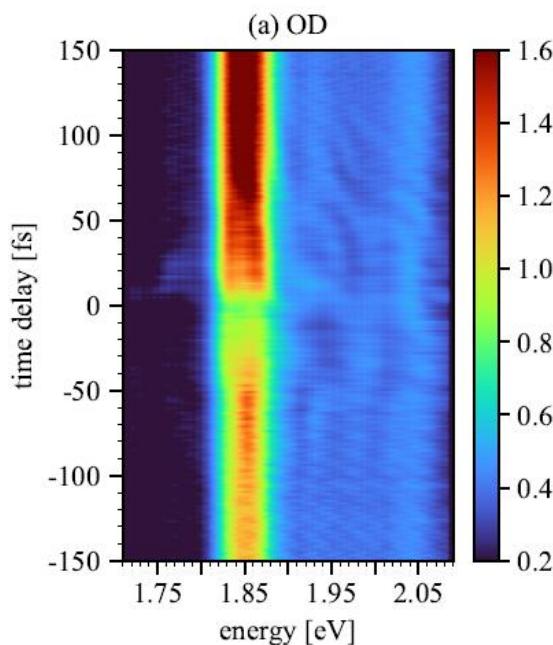
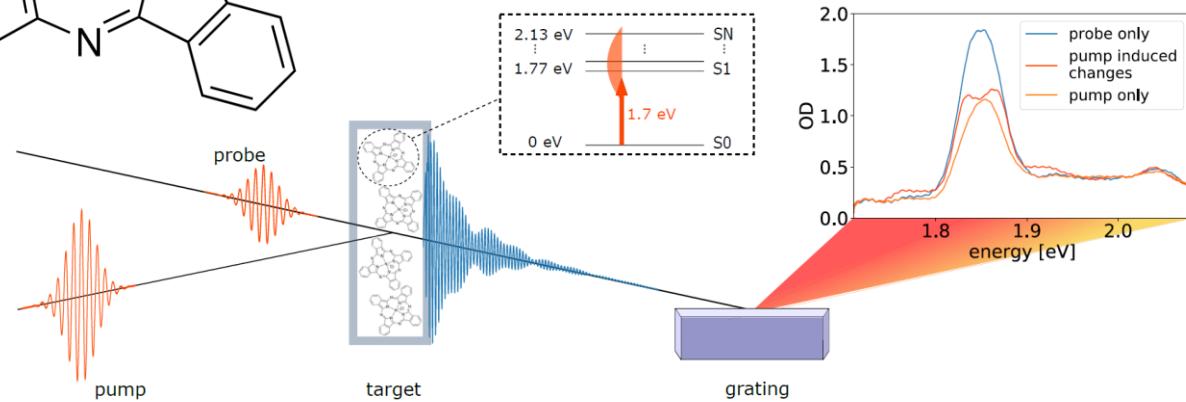
Laser Dye IR144 in methanol





Recent measurements in Aluminum-phthalocyanine-chloride (AlPhCl)

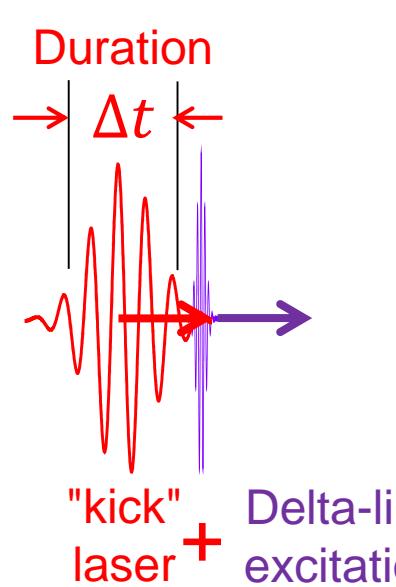
Carina da Costa Castanheira *et al.* (in preparation)



Time-domain, *impulsive*, resonance control

Variation of intensity: tuning of coupling strength, phase

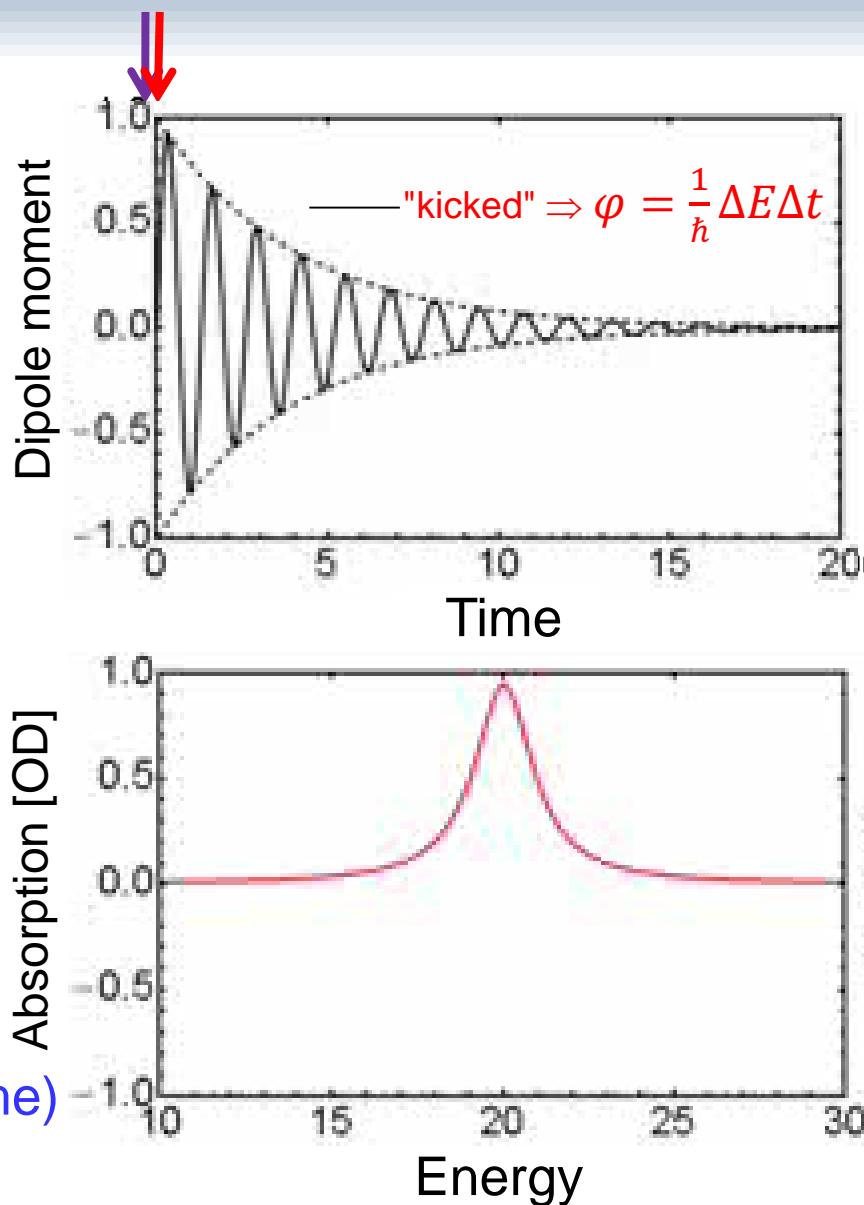
Science 340, 716 (2013)



Tuning from
absorption to gain!

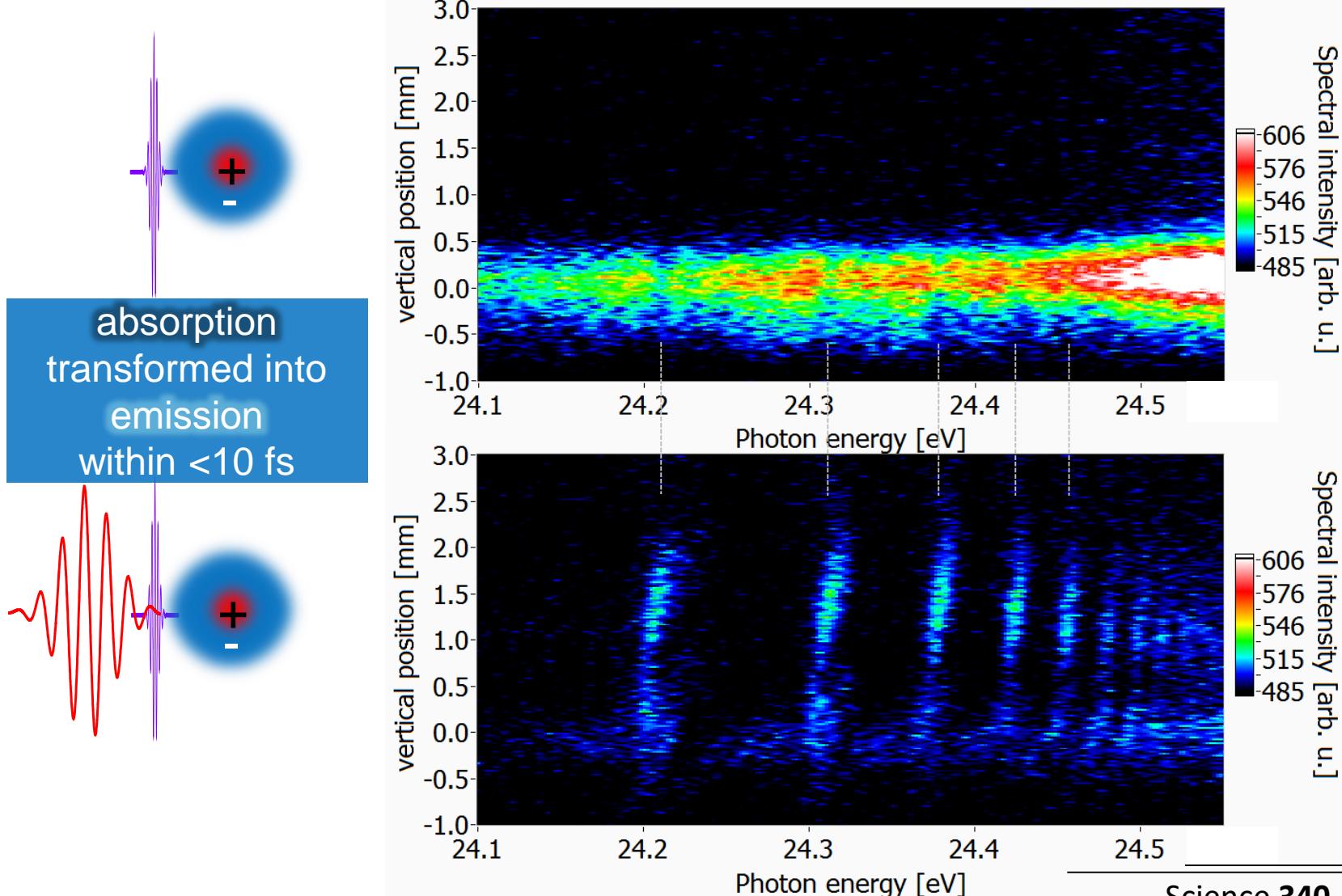
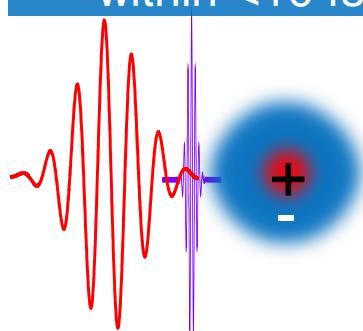
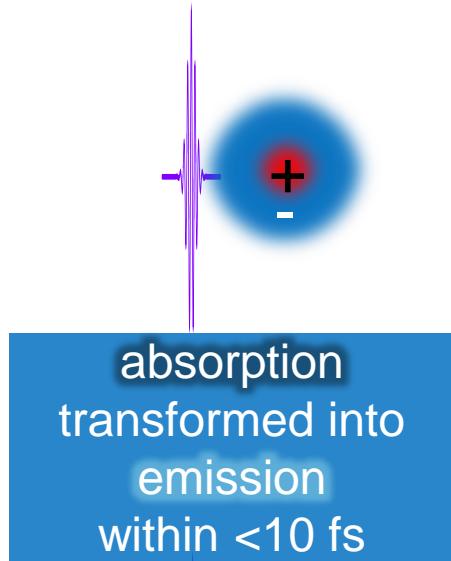
... in the "impulsive" limit!

(much shorter than the lifetime)

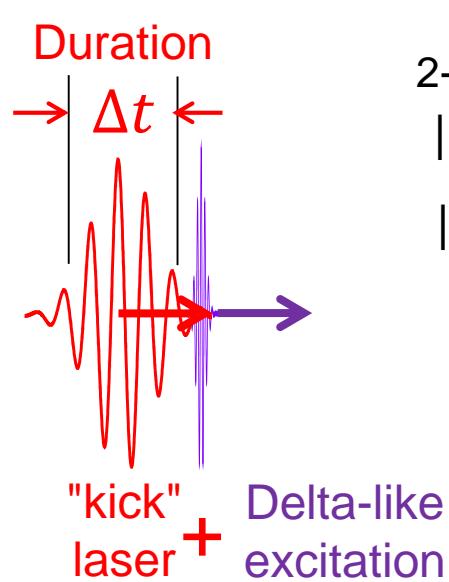


Proof-of-Principle: Observation of Resonant Gain

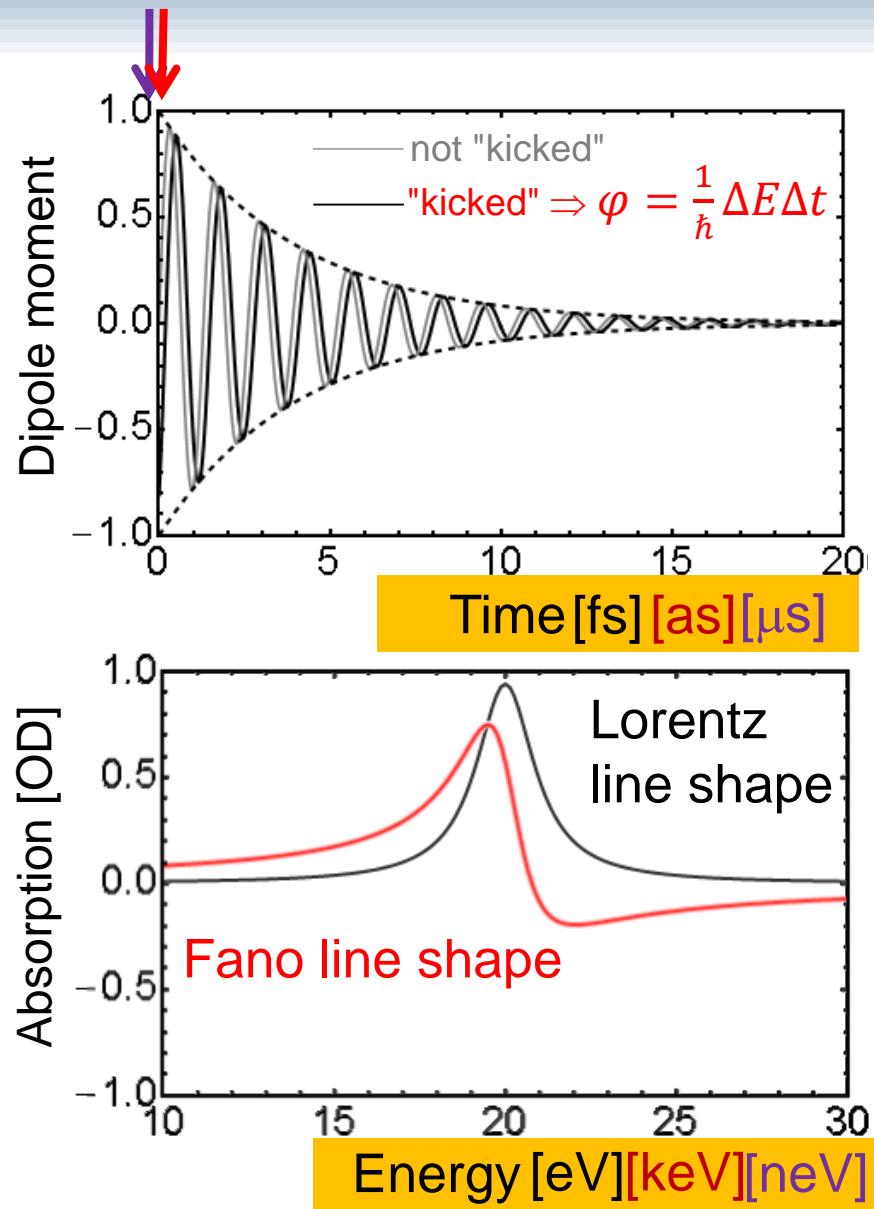
in singly-excited helium



The Beauty of a Fundamental Mechanism



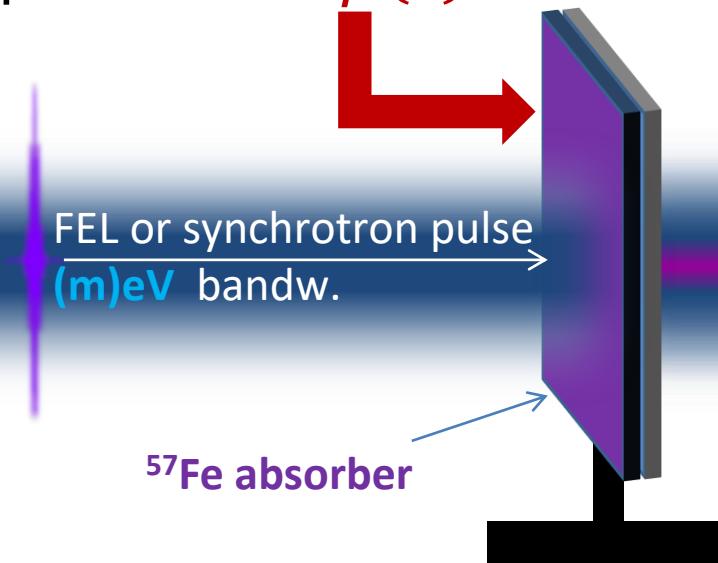
applies to all
time scales and
photon energies...



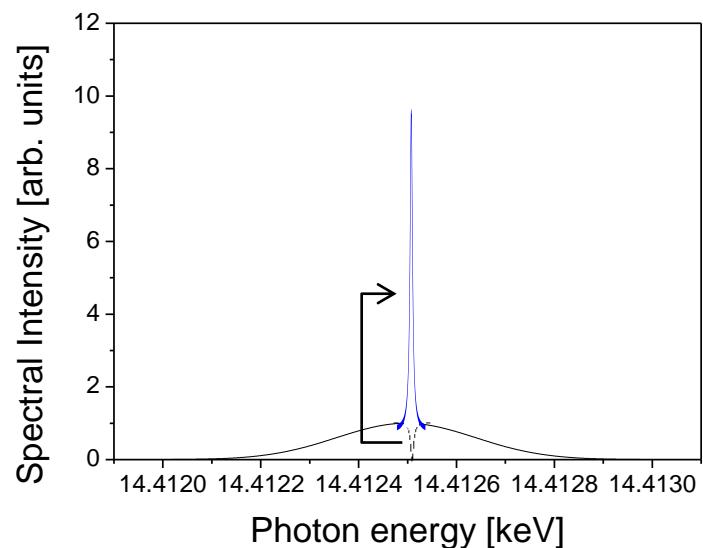
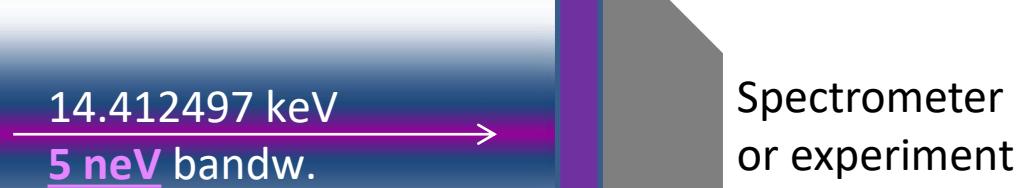
Resonant gain for hard x-rays

^{14.4 keV,}
 ^{^{57}Fe} Mössbauer

phase shift $\varphi(t)$ at $t = 0$



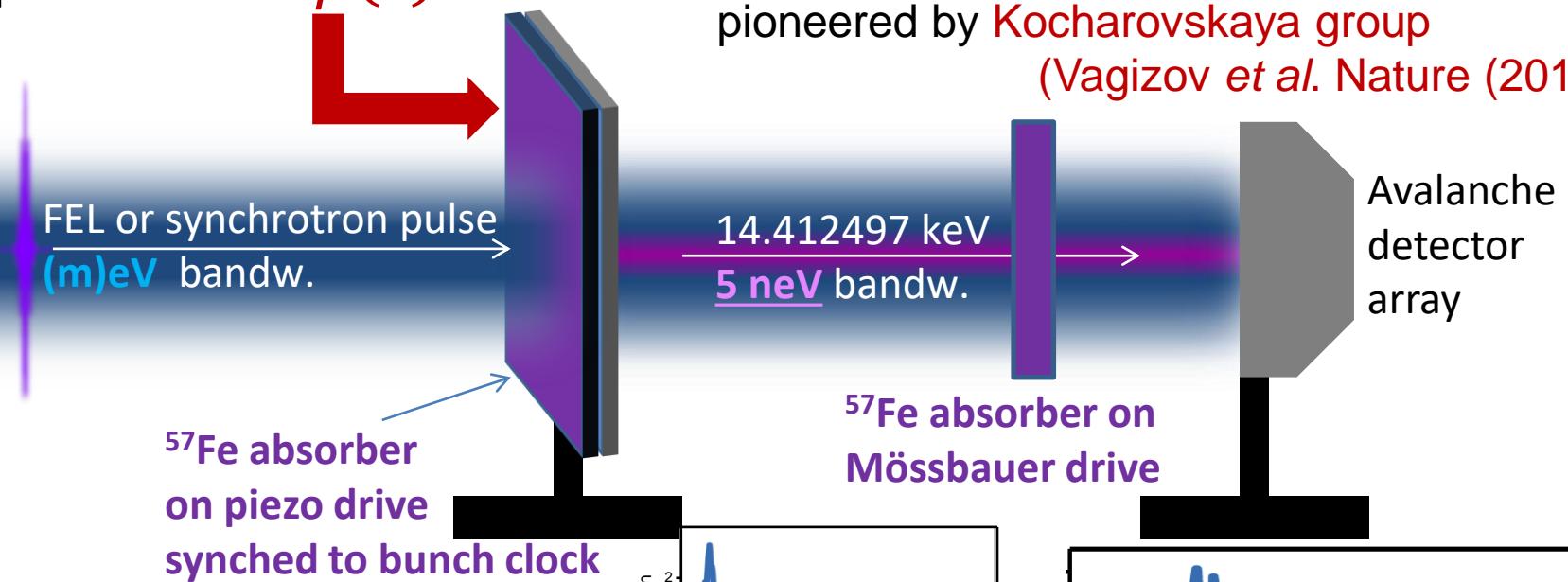
Laser-like
narrowband
line emission
at hard x-rays



Resonant Mössbauer "gain" @ ESRF, ID18



Heeg *et al.* Science (2017) cooperation with groups of J. Evers, R. Röhlsberger, R. Rüffer phase shift $\varphi(t)$ at $t = 0$ using a Mössbauer Coherent Control method pioneered by Kocharovskaya group (Vagizov *et al.* Nature (2014))

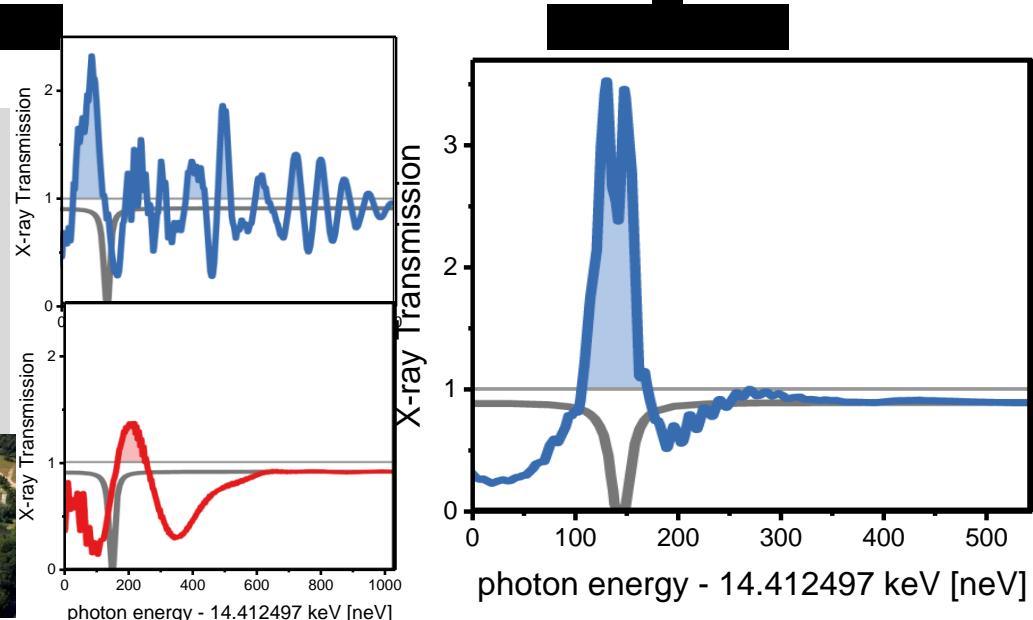


Now: Mechanical motion for phase shift
=> nanosecond response

Future: Optical control of phonons
=> femtosecond response

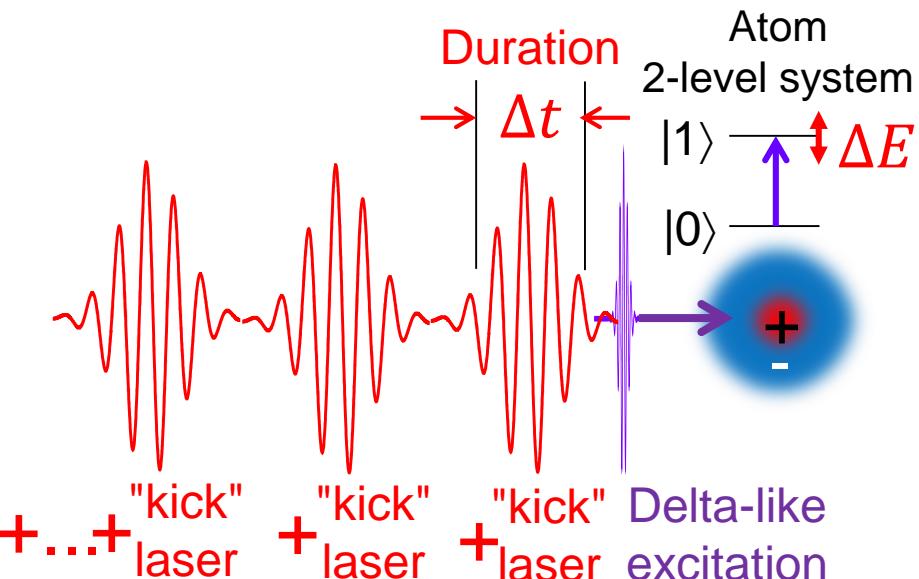
X-ray pulse shaping?

Experiment@PETRAIII



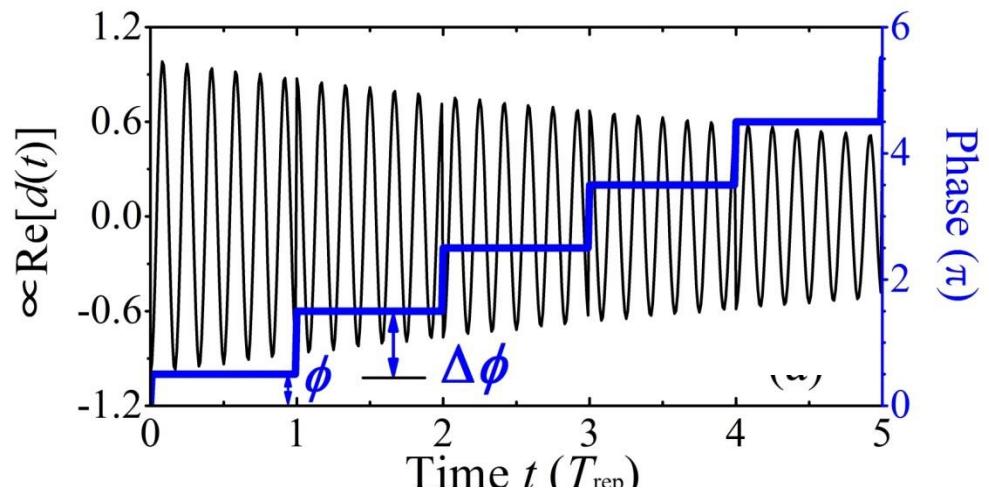
Towards x-ray frequency combs

Cooperation with group of C. H. Keitel (MPIK): Cavaletto, Harman *et al.*, Nat. Photonics (2014)
Liu, Ott, *et al.*, New. J. Phys. (2014)

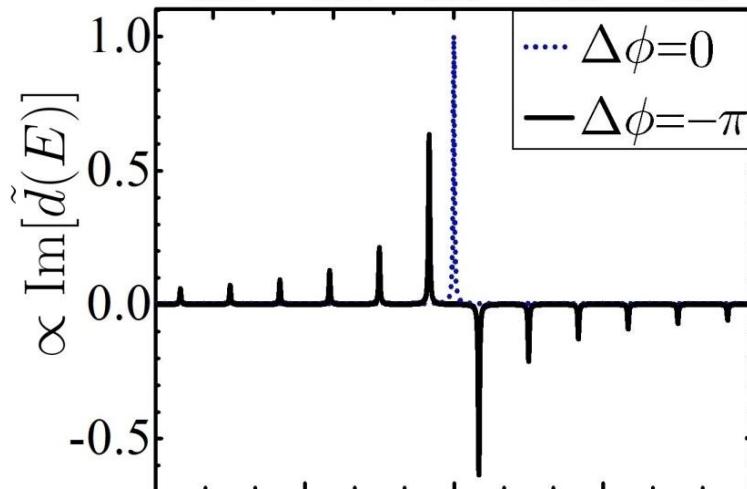


Using a **pulse train** for
periodic phase shifting ...

... creates a
frequency comb
precisely locked to
an atomic transition



$E - E_r (\text{meV})$

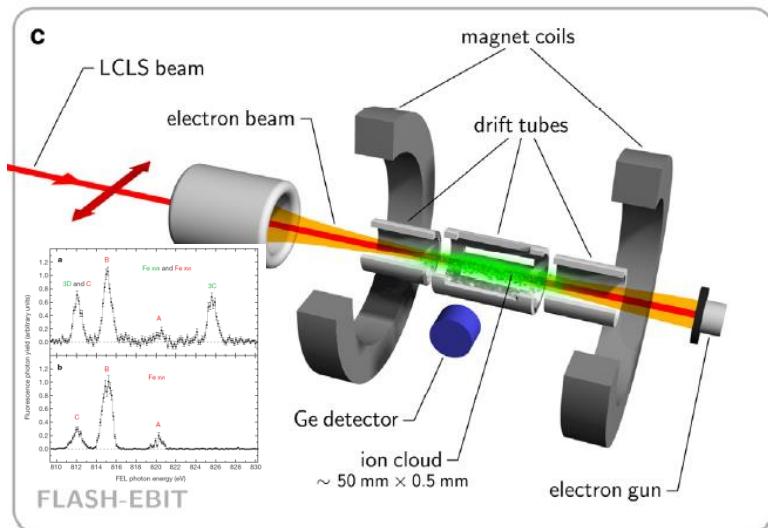


Towards x-ray precision metrology of highly-charged ions

Crespo group (MPIK)

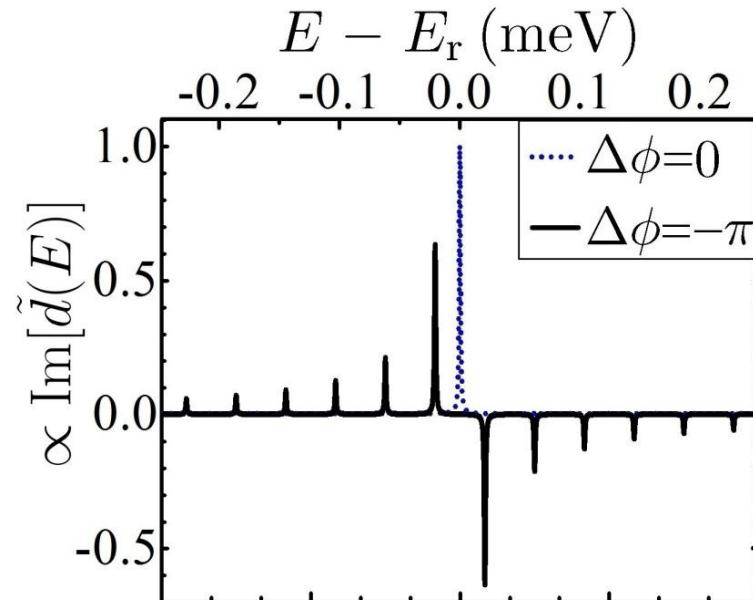
(image modified from:

Bennett et al. Nature 492, 225 (2012))



High-resolution spectroscopy of long-lived transitions in highly-charged ions:

- Precision tests of theories in heavy ions (QED, nuclear&relativistic-electronic structure)
- linking nuclear/electronic x-ray transitions: variation of coupling "constants"?



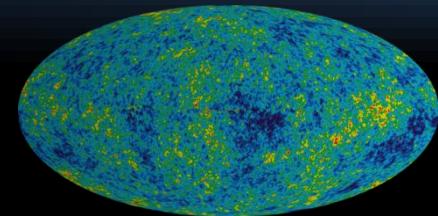
time scales in science



shortest man-made
flash/pulse of light
~50 attoseconds



1 second



age of the universe
14 billion years



as fs ps ns μ s ms

10^{-18} 10^{-12} 10^{-6} 1 10^6 10^{12} 10^{18}

electronic
timescale

human/biological
timescale

nuclear/atomic
timescale

molecular
timescale

geological/astronomical
timescale

cameras

laser pulses



Ultra slow dynamics...



shortest man-made
flash/pulse of light
~50 attoseconds



as fs ps ns μ s ms

10^{-18} 10^{-12}

electronic
timescale

nuclear/atomic
timescale

molecular
timescale

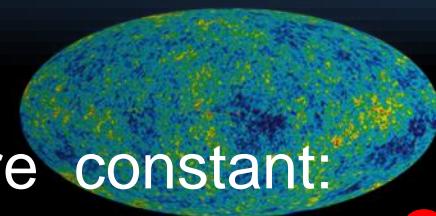
cameras



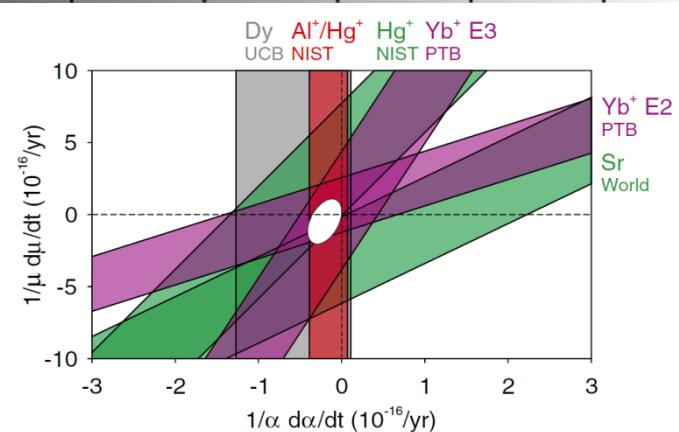
α
1 second



$$\alpha \approx \frac{1}{137.035999139(31)} + \dot{\alpha} \times t$$



age of the universe?
14 billion years



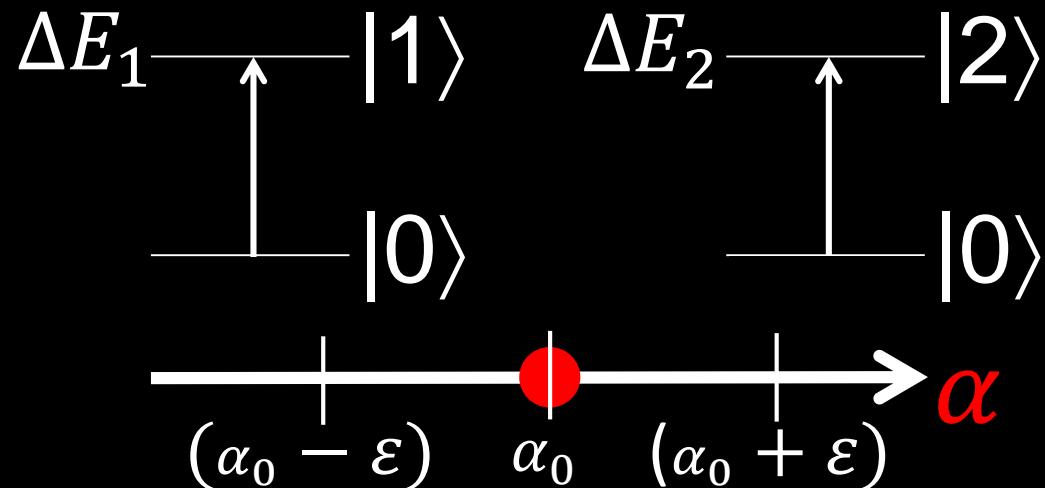
Godun *et al.* PRL **113**, 210801 (2014)

Huntemann *et al.* PRL **113**, 210802 (2014)

sensitivity to α variation

Dzuba, Flambaum, Webb
PRL 82, 888 (1999)

$$\Delta E_n = \frac{E_n}{\nu} (Z\alpha)^2 \left[\frac{1}{j + 1/2} - C(j, l) \right]$$

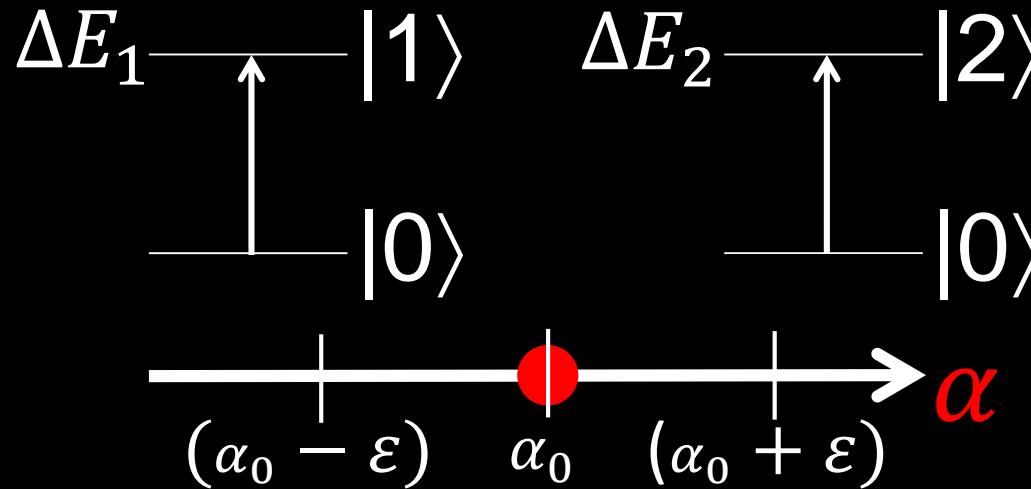


The highly charged ion (HCl) advantage: ***Enhanced*** sensitivity to α variation

Dzuba, Flambaum, Webb
PRL 82, 888 (1999)

$$\Delta E_n = \frac{E_n}{\nu} (\mathbf{Z}\alpha)^2 \left[\frac{1}{j + 1/2} - C(j, l) \right]$$
$$= 2 q \Delta\alpha/\alpha + \text{const.}$$

$\text{Hg}^+ : q \sim 52\,200 \text{ cm}^{-1}$
 $\text{Ir}^{17+} : q \sim 740\,000 \text{ cm}^{-1}$



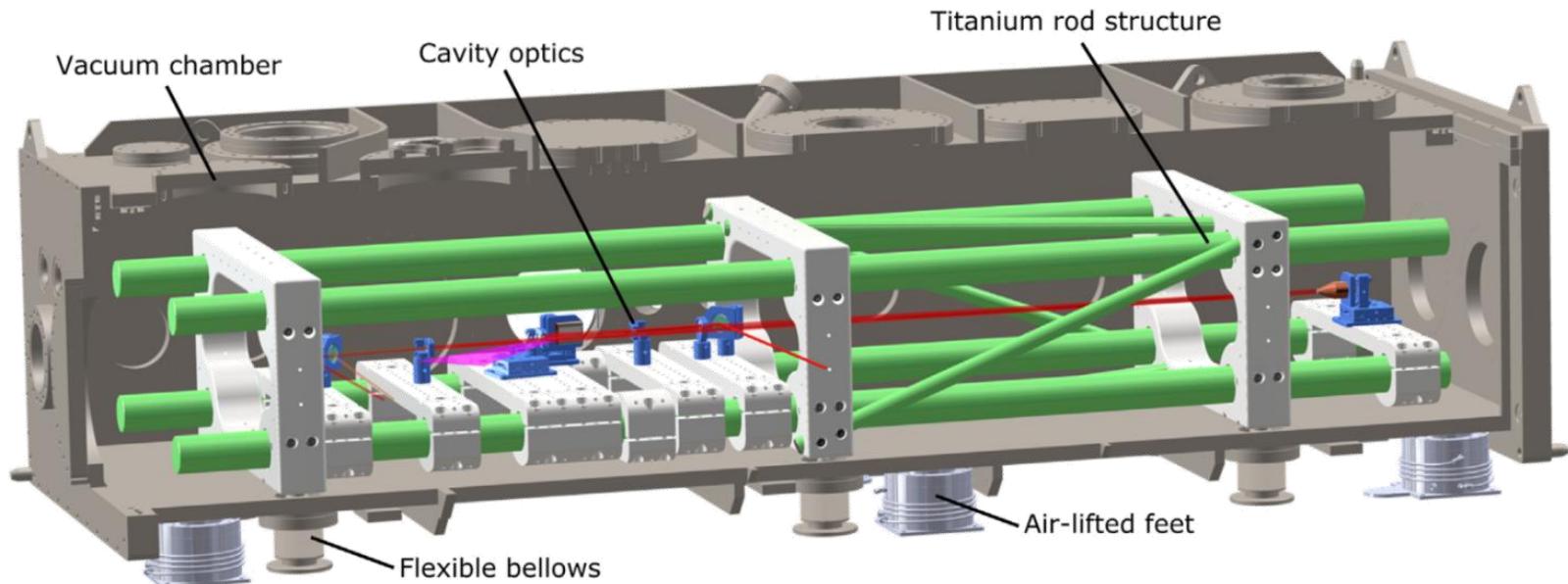
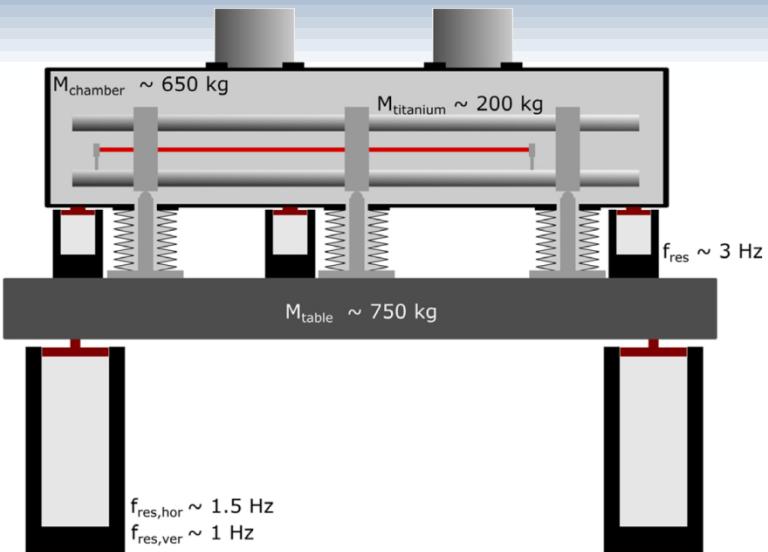
At some point,

controlled/precision VUV/xUV/xray light may be helpful...

Can we generate VUV frequency combs?

Design of enhancement/HHG cavity

- Enhancement cavity mounted on high-stiffness titanium frame on optical table
- Pump vibrations absorbed through mechanical low-pass filter, factor 10 reduction in amplitude



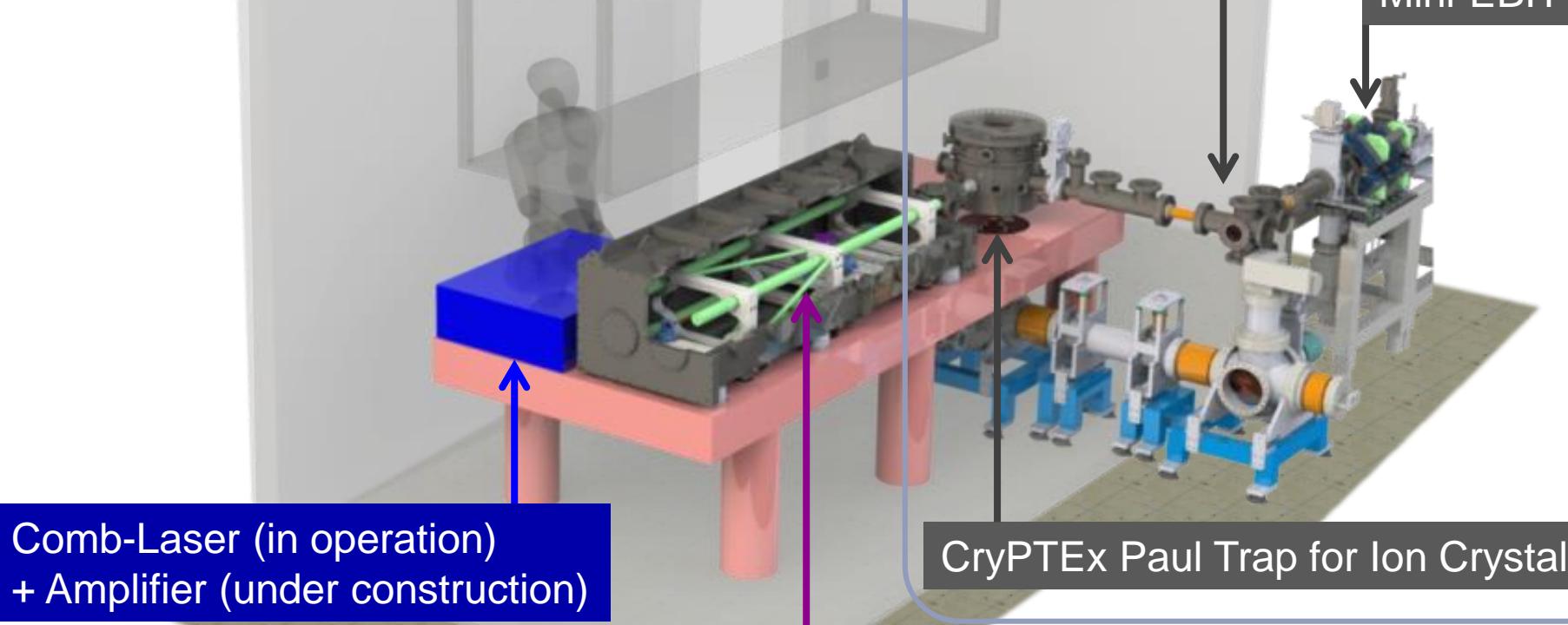
VUV-Comb-in-a-box (planning: -3 yrs)

Container enclosure for
Temperature stabilization
Noise isolation

Lisa Schmöger et al., Science (2015)

HCI deceleration/transfer line

Mini-EBIT

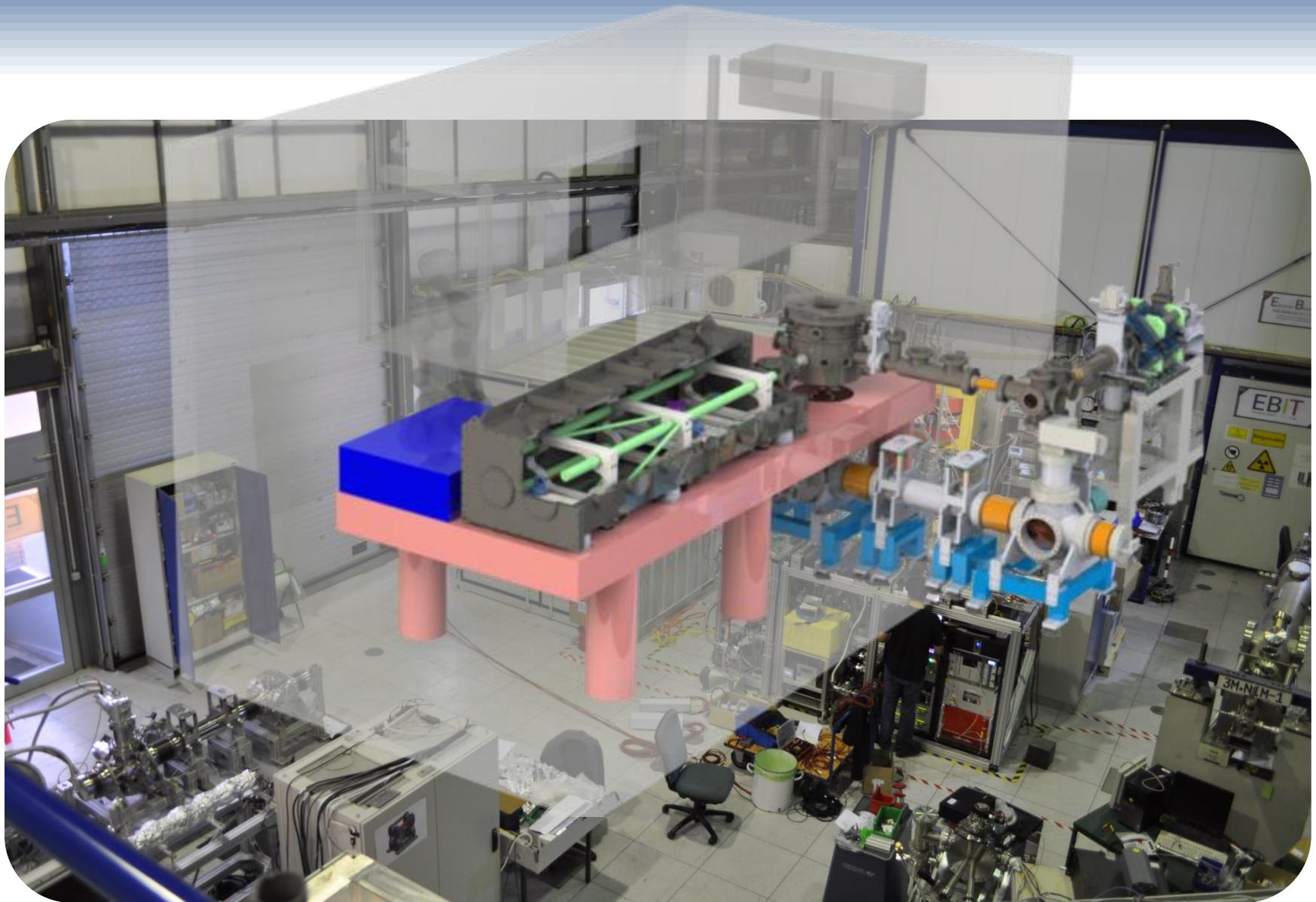


Comb-Laser (in operation)
+ Amplifier (under construction)

CryPTEEx Paul Trap for Ion Crystal

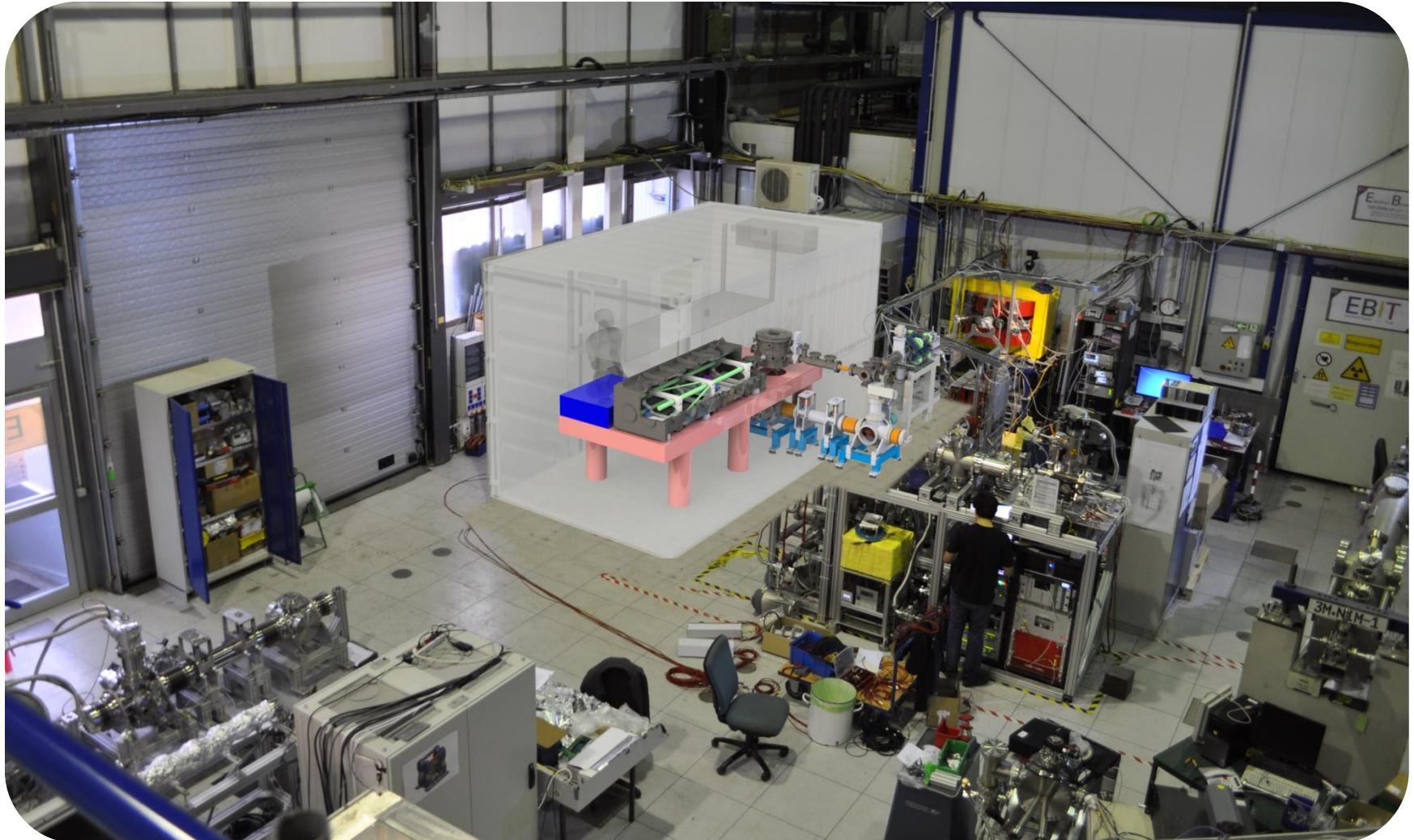
HHG-comb enhancement cavity on stabilized optical table
mechanically decoupled from vacuum system
(under construction)

VUV-Comb-in-a-box



VUV-Comb-in-a-box

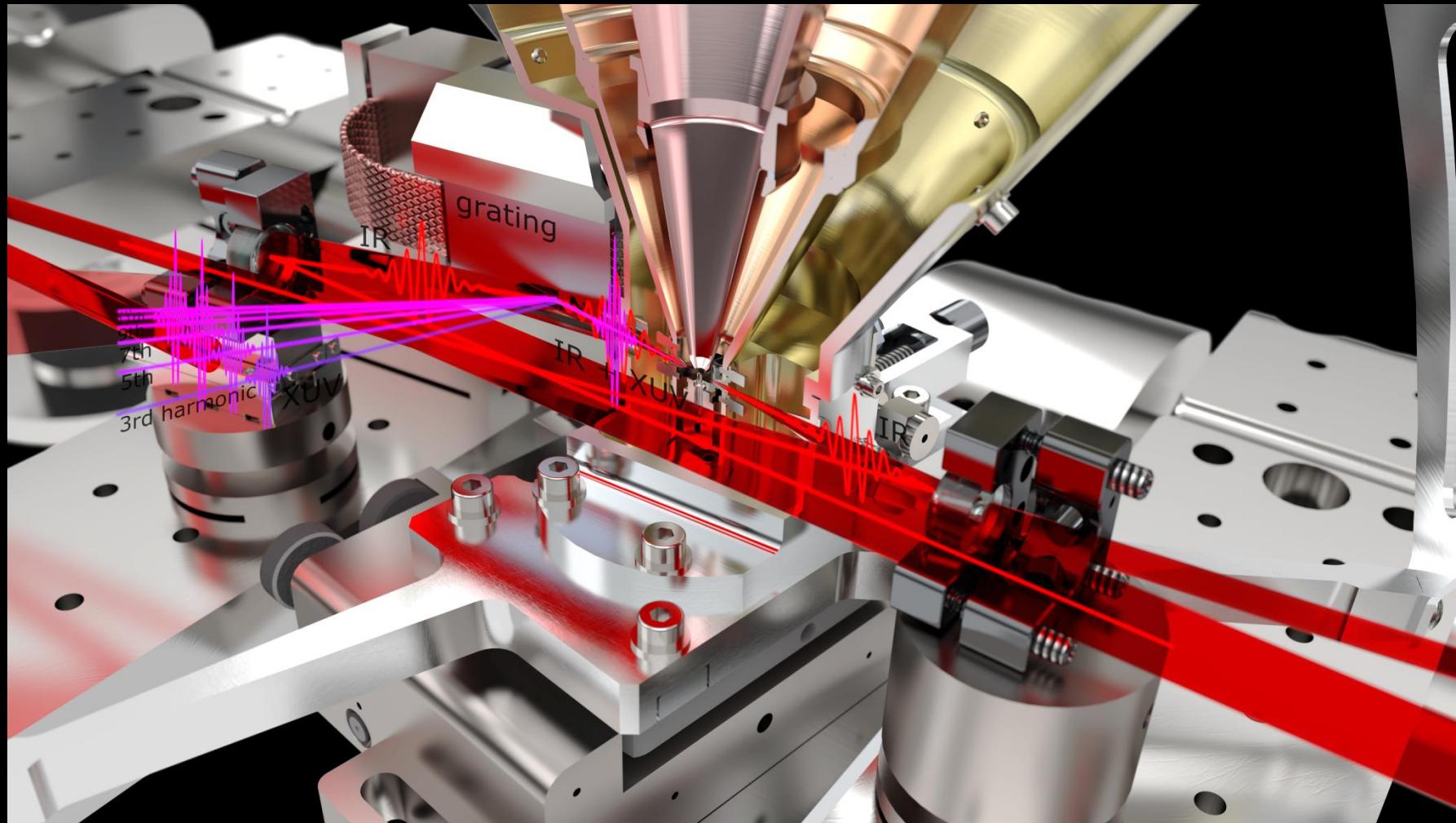
EBIT Hall Area



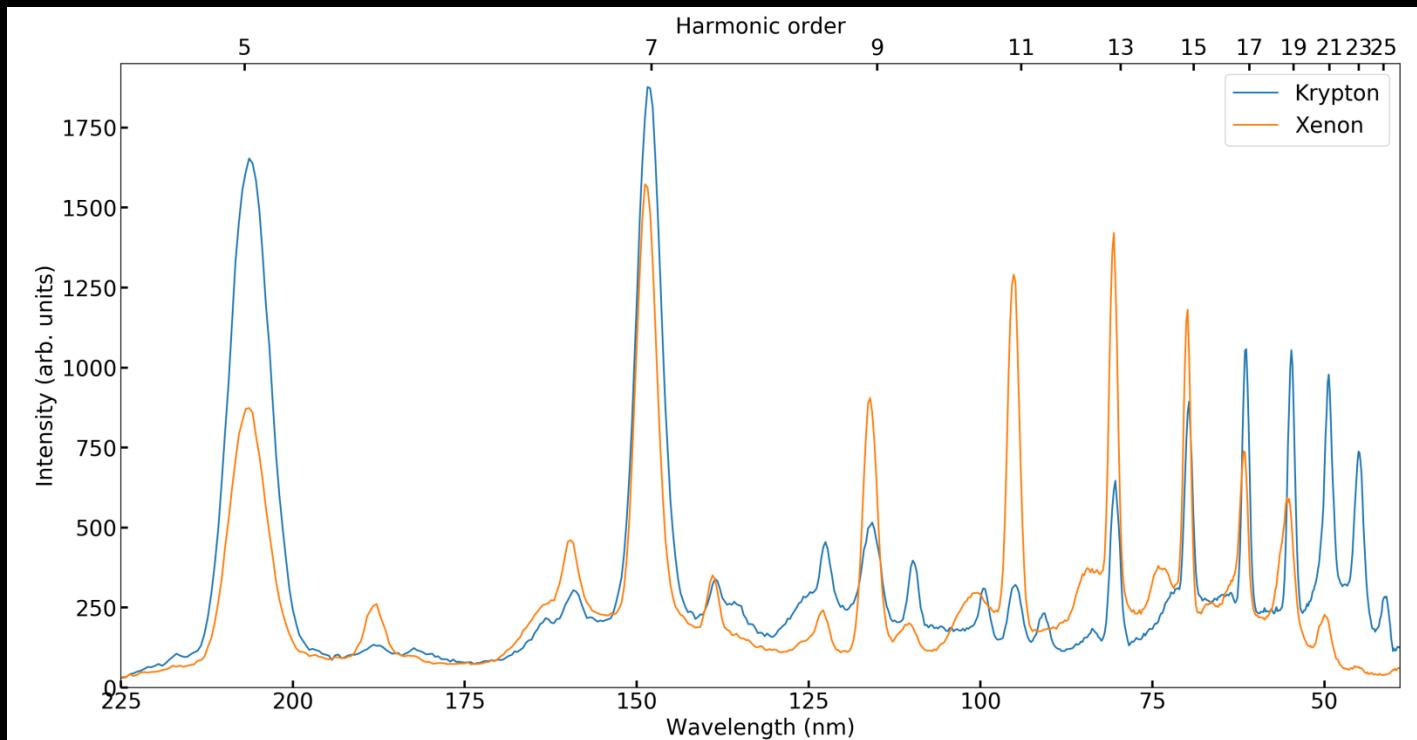
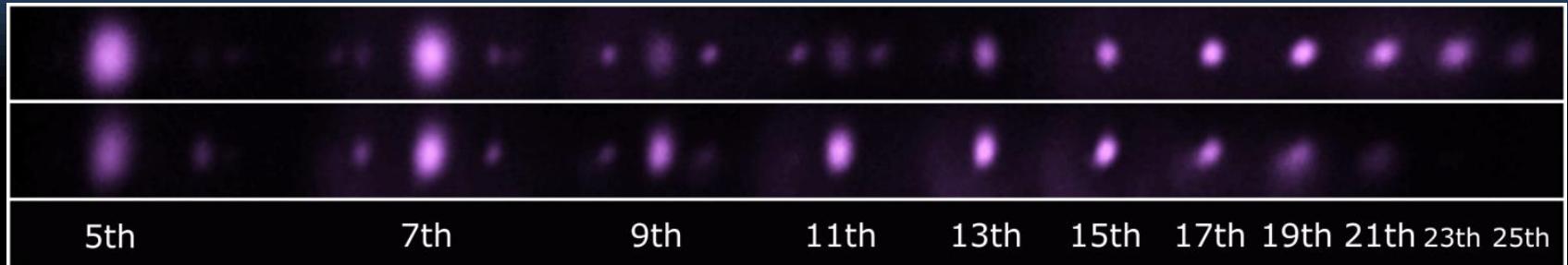
Assembling the cavity vacuum chamber



Design of the HHG interaction region



VUV-comb experimental results



25th harmonic observed with high-density jet on fluorescent screen
(Janko Nauta, Jan-Hendrik Oelmann, José Crespo *et al.*)

VUV-comb experimental results

15th 13th 11th 9th 7th 5th

Cavity scanning, 2.5 kW, Xenon 1.6 bar

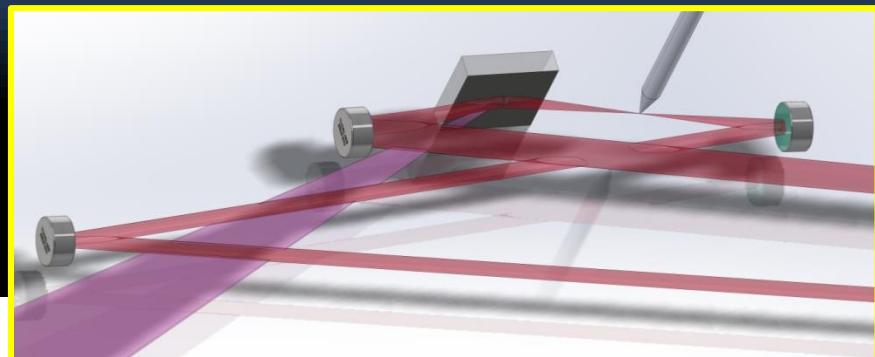
7th 9th 11th 13th 15th 17th 19th 21th 23th 25th 27th 29th 31th 35th

Ar

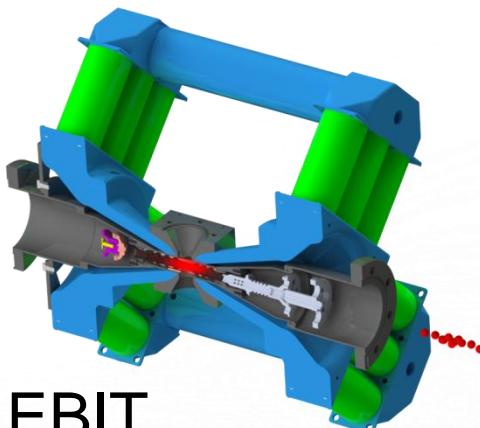
Kr

Xe

Next-step VUV-Spectroscopy of HCl

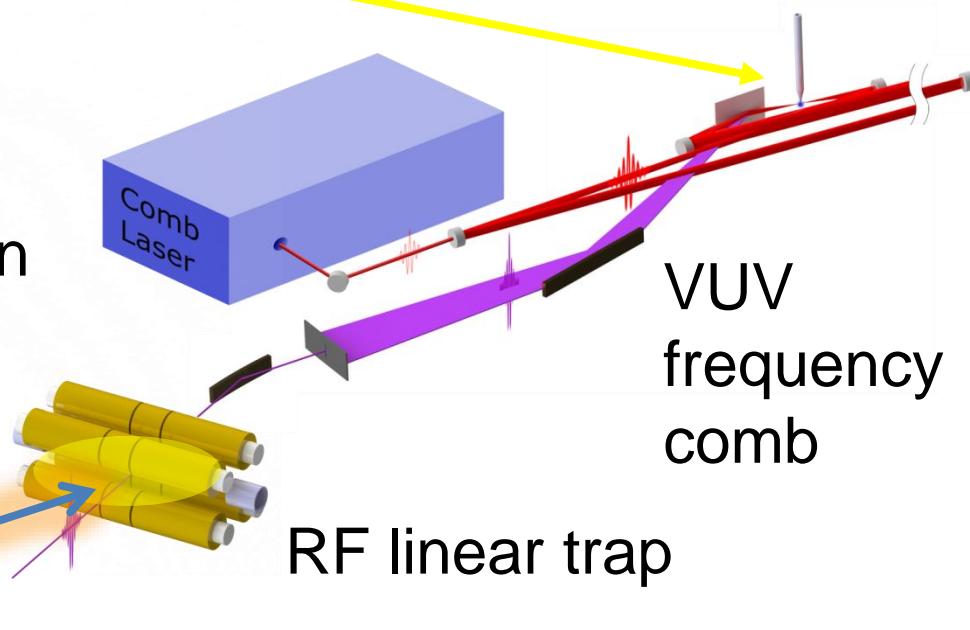
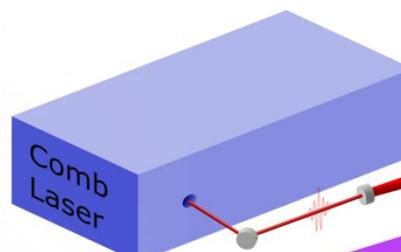
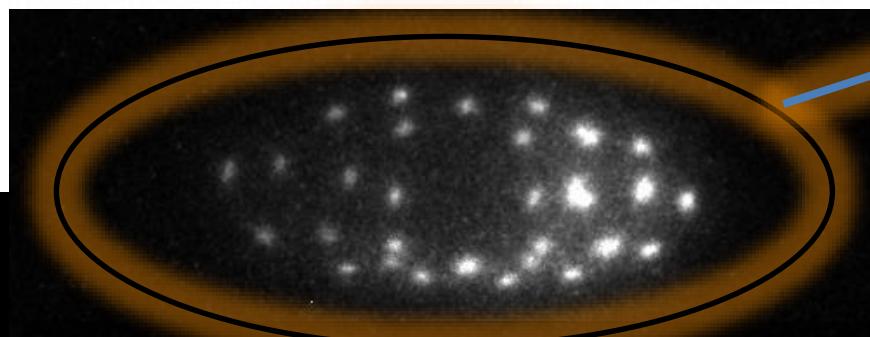


- In-vacuo enhancement cavity
- In $15 \mu\text{m}$ focus: $\approx 10^{13} \text{ W/cm}^2$
- 100 MHz repetition rate

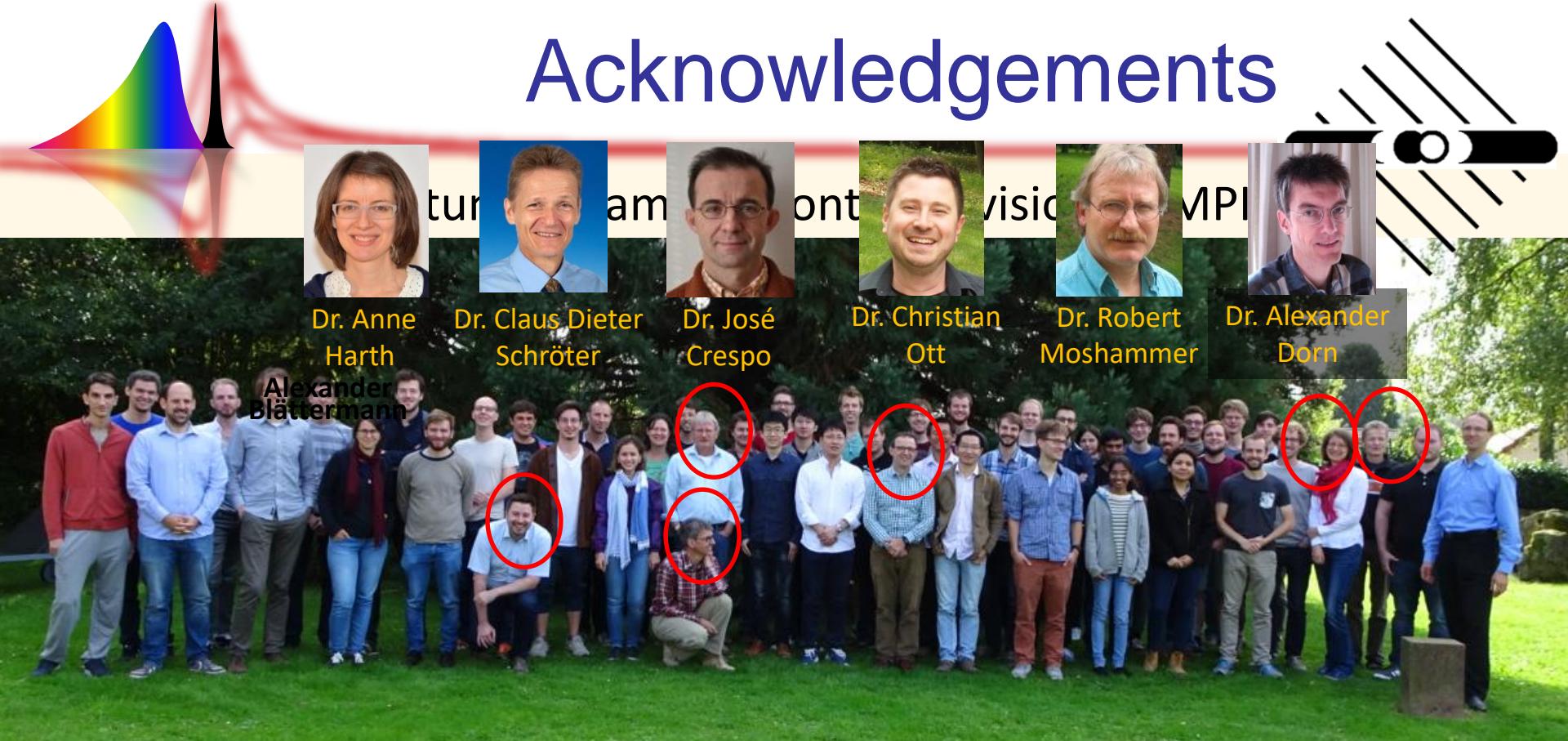


Deceleration

EBIT



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G. Brenner, M. Braune (DESY): FLASH FEL exp.

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Z.-H. Loh (NTU, Singapore): FLASH exp.

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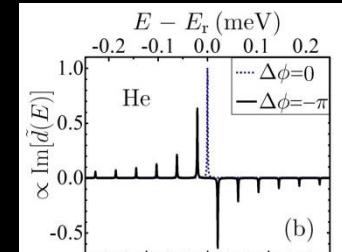
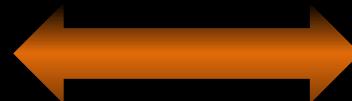
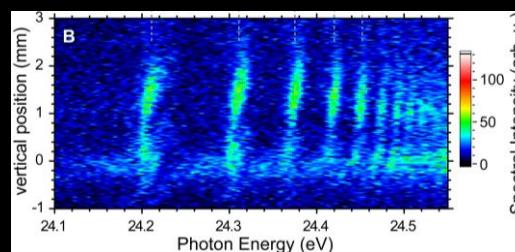
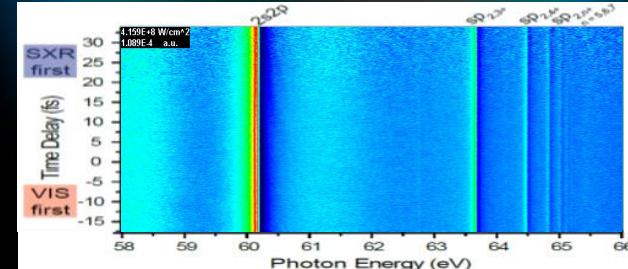


erc
X-MuSiC

Heidelberg Center for
Quantum Dynamics

Summary

- Ultrafast **Laser** quantum control of **small atomic/molecular quantum systems** (natural, well-defined "Å-labs") for fundamental-physics experiments
- Phase **information** is key to quantum dynamics and can be retrieved by time—"resolved" experiments
- qualitatively new insights into fundamental processes (e.g. Fano \leftrightarrow Lorentz, Absorption \leftrightarrow Gain)
=> New ideas...



Understand, and modify, general mechanisms of Quantum Dynamics
Unifying Ultrafast Control and Precision Physics