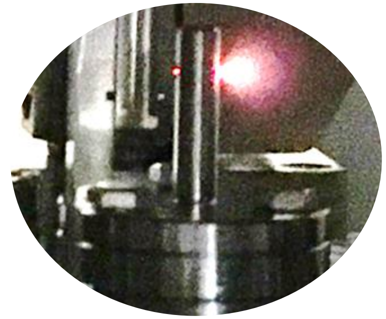
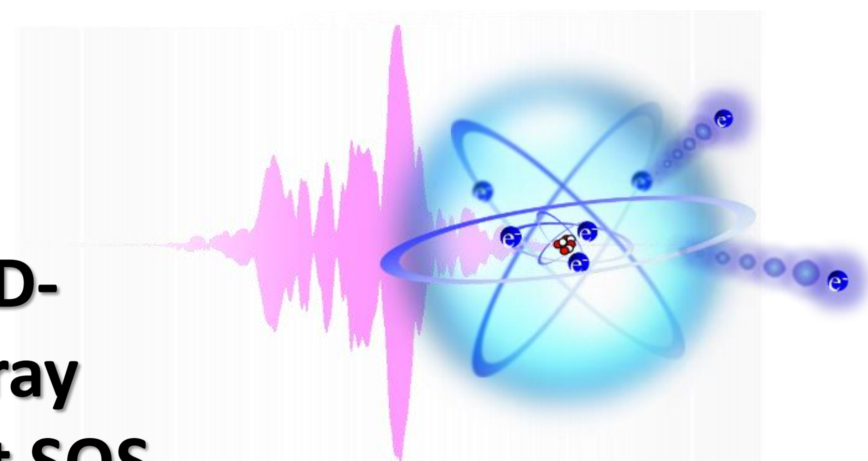
MAX-PLANCK-INSTITUT
FÜR KERNPHYSIK
HEIDELBERG



Absorption response of resonant atomic electron transitions to intense XUV electric fields

MAX-PLANCK-GESELLSCHAFT

Christian Ott



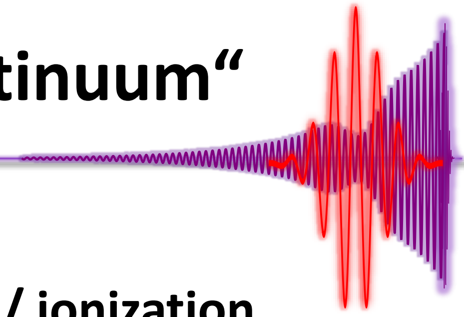
Workshop on 1D-imaging soft X-ray Spectroscopy at SQS

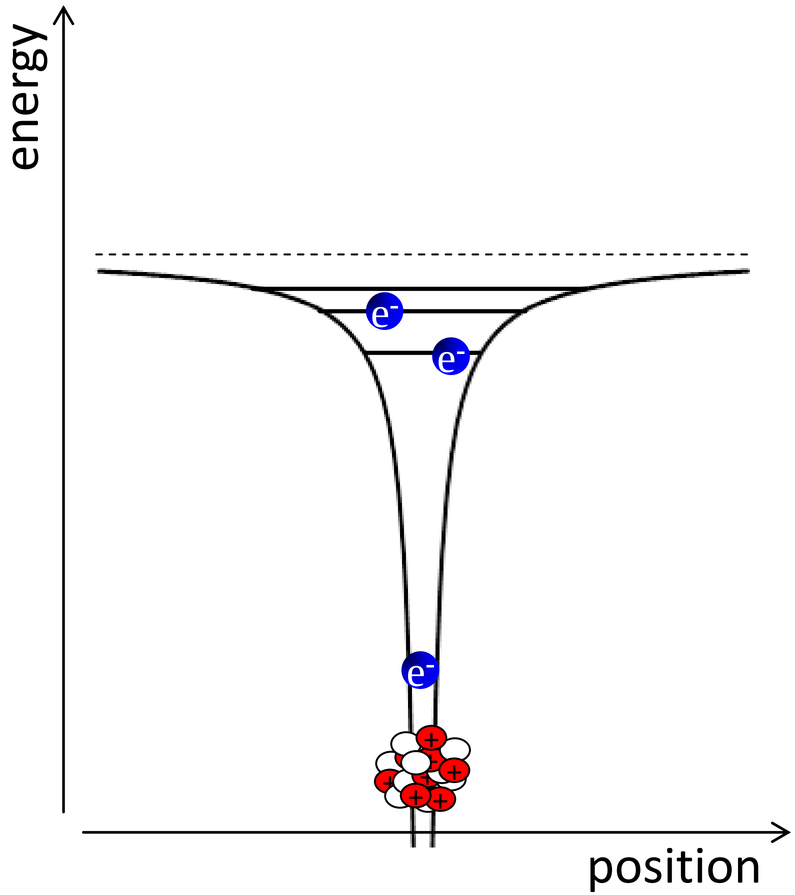
21./22.10.2021

christian.ott@mpi-hd.mpg.de

https://www.mpi-hd.mpg.de/mpi/en/research/scientific-divisions-and-groups/quantum-dynamicscontrol/research/excited-atomsmolecules-in-strong-fields-ag-ott

XUV and x-ray wavelengths: „living in the continuum“





- **Inner-shell excitation / ionization**
- **Auger-Meitner / autoionization (also inter-atomic in molecule)**

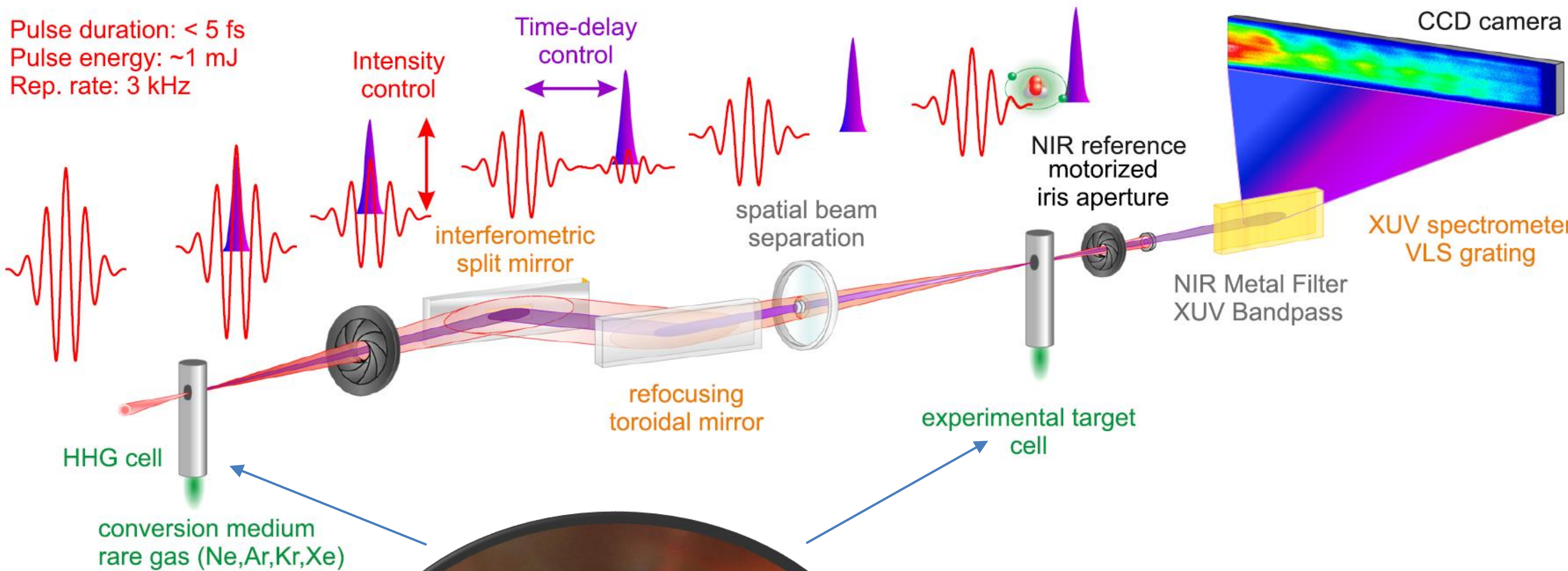
  “carries away” the coherence

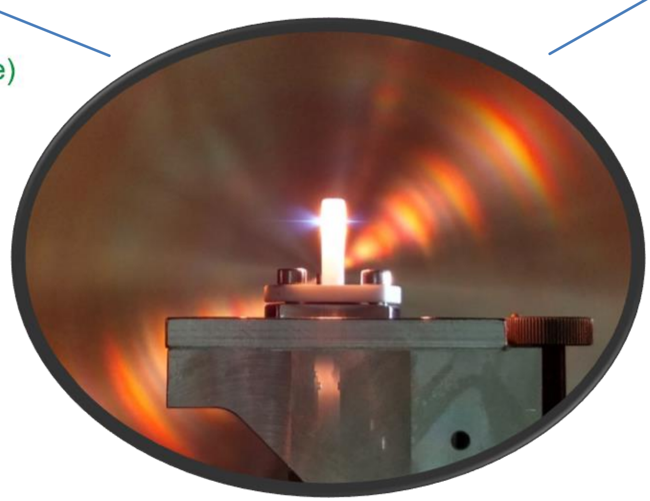
➢ **Typically ultrashort coherence times on few-femtosecond (even attosecond) timescale**

Resonant nonlinear XUV / x-ray light-matter interaction: probing and controlling the coherent ultrafast electronic dynamics in atoms and molecules

(NIR)-Laser-Control of XUV resonances

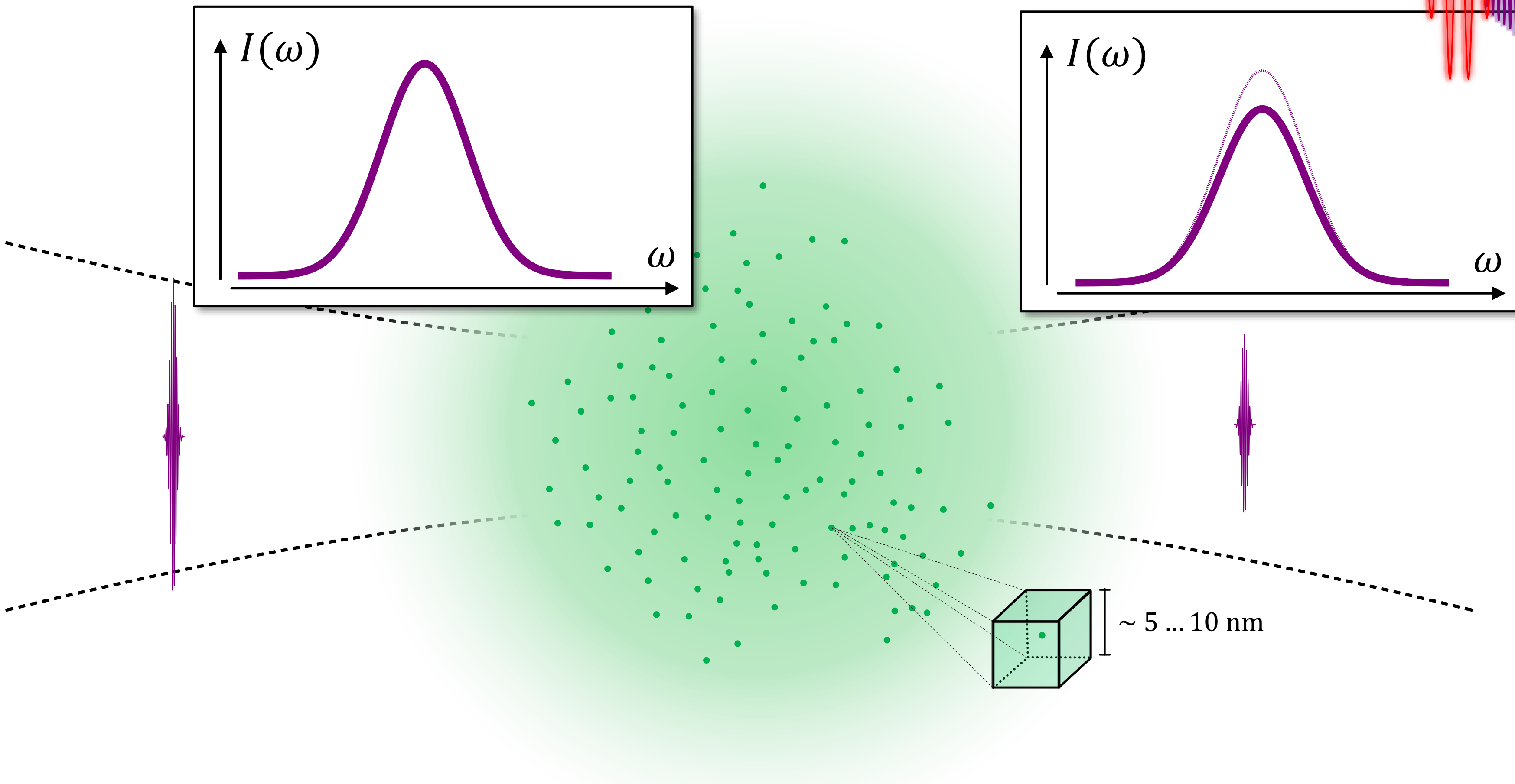
Experimental Setup: V. Stooß et al., Rev. Sci. Instrum., (2019), **90**, 053108





Laser control of attosecond electron dynamics near XUV resonant transitions

Few basics of light-matter interaction in dense media

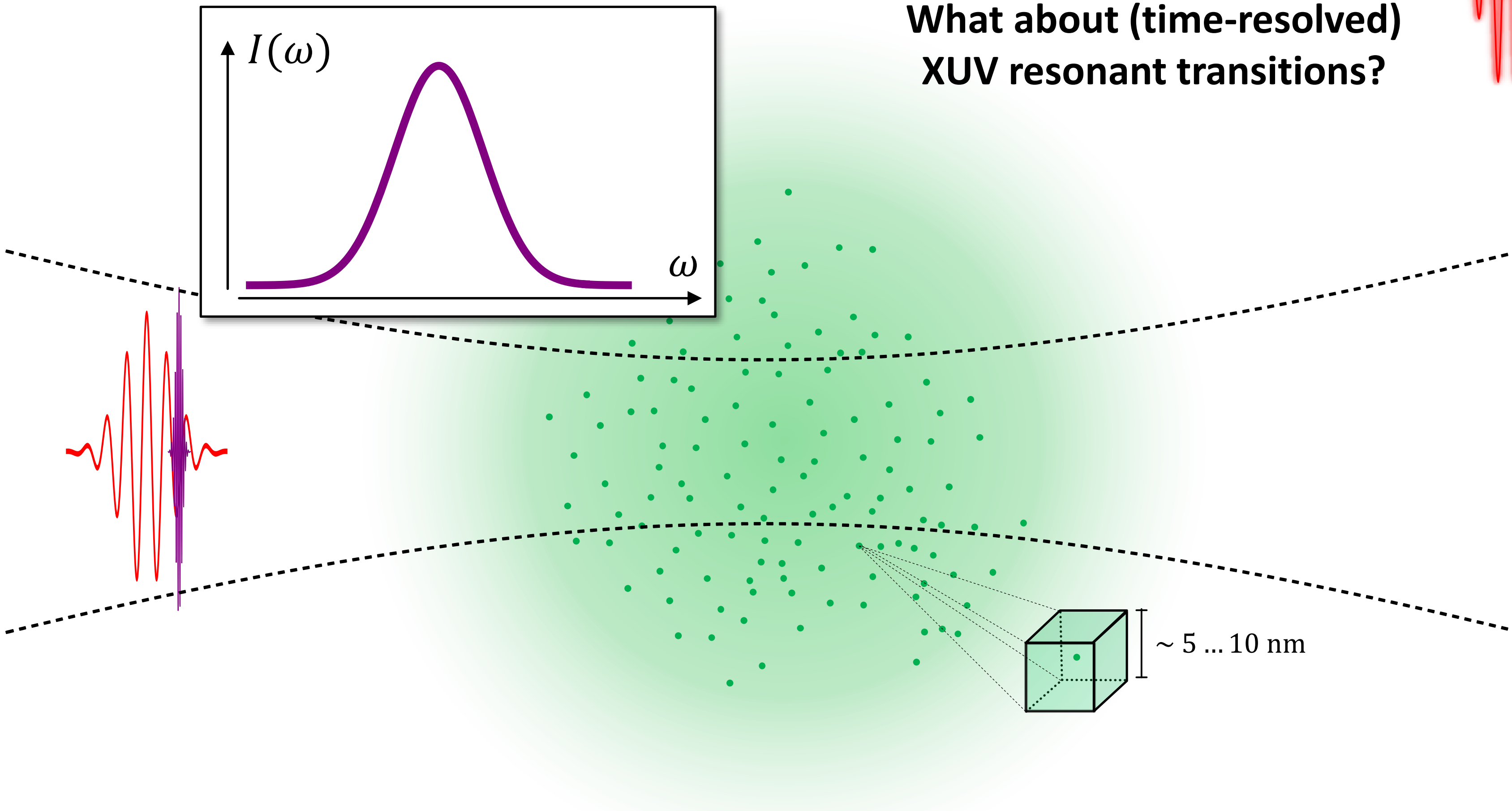


Beer-Lambert law of attenuation:

$$I(\omega) = I_0(\omega)e^{-\rho L\sigma(\omega)}$$

Few basics of light-matter interaction in dense media

What about (time-resolved) XUV resonant transitions?

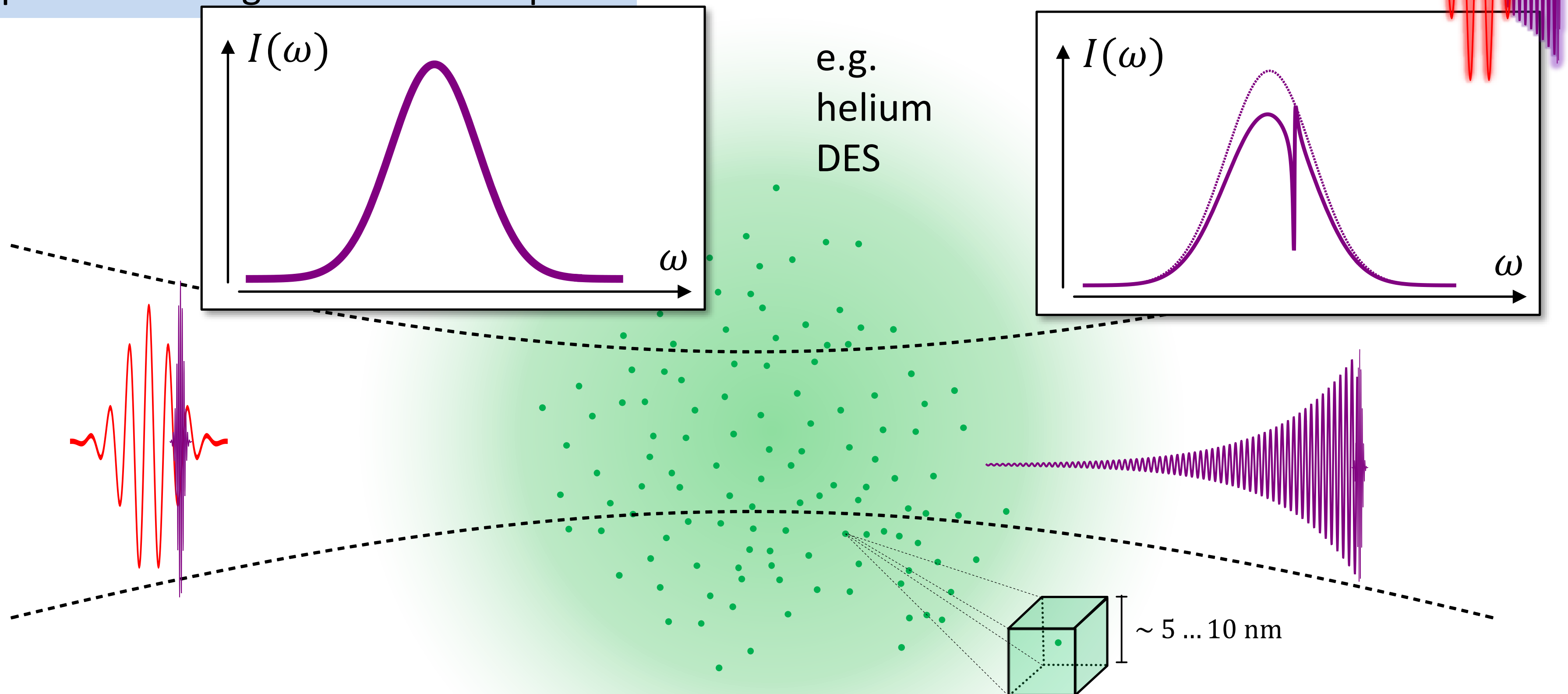


Beer-Lambert law of attenuation:

$$I(\omega) = I_0(\omega) e^{-\rho L \sigma(\omega)}$$

Controlling the dipole response function

Example: Controlling resonant absorption



$\vec{d}(t) = \langle \Psi(t) | \vec{\mu} | \Psi(t) \rangle$

Optical Density $\propto \Im[n(\omega)] \propto \Im[\chi(\omega)] \propto \Im[\tilde{d}(\omega)]$

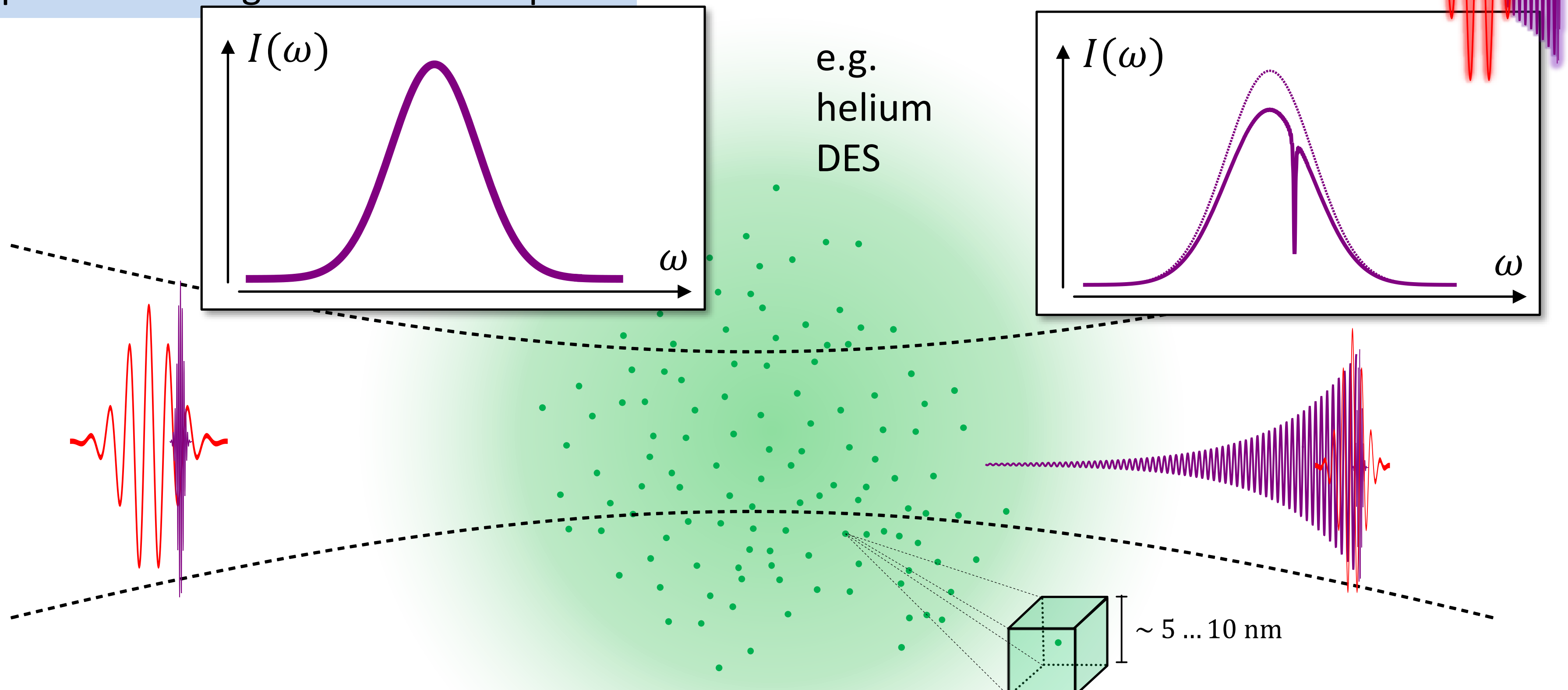
$$\mathrm{OD}(\omega) = \log\left[\frac{I_0(\omega)}{I(\omega)}\right] = \frac{\rho L}{\ln(10)} \frac{\omega}{\varepsilon_0 c} \Im\left[\frac{\tilde{d}(\omega)}{\tilde{\mathcal{E}}(\omega)}\right]$$

(for dilute media, i.e. not too dense)

Maxwell's propagation of radiation fields; refractive index; phase matching and phase sensitivity; interference!

Controlling the dipole response function

Example: Controlling resonant absorption



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(for dilute media, i.e. not too dense)

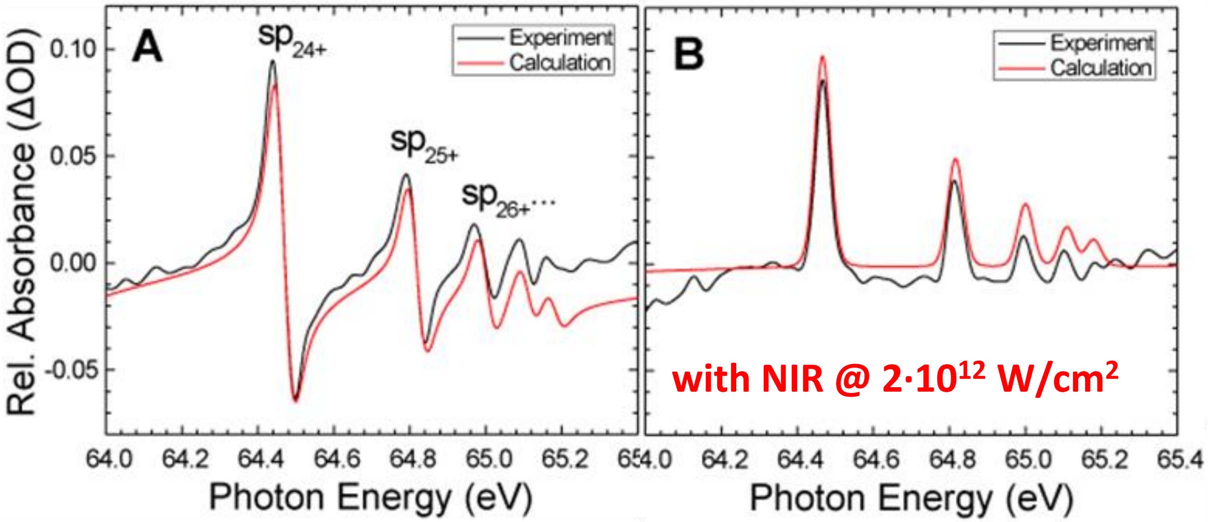
Maxwell's propagation of radiation fields; refractive index; phase matching and phase sensitivity; interference!

Controlling spectral lineshapes (Fano <> Lorentz)

in the helium atom

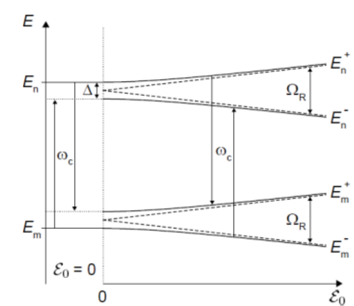
Control the phase of autoionizing two-electron (Fano) dipole response

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

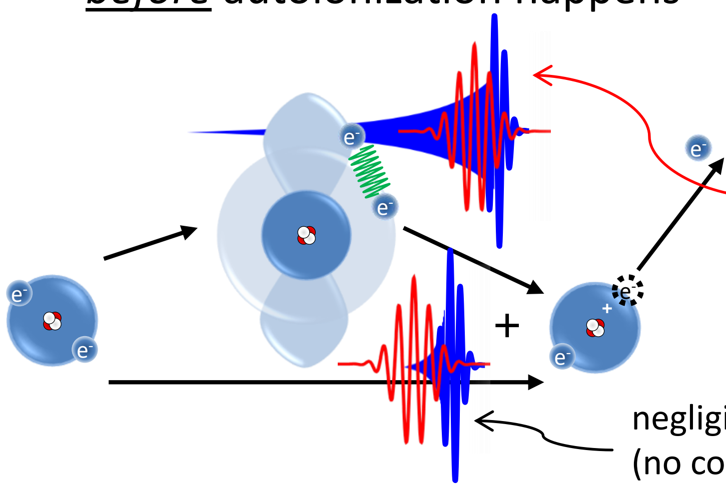


$$\varphi(q) = 2\arg(q - i)$$

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



- selectively modify the two-electron excited state *before* autoionization happens



coherent sum:
bound & continuum XUV dipole response

NIR-induced energy shift $\Delta E(t)$ [e.g., AC Stark, Rabi, **strong-field effects**] of excited-state (dipole reponse):

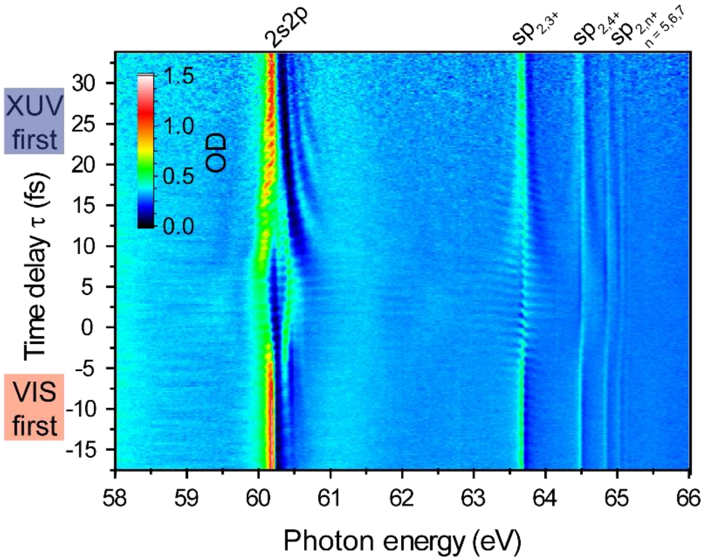
$$\Delta\varphi \sim \int_{T_{\mathrm{NIR}}} \mathrm{d}t\, \Delta E(t)$$

negligible
(no continuum dipole during NIR)

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

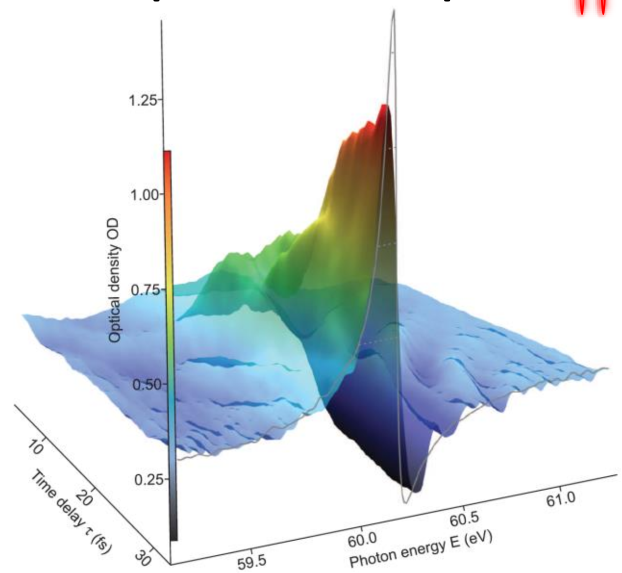
A real-time view into two-electron dynamics

Observing and controlling a two-electron wavepacket



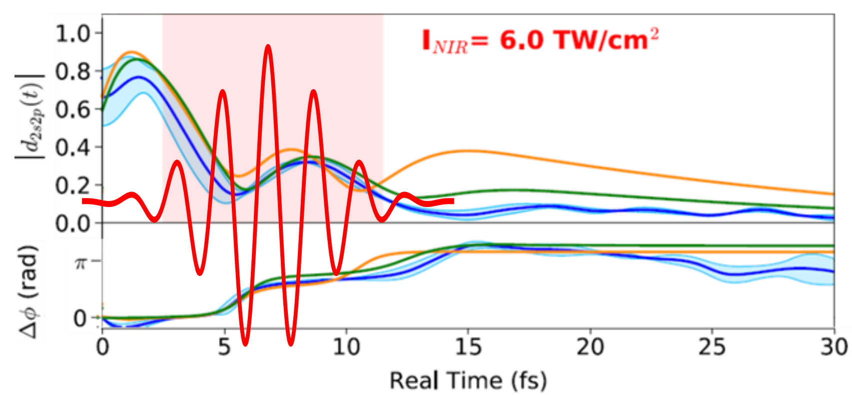
C. Ott et al., Nature **516**, 374 (2014)

Buildup of a Fano spectral lineshape



A. Kaldun, A. Blättermann et al., Science **354**, 738 (2016)

A view into laser-driven (Rabi cycling) two-electron dynamics

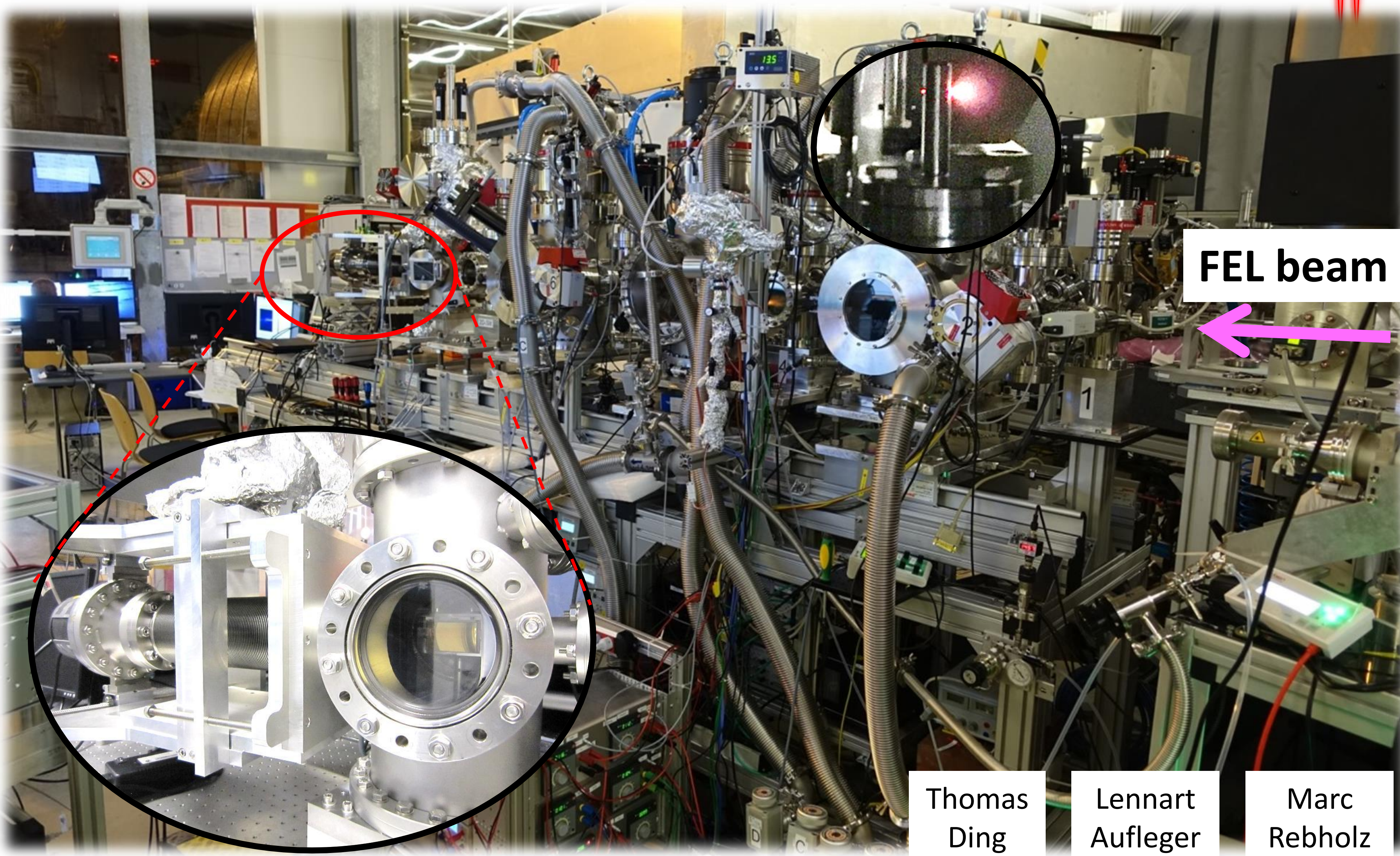


V. Stooß et al., PRL **121**, 173005 (2018)

How do atom-specific resonances look when viewed through the interaction with intense XUV-only radiation fields?

The experimental apparatus @ FLASH, BL2

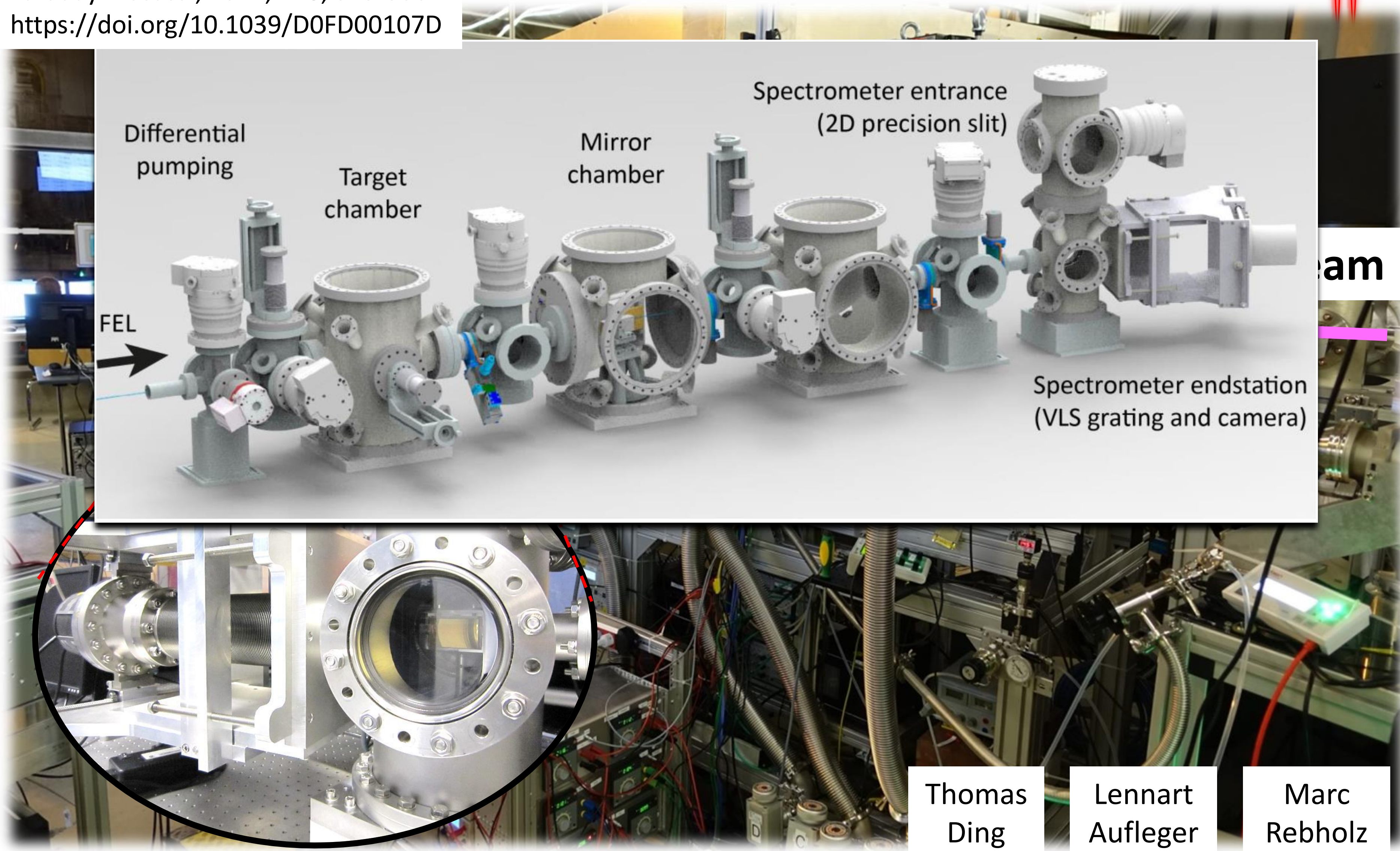
Free-Electron-Laser @DESY in Hamburg, Germany



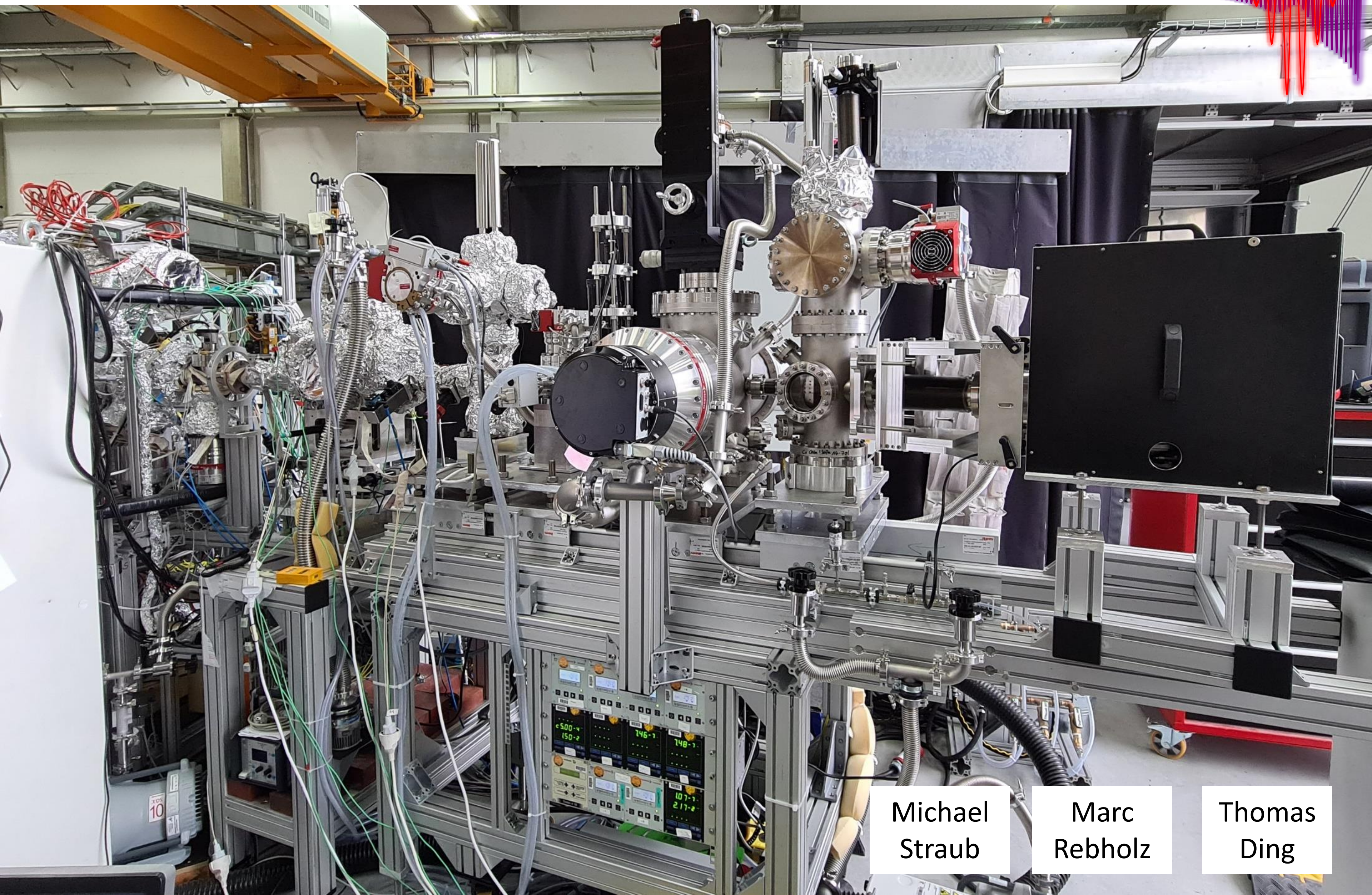
The experimental apparatus @ FLASH, BL2

T. Ding, M. Rebholz, L. Aufleger, et al.,
Faraday Discuss., 2021, **228**, 519-536
https://doi.org/10.1039/D0FD00107D

Free-Electron-Laser @DESY in Hamburg, Germany



… now installed at FL26 behind REMI at FLASH2

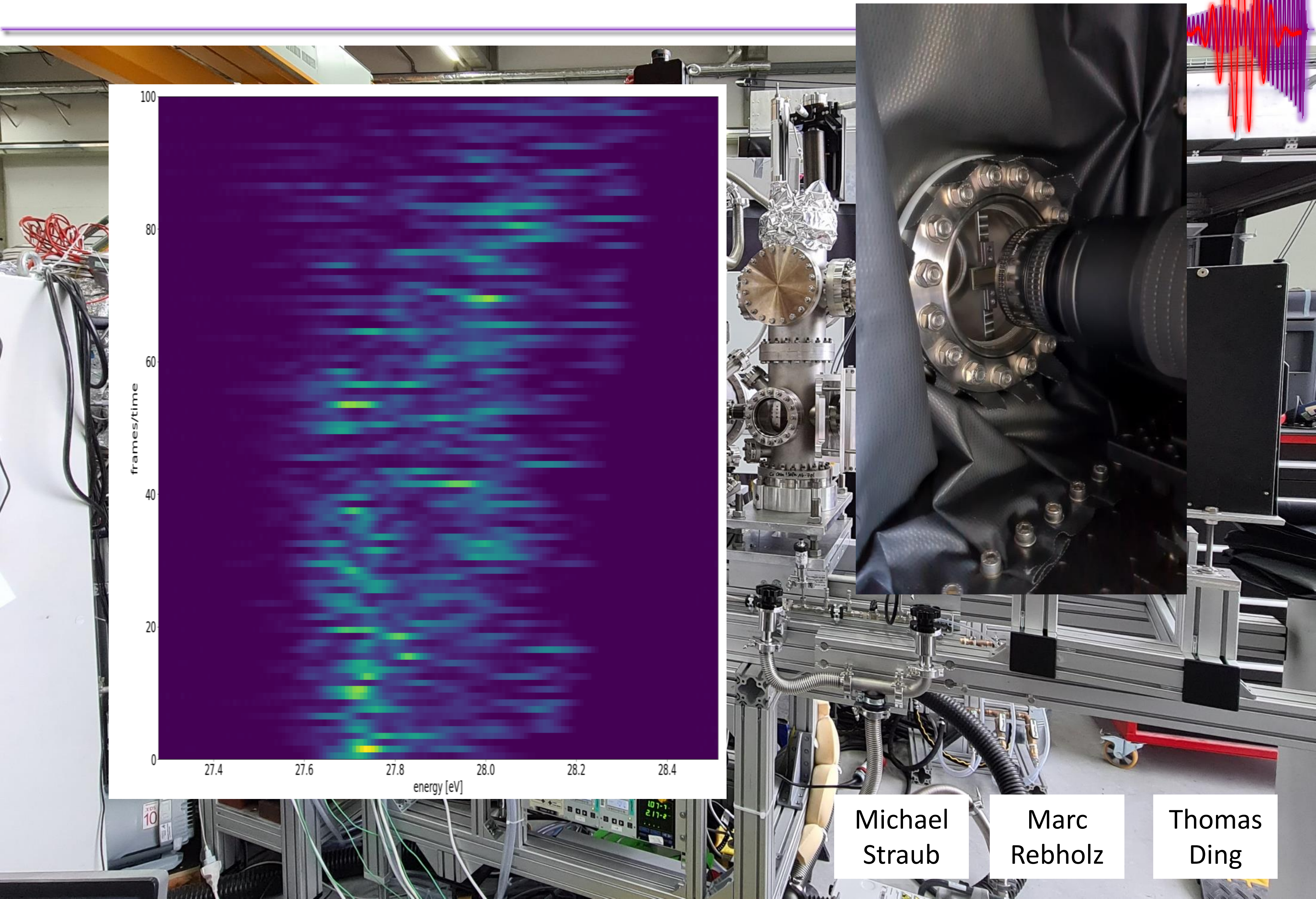


Michael Straub

Marc Rebholz

Thomas Ding

… now installed at FL26 behind REMI at FLASH2

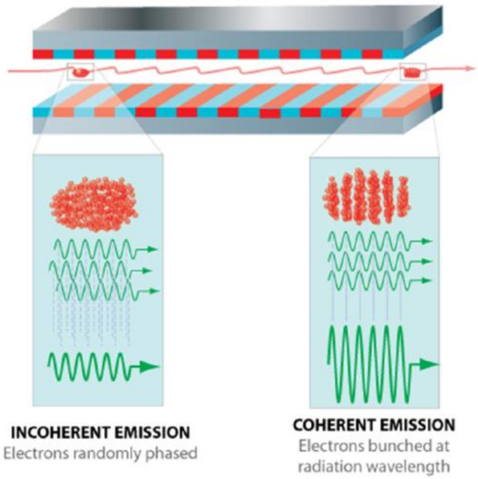


Michael Straub

Marc Rebholz

Thomas Ding

Partially coherent SASE FEL Pulses

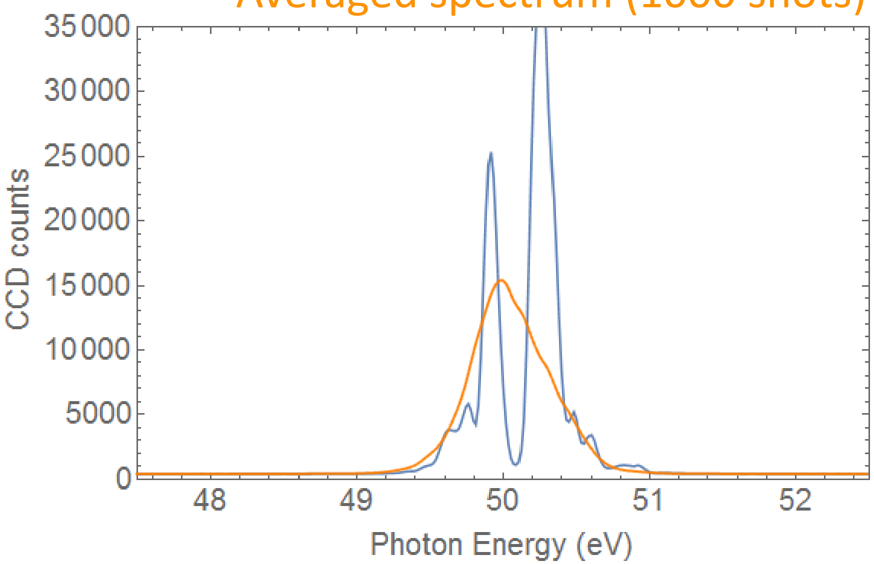


Self-amplified spontaneous emission: XUV-FEL pulses amplified from noise, temporally constrained to electron bunch duration (-> partial coherence)

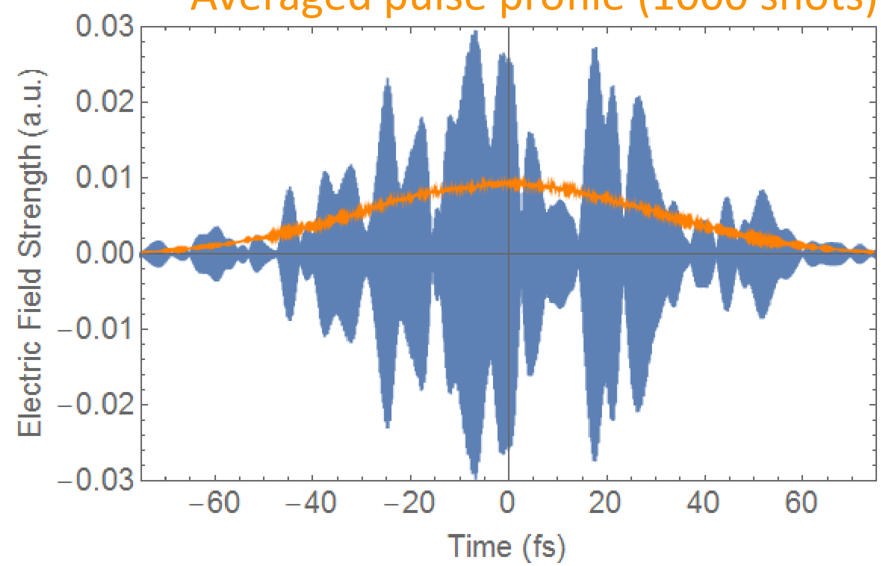
Seddon *et al* 2017 *Rep. Prog. Phys.* **80** 115901

*Partial-coherence model:
T. Pfeifer *et al.,* Opt. Lett. **35**, 3441-3443 (2010)

Single SASE spectra ***measured @ FLASH***
Averaged spectrum (1000 shots)

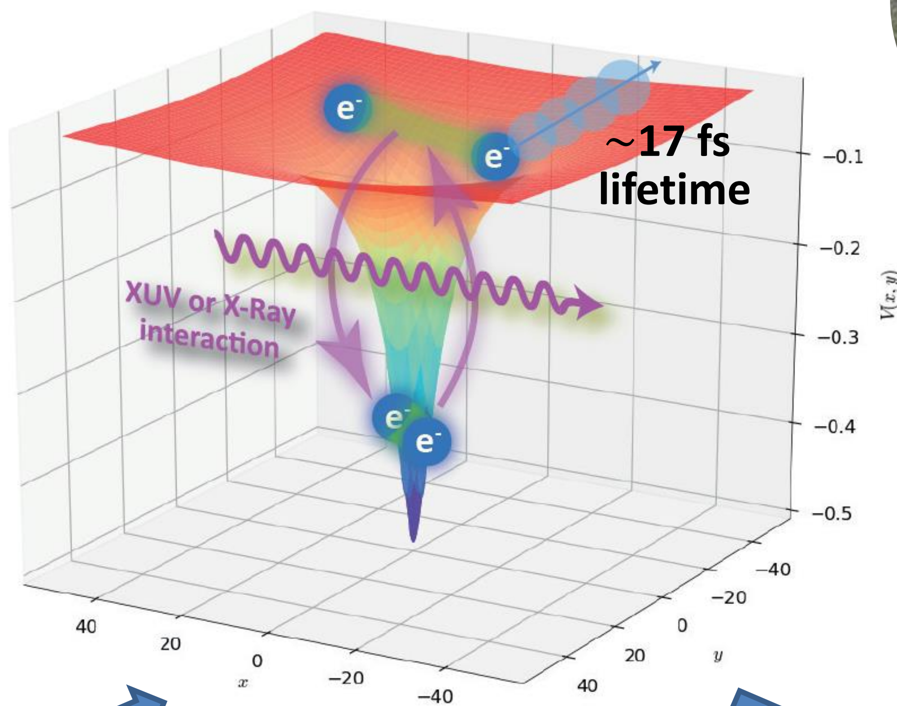


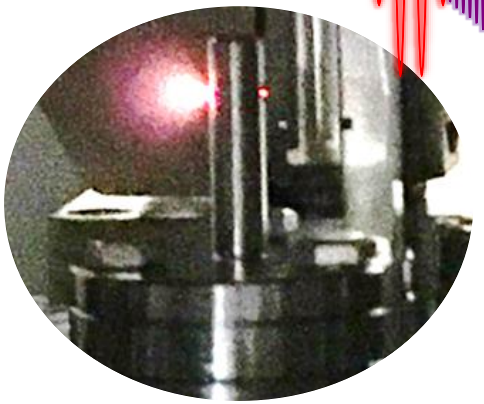
Single SASE pulses ***(simulated*)***
Averaged pulse profile (1000 shots)

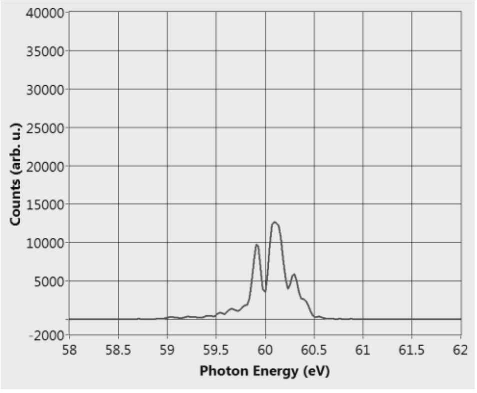


Intense XUV at Free-Electron Laser in Hamburg (FLASH)

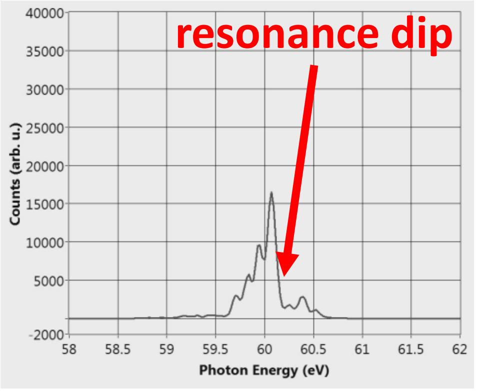
A one-photon two-electron transition in helium viewed in intense XUV light







incident



transmitted

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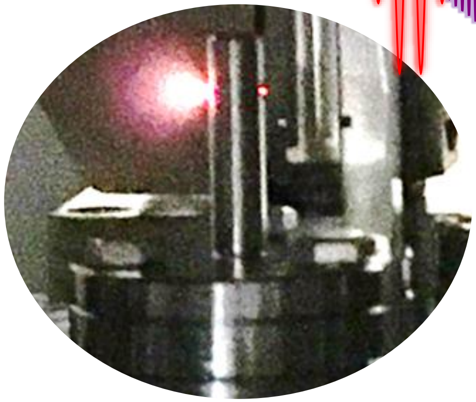
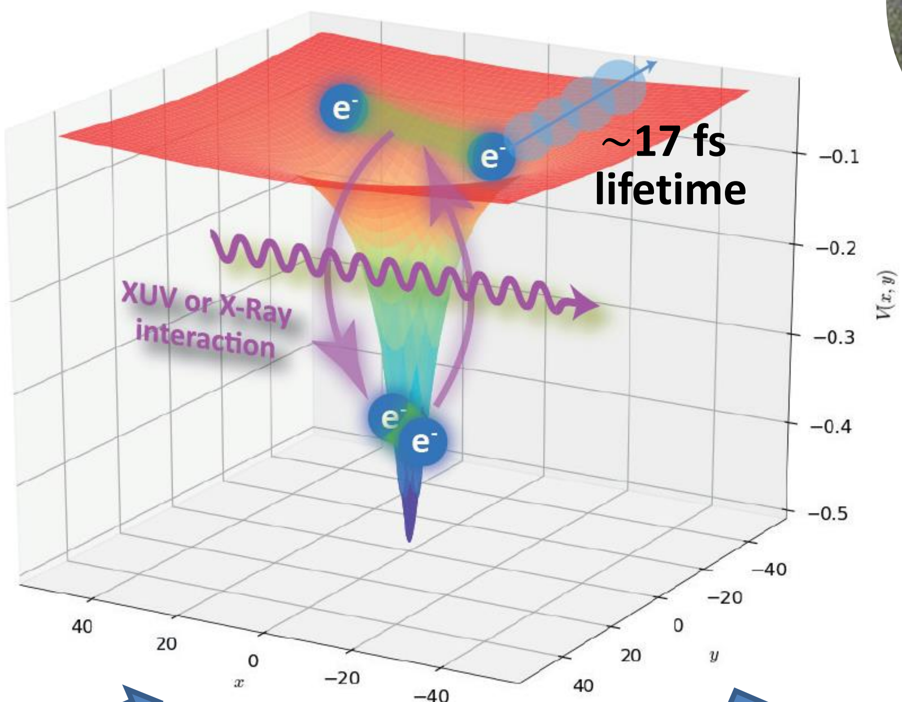
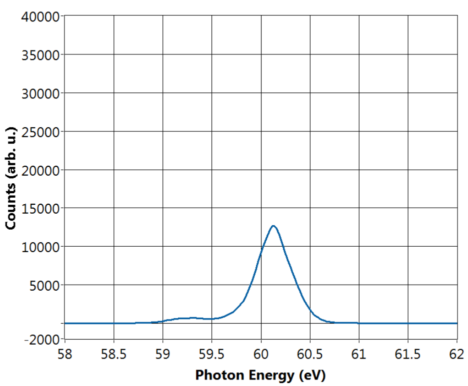
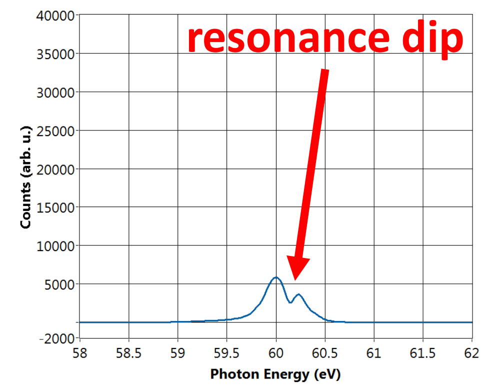






incident



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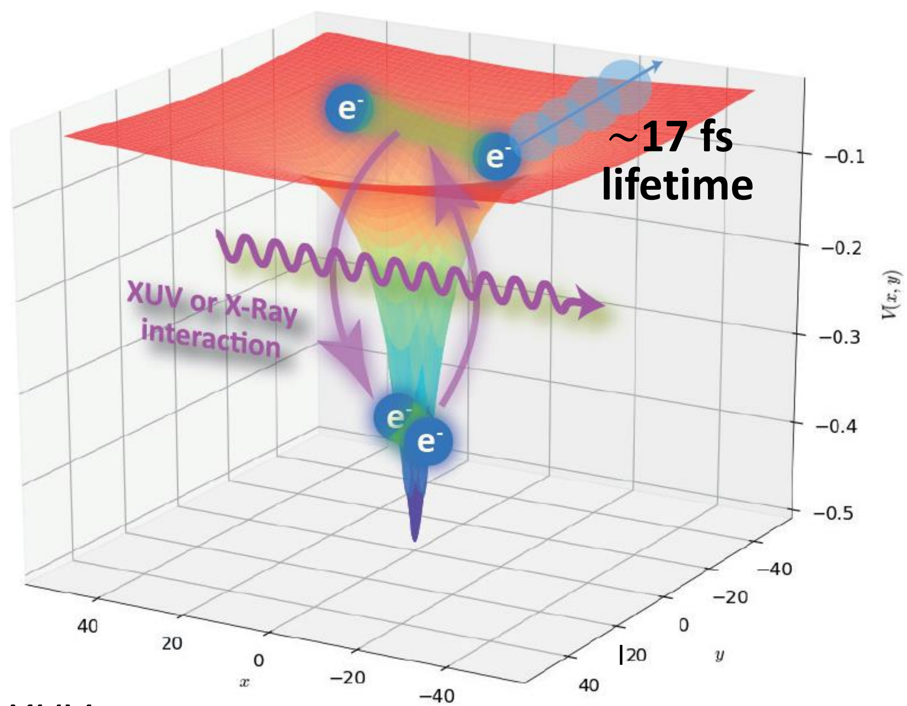
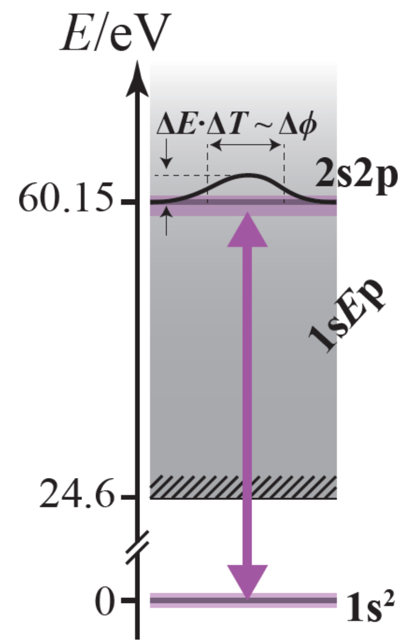
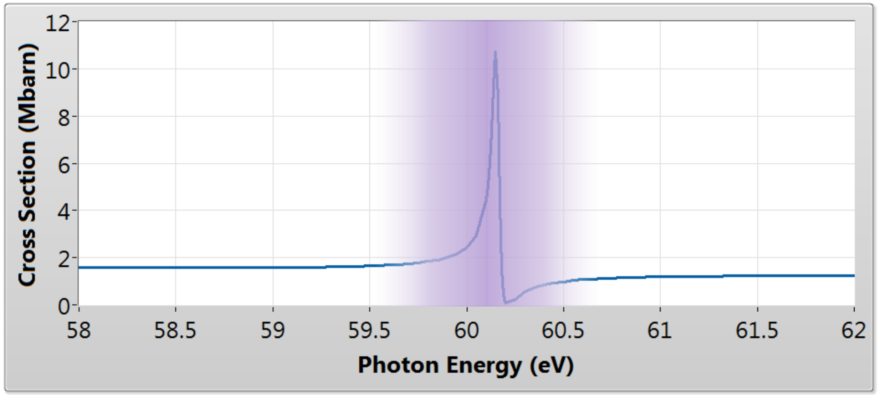
Intense XUV at Free-Electron Laser in Hamburg (FLASH)





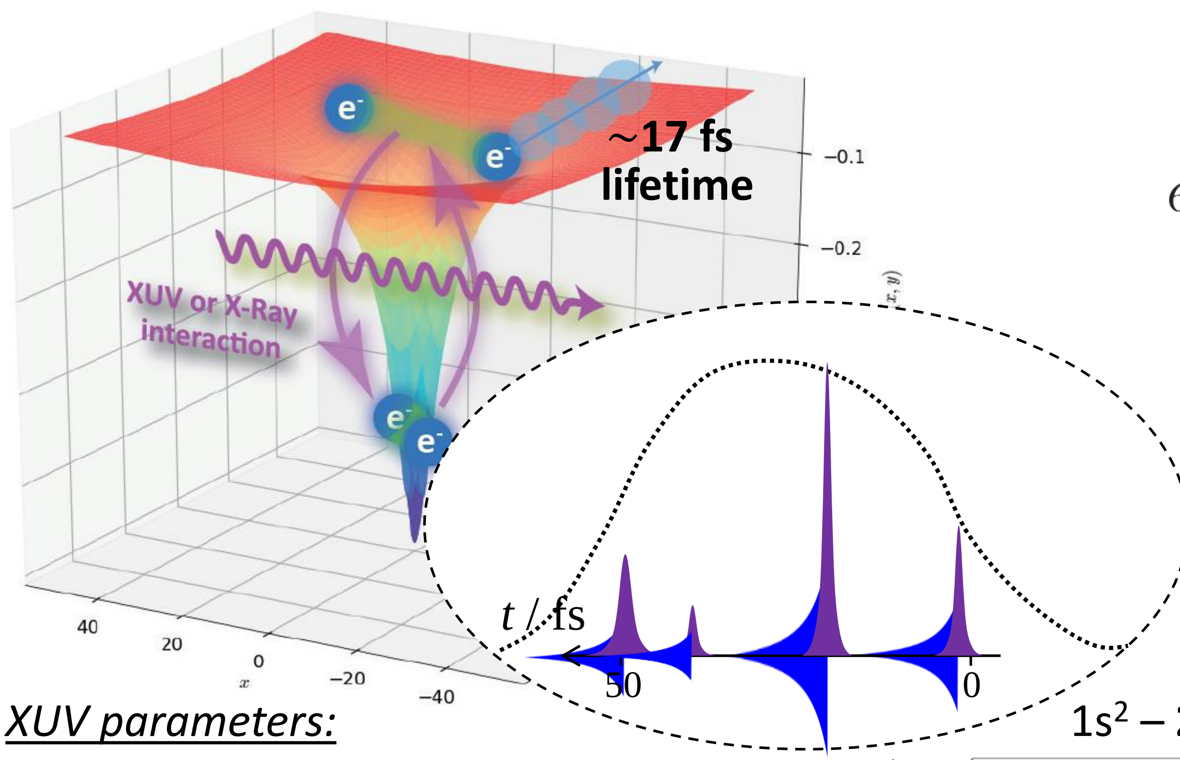
$1s^2$ – **2s2p** double excitation in **helium**

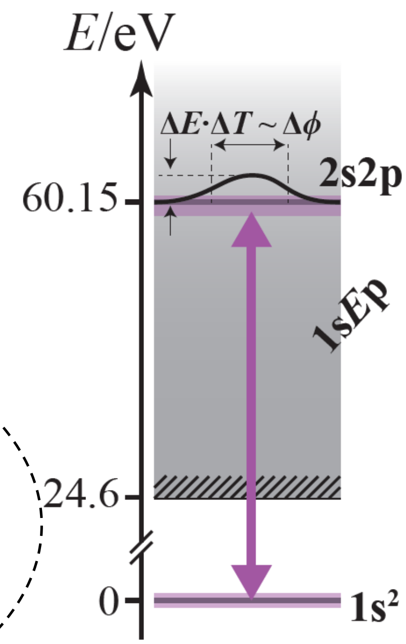


XUV parameters:

rep. rate:	single bunch (10 Hz)
pulse energy:	up to ~50 μJ on target
focused beam size:	~25 μm
photon fluence:	→ up to ~8 J/cm²
photon energy:	60.1 eV
SASE bandwidth:	~0.4 eV
avg. duration:	~50 … 100 fs

Intense XUV at Free-Electron Laser in Hamburg (FLASH)





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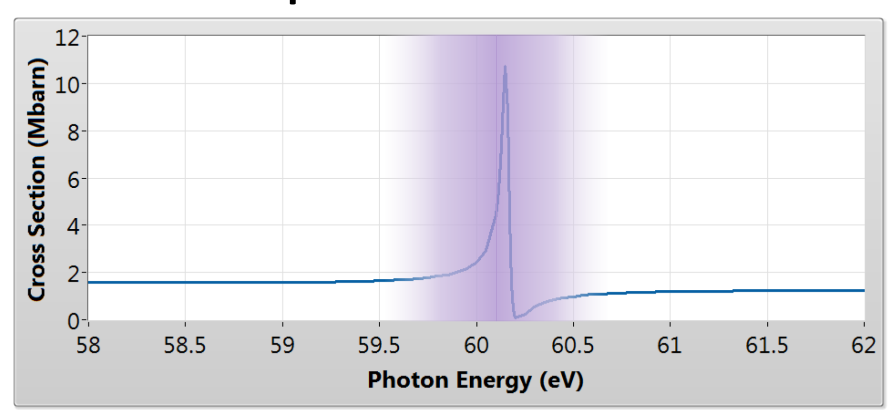
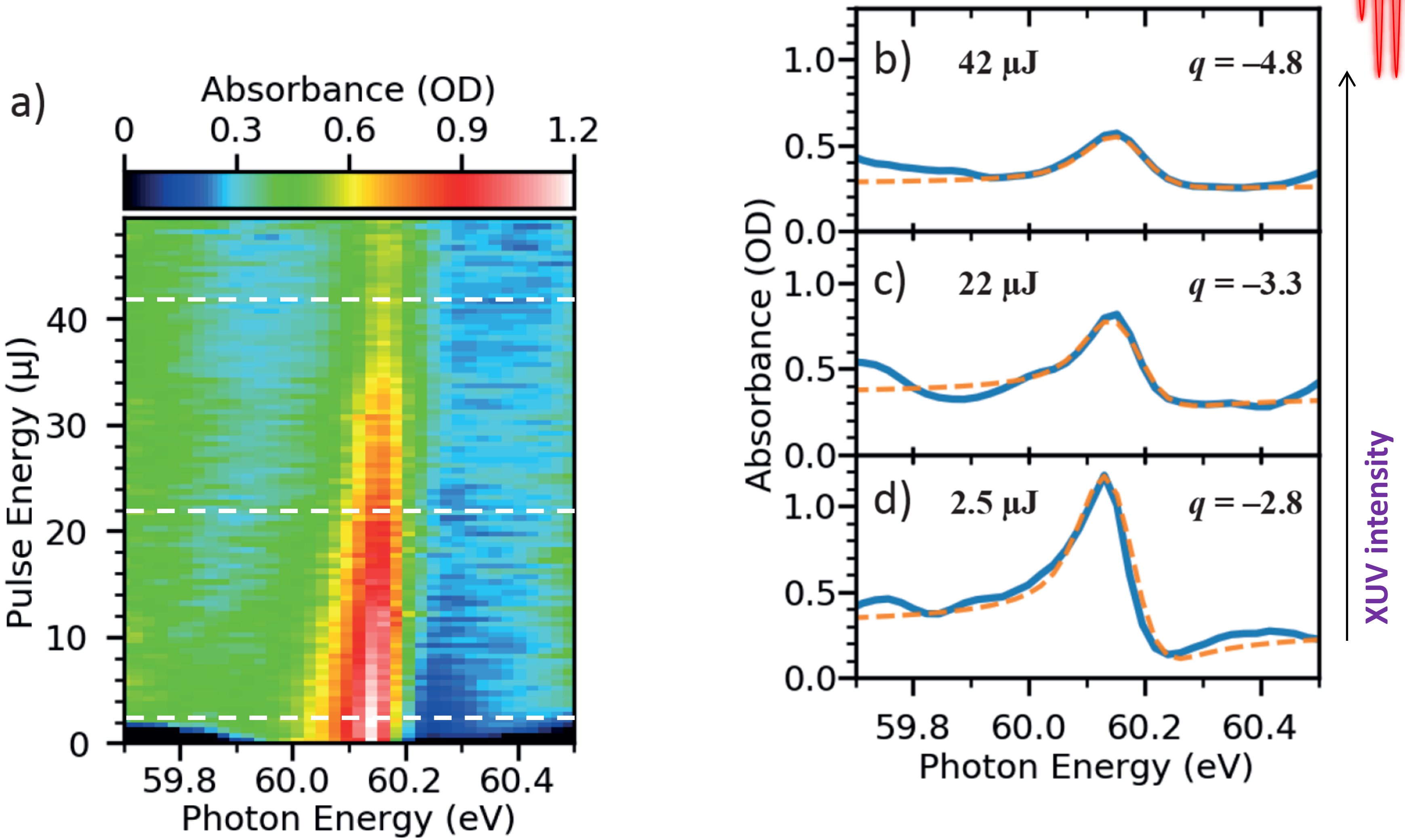


Transient Absorption at high XUV intensity

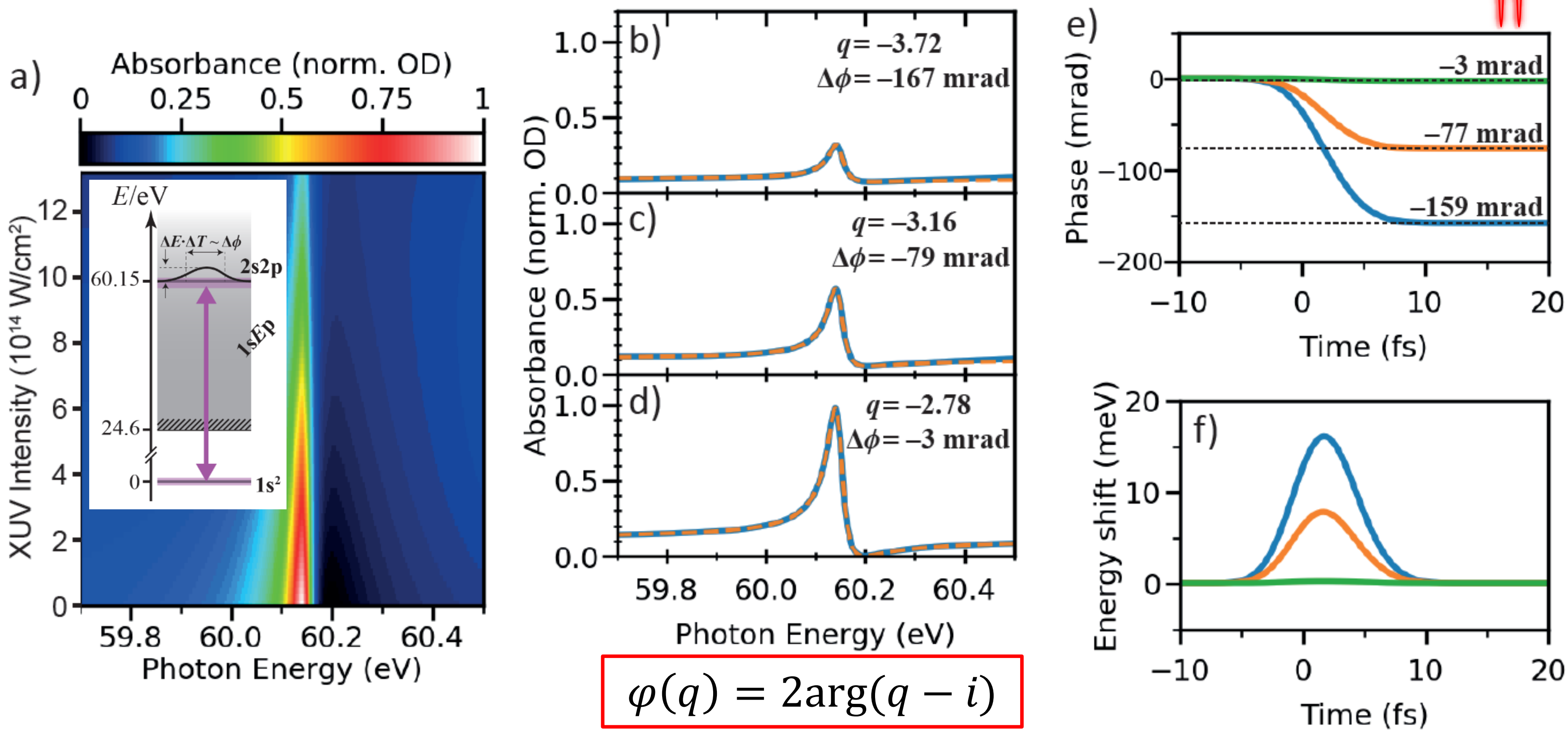


- observe systematic trend to more symmetric lineshape

C Ott et al., PRL 123, 163201 (2019)

XUV-Strong-Coupling of 1s² – 2s2p

- Model with 5-fs (FWHM) Gaussian-shaped pulse centered at 60.10 eV



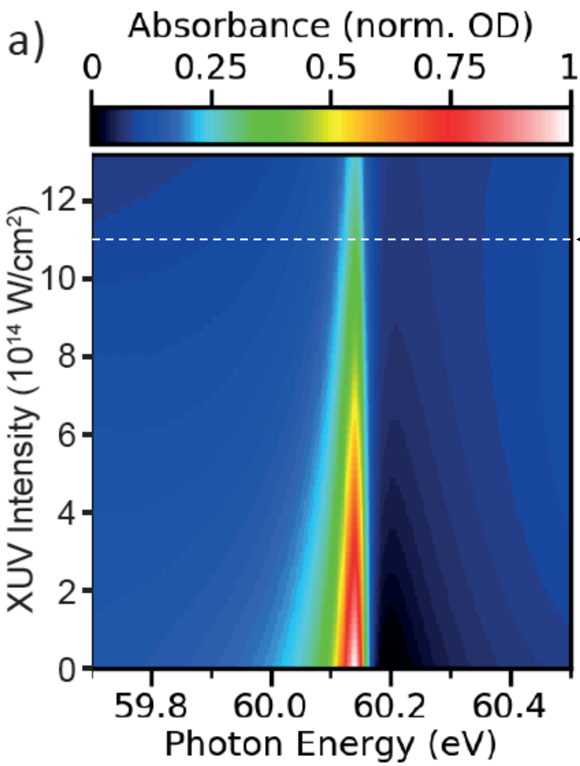
$$\varphi(q) = 2\arg(q - i)$$

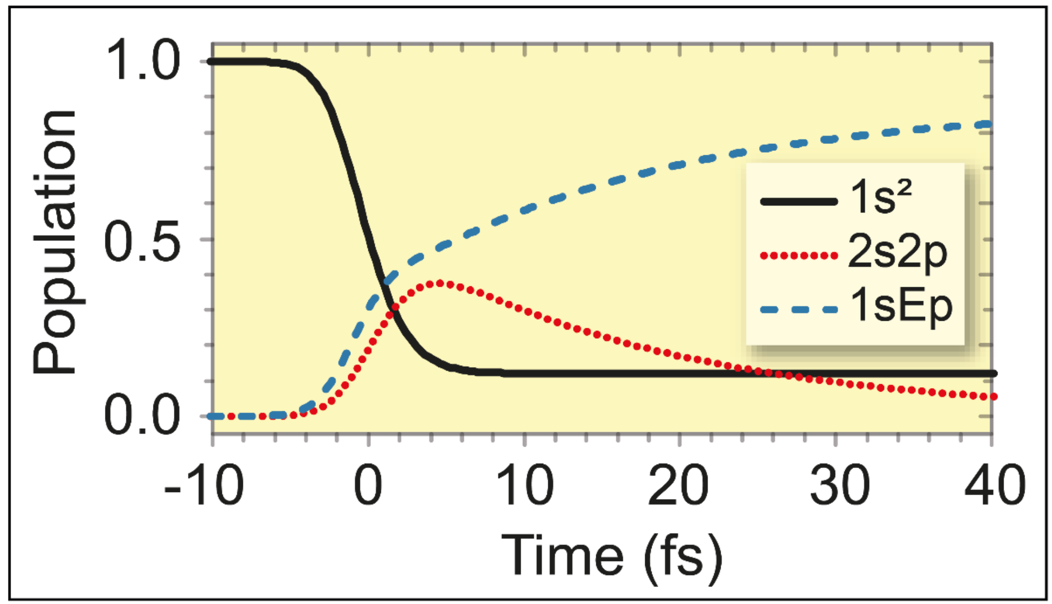
- XUV-induced dressing leads to transient energy shifts during short pulse
- accumulates into phase shift of the 1s² – 2s2p dipole response, which can be measured through the line-shape asymmetry in transient absorption

C Ott et al., PRL 123, 163201 (2019)

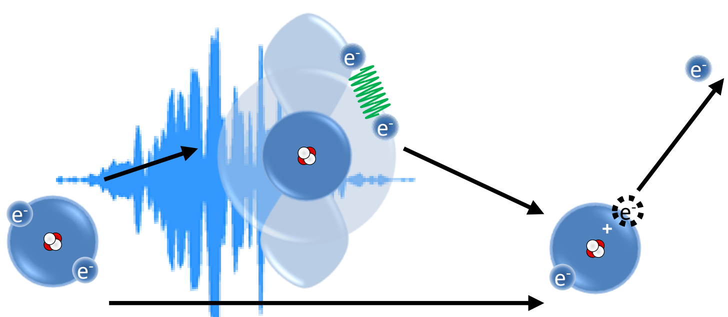
XUV-Strong-Coupling of 1s² – 2s2p

- Model with 5-fs (FWHM) Gaussian-shaped pulse centered at 60.10 eV





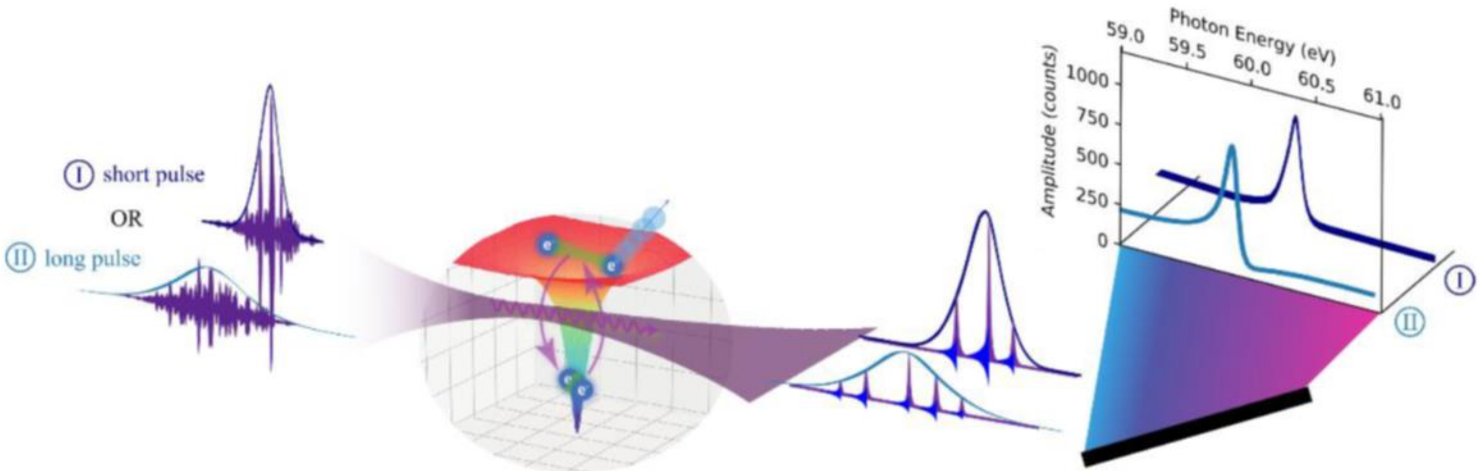
predicted by model simulation

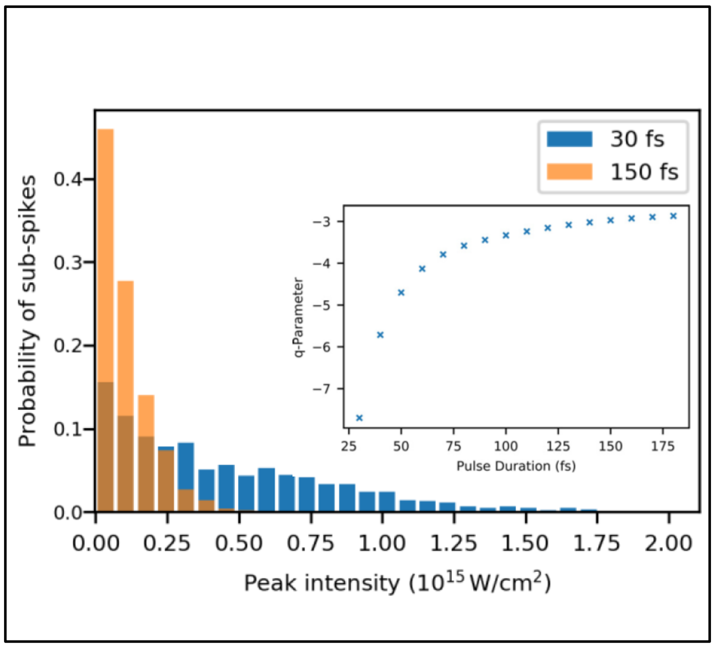


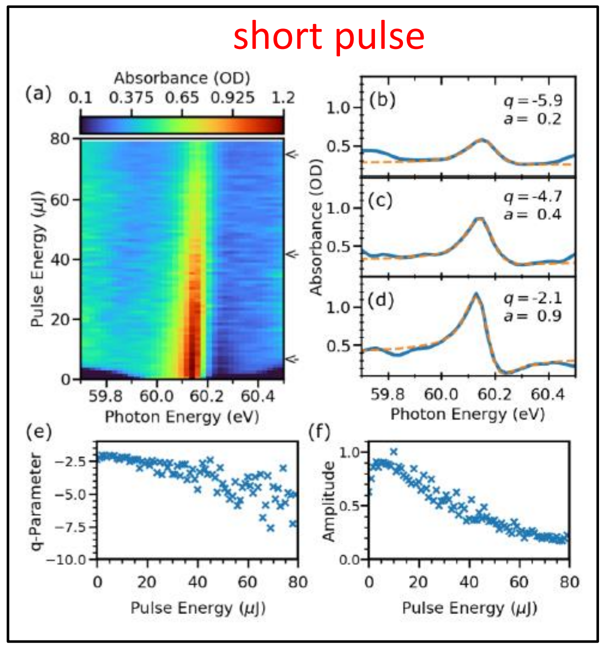
Quantum state population control of an autoionizing two-electron system

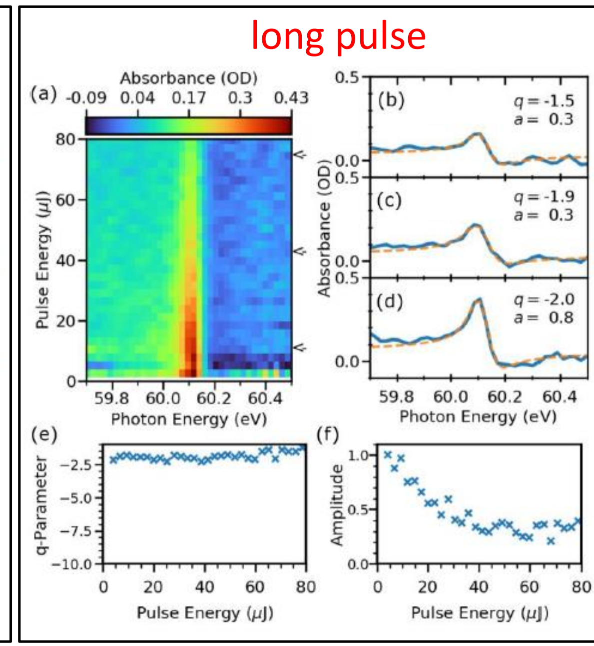
C Ott et al., PRL 123, 163201 (2019)

The impact of the (SASE) FEL pulse duration



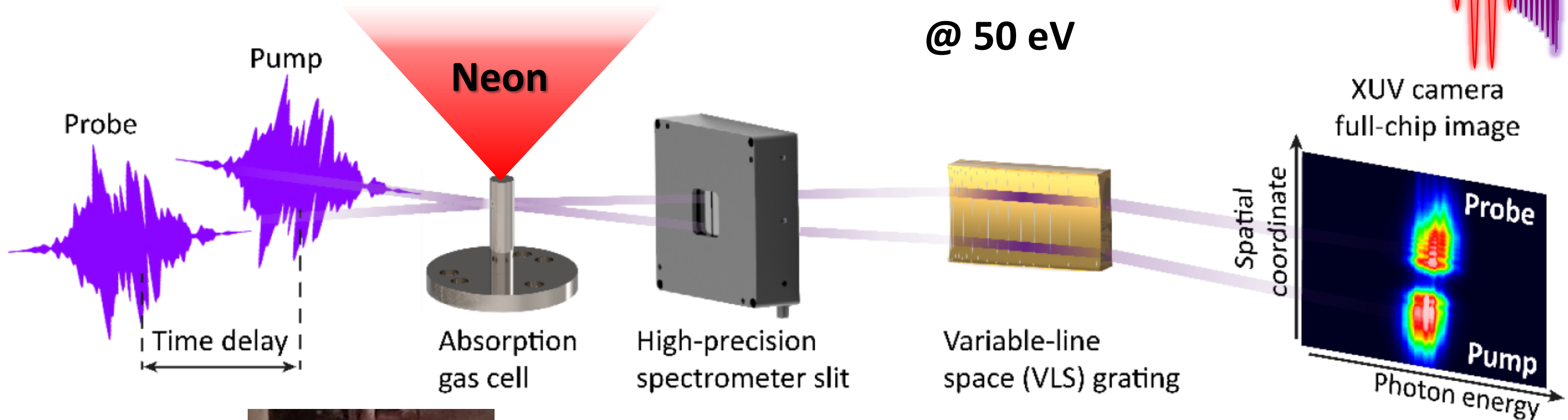


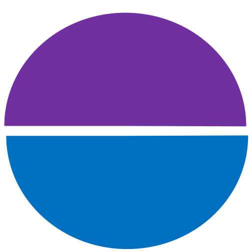


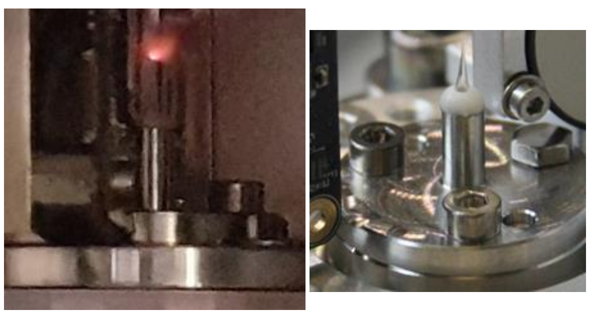


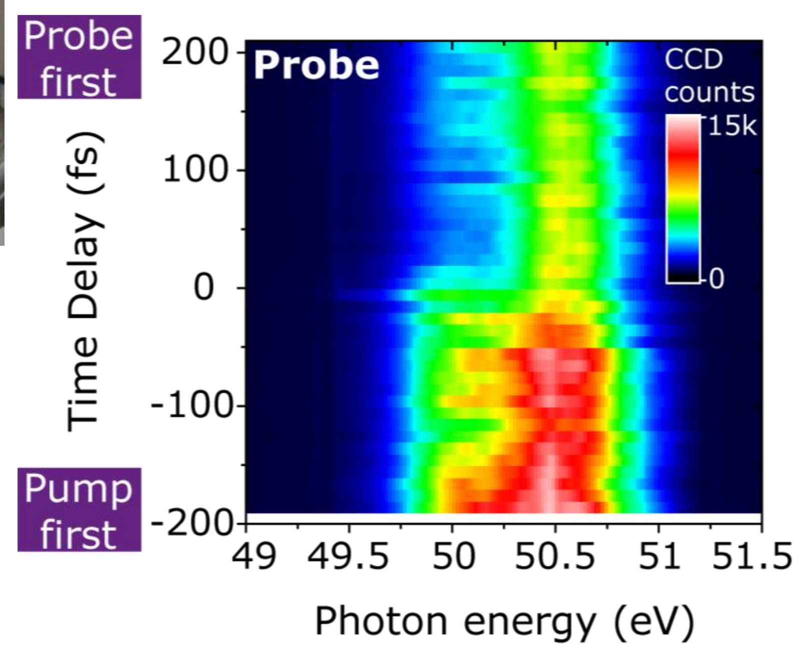
L Aufleger et al., J Phys B, 53, 234002 (2020)

Time-resolving resonant XUV atomic resonances in Ne





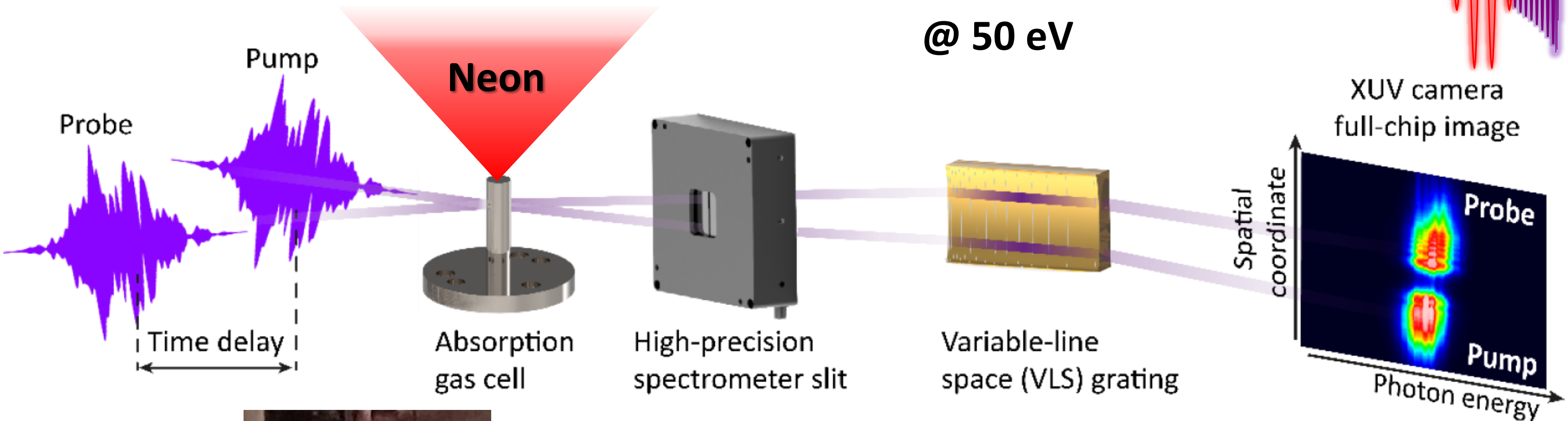


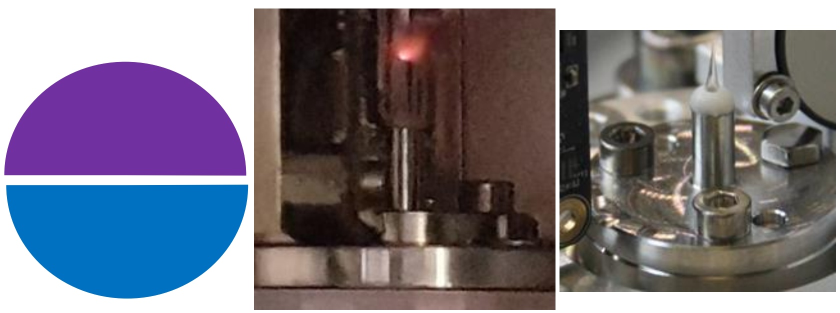


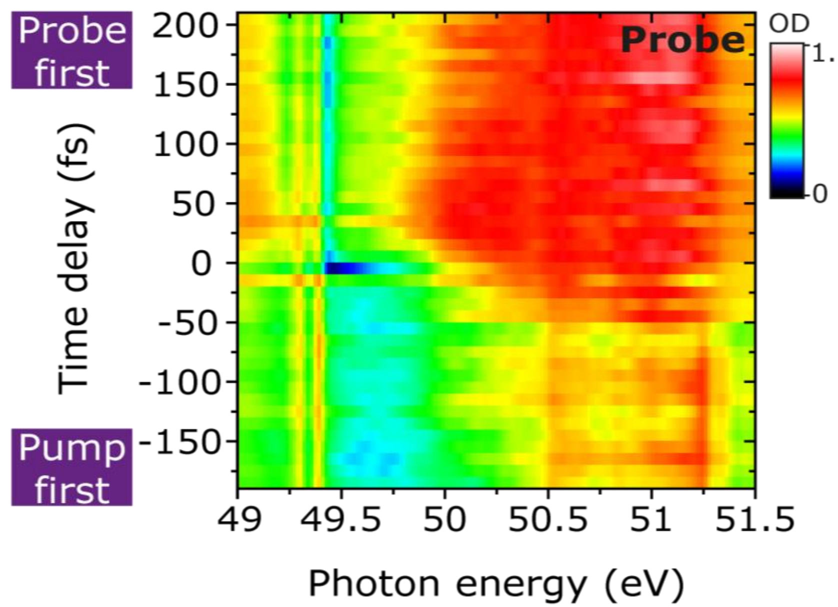
XUV Split & Delay Unit @BL2,
H. Zacharias, S. Roling *et al.*
(University of Münster)

[M Wöstmann et al.,
J Phys B 46, 164005 (2013)]

Time-resolving resonant XUV atomic resonances in Ne







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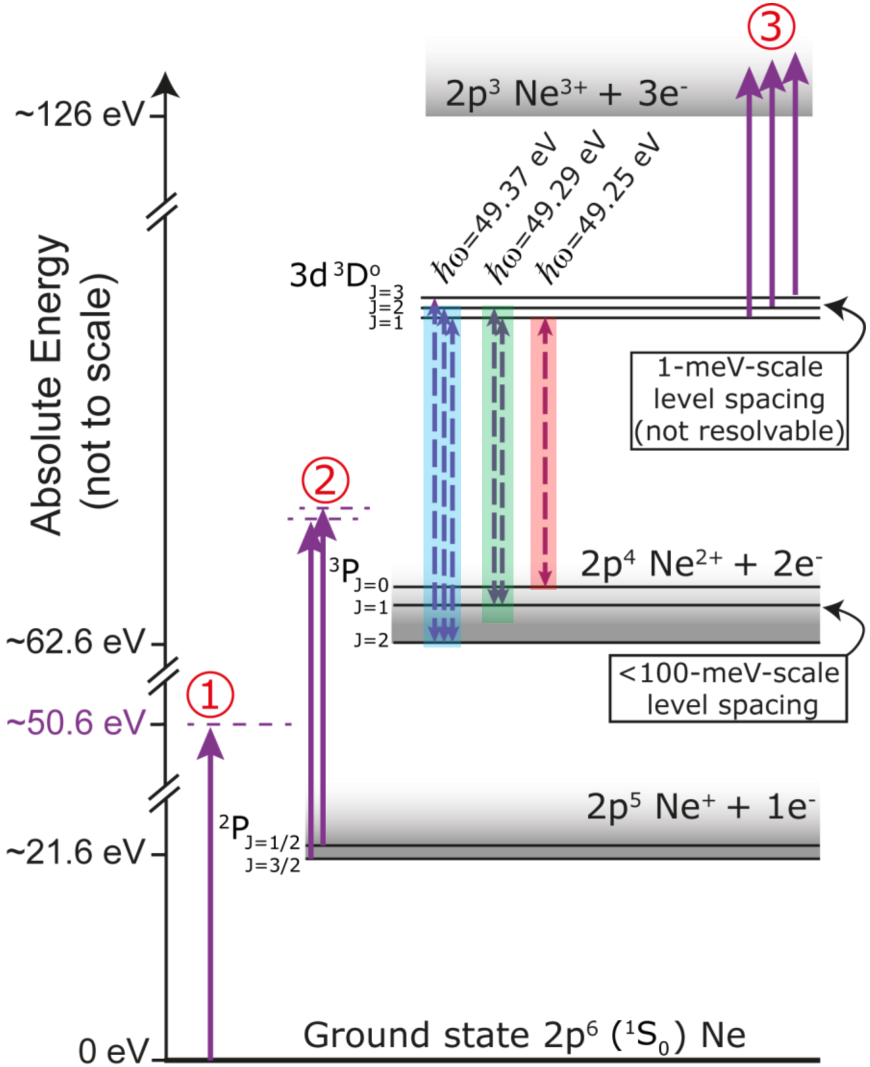
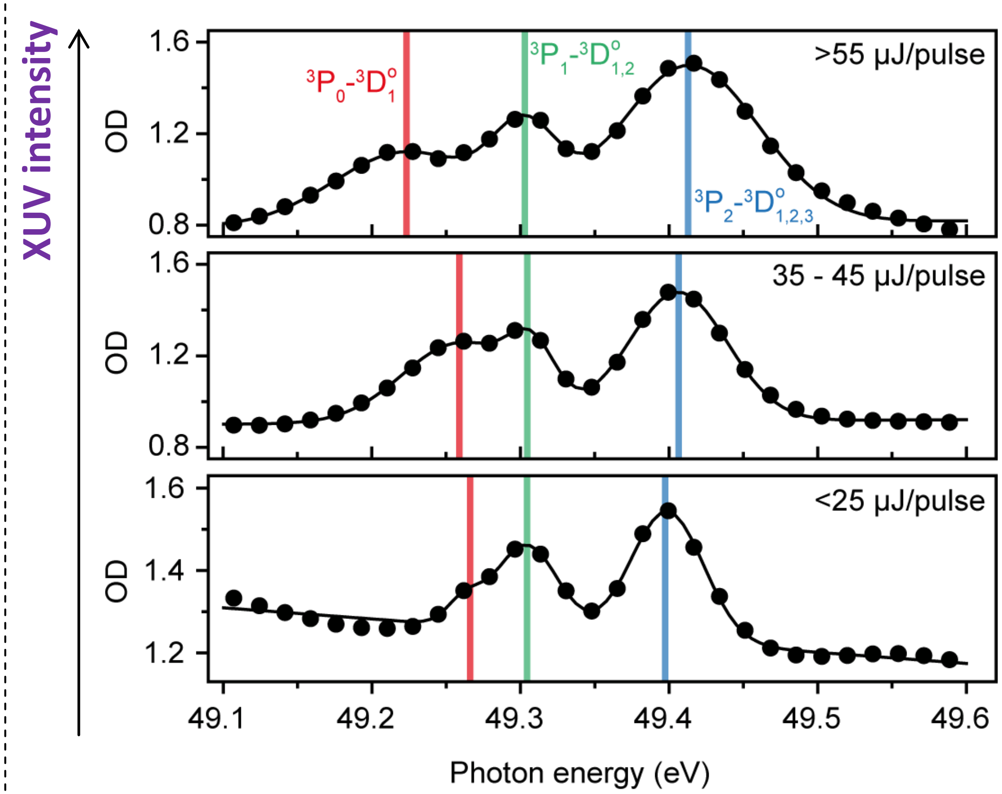
Neon Ne^{2+} ions: strongly coupled resonances @ 50 eV



Directly identify resonances in transient ionic species, and their Stark shift

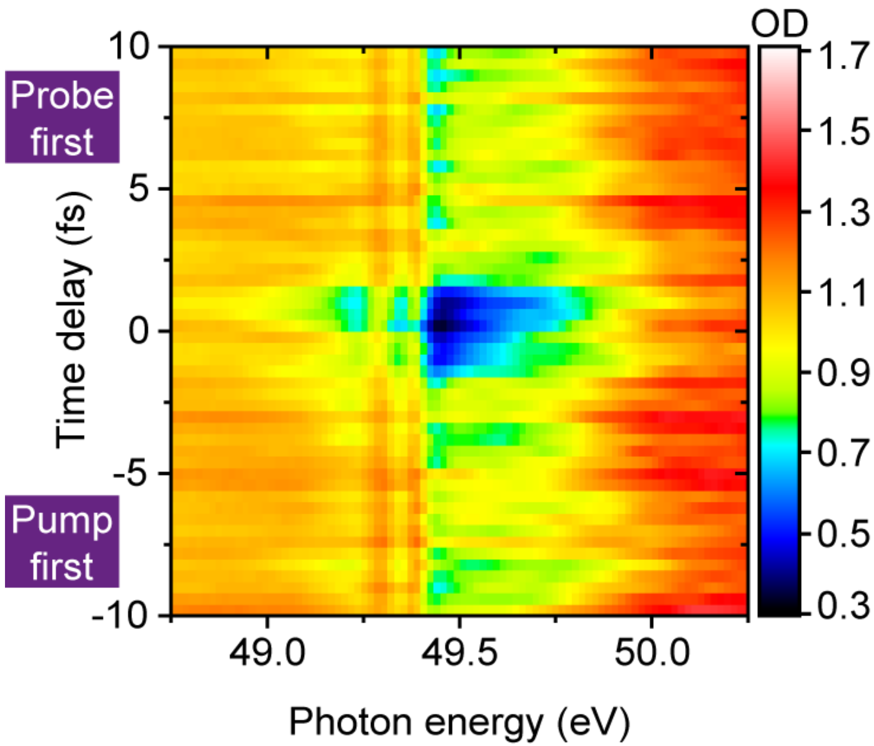
- **Observation of intensity dependent resonance shifts (AC Stark effect)**

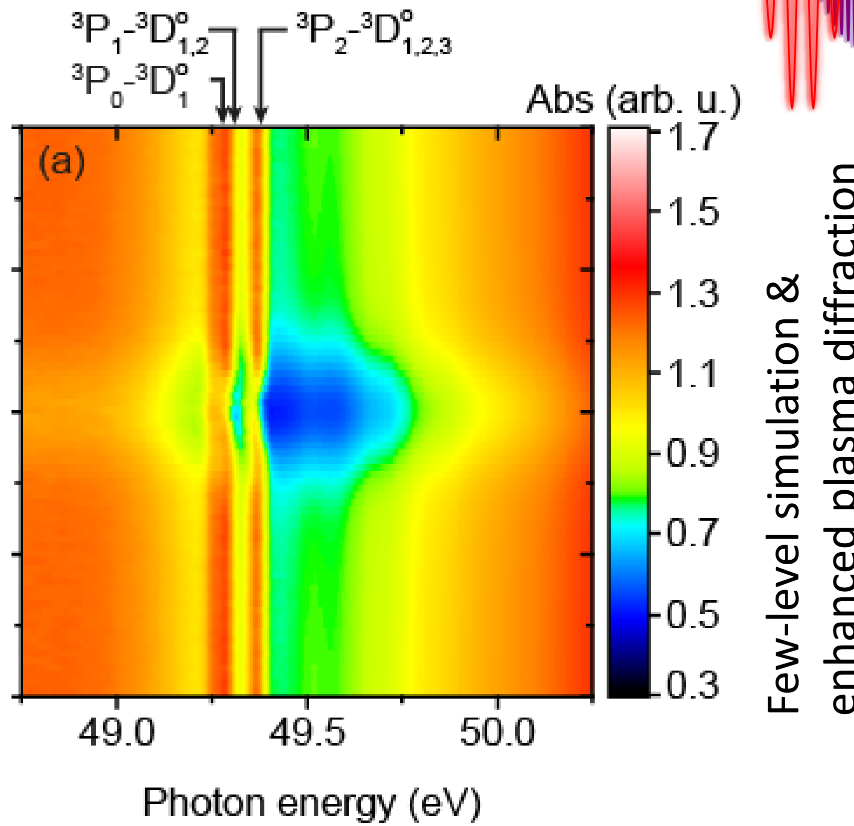


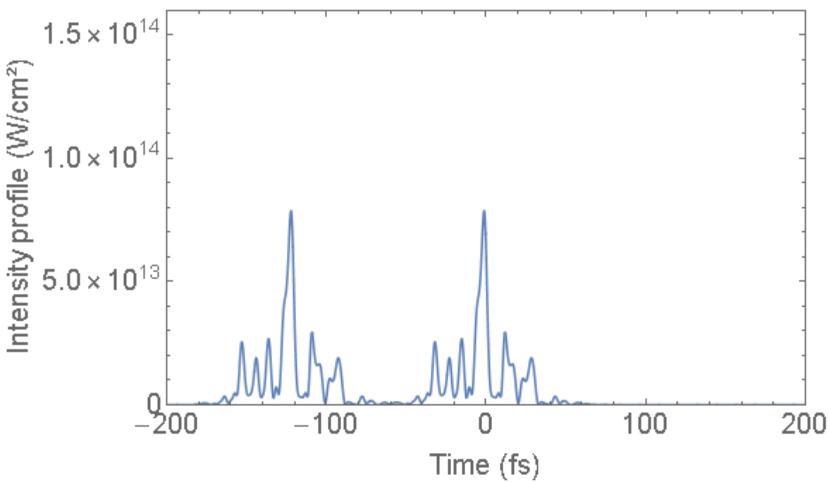
- **XUV-induced strong coupling of ^3P – ^3D spin-orbit multiplet in Ne^{2+}**

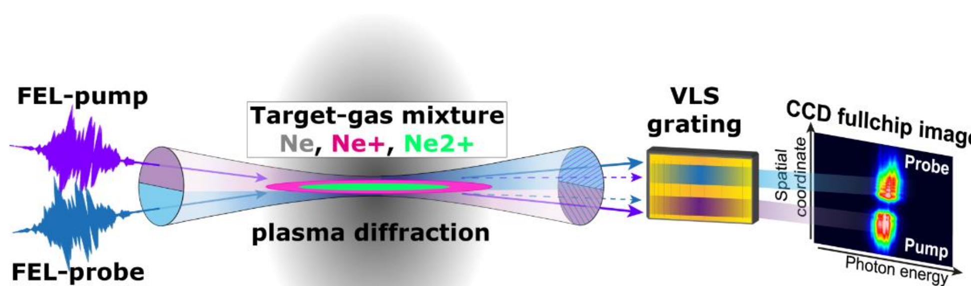
T Ding et al., PRL 123, 103001 (2019)

Coherence enhancement in temporal overlap region



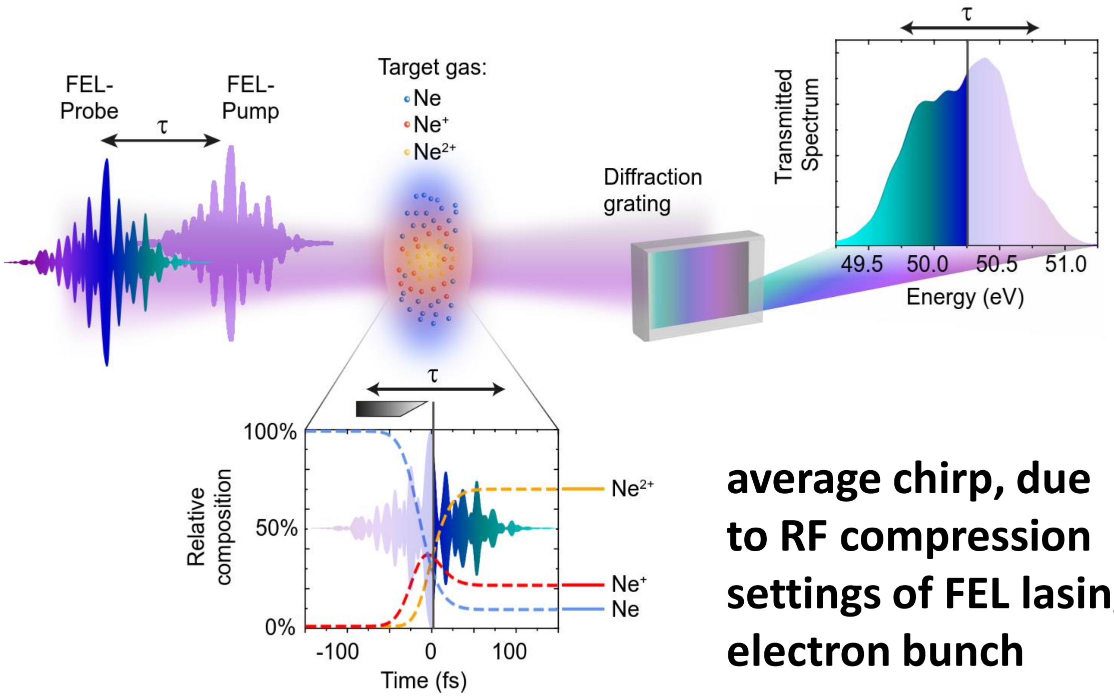


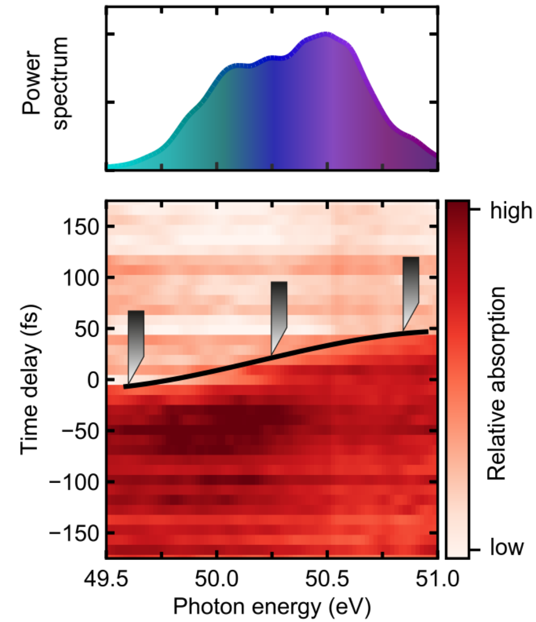




T Ding et al., PRL 123, 103001 (2019)

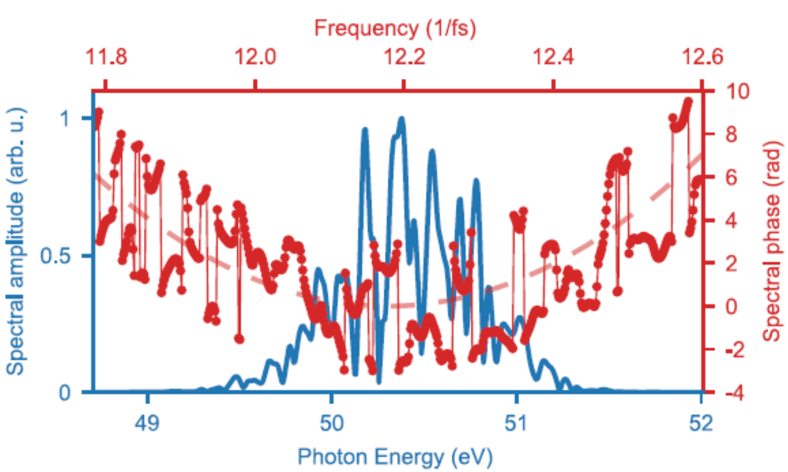
Spectro-temporal sensitivity to chirp of SASE FEL pulses





average chirp, due to RF compression settings of FEL lasing electron bunch

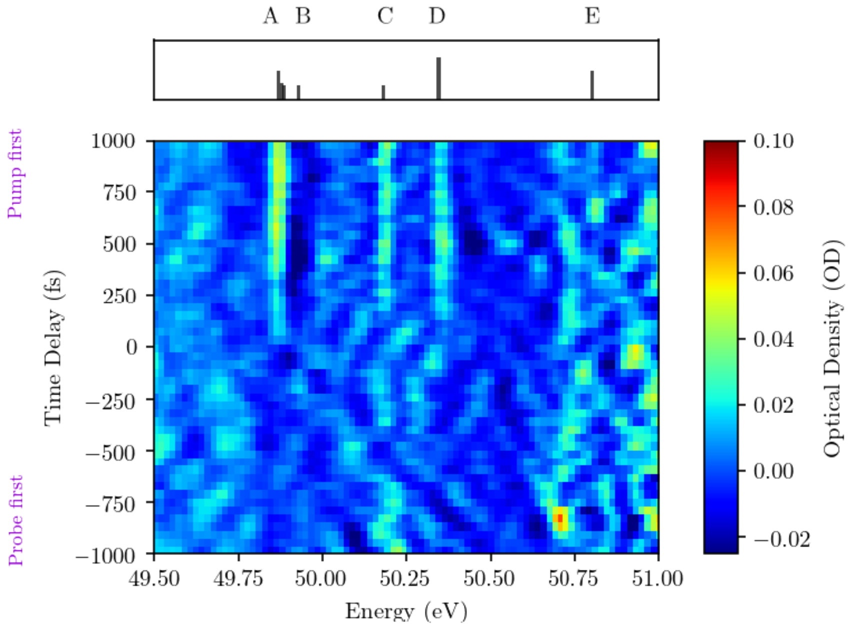
→ (32±1) fs² & (–8±2) fs³ obtained from data



Simulated SASE pulse (single shot) contains stochastic phase (partial coherence model) with superimposed systematic phase due to chirp

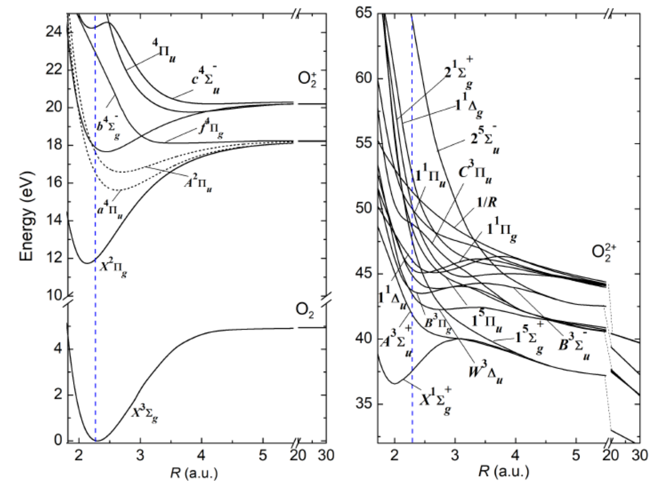
T. Ding et al.,
Nature Communications (2021) 12:643
https://doi.org/10.1038/s41467-020-20846-1

XUV-initiated O_2 dissociation dynamics



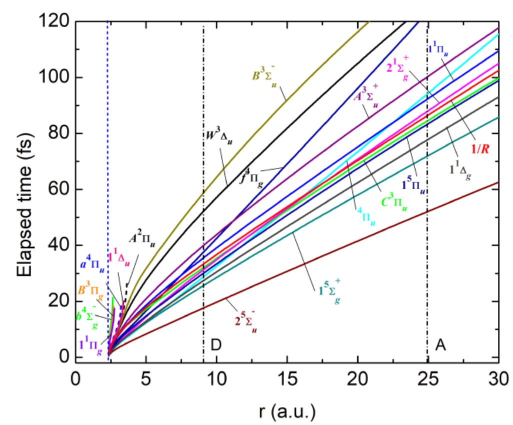
[Magrakvelidze et al., PRA (2012), 86, 1 – 11] (ion KER XUV-pump-XUV-probe, related experiment)

collaboration with U. Thumm (KSU),
M. Magrakvelidze (Cabrini U)



Spectroscopic identification: atomic O^{2+} resonant transitions after dissociation

Transition	Term Scheme	Energy (eV)
A: $2p^2 - 2p5d$	$(g^3P_2) - (^3D^o_3)$	49.87
A: $2p^2 - 2p5d$	$(g^3P_1) - (^3D^o_2)$	49.88
A: $2p^2 - 2p5d$	$(g^3P_0) - (^3D^o_1)$	49.89
B: $2p^2 - 2p7d$	$(^1D_2) - (^1F^o_3)$	49.93
C: $2s^22p^2 - 2s2p^23p$	$(^1D_2) - (^1F^o_3)$	50.18
D: $2s^22p^2 - 2s2p^23p$	$(^1D_2) - (^1D^o_2)$	50.35
E: $2s^22p^2 - 2s2p^23p$	$(^1D_2) - (^1P^o_1)$	50.80



Ding et al. Faraday Discuss., 2021, **228**, 519-536

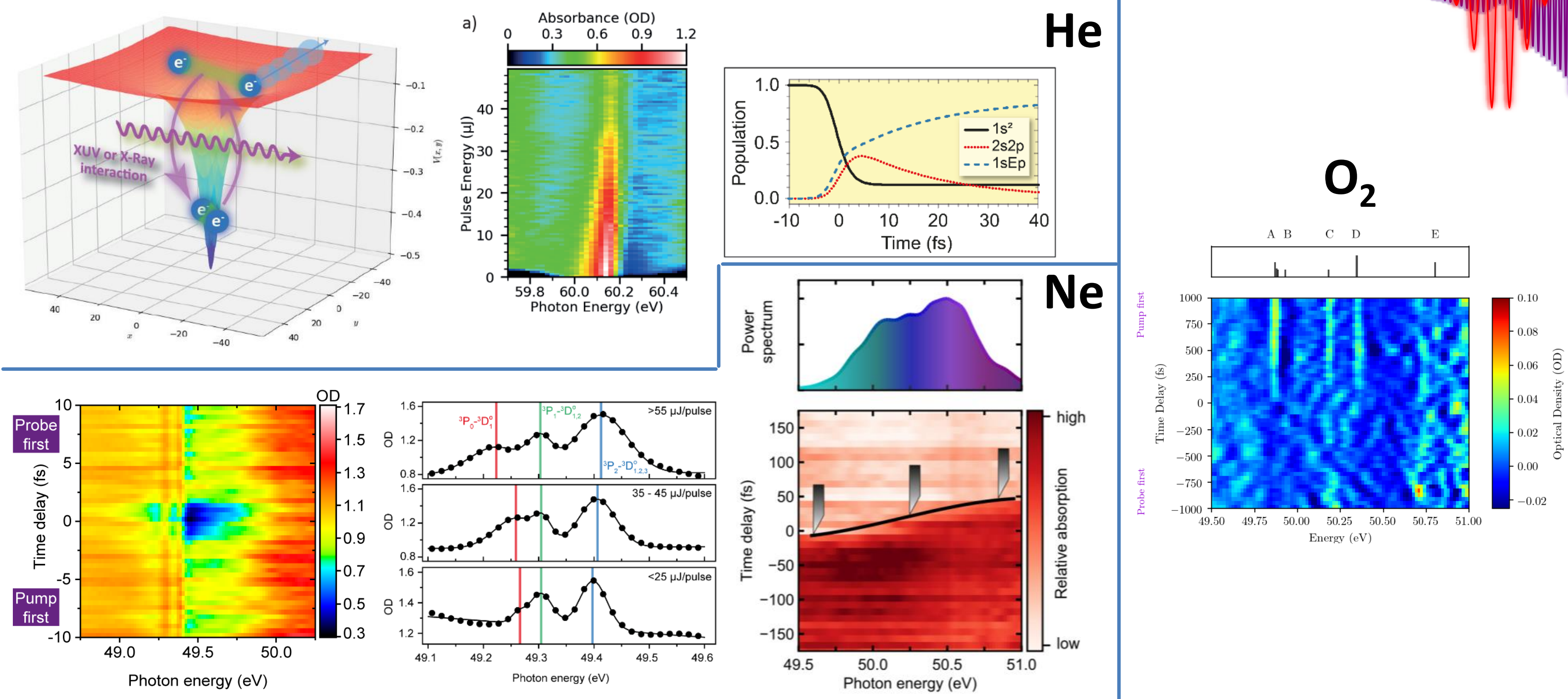
Rebholz et al., submitted manuscript (2021)

Classical propagation along O_2^+ and O_2^{2+} PECs

Predicted dissociation times shorter than observed

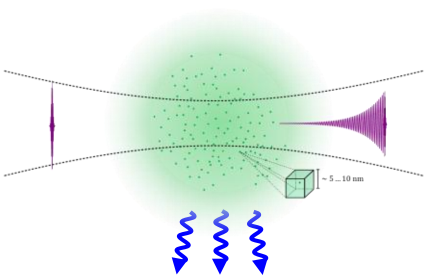
Calls for delayed mechanism, possibly involving predissociating states

Summary & Conclusion

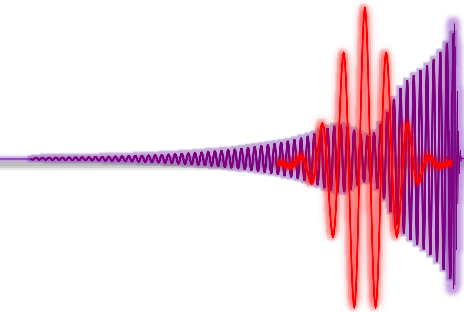


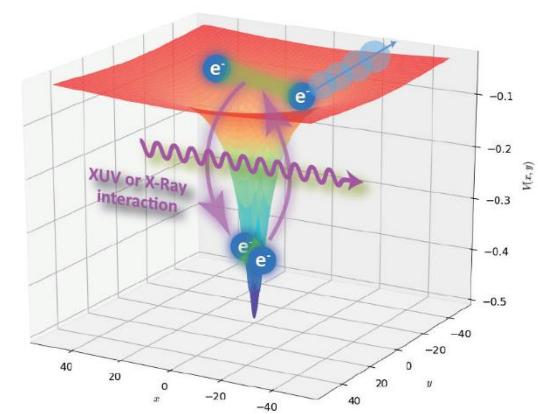
Site-specific nonlinear light-matter interaction with intense XUV/x-ray light (light in – light out)

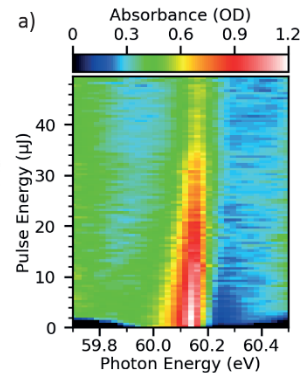
- ***Forward direction:*** **absorption & stimulated emission (coherent interactions; direct sensitivity to amplitudes and coherences)**
- ***Perpendicular direction:*** **incoherent flourescence decay (direct sensitivity to site-specific population dynamics)**



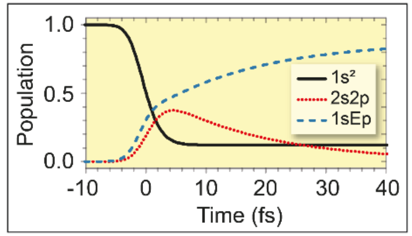
Summary & Conclusion



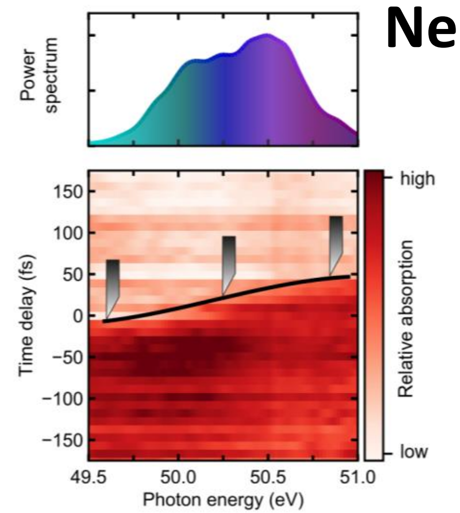




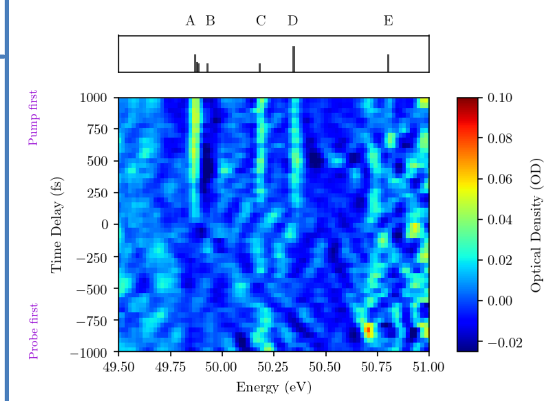
He

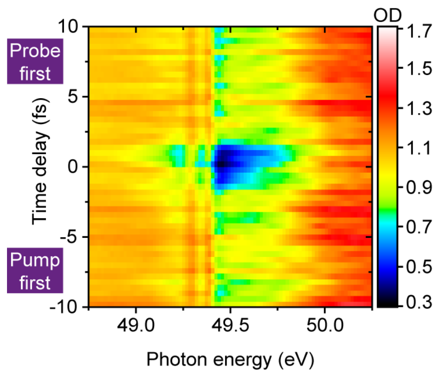


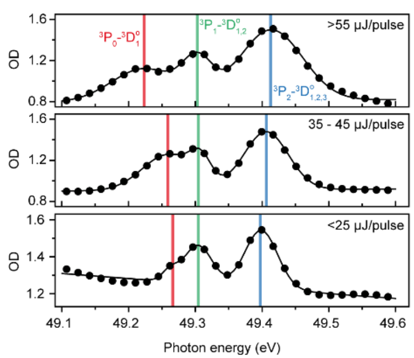
Ne



O_2



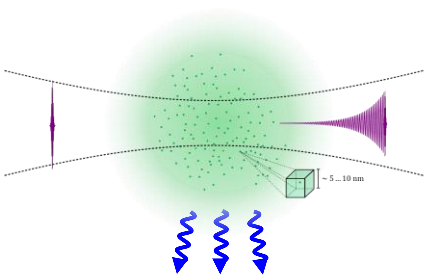




Site-specific nonlinear light-matter interaction with intense XUV/x-ray light (light in – light out)

- *Forward direction:* absorption & stimulated emission
  (c... direct sensitivity
  to ampl...
- *Perpendicular direction:* incoherent ...
  (direct sensitivity to site-specific population dynamics)

everything dressed in intense XUV/x-ray electric fields
adressing & controlling individual atomic resonances



Quantum Dynamics & Control Division
Prof. Thomas Pfeifer

MAX-PLANCK-GESELLSCHAFT

MAX-PLANCK-INSTITUT FÜR KERNPHYSIK

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