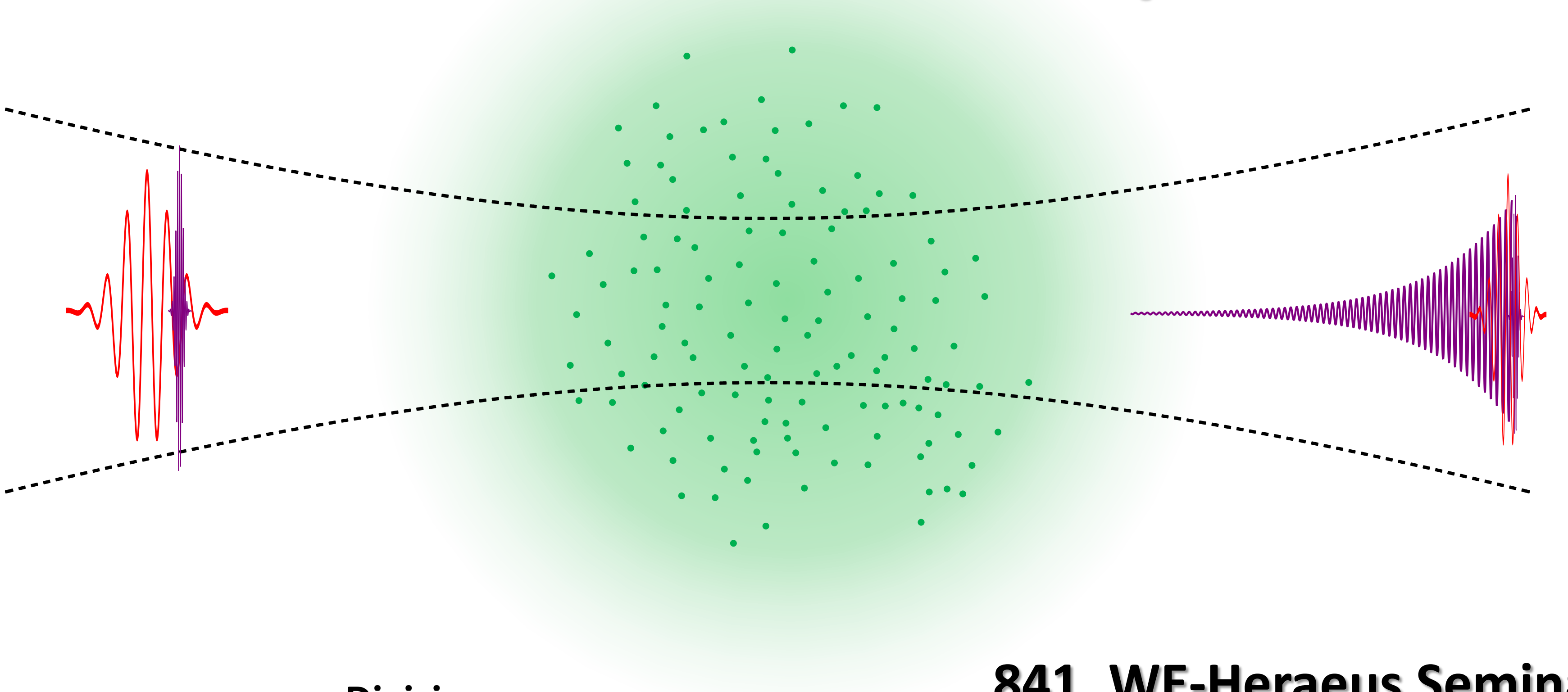
MAX-PLANCK-INSTITUT
FÜR KERNPHYSIK
HEIDELBERG

State-resolved quantum dynamics in atoms and molecules with femtosecond and attosecond laser pulses

MAX-PLANCK-GESELLSCHAFT

Christian Ott

03.09.2025



✉ christian.ott@mpi-hd.mpg.de

Division
Quantum Dynamics & Control
Prof. Dr. Thomas Pfeifer

841. WE-Heraeus Seminar:
Important Quantum Technologies -
Origins and Applications

Outline

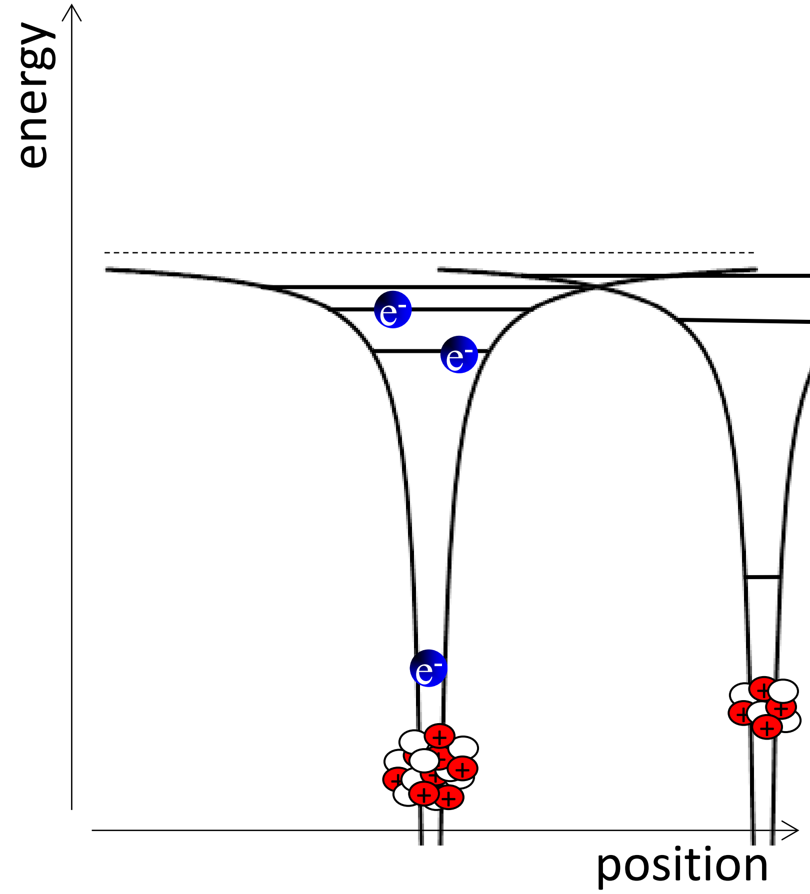
1) Introduction: a time-domain view into absorption spectroscopy

2) Learning from laser-controlled spectral line shapes of doubly excited states in helium

3) Electronic-state-resolved dynamics in small molecules and their laser control

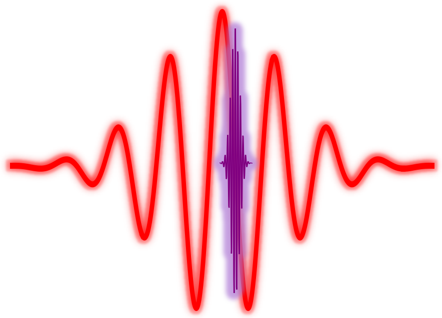
4) Conclusion

Ultrafast XUV/x-ray dynamics through multi-electron interaction



- Interaction with XUV/x-ray pulses:
  - Inner-shell excitation & ionization
- Through multi-electron interaction:
  - Ultrafast (femto-/attosecond) relaxation via Auger-Meitner decay / autoionization (in molecule also inter-atomic)
- How do atoms & molecules coherently respond to ultrashort pulses of … ?

intense XUV FEL

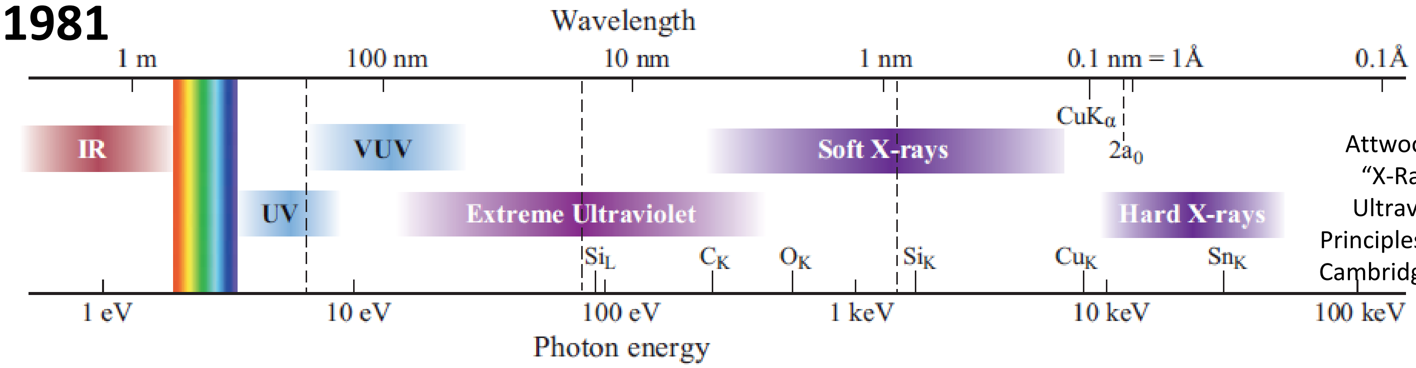


XUV HHG & intense NIR

State-specific XUV/x-ray resonant transitions

Kai M. Siegbahn: Nobel Prize in Physics 1981

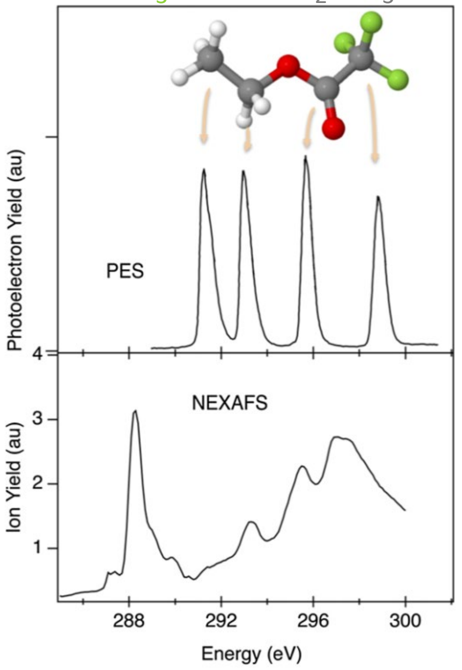
"for his contribution to the development of high-resolution electron spectroscopy."

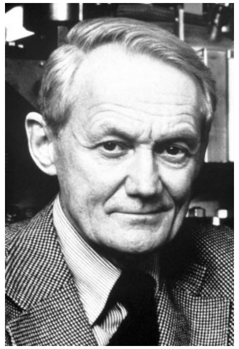


Attwood & Sakdinawat, "X-Rays and Extreme Ultraviolet Radiation – Principles and Applications", Cambridge Univ. Press 2016.

ethyl trifluoroacetate

CF_3-CO-O-CH_2-CH_3





XUV/x-ray key properties:

- Resonant site-specific electronic transitions
- Local probe of ultrafast molecular dynamics

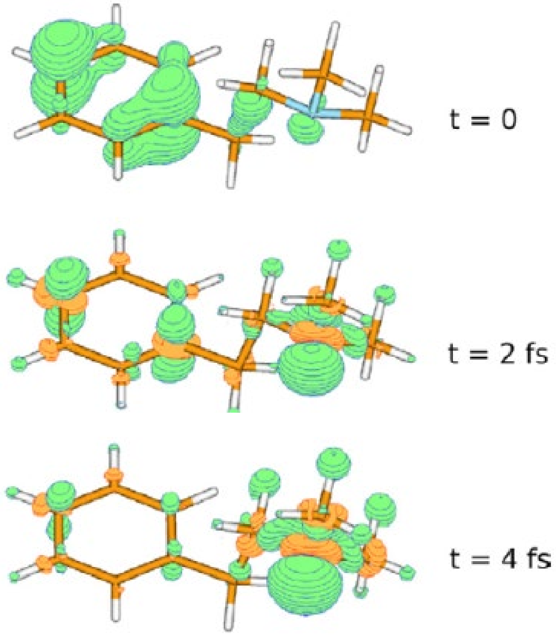
Natural timescale of electron motion:

- Typically femtosecond to attosecond timescale
- Inverse energy level spacing $T \sim \frac{h}{\Delta E} \sim \frac{4.1}{\Delta E\,[\mathrm{eV}]}\,[\mathrm{fs}]$

Our scientific goals:

Observe, understand and control ultrafast state-specific multi-electron dynamics in natural quantum systems (atoms & molecules) and their interaction with intense radiation fields

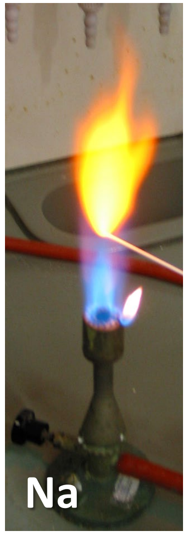
2-phenylethyl-*N*,*N*-dimethylamine (PENNA)



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

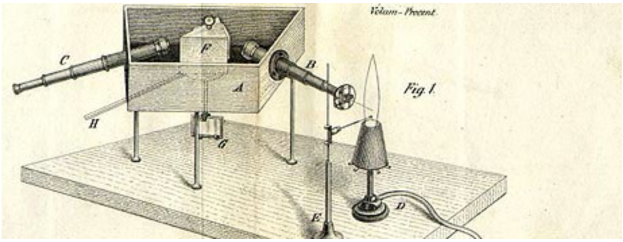
Sorensen et al., JPhysB 53 244011 (2020).
Travnikova et al., JESRP 185, 191 (2012).
Siegbahn, "ESCA Applied to Free Molecules", North-Holland Publishing Company (1971).

Absorption spectroscopy with broadband light sources



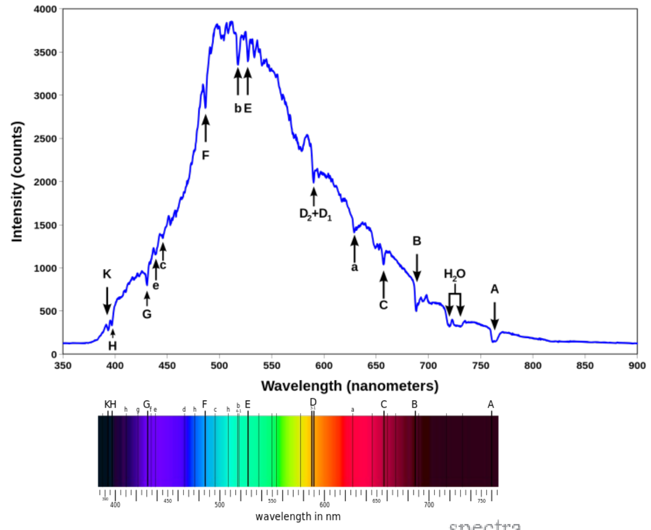
https://de.wikipedia.org/
wiki/Flammenfärbung

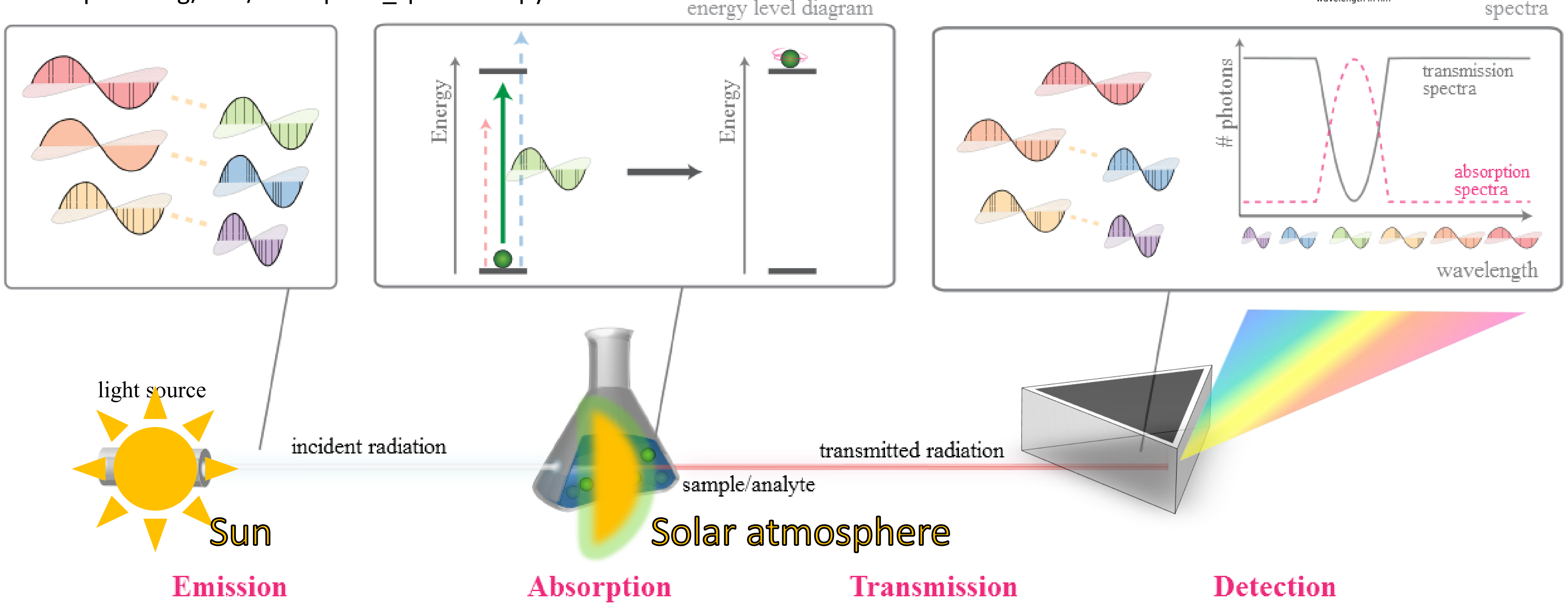
Wollaston (1802), Fraunhofer (1814):
observe lines in solar spectrum



Kirchhoff, Bunsen (1859):
systematic assignment of
emission lines
"spectral analysis"

https://en.wikipedia.org/wiki/Absorption_spectroscopy

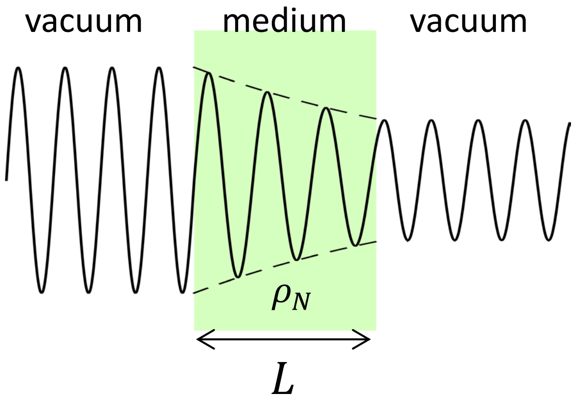




Maxwell‘s theory of light-matter interaction

light-matter response determined by the medium’s **complex refractive index $\tilde{n}(\omega) = n(\omega) + i\kappa(\omega)$**

travelling wave: $\overrightarrow{e^{i(kx-\omega t)}}$



$$\rightarrow \left(e^{-\frac{\omega\cdot\frac{1}{2}\tilde{\chi}_i(\omega)\cdot L}{c}}\right)^2 \left|\tilde{E}_{in}(\omega)\right|^2 = I_{in}(\omega)e^{-\sigma(\omega)\rho_N L}$$

$$\rightarrow \sigma(\omega) = \frac{\omega}{c\varepsilon_0}\Im\left[\frac{\tilde{d}(\omega)}{\tilde{E}_{in}(\omega)}\right]$$

propagation through medium:

$$e^{ik\cdot\tilde{n}(\omega)\cdot L} = e^{i\frac{\omega\cdot n(\omega)\cdot L}{c}}\cdot e^{-\frac{\omega\cdot\kappa(\omega)\cdot L}{c}}$$

with Beer–Lambert‘s law:

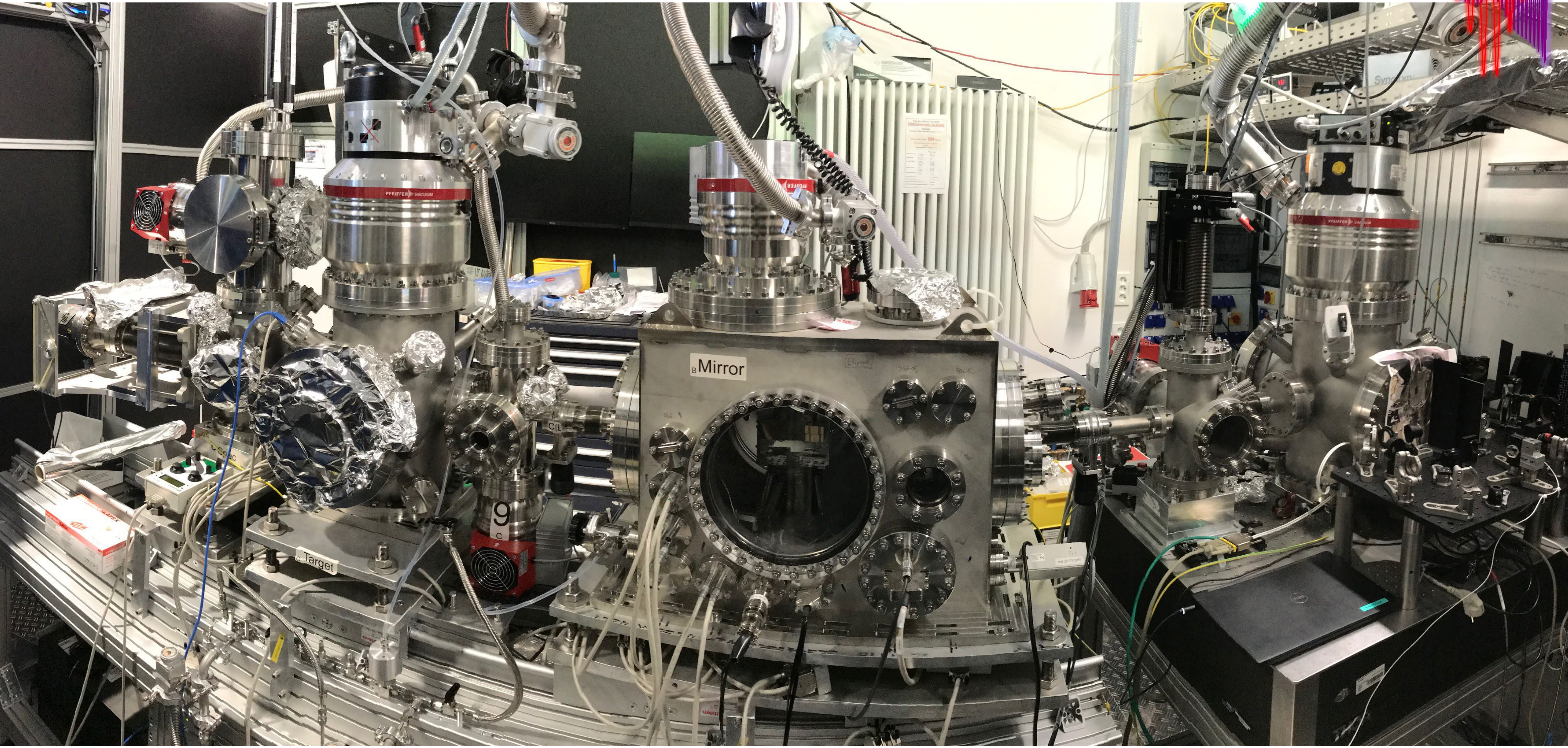
$$I_{out}(\omega) = I_{in}(\omega)\cdot e^{-\sigma(\omega)\rho_N L}$$

using:

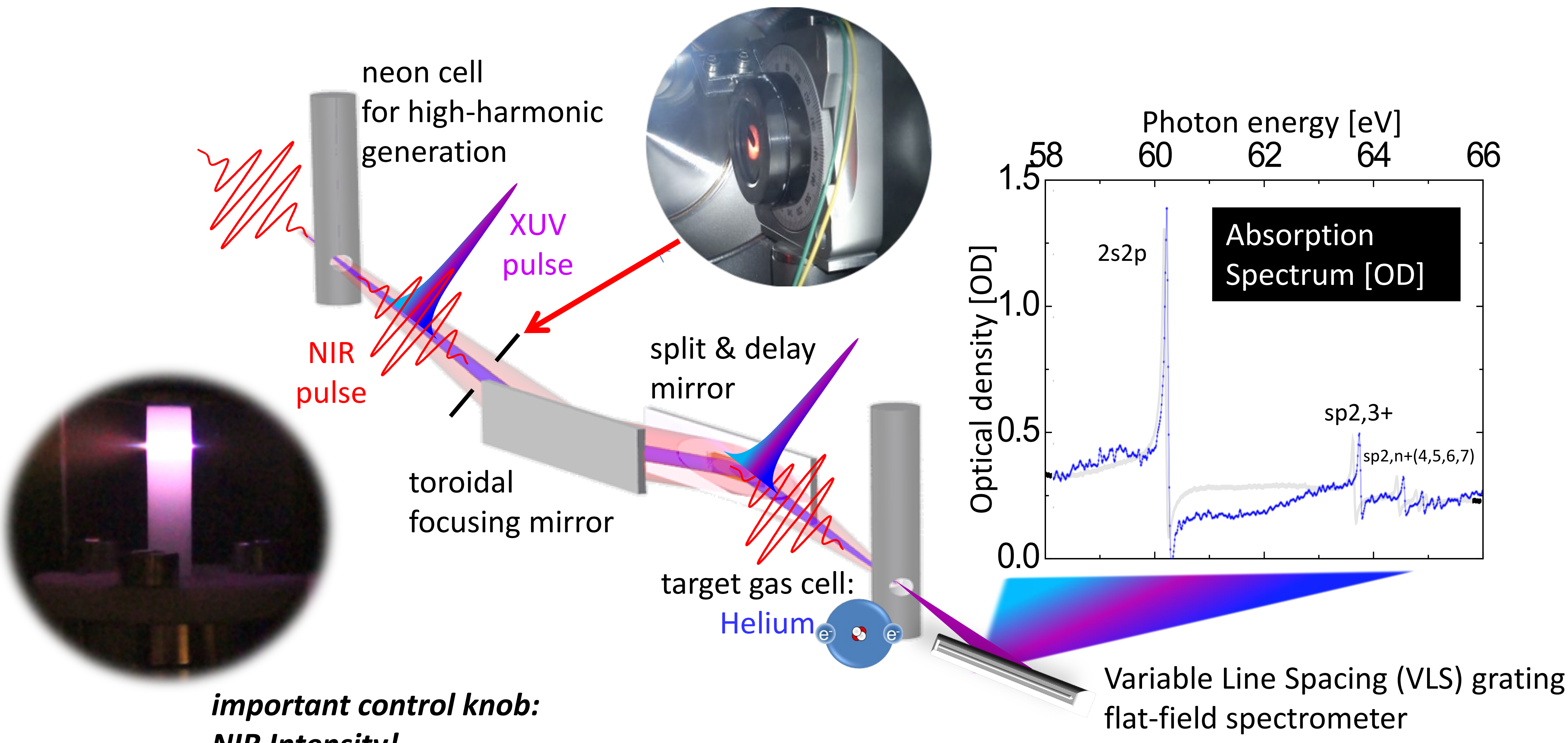
- $\tilde{n}(\omega) = \sqrt{1+\tilde{\chi}(\omega)} \approx 1 + \frac{1}{2}\tilde{\chi}_r(\omega) + i\frac{1}{2}\tilde{\chi}_i(\omega)$
- $I(\omega) \propto \left|\tilde{E}(\omega)\right|^2$
- $\tilde{P}(\omega) = \varepsilon_0\tilde{\chi}(\omega)\tilde{E}(\omega)$
- $\tilde{P}(\omega) = \rho_N\tilde{d}(\omega)$

- Absorption of light microscopically linked to system‘s dipole response $\tilde{d}(\omega)$
- Straightforwardly generalized to nonlinear laser-induced dynamics with laser-controlled $\tilde{d}[\omega; I(\tau)]$

Experimental setup of laser-controlled absorption spectra

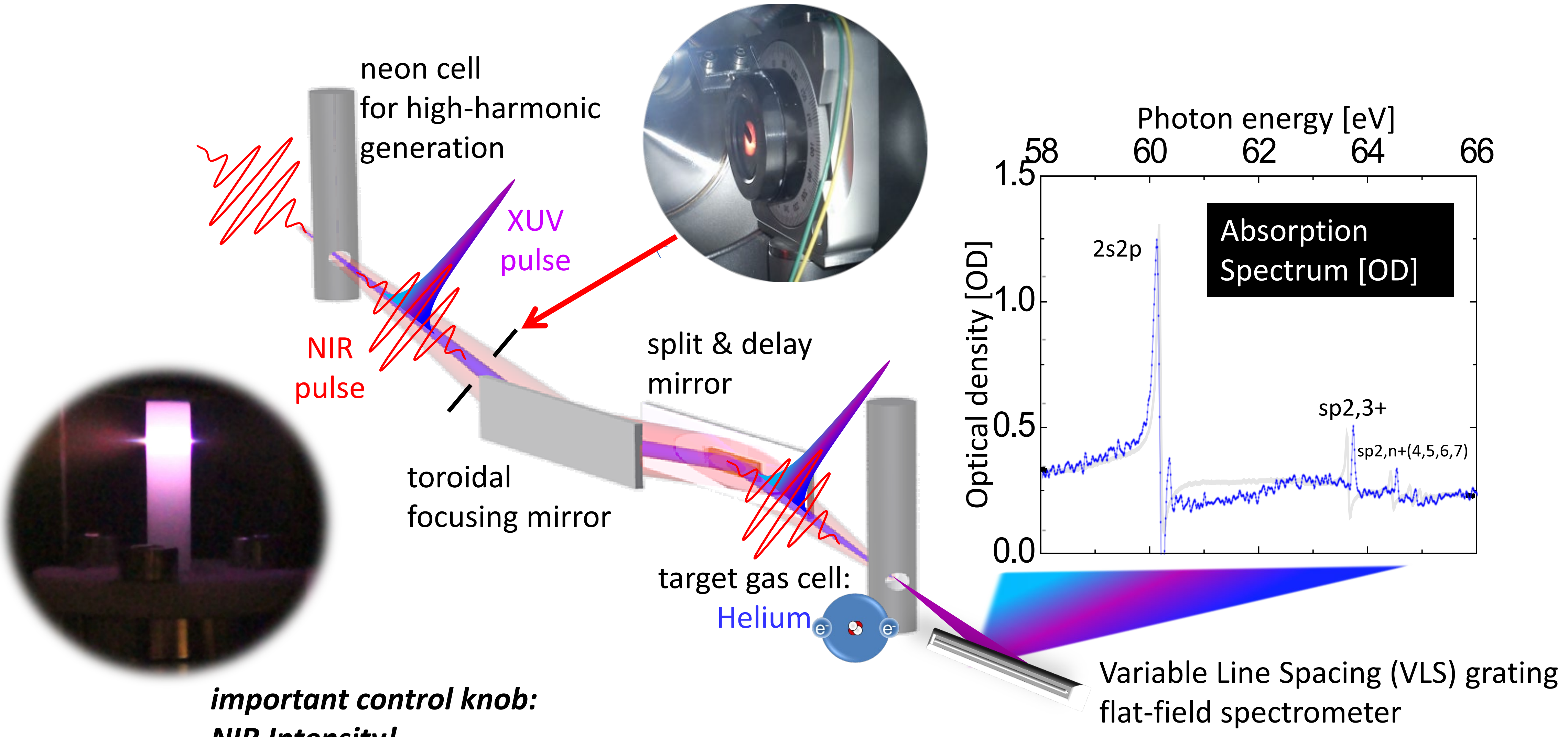


Experimental scheme of laser-controlled absorption spectra



important control knob:
NIR Intensity!

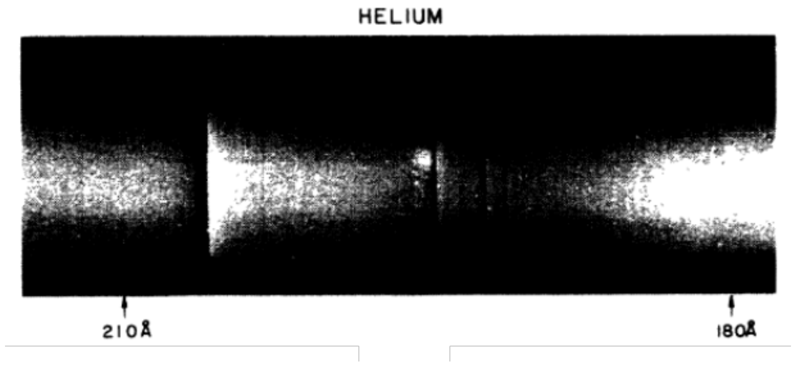
Experimental scheme of laser-controlled absorption spectra

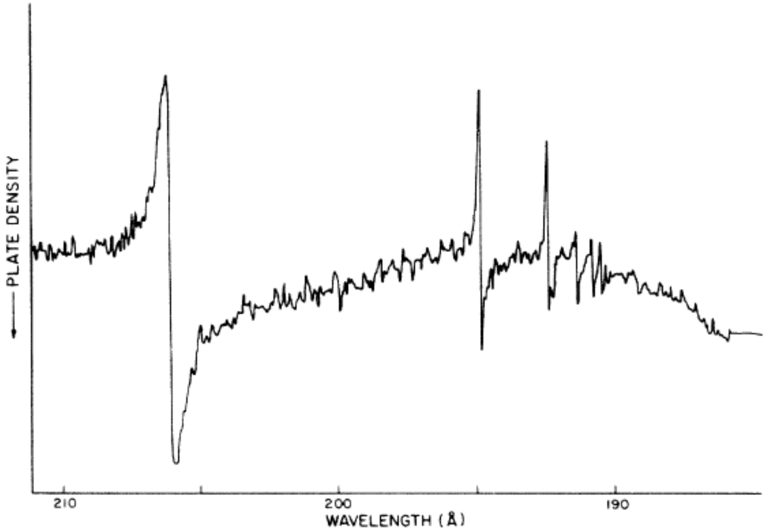


important control knob:
NIR Intensity!

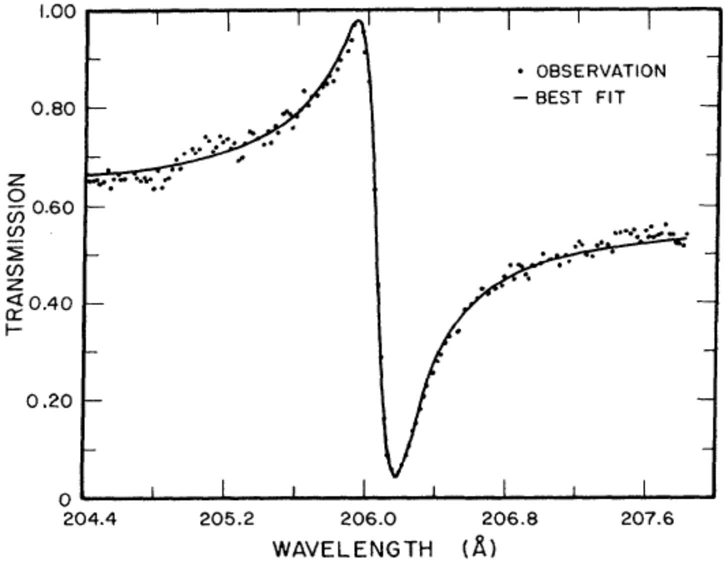
Absorption in Helium with XUV light

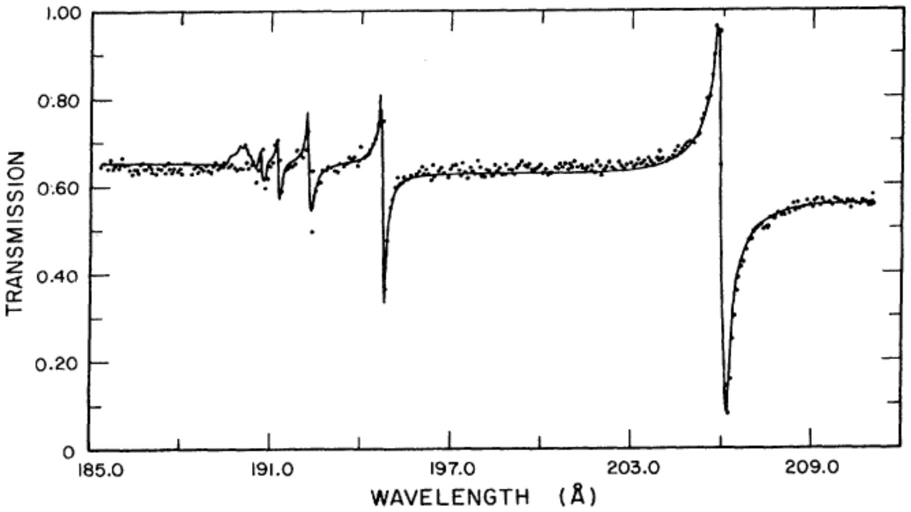
- Synchrotron „white light“ with bending magnets
- Illumination of photo plates after transmission through helium and spectrometer





Madden&Codling, PRL 10, 516-518, (1963).

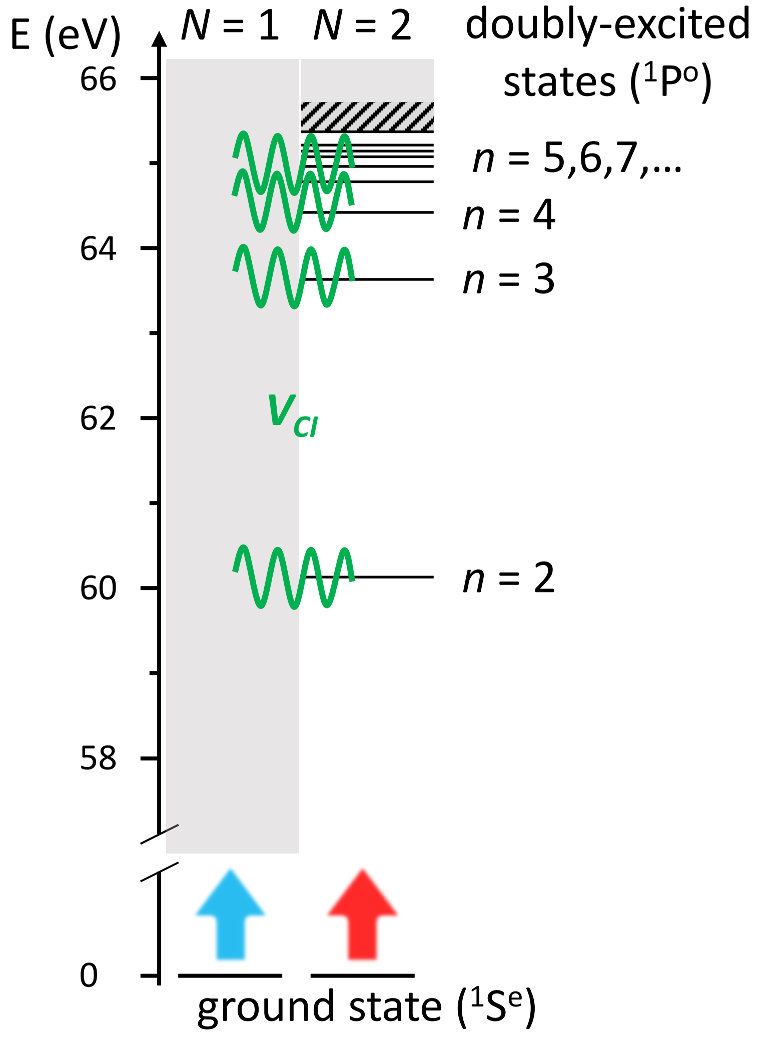




Morgan&Ederer, PRA 29, 1901-1906, (1984).

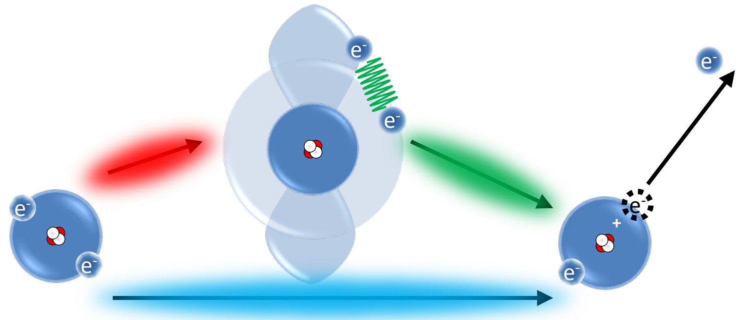
Doubly excited states in helium: a natural quantum interferometer

$$\hat{H} = -\frac{1}{2}\vec{\nabla}_1^{\,2} - \frac{2}{|\vec{r}_1|} - \frac{1}{2}\vec{\nabla}_2^{\,2} - \frac{2}{|\vec{r}_2|} + \frac{1}{|\vec{r}_{12}|}$$



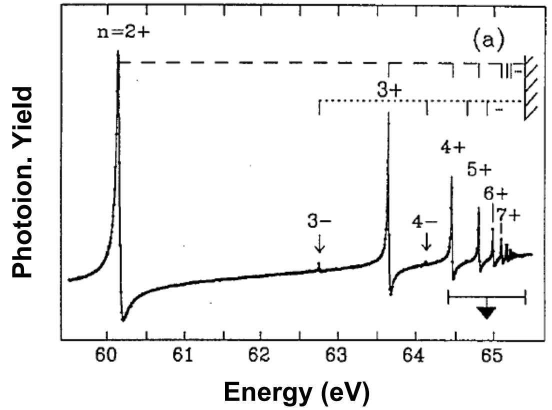
U. Fano Phys. Rev. **124**, 1866 (1961)

life time: few 10 fs to 100 fs



line width:
Few 10 meV to few meV

from ground state to (auto-)ionization:
quantum „Fano“ interferometer



asymmetric line shapes
(due to interference)

parametrized by Fano
q - parameter

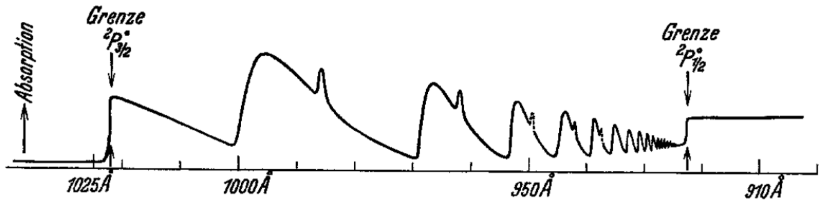
$$q = \frac{\langle \phi | \hat{\mathbf{T}} | g \rangle}{\pi V_E^* \langle \chi_E | \hat{\mathbf{T}} | g \rangle}$$

M. Domke *et al.*, Phys. Rev. A **53**, 1424 (1996)

Beutler – Fano line shapes

H. A. Beutler Zeitschrift für Physik **93**, 177–196 (1935)

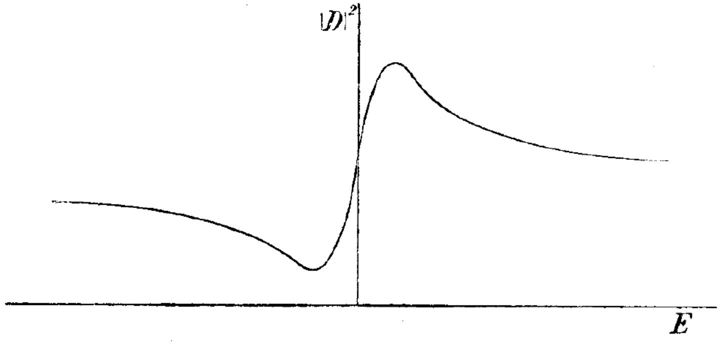
Über Absorptionsserien von Argon, Krypton und Xenon zu Termen zwischen den beiden Ionisierungsgrenzen $^2P^o_{3/2}$ und $^2P^o_{1/2}$.



U. Fano Nuovo Cimento **12**, 154–161 (1935)

SULLO SPETTRO DI ASSORBIMENTO
DEI GAS NOBILI
PRESSO IL LIMITE DELLO SPETTRO D'ARCO

Nota di Ugo Fano



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

American Physical Society
https://link.aps.org › doi › PhysRev.124.1866

Effects of Configuration Interaction on Intensities and Phase ...

by U Fano · 1961 · Cited by 14441 — The interference of a discrete autoionized state with a continuum gives rise to characteristically asymmetric peaks in excitation spectra.

Accessed by Google on July 1st, 2025

Fano resonances are everywhere

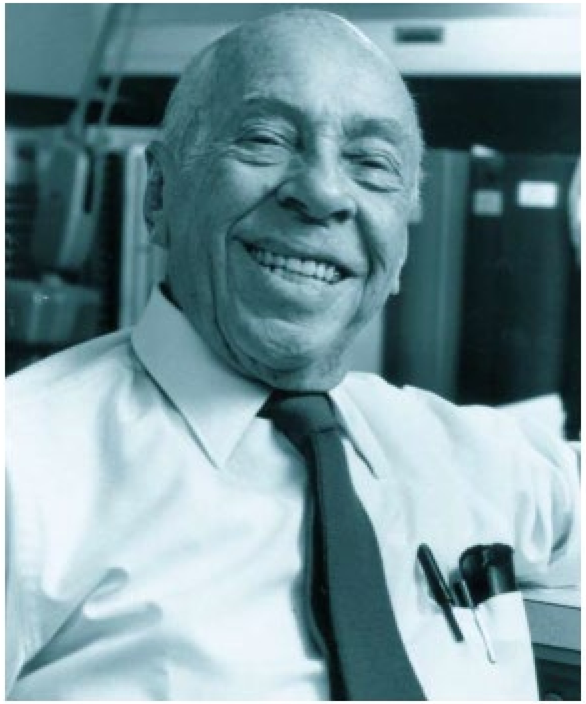
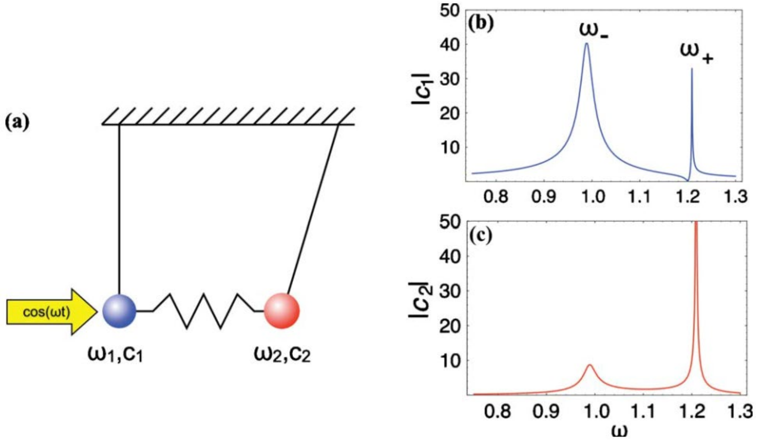
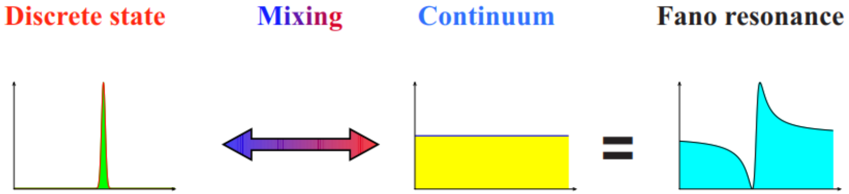
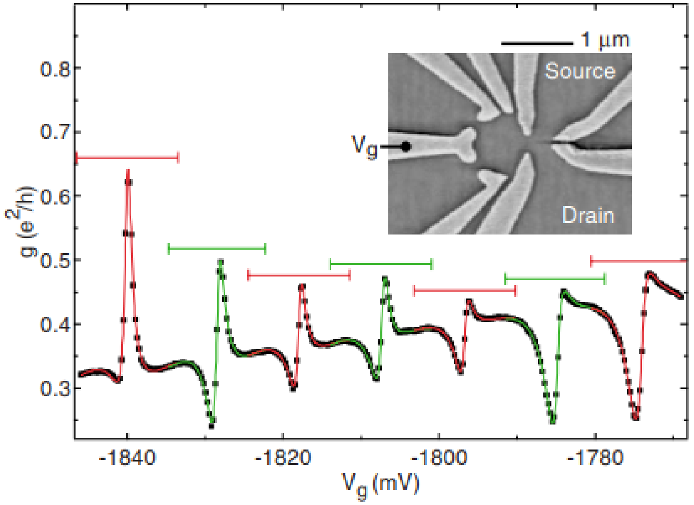


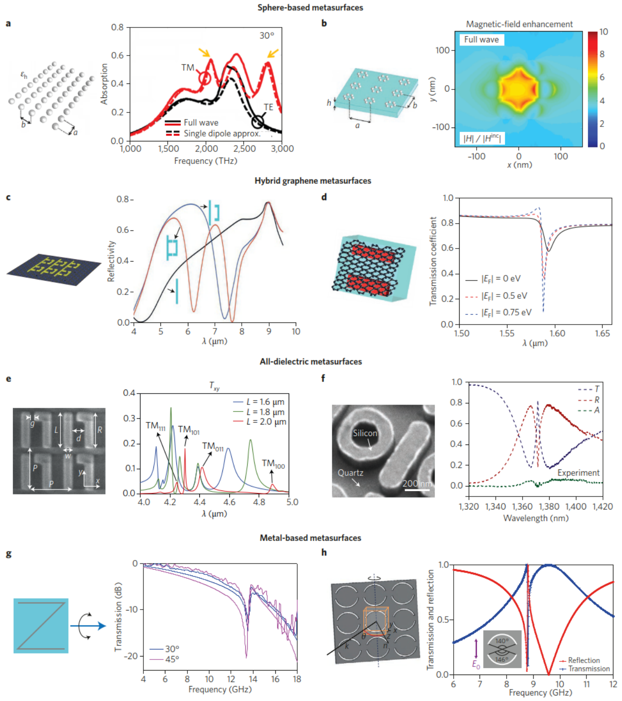
FIG. 1. (Color online) Ugo Fano (1912–2001). "Outstanding interpreter of how radiation interacts with atoms and cells" (Clark, 2001) and much more (this review).





$$\frac{q^2-1}{\epsilon^2+1} + \frac{2q\epsilon}{\epsilon^2+1} + 1 = \frac{(\epsilon+q)^2}{\epsilon^2+1}$$





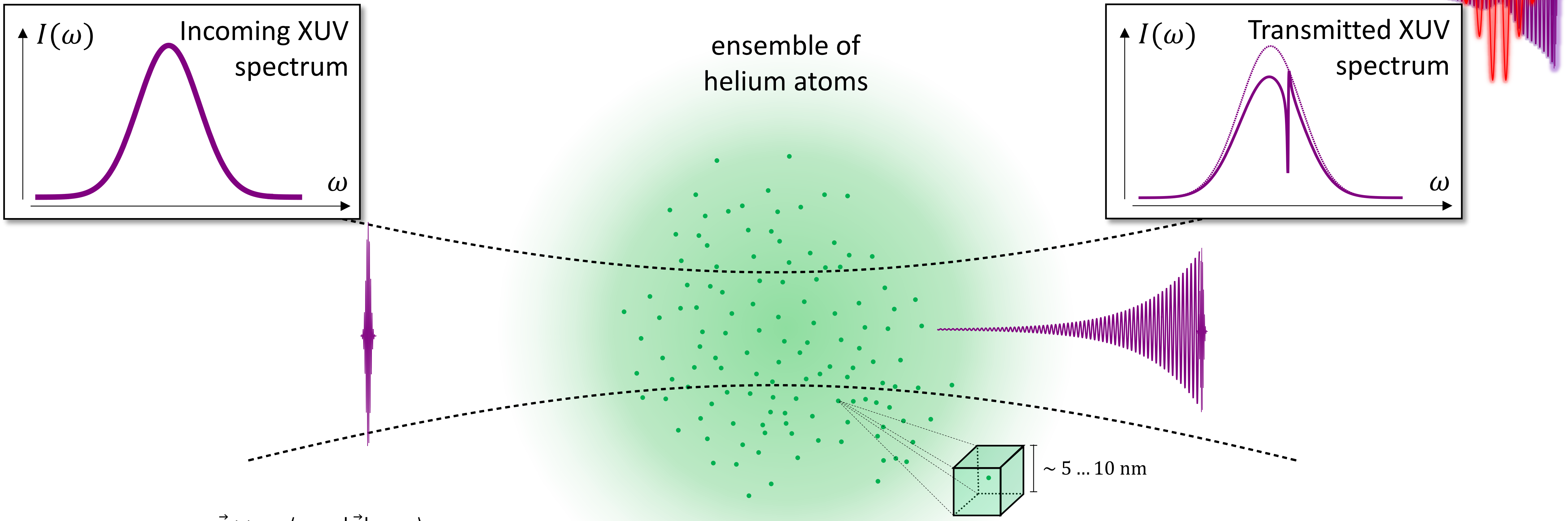
Selected Review Articles:

Limonov et al., Nature Photonics **11**, 543 – 554, (2017).

Miroshnichenko et al., Rev. Mod. Phys. **82**, 2257 – 2298, (2010).

Many applications in nanoscale structures

Controlling the coherent (XUV) dipole emission with laser pulses



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

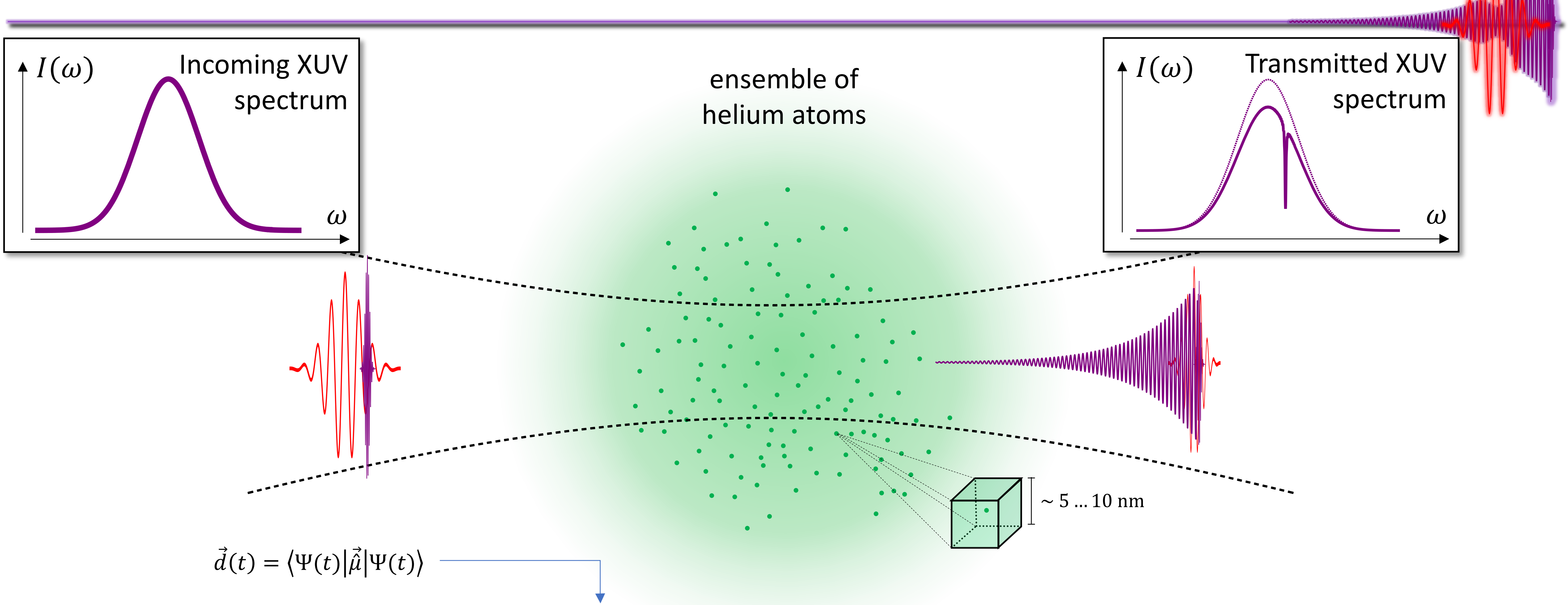
Optical Density $\propto \Im[n(\omega)] \propto \Im[\chi(\omega)] \propto \Im[\tilde{d}(\omega; E_{NIR}(t,\tau))]$

$$\mathrm{OD}(\omega) = \log\left[\frac{I_0(\omega)}{I(\omega)}\right] = \frac{\rho L}{\ln(10)} \frac{\omega}{\varepsilon_0 c} \Im\left[\frac{\tilde{d}(\omega; E_{NIR}(t,\tau))}{\tilde{\mathcal{E}}_{XUV}(\omega)}\right]$$

(for dilute media, i.e. optically thin)

- **Maxwell's propagation of radiation fields (refractive index)**
- **Phase matching and phase sensitivity of coherent excitations**
- **Interference of radiation fields are important!**

Controlling the coherent (XUV) dipole emission with laser pulses



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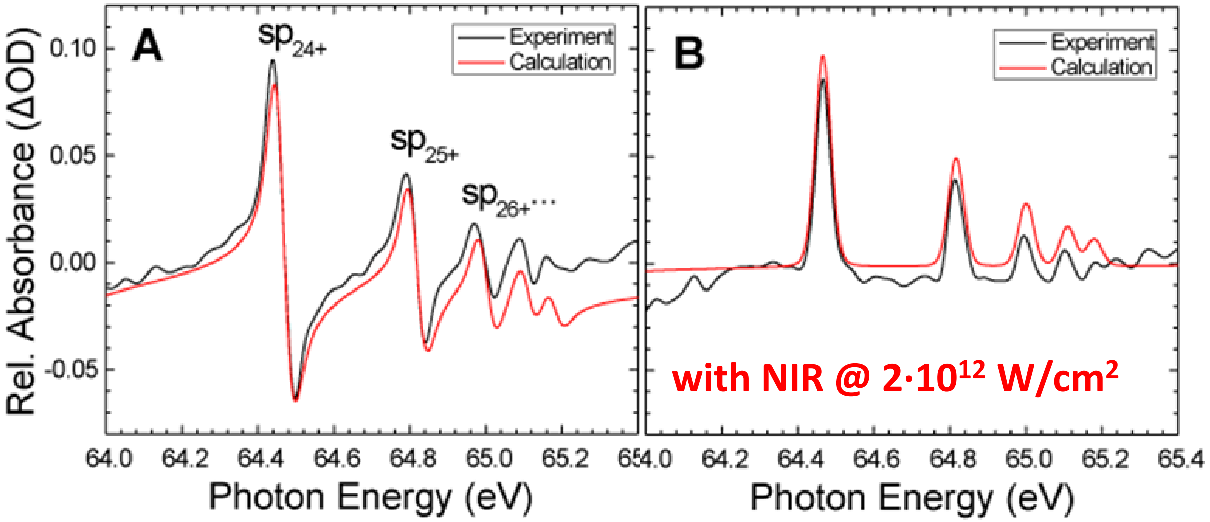
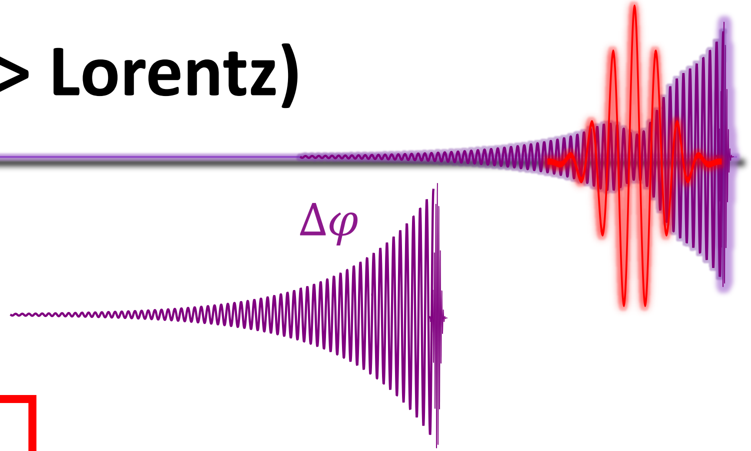
Laser control of spectral lineshapes (Fano <> Lorentz)

Time-domain phase control of spectral line shapes



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



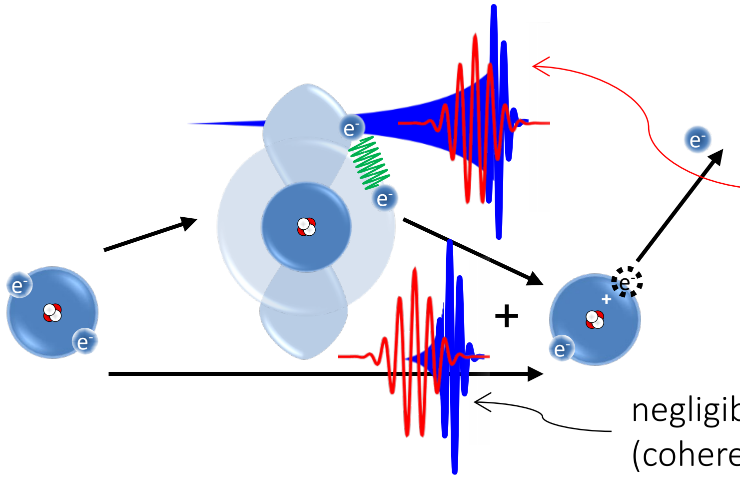
$$\varphi(q) = 2\arg(q-i)$$

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

➢ Time-domain picture of the Fano absorption lineshape: impulsively modified dipole response

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

➢ selectively modify the two-electron excited-state quantum pathway ***before*** its decay (autoionization)



coherent sum:
bound & continuum XUV dipole response

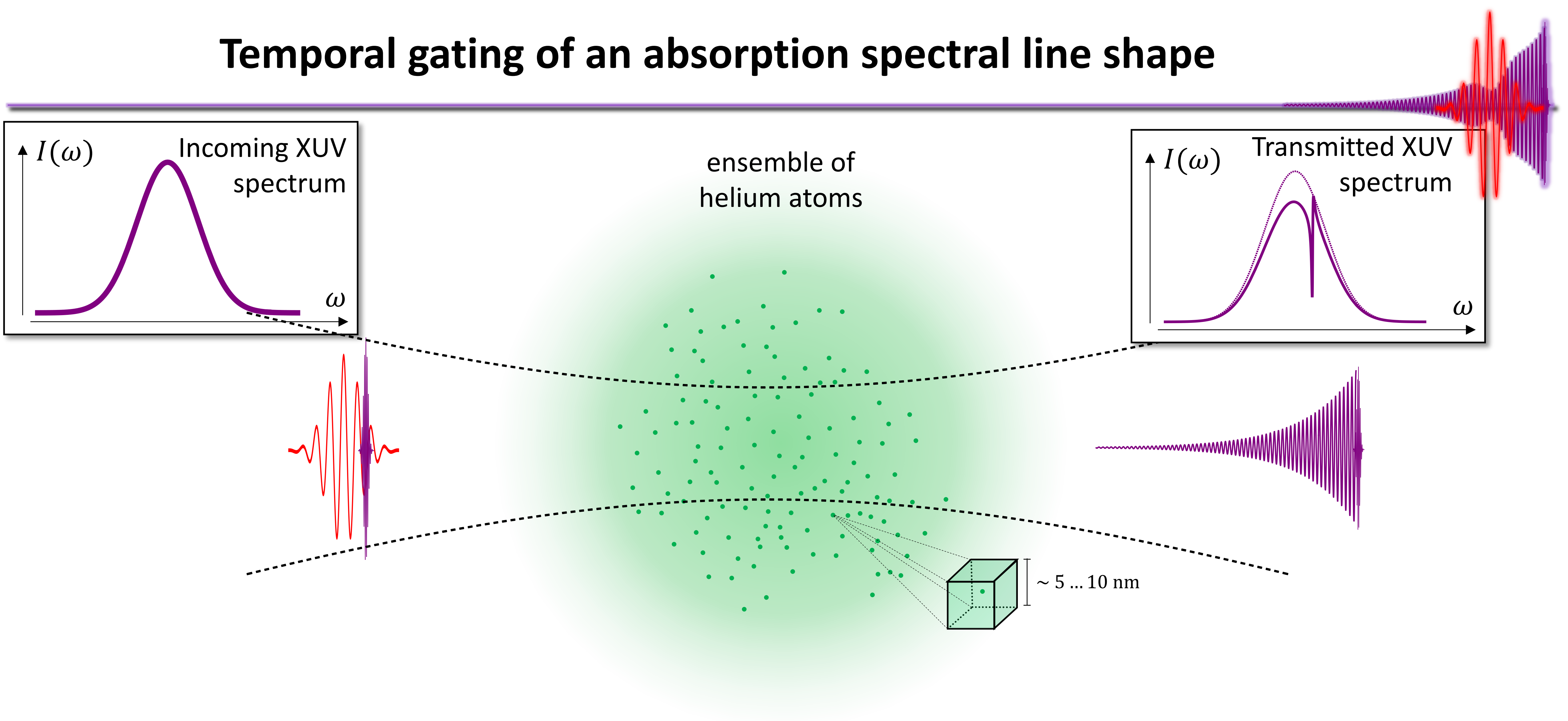
Laser-induced energy shift $\Delta E(t)$ [e.g., AC Stark, **strong-field effects**] of excited-state (coherent XUV dipole reponse):

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

negligible
(coherent control of XUV continuum dipole only in pulse temporal overlap)

Collab.: Greene, Keitel, Evers

Temporal gating of an absorption spectral line shape



$$\mathrm{OD}(\omega) = \frac{\rho L}{\ln(10)} \frac{\omega}{\varepsilon_0 c} \Im\left[\frac{\tilde{d}\big(\omega; E_{NIR}(\tau)\big)}{\tilde{\mathcal{E}}_{XUV}(\omega)}\right]$$

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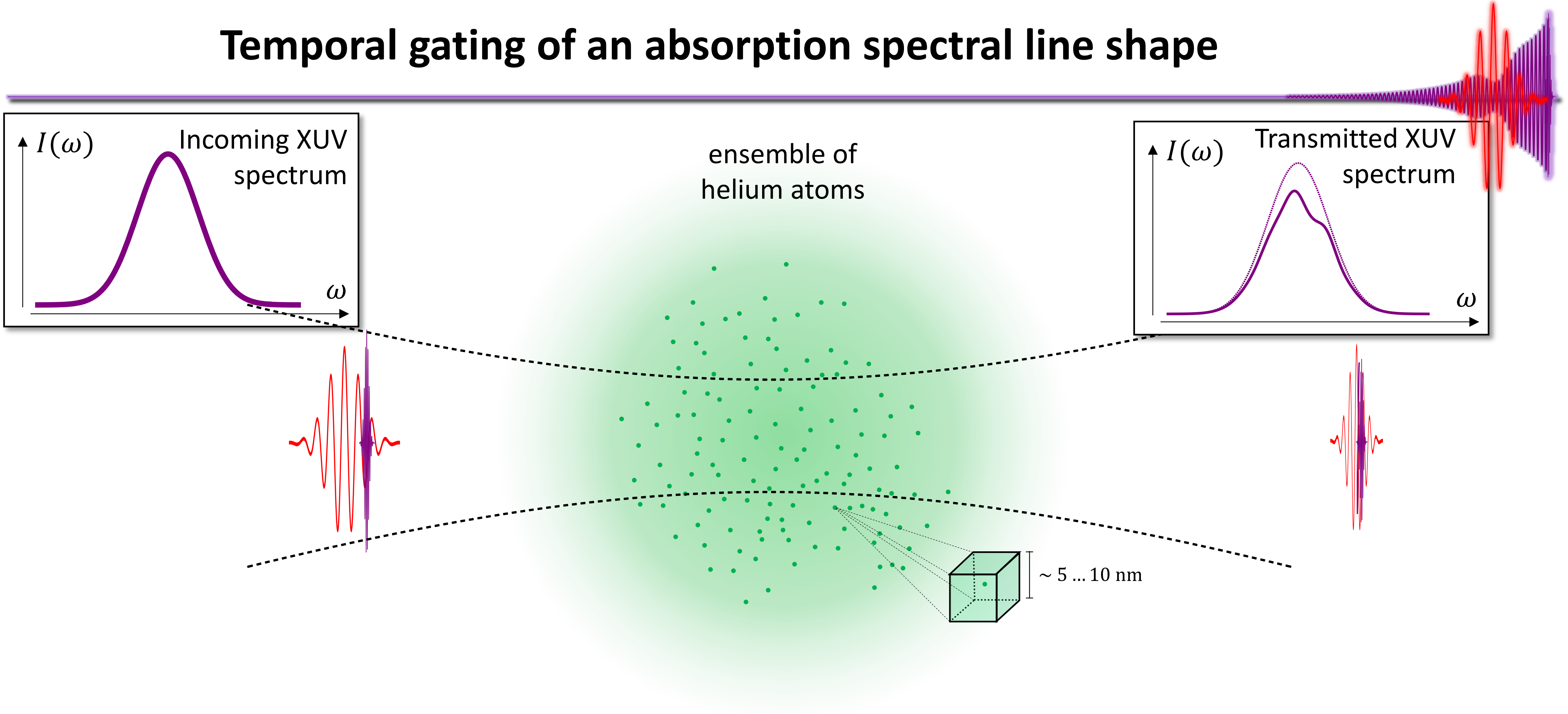

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- **A time-delayed strong NIR pulse interrupts the coherent XUV dipole emission**

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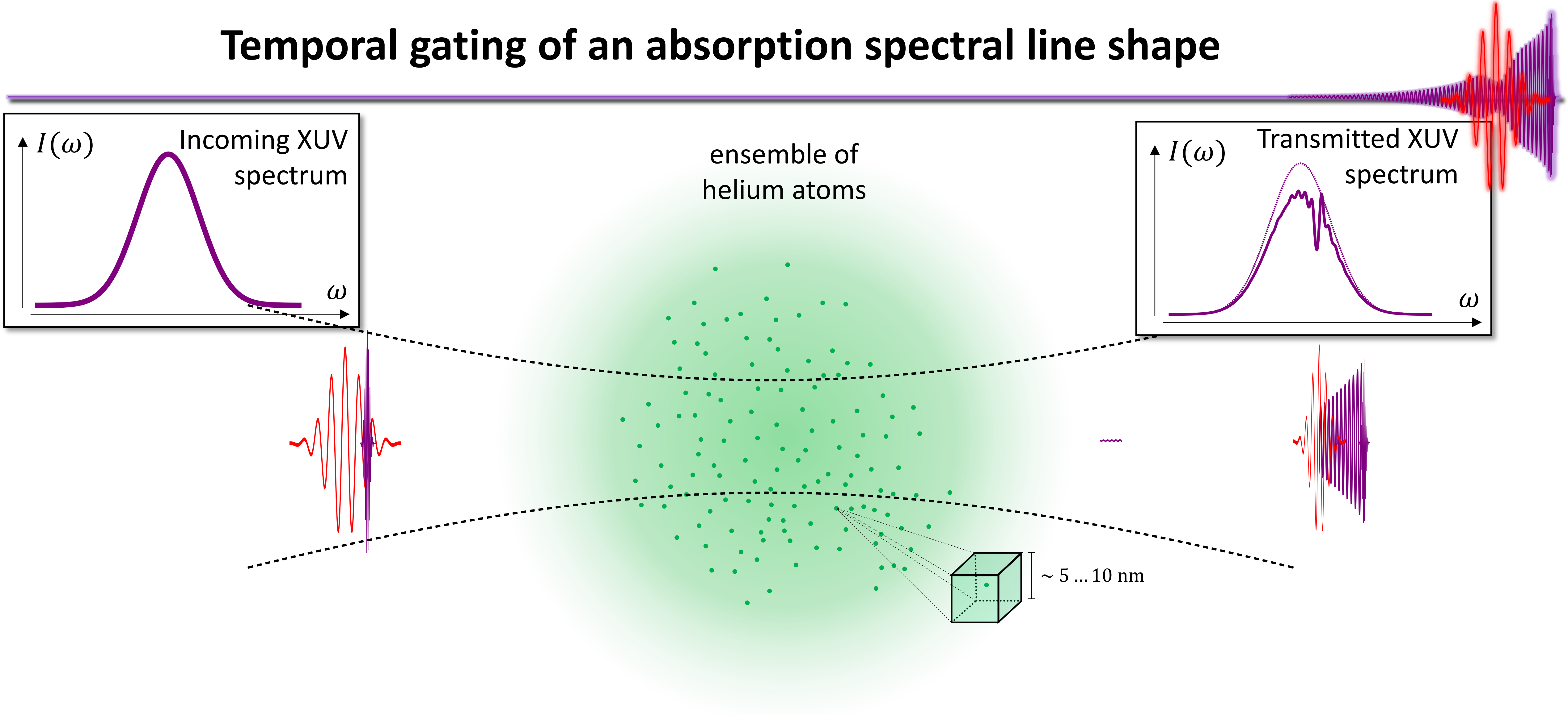

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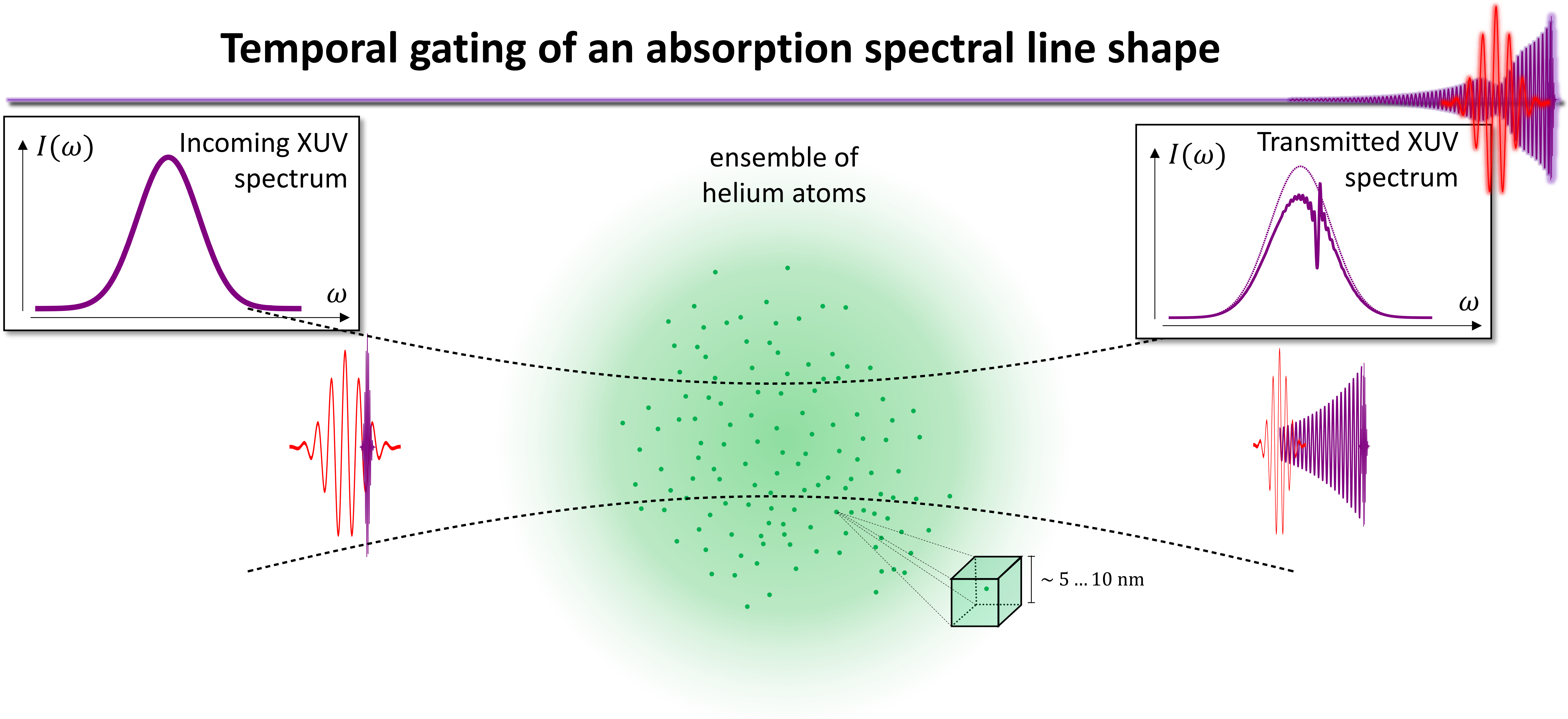

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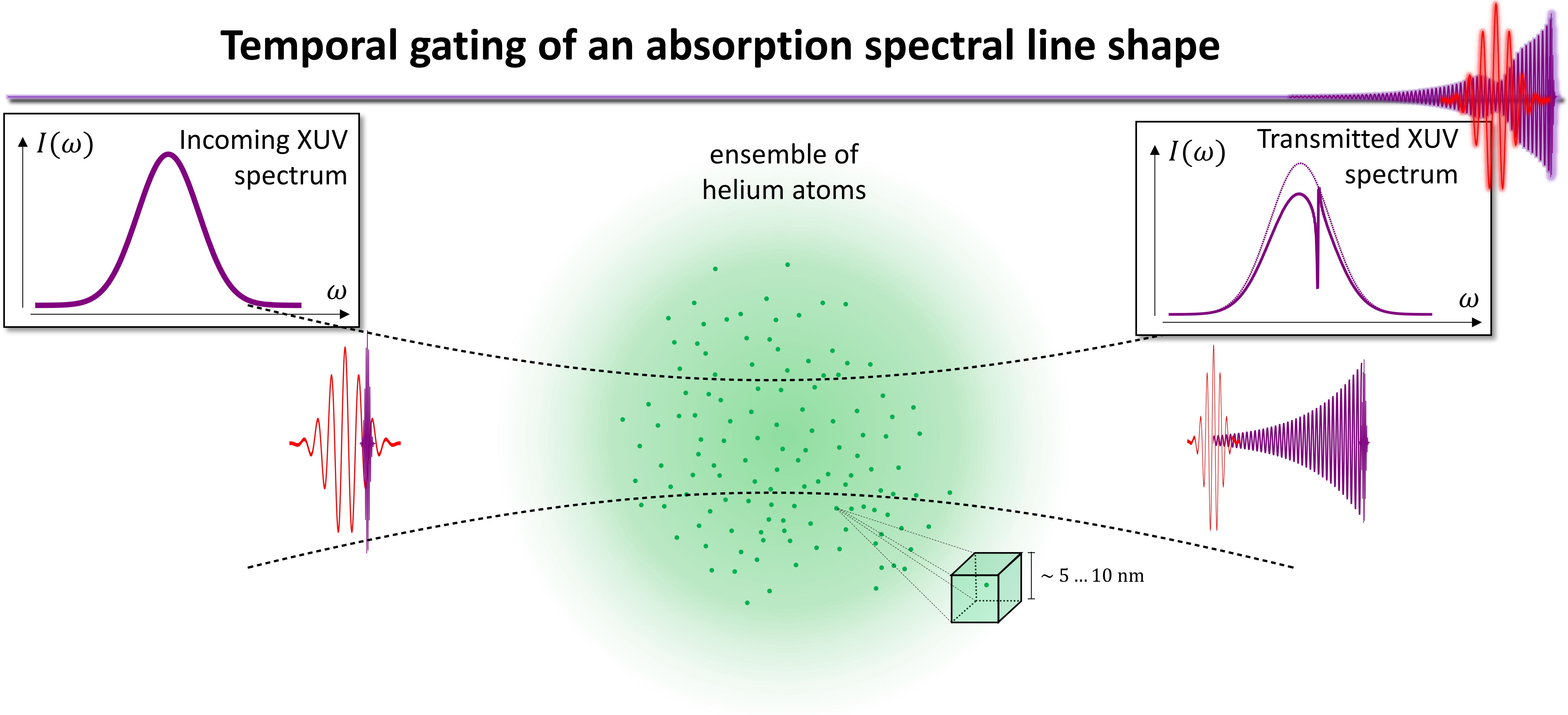

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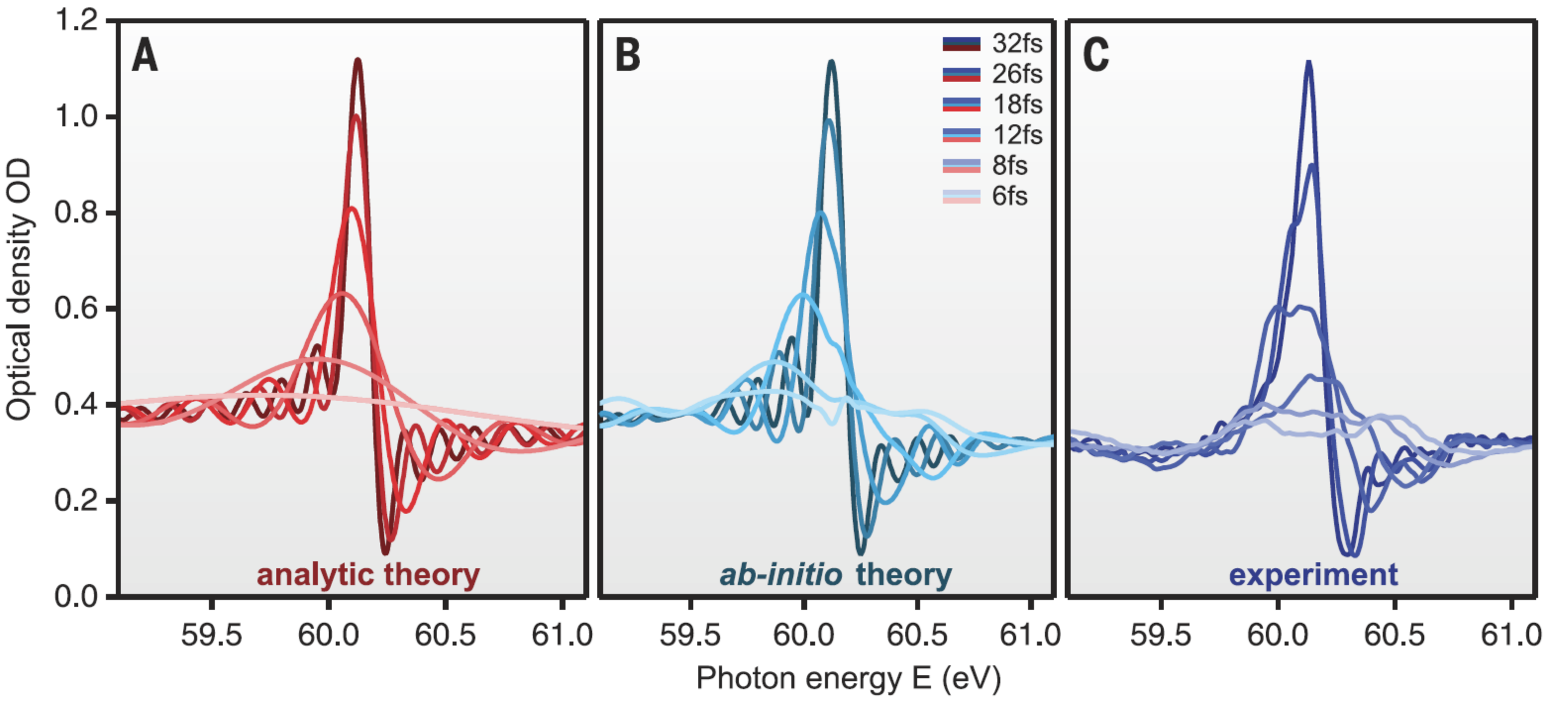
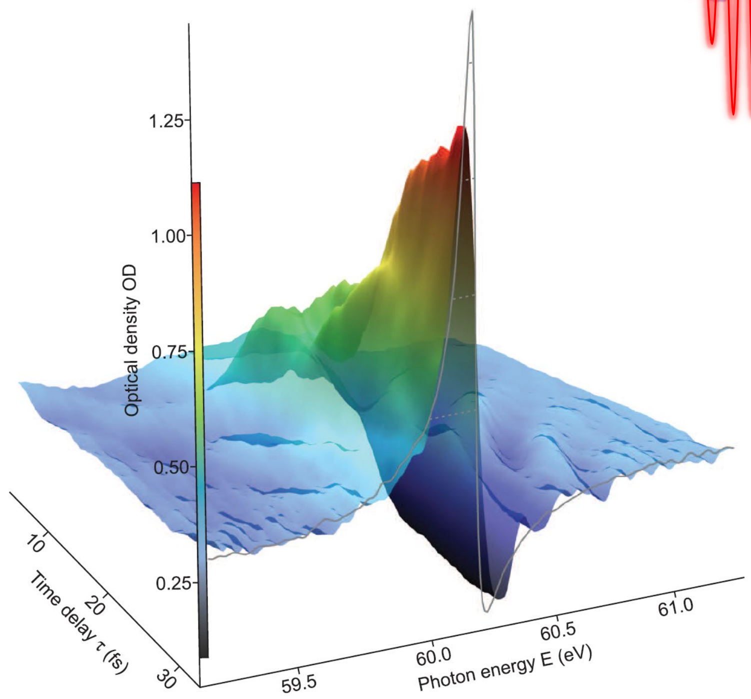
Buildup of a Fano absorption resonance





Collab.: C. D. Lin, J. Burgdörfer

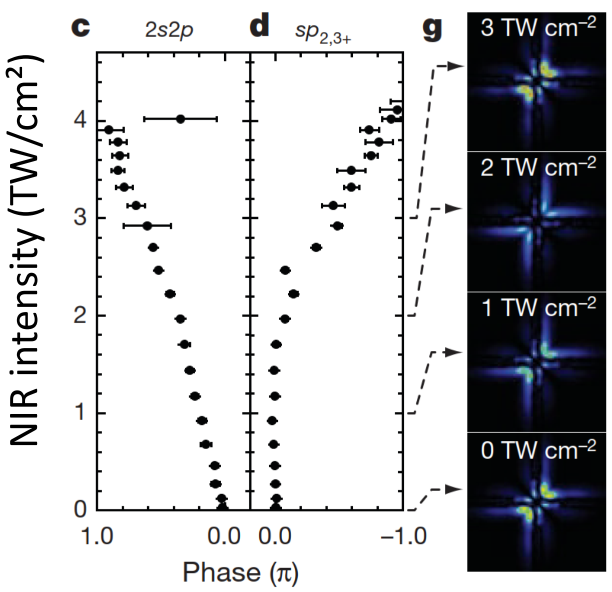
- The Fano lineshape encodes the **correlated electron-electron interaction**
- Direct measurement of how this interaction develops in real-time and their **control with strong laser fields**

Ultrafast quantum control of
coherent two-electron dynamics
of a bound quantum state
(„inside the atom“)

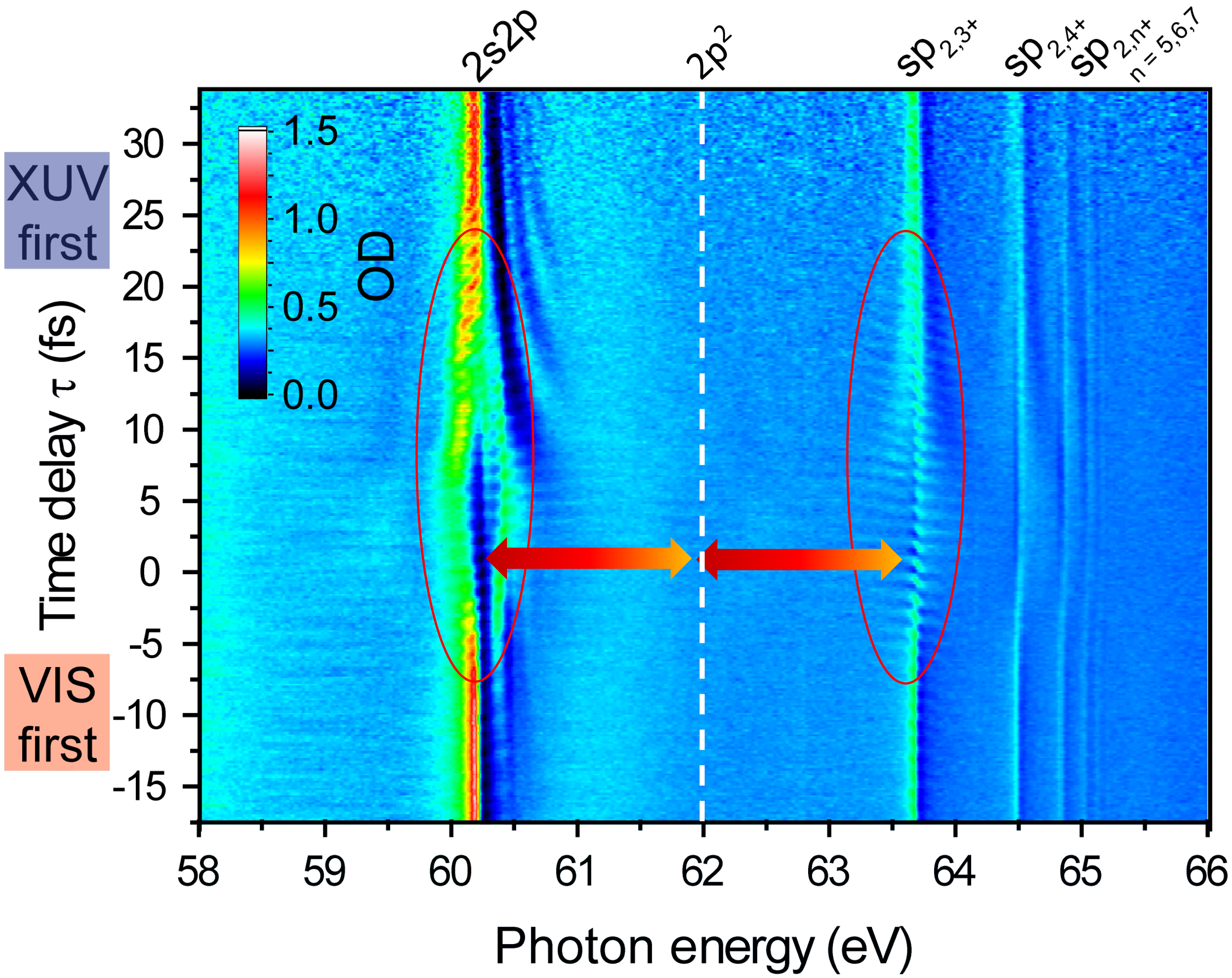
Kaldun, Blättermann et al., Science 354, 738 (2016)

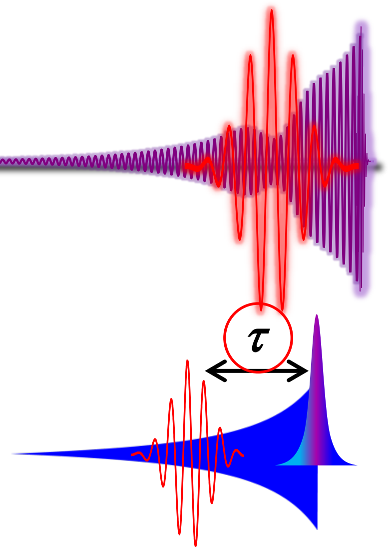
Complementary „monitoring birth of a photoelectron“ in: Gruson et al., Science 354, 734 (2016)

Coherent laser-control of a two-electron wave packet



full 2π phase tunability of a short-lived bound-state two-electron wave packet





2s2p – $sp_{2,3+}$: coherent 1.2 fs wavepacket beating

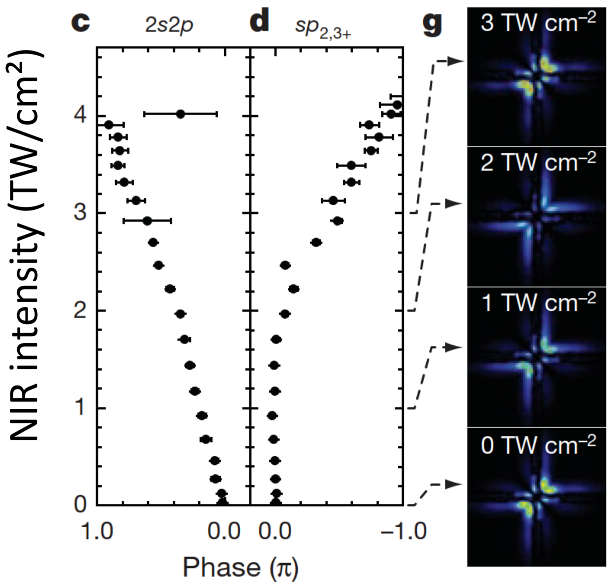
$$\propto e^{i\frac{\Delta E}{\hbar}\tau}$$

phase information through state-resolved interference of correlated two-electron states

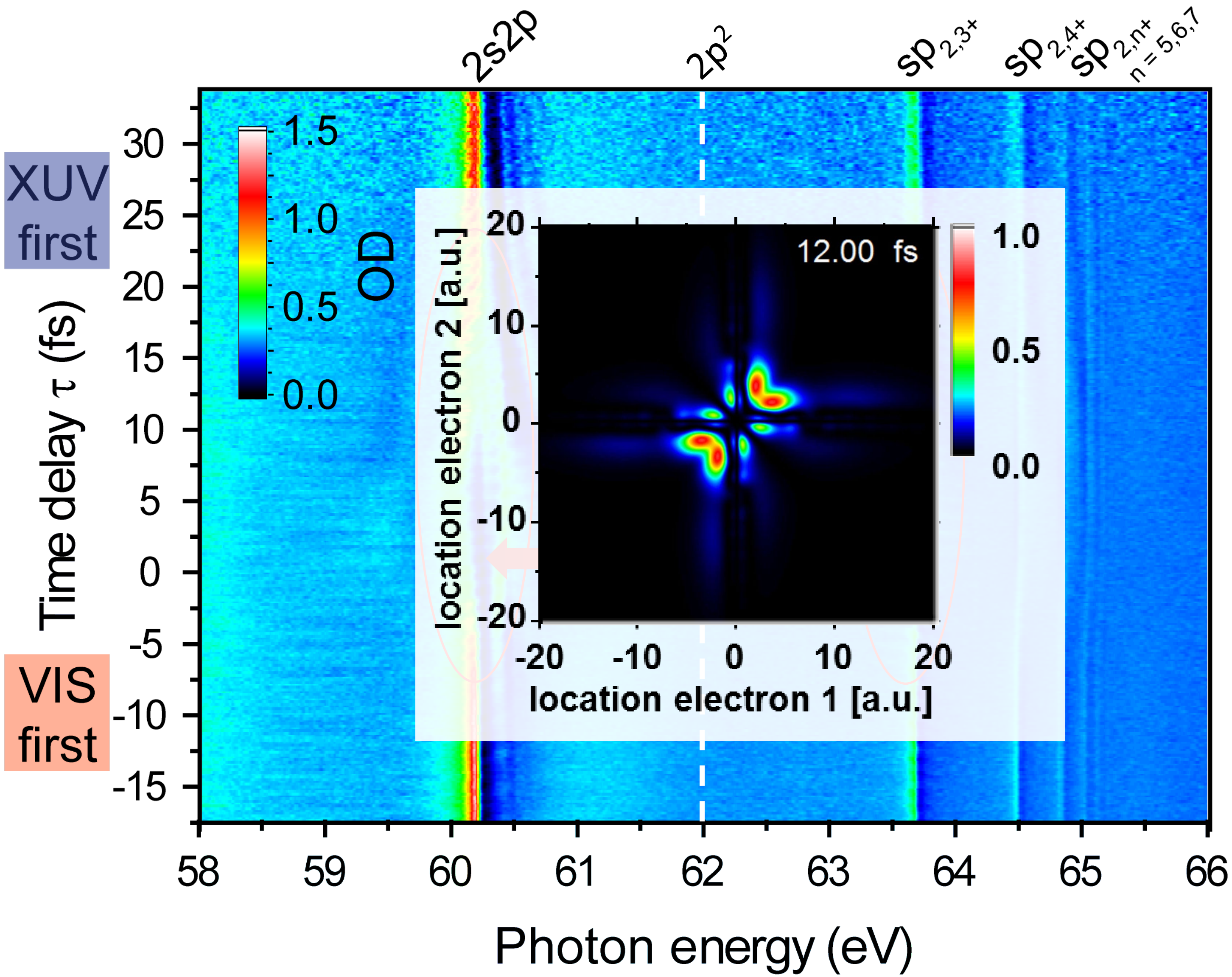
Ott et al., Nature **516**, 374 (2014)

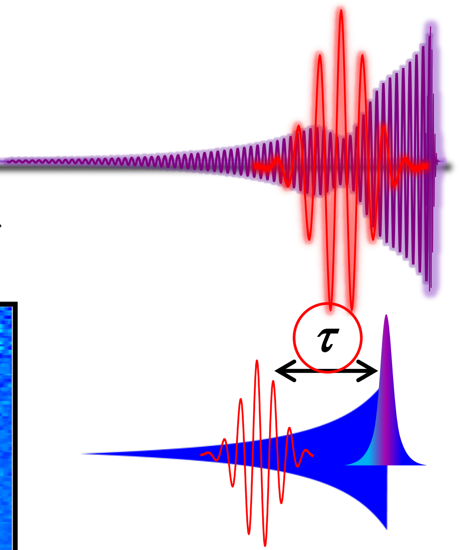
Collab.: L. Argenti, F. Martín

Coherent laser-control of a two-electron wave packet



full 2π phase tunability of a short-lived bound-state two-electron wave packet





$2s2p - sp_{2,3+}$: coherent 1.2 fs wavepacket beating

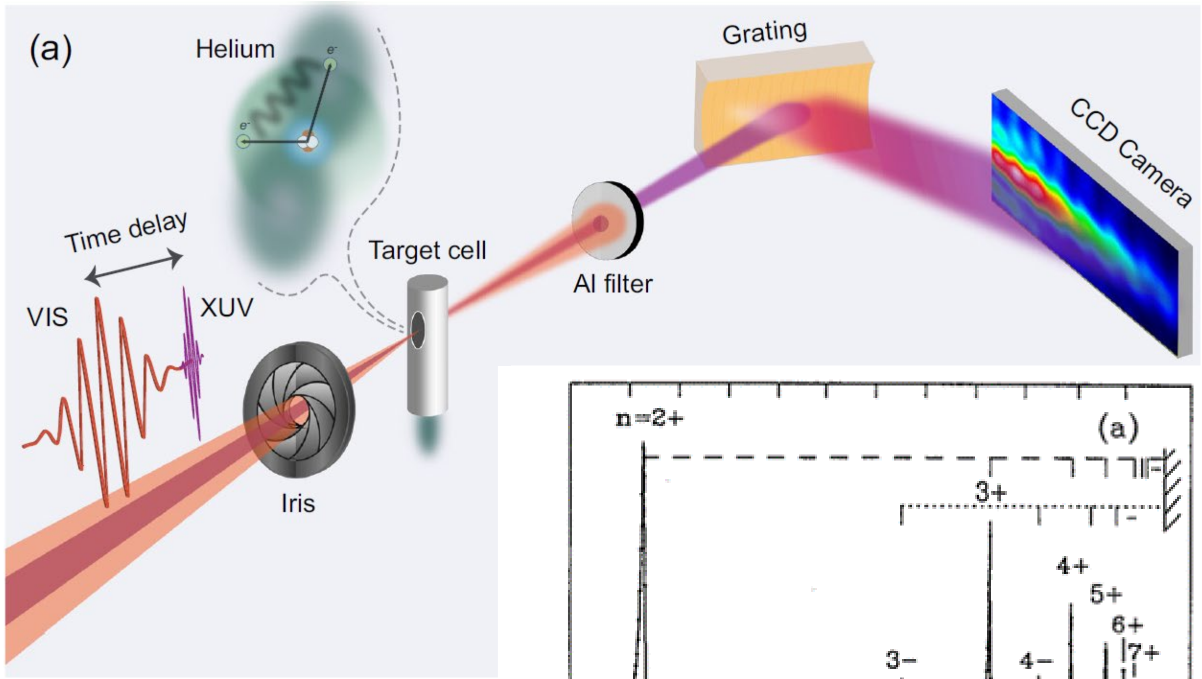
$$\propto e^{i\frac{\Delta E}{\hbar}\tau}$$

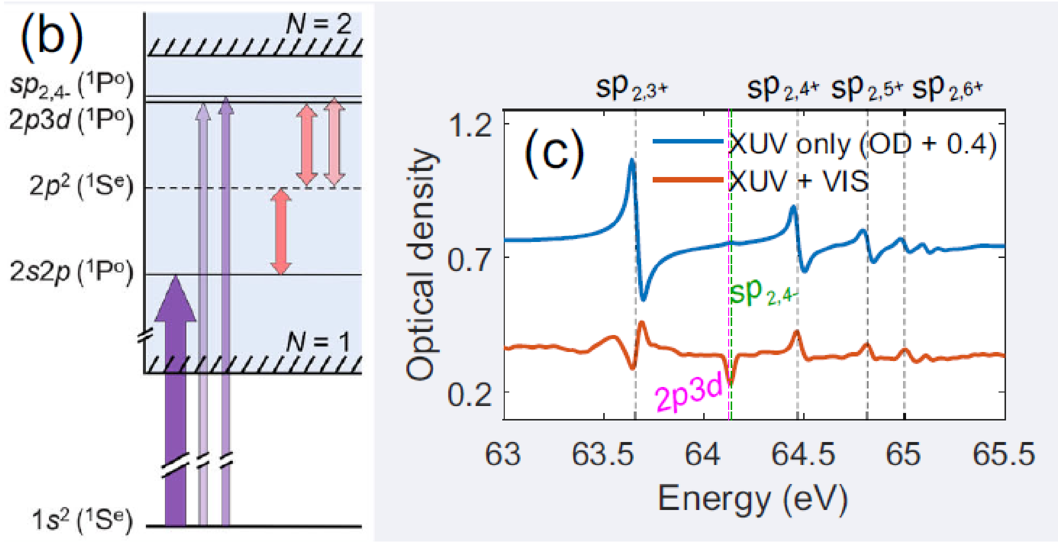
phase information through state-resolved interference of correlated two-electron states

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

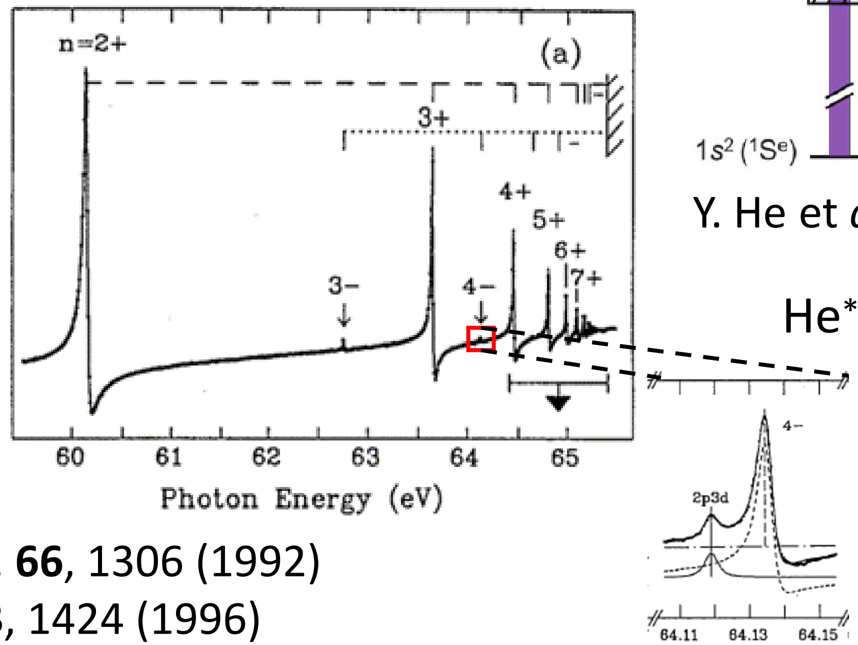
Collab.: L. Argenti, F. Martín

Bringing weak two-electron transitions in helium to light





Y. He et *al.*, Nature Communications **16**, 5322, (2025).



all 3 series only resolved in 1990's with high-sensitivity synchrotron experiments

He** $^1P^o$ states converging to He++ $(N = 2)$

$2s \rightarrow np$ $\quad 2p \rightarrow ns,\ nd$

⇒ three distinct series of He** ($^1P^o$) two-electron states

$2snp + 2pns \equiv sp_{2n+}$

$2snp - 2pns \equiv sp_{2n-}$

$2pnd$

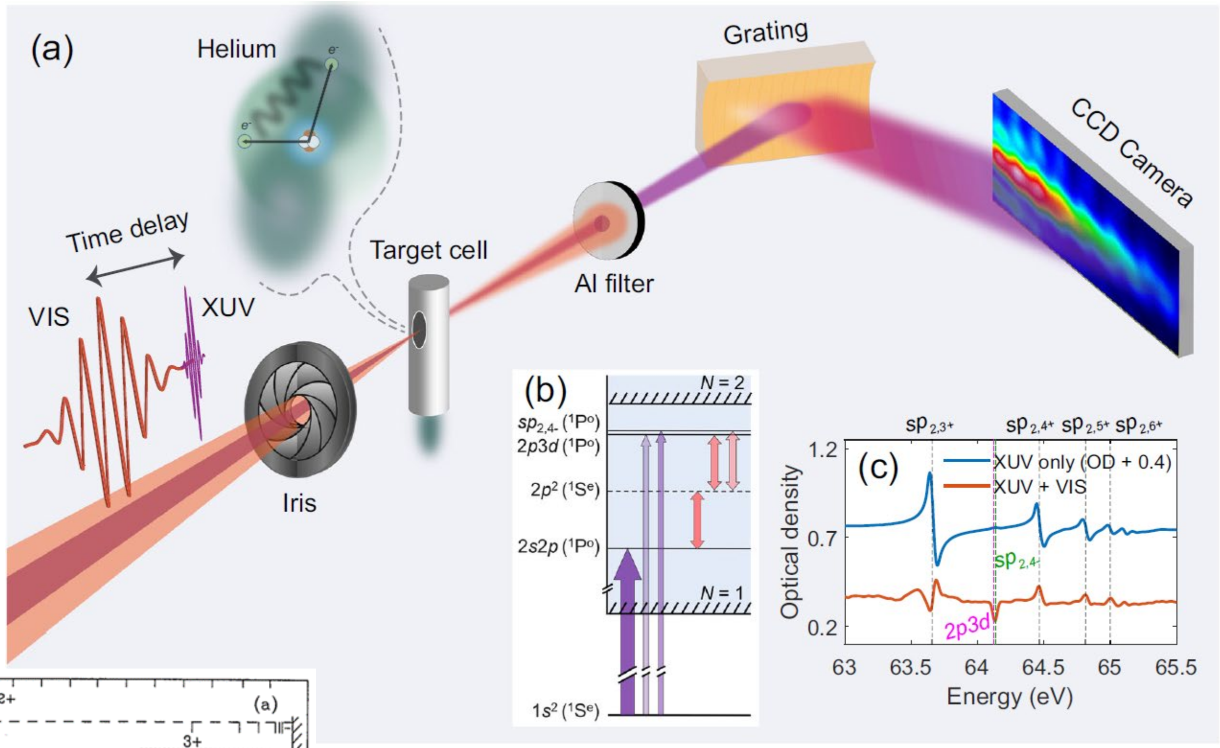
M. Domke *et al.*, Phys. Rev. Lett. **66**, 1306 (1992)

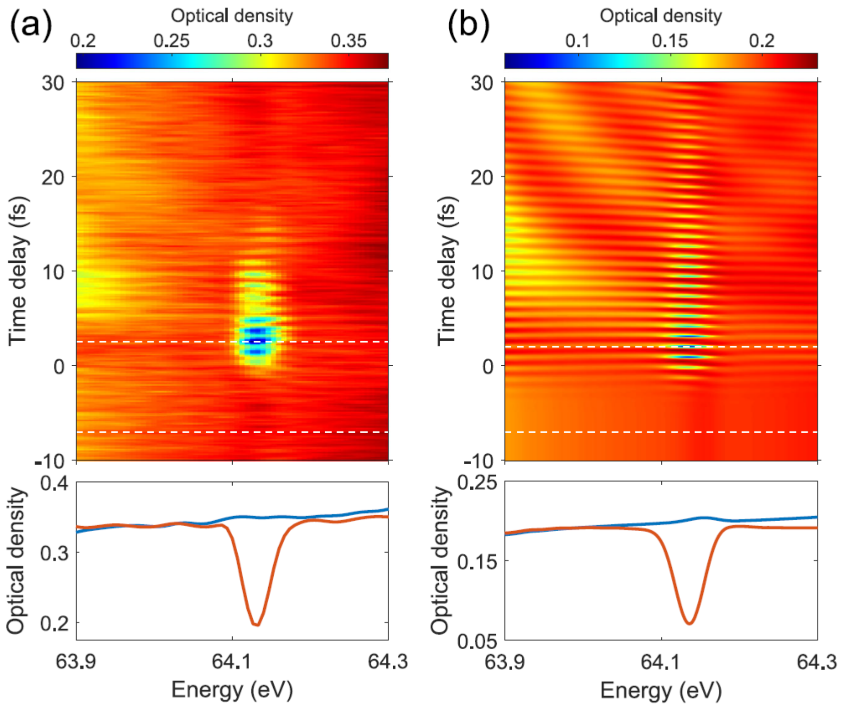
M. Domke *et al.*, Phys. Rev. A **53**, 1424 (1996)

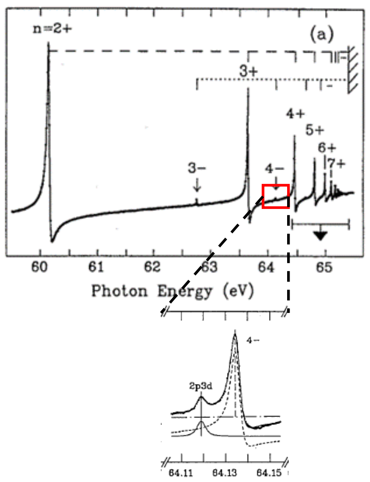
C. D. Lin, "Hyperspherical coordinate approach to atomic and other Coulombic three-body systems", Physics Reports **257**, 1-83, (1995).

G. Tanner, K. Richter, J.-M. Rost, "Theory of two-electron atoms", Rev. Mod. Phys. **72**, 497-544, (2000).

Bringing weak two-electron transitions in helium to light

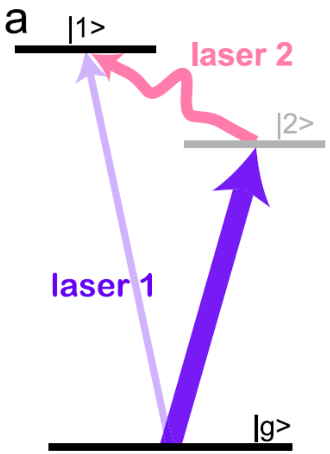


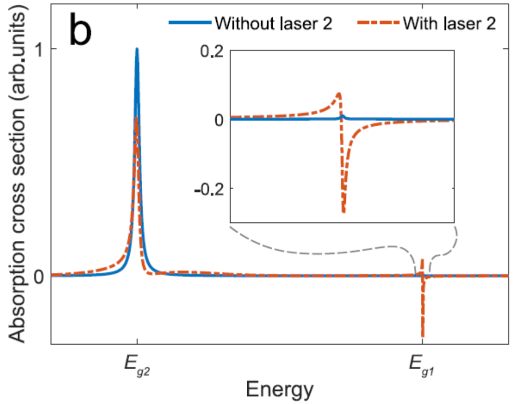




Attosecond time-resolved enhancement of doubly excited states that are only very weakly coupled from the ground state

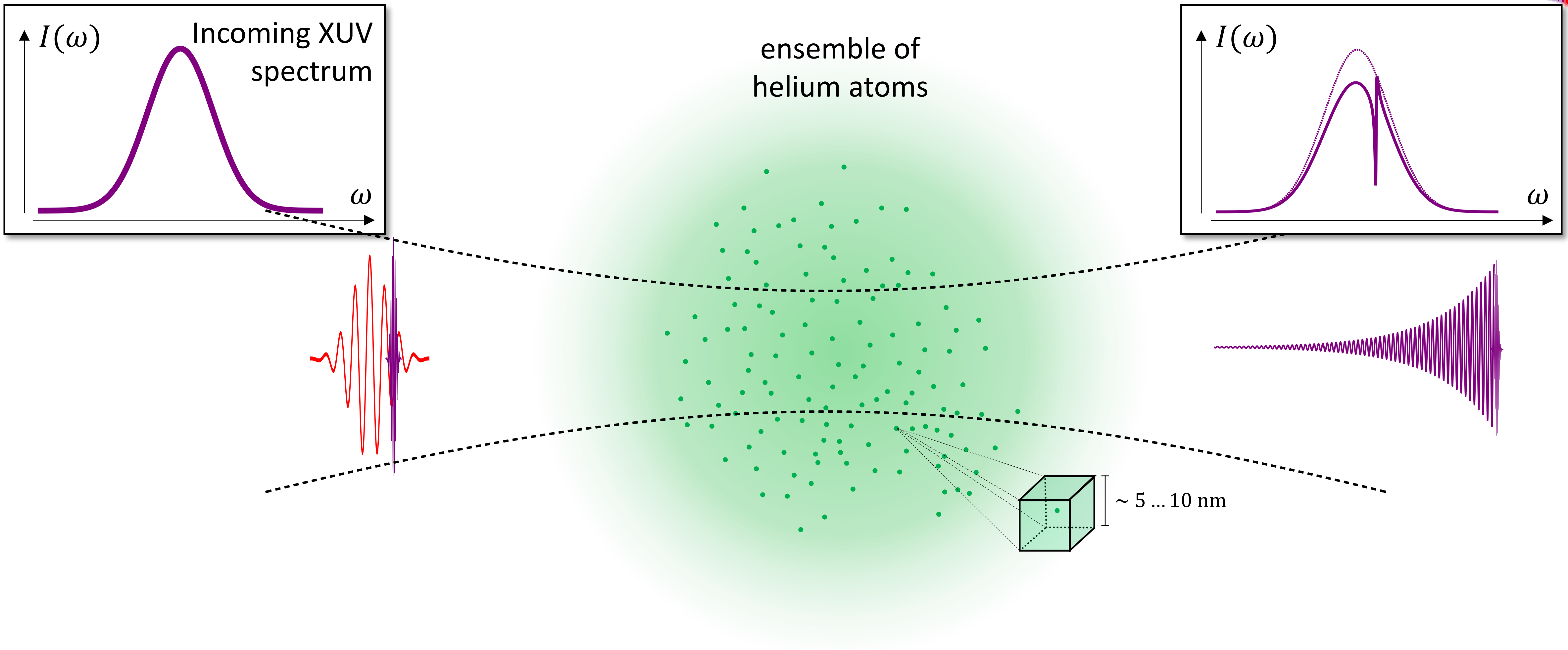
Collab.: X. M. Tong





Y. He et *al.*, Nature Communications **16**, 5322, (2025).

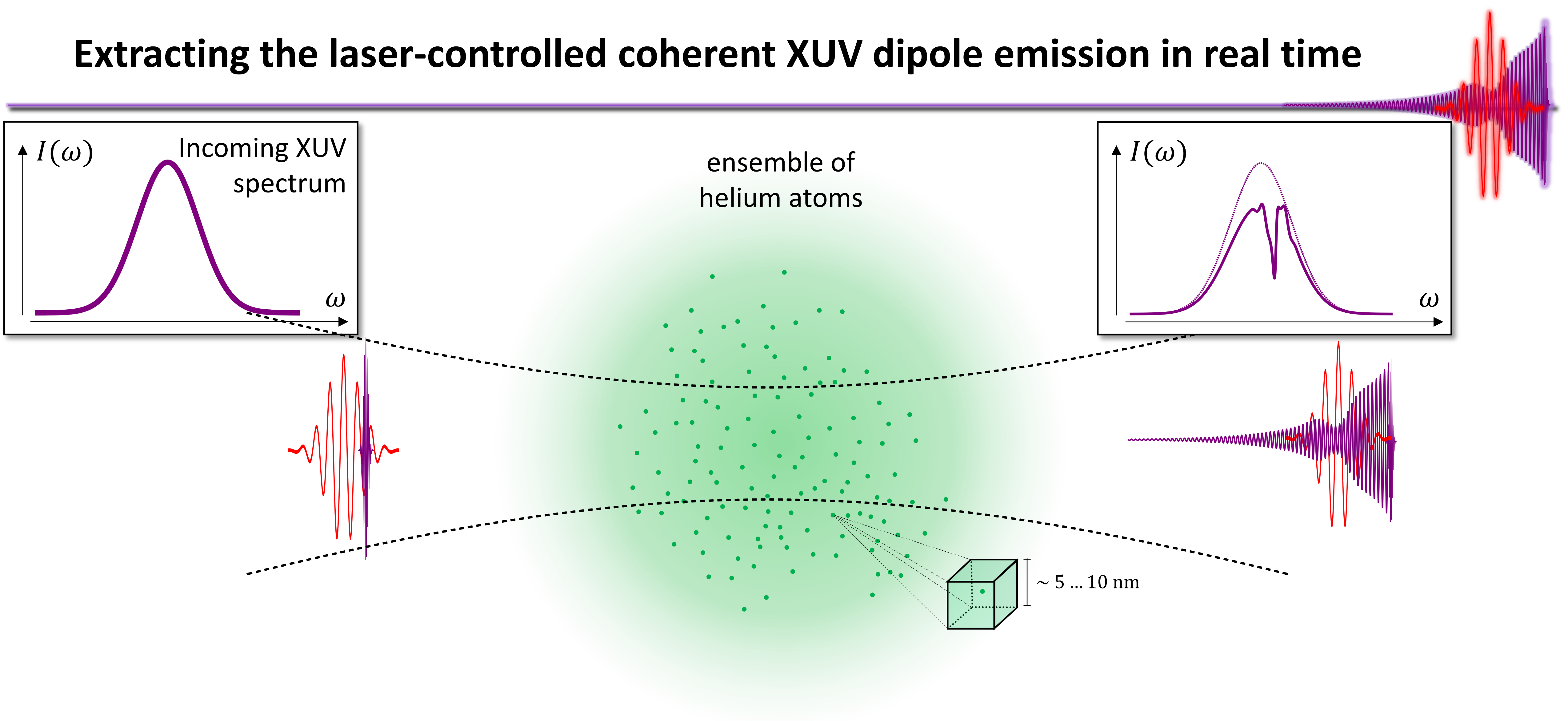
Extracting the laser-controlled coherent XUV dipole emission in real time



$$\mathrm{OD}(\omega) = \frac{\rho L}{\ln(10)} \frac{\omega}{\varepsilon_0 c} \Im\left[\frac{\tilde{d}(\omega; E_{NIR}(\tau))}{\tilde{\mathcal{E}}_{XUV}(\omega)}\right]$$

- **A time-delayed strong NIR pulse imprints ultrafast dynamics into the coherent XUV dipole emission**

Extracting the laser-controlled coherent XUV dipole emission in real time



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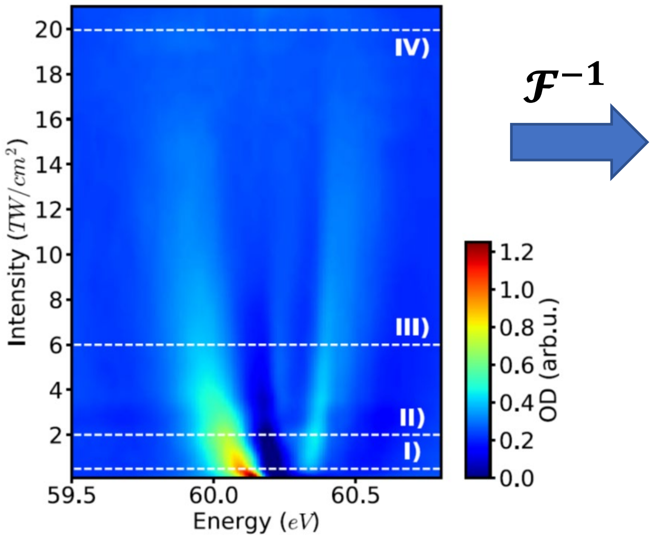
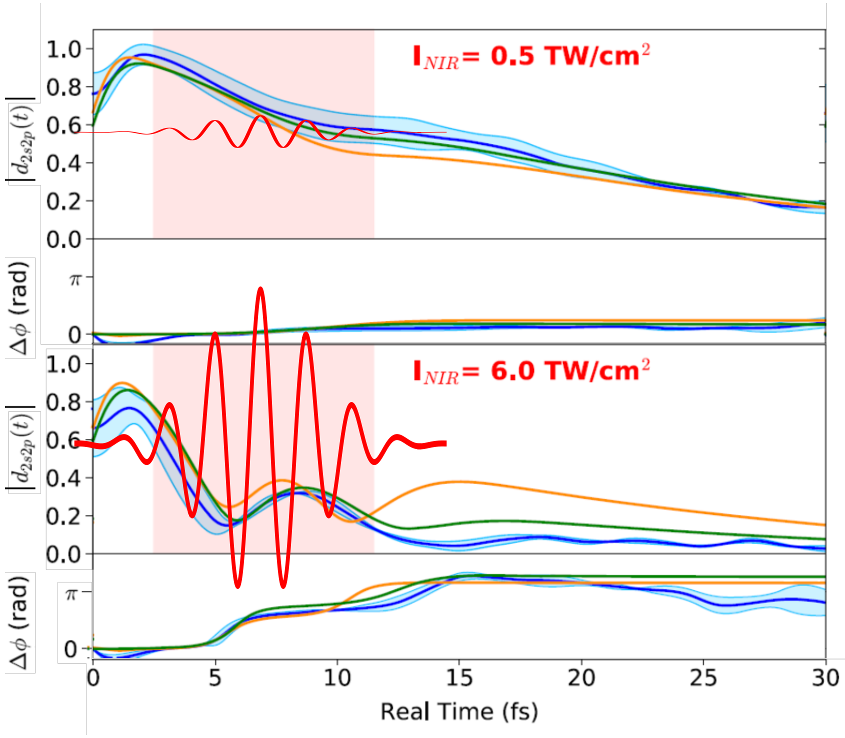
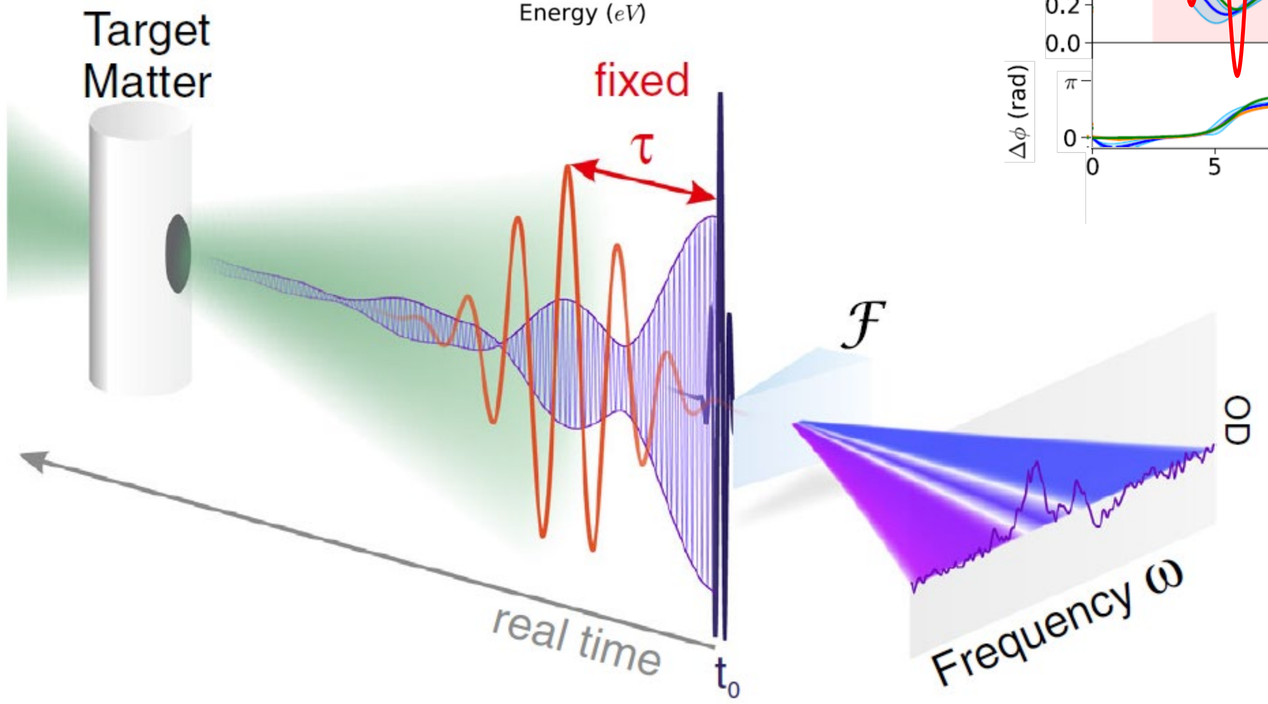




A real-time view into the strong-coupling dynamics of a superposition of two-electron states

Collab.: J. Burgdörfer



experimental realization of impulsive „Dirac delta“ coherent response triggered by attosecond XUV pulse

$$d(t) \propto \mathfrak{F}^{-1}\left[iA\left(\omega; \delta_{XUV}(t-t_0) \& E_{NIR}(t,\tau)\right)\right](t) \quad \text{for } t > t_0$$

$$= \frac{1}{2\pi}\int_{-\infty}^{+\infty} iA\left(\omega; \delta_{XUV}(t-t_0) \& E_{NIR}(t,\tau)\right) e^{-i\omega t} d\omega$$

Stooß et al., Phys. Rev. Lett. **121**, 173005 (2018)

Outline

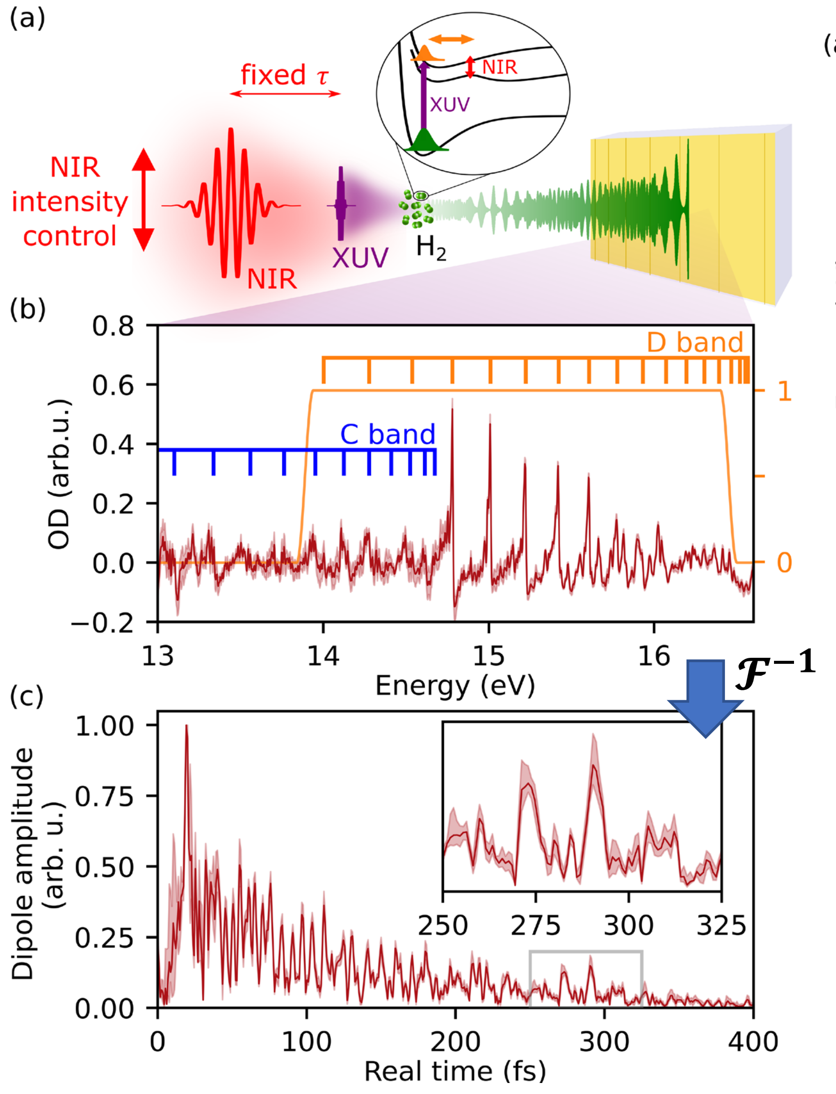
1) Introduction: a time-domain view into absorption spectroscopy

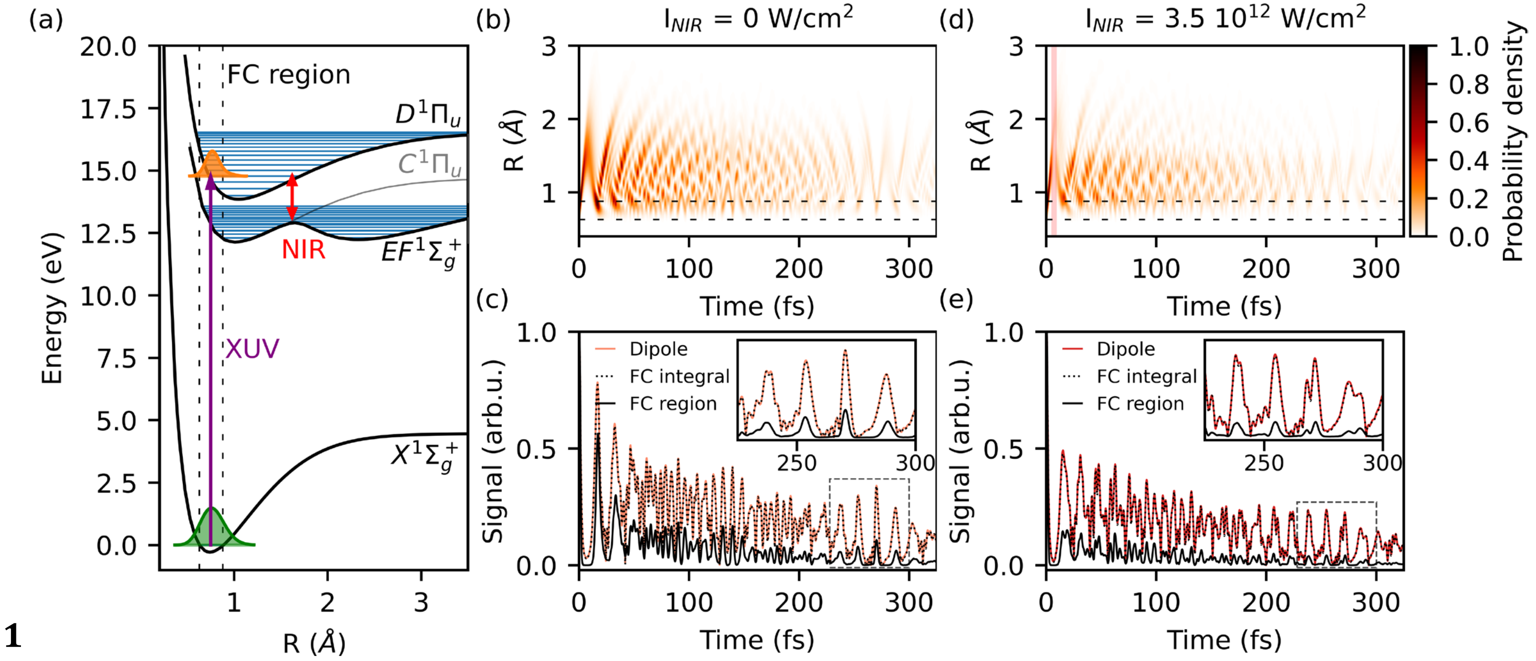
2) Learning from laser-controlled spectral line shapes of doubly excited states in helium

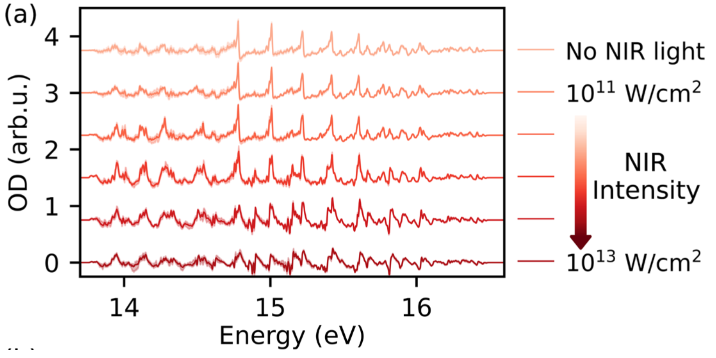
3) Electronic-state-resolved dynamics in small molecules and their laser control

4) Conclusion

Coherent control of an ultrafast vibration in intact (neutral) H_2 molecule



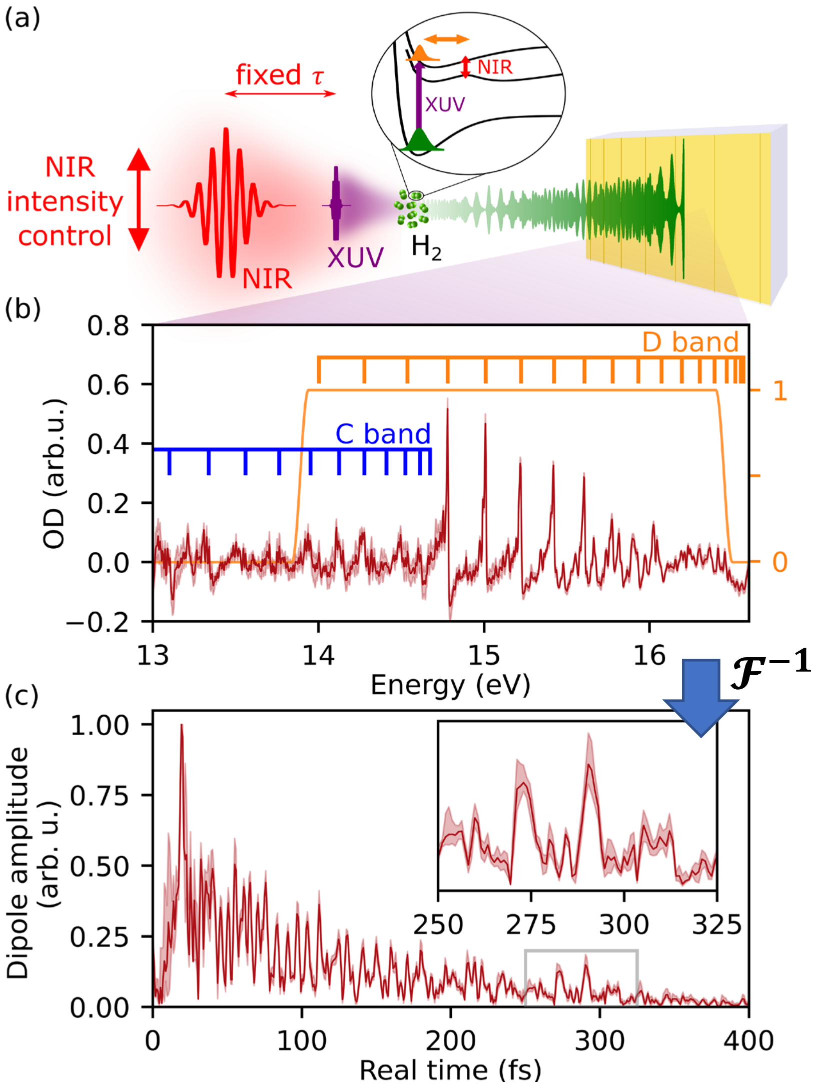


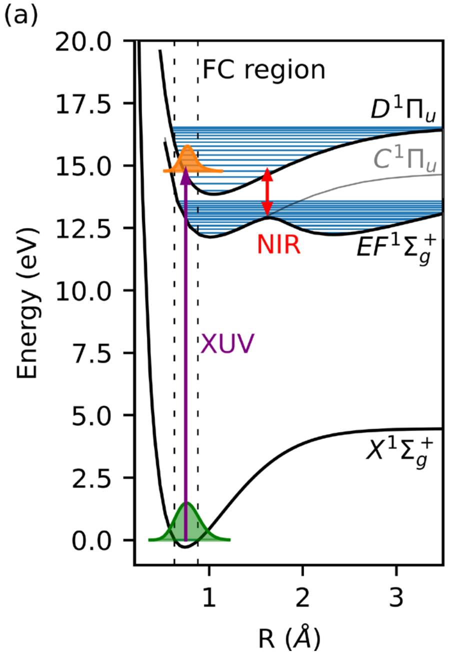


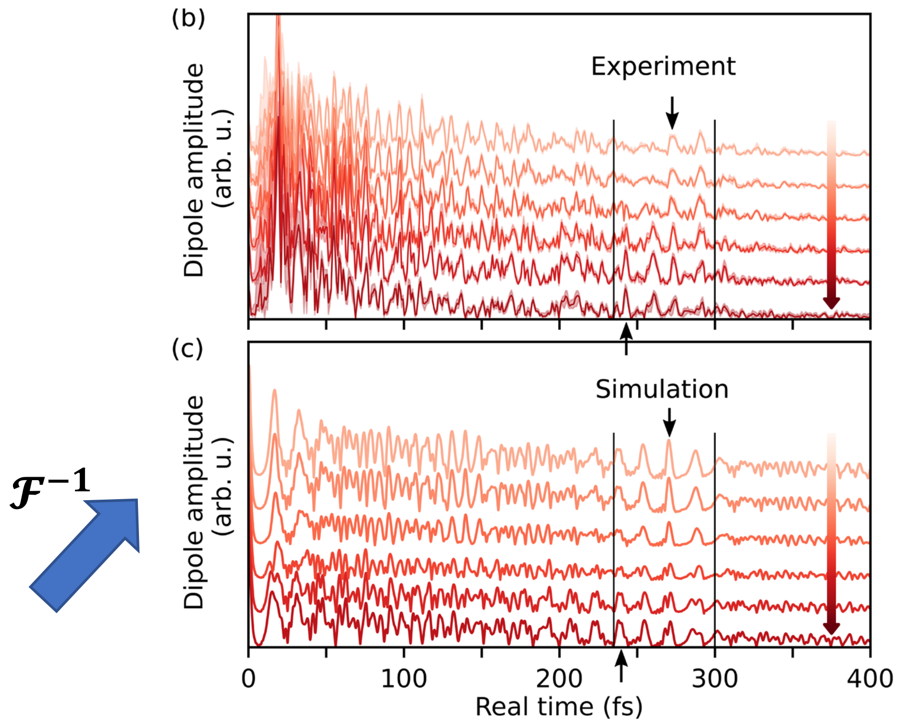
Collab.: A. Saenz

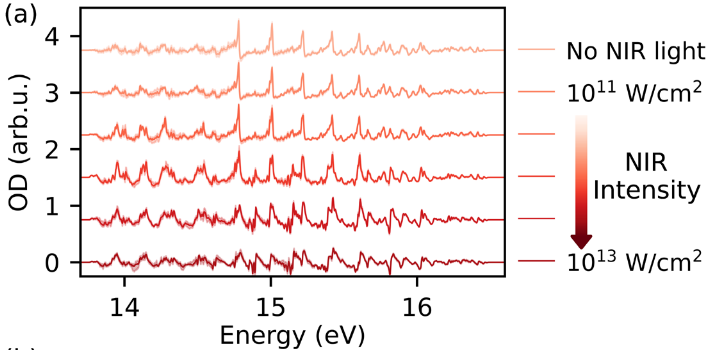
Borisova et al., Phys. Rev. Research **6**, 033326 (2024)

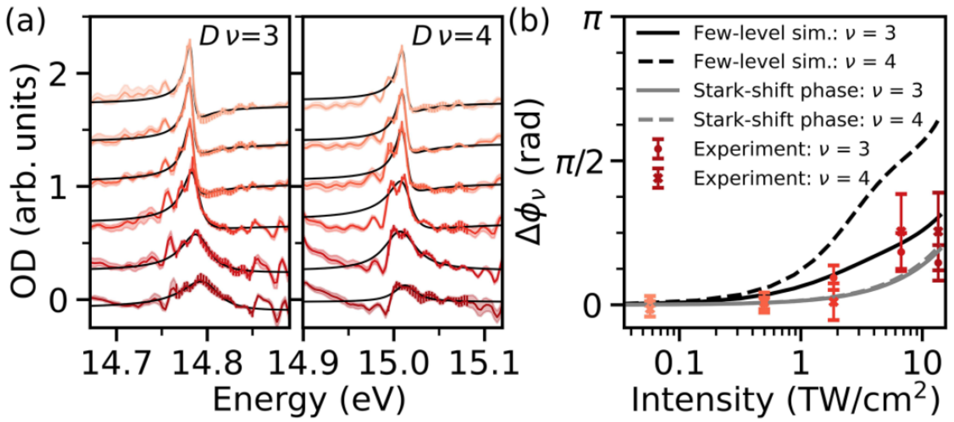
Coherent control of an ultrafast vibration in intact (neutral) H_2 molecule









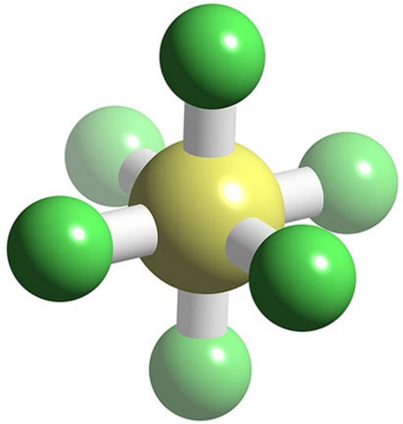


with state-resolved extraction of ultrafast quantum phase shifts

Collab.: A. Saenz

Borisova et al., Phys. Rev. Research **6**, 033326 (2024)

X-ray absorption spectroscopy of SF_6 (the sulfur $L_{2,3}$ edge)



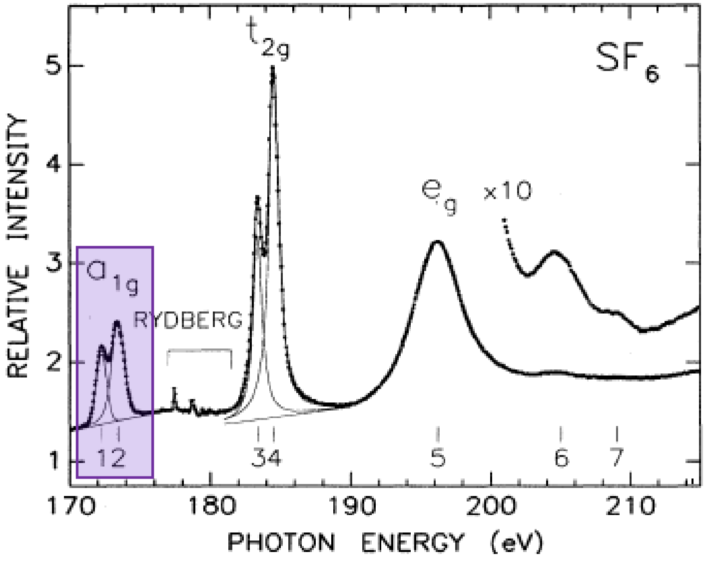
https://www.chemtube3d.com/sf6/

Octohedral geometry

„molecular noble gas“

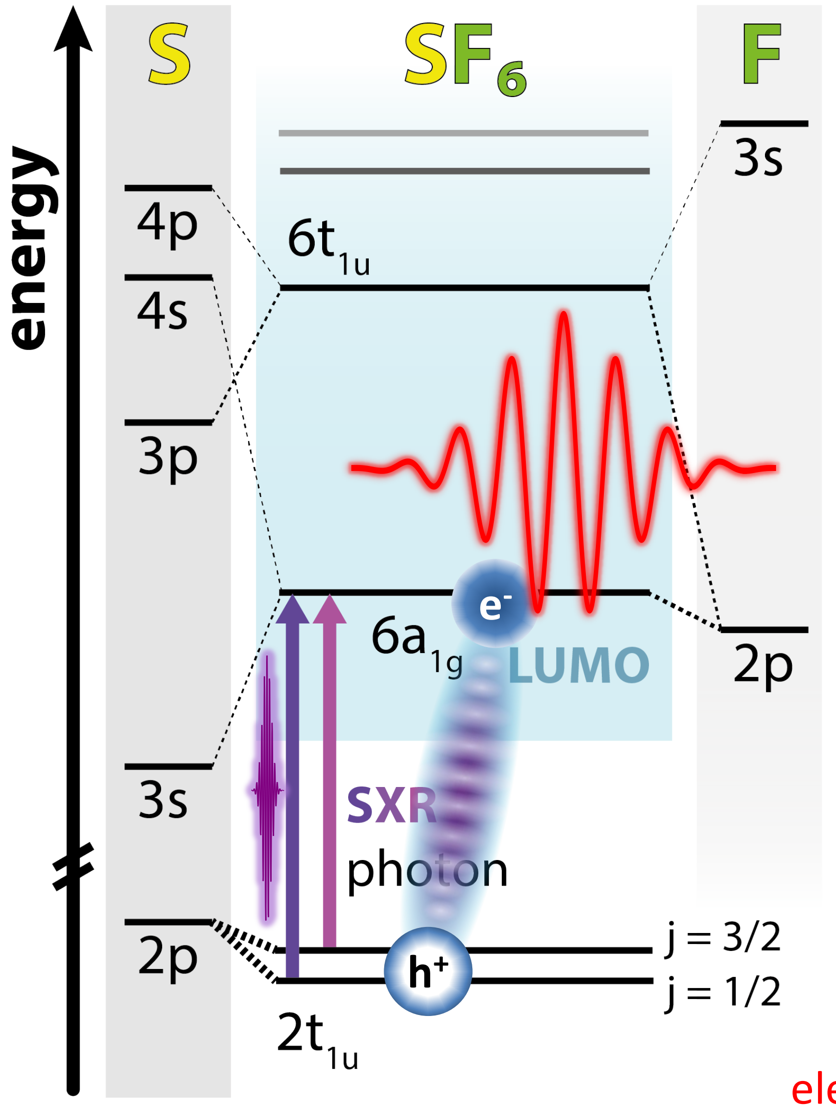
Sulfur: $1s^2\ 2s^2\ 2p^6\ 3s^2\ 3p^4$

Fluorine: $1s^2\ 2s^2\ 2p^5$



Hudson et al. PRA 1993

Energy level scheme according to Dehmer, J. Chem. Phys. 1972



Multi-electron orbitals with many-body wavefunctions (mediated by Coulomb interaction)

$$\iint \psi_h^*(\vec{r_1})\,\psi_e^*(\vec{r_2})\,\frac{1}{r_{12}}\,\psi_h(\vec{r_2})\,\psi_e(\vec{r_1})\,d\vec{r_1}\,d\vec{r_2}$$

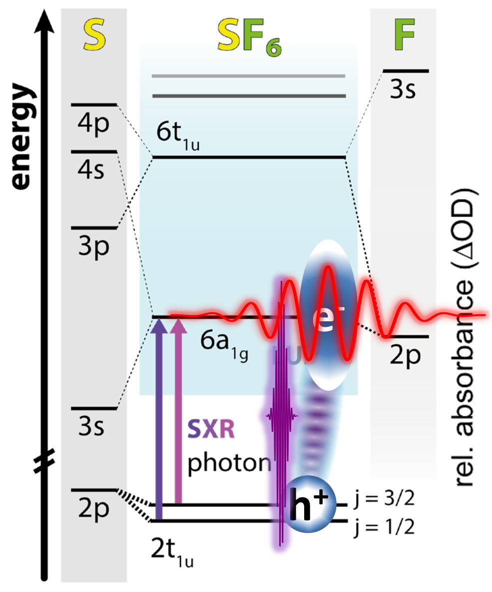
Exchange interaction due to Pauli principle with indistinguishable fermions

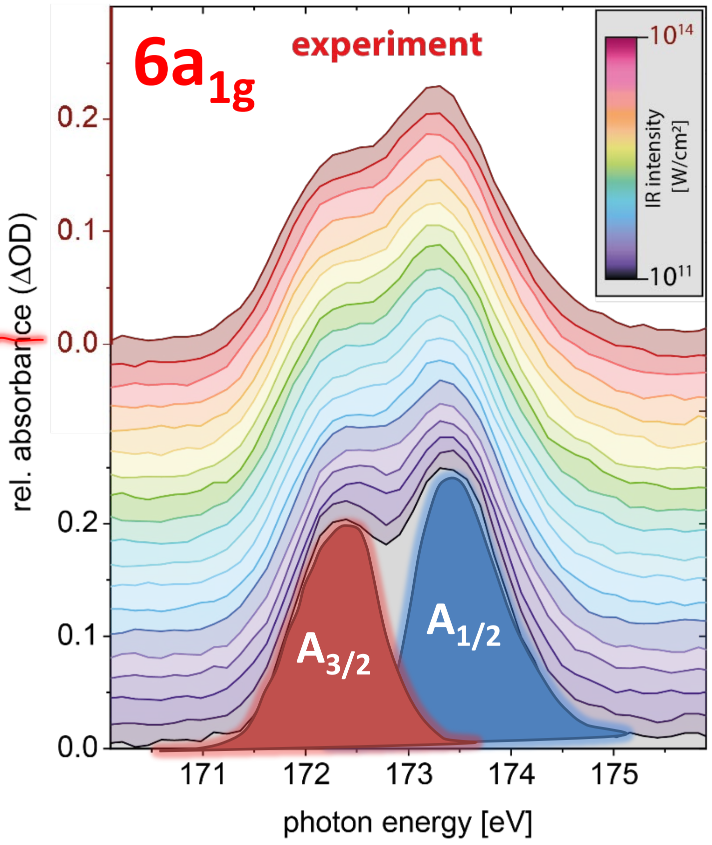
Area ratio deviates significantly from 2:1 (→ sensitive probe of **h^+ – e^-** exchange interaction)

Onodera & Toyozawa, J. Phys. Soc. Jpn., **22** 833 (1967).

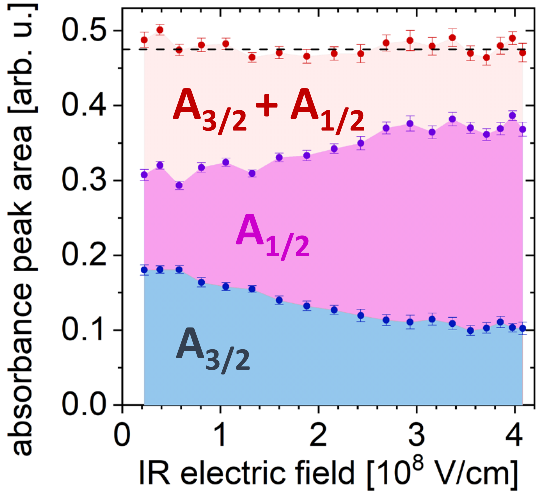
What happens when a strong laser electric field interacts with this system?

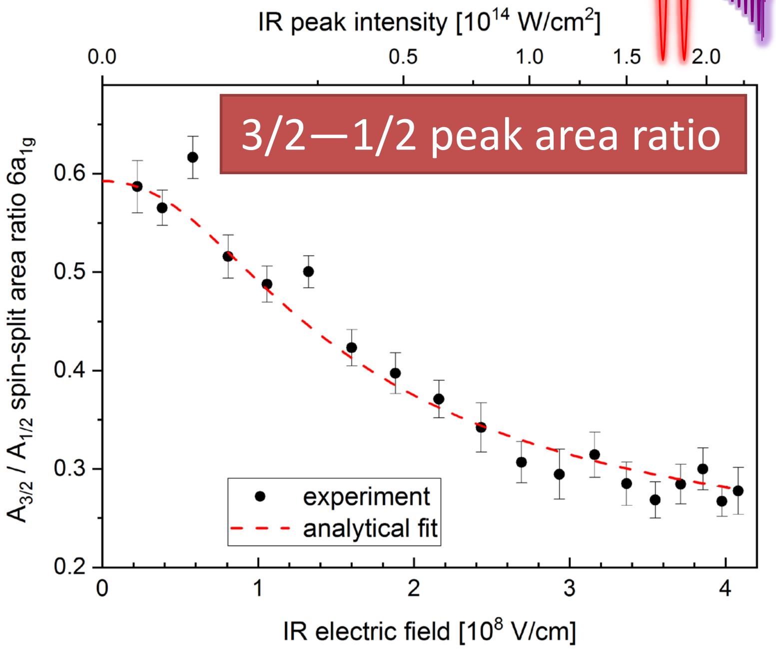
Laser control of coherent x-ray dipole response enhanced by exchange interaction





- sum of areas constant





- **$h^+ – e^-$** (3/2 : 1/2)
  relative spectral weight decreases further with intense IR

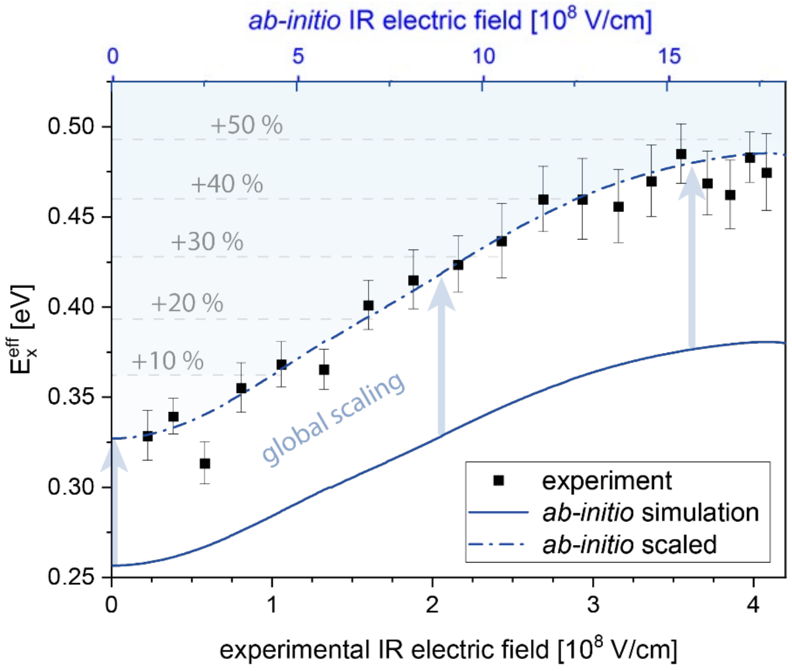
- **Can be understood with an analytical model of a polarized electron in the valence-excited orbital**
- **Analytical fit: Few-level system of dipole-coupled many-body states**
- **Including quantum exchange interaction (indistinguishable fermions) of many-particle wave function**

P. Rupprecht et al., Phys. Rev. Lett. **128**, 153001 (2022).

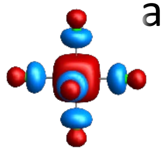
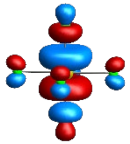
Laser-control of the effective exchange energy in core-excited SF_6

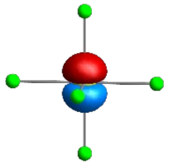
Collab.: M. Haverkort

- Analytical fit with effective **polarized valence orbital**
- Compare with *ab-initio* calculation: restricted active space (QUANTY)



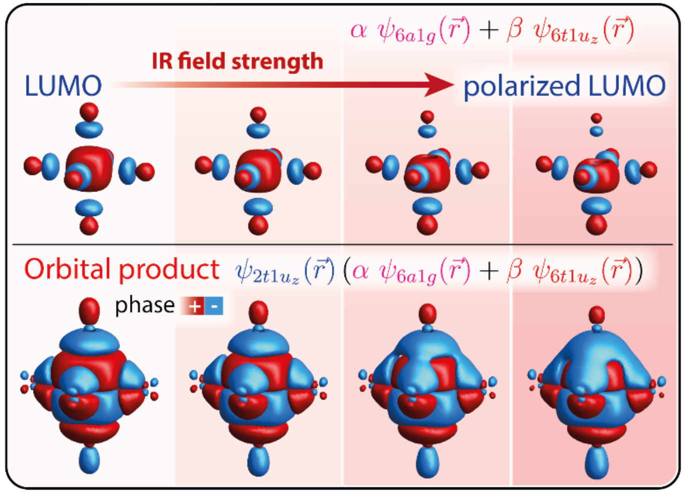
Increase of effective core-hole—excited-electron **exchange energy** by up to **50%** via IR pulses

Polarized valence orbital:
→ mixing of **$6a_{1g}$**  and **$6t_{1u}$** 



core orbital
$2t_{1u}$

Parity (g/u) of dipole-coupled orbitals



- Looking at Coulomb exchange integral:

$$\iint \psi_h^*(\vec{r_1})\ \psi_e^*(\vec{r_2})\ \frac{1}{r_{12}}\ \psi_h(\vec{r_2})\ \psi_e(\vec{r_1})\ d\vec{r_1}\ d\vec{r_2}$$

core hole

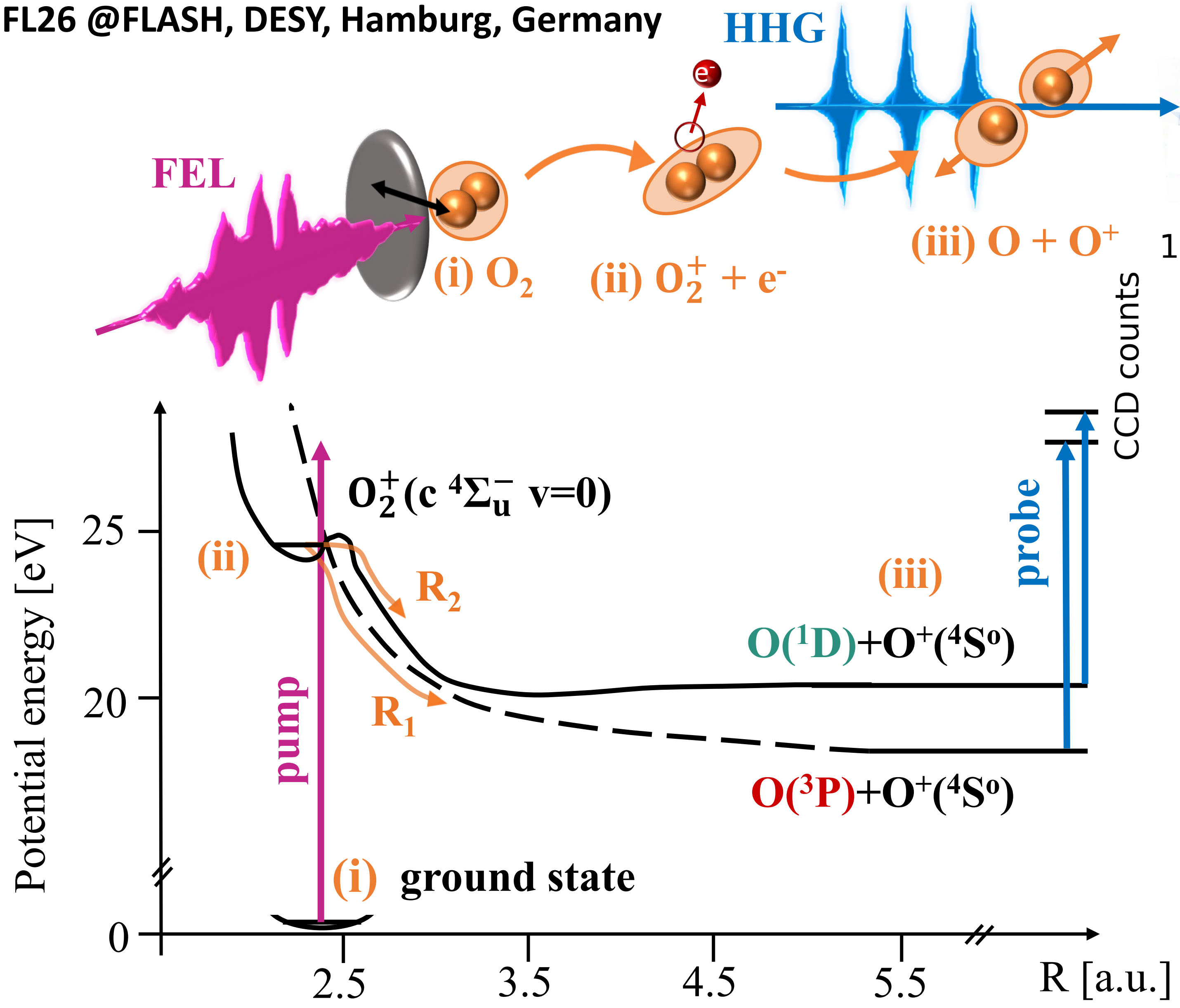
polarized valence orbital

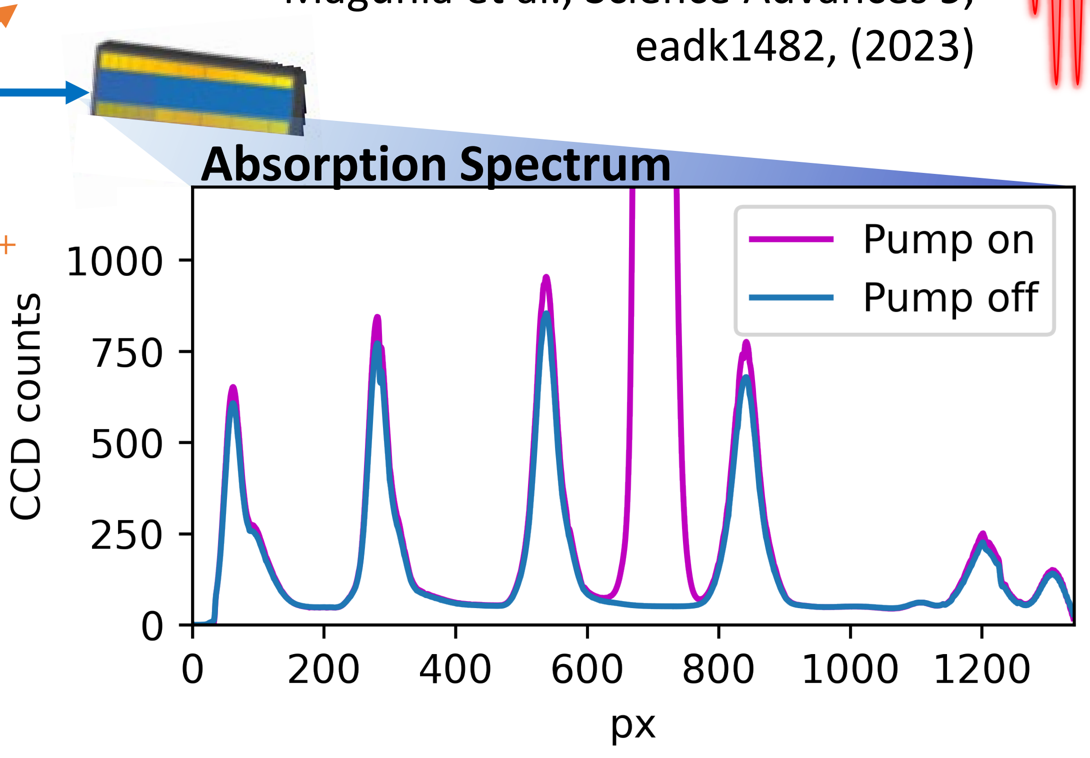
P. Rupprecht et al., Phys. Rev. Lett. **128**, 153001 (2022).

Time-resolving state-specific ultrafast quantum dynamics in O_2 molecule

FL26 @FLASH, DESY, Hamburg, Germany

Magunia et al., Science Advances **9**, eadk1482, (2023)





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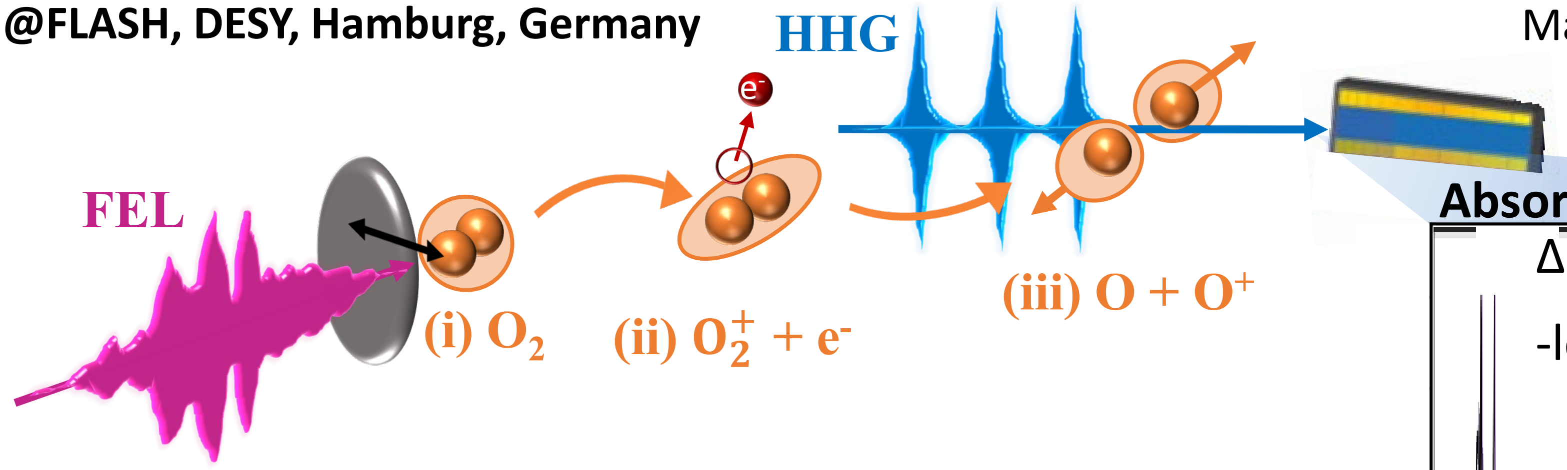
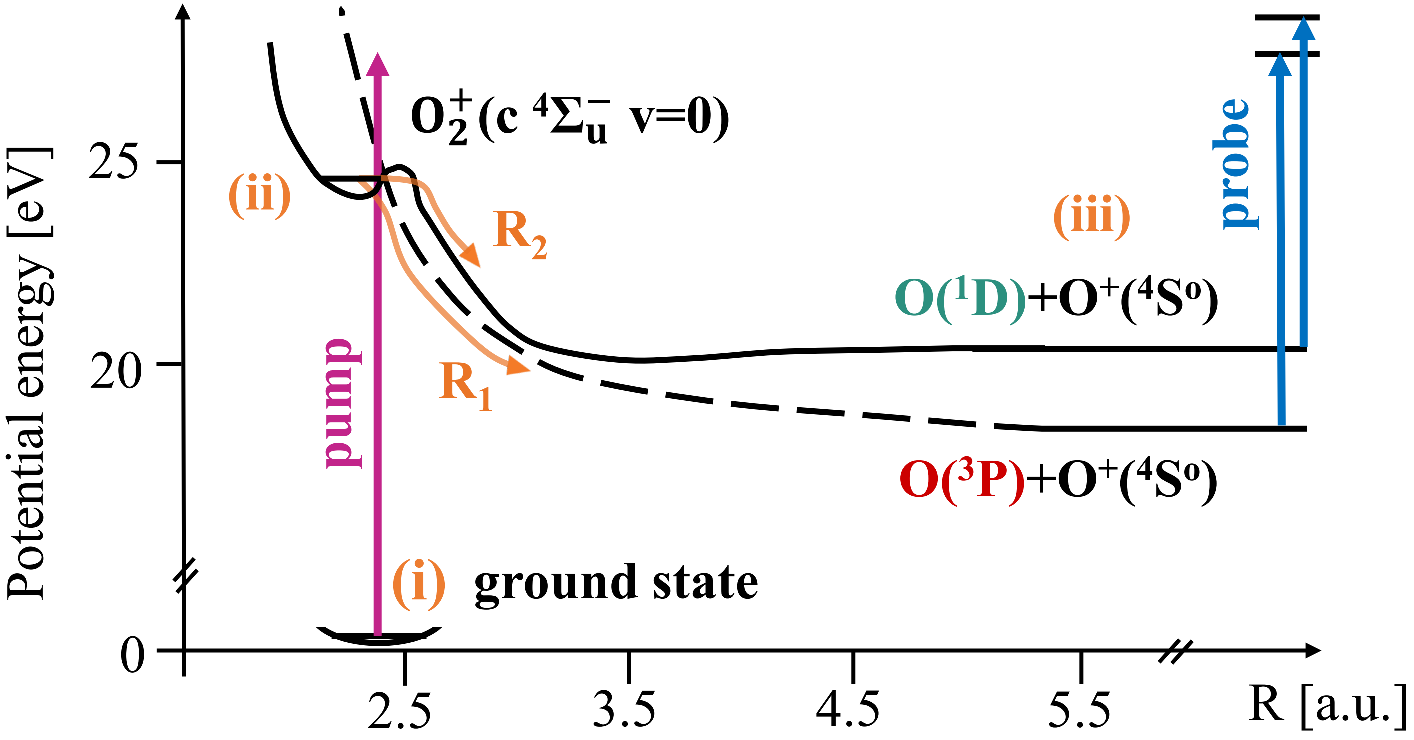
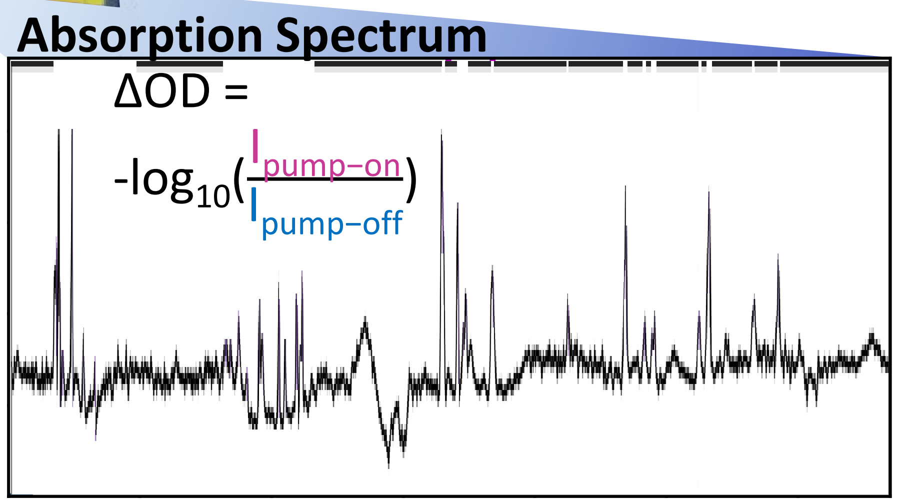






Quantifying coherent molecular dynamics:

tunneling dissociation (R_2)

vs.

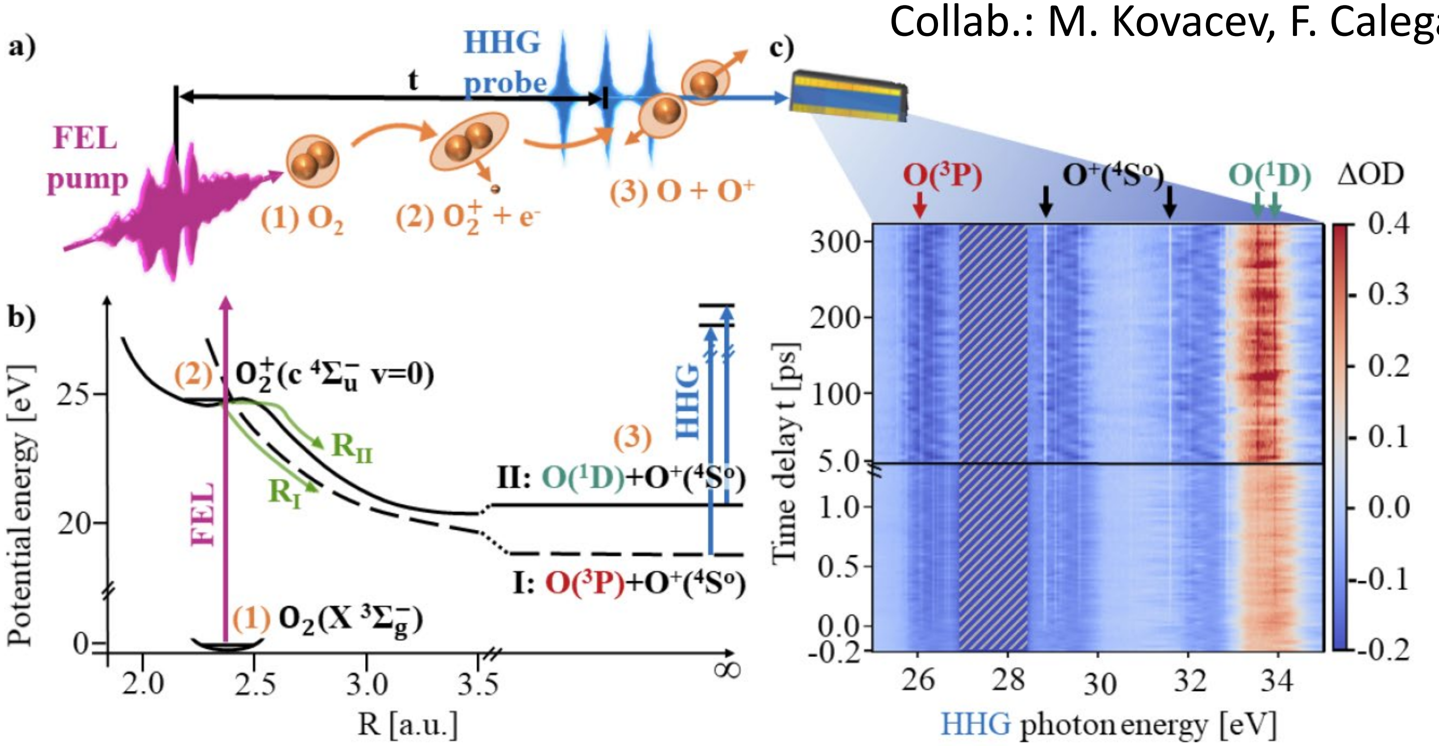
non-adiabatic couplings (R_1)
(pre-dissociation)

with specificity to internal electronic excitation in ionic & neutral fragments

Time-resolving state-specific ultrafast quantum dynamics in O_2 molecule

Collab.: M. Kovacev, F. Calegari

Magunia et al., Science Advances **9**, eadk1482, (2023)



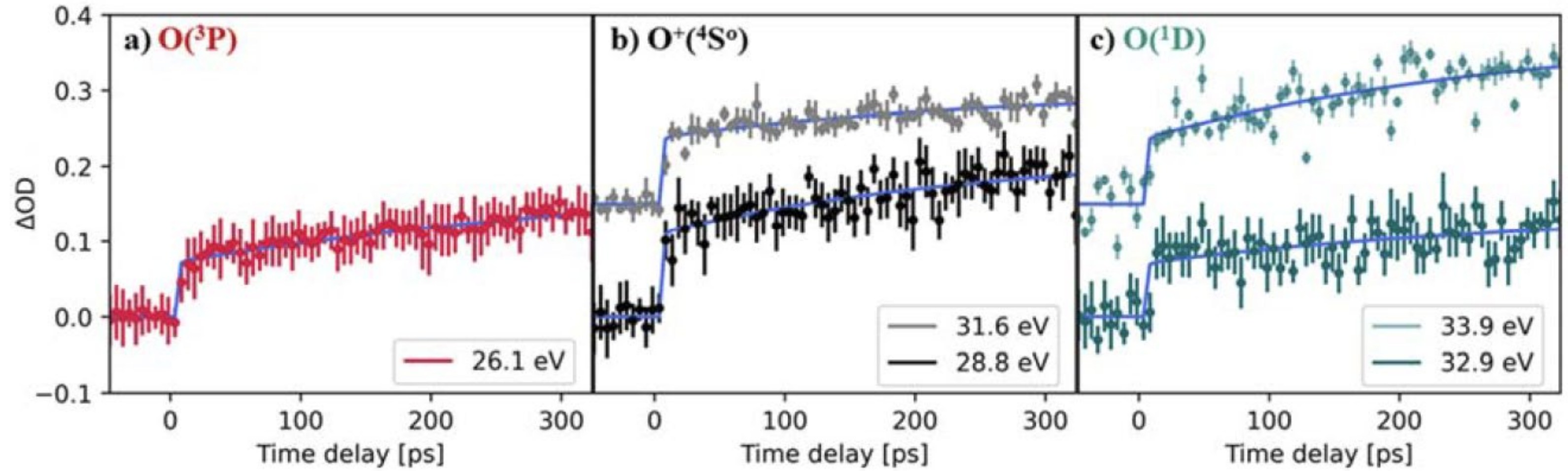
rate equation model of parallel dissociation dynamics

$$N_{O^+(^4S)}(t) = 1 - e^{-t/\tau},$$

$$N_{O(^1D)}(t) = \frac{R_{II}}{R_I + R_{II}}\left(1 - e^{-t/\tau}\right),$$

$$N_{O(^3P)}(t) = \frac{R_I}{R_I + R_{II}}\left(1 - e^{-t/\tau}\right),$$

$$\tau = \frac{1}{R_I + R_{II}}$$



exponential-fit results:

$\bar{\tau}$ = (280 ± 160) ps

$\tau_{O(^3P)}$ = (280 ± 120) ps

$\tau_{O^+(^4S^o)}$ = (290 ± 170) ps

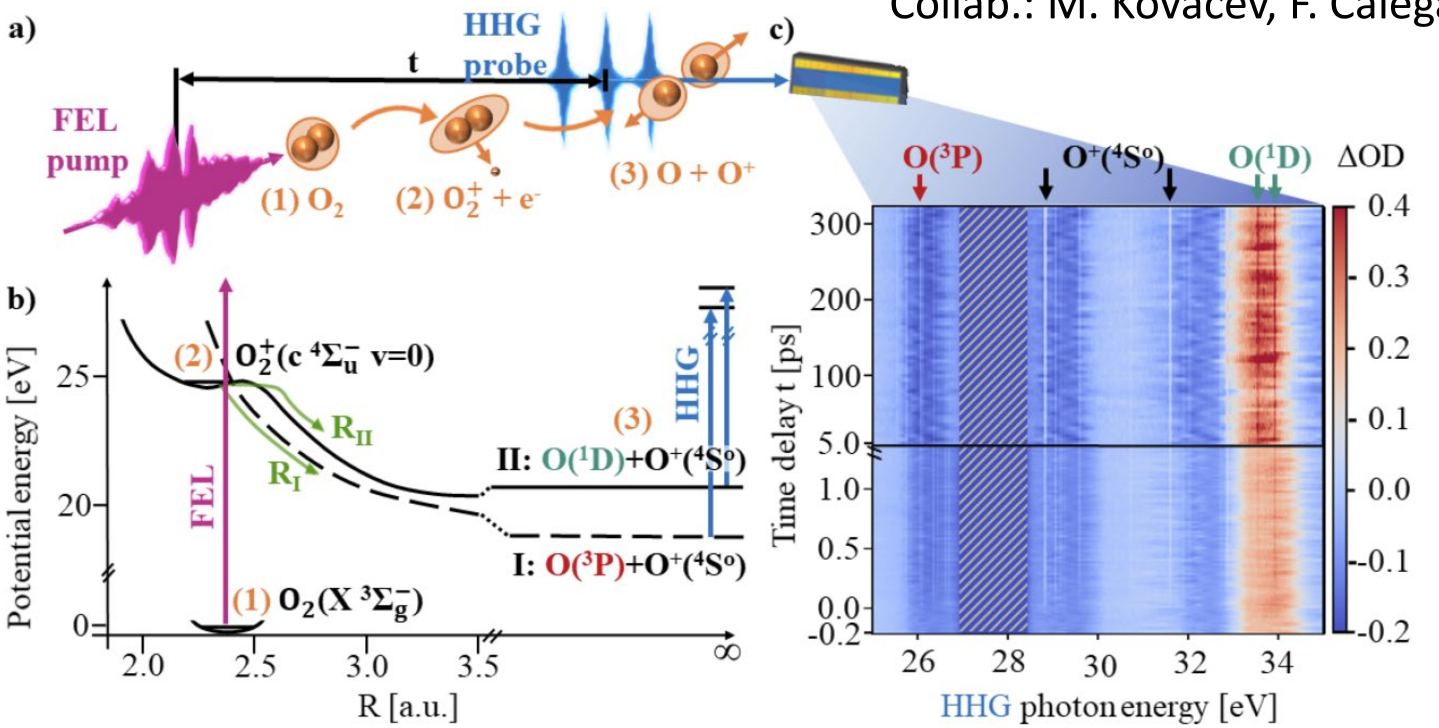
$\tau_{O(^1D)}$ = (280 ± 200) ps

in literature: ranging from picoseconds to nanoseconds

Time-resolving state-specific ultrafast quantum dynamics in O_2 molecule

Collab.: M. Kovacev, F. Calegari

Magunia et al., Science Advances **9**, eadk1482, (2023)



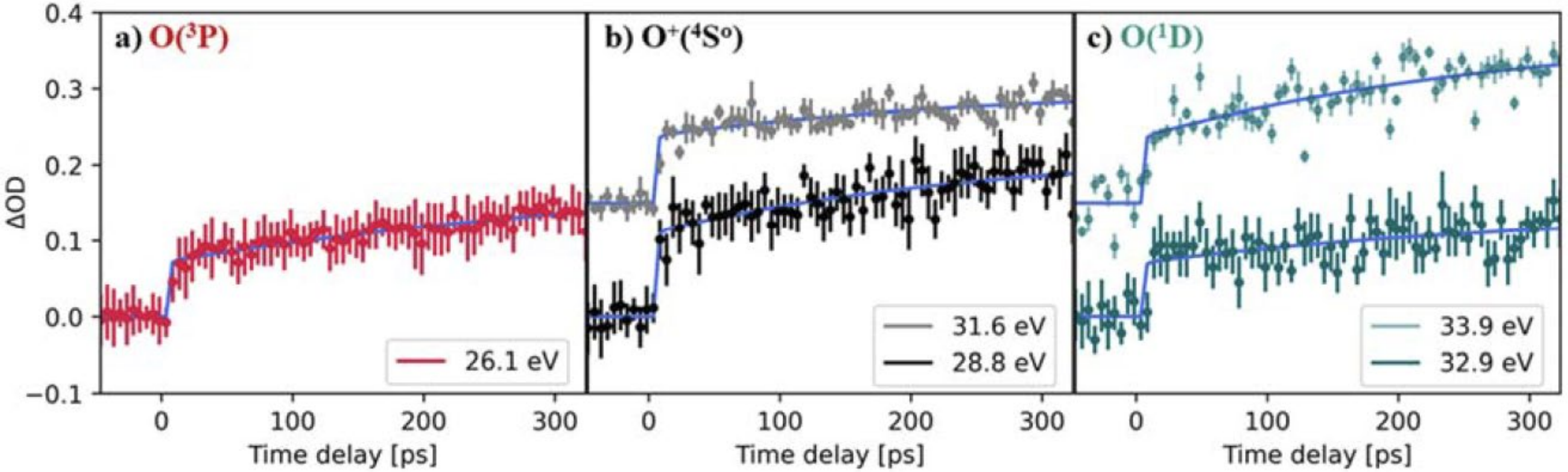
evaluating transition strengths

$\bar{\tau}$ = (280 ± 160) ps

R_I = (1.0 ± 0.7) ns^{-1}

R_{II} = (2.6 ± 2.2) ns^{-1}

$\frac{R_{II}}{R_I}$ = (2.6 ± 1.1)



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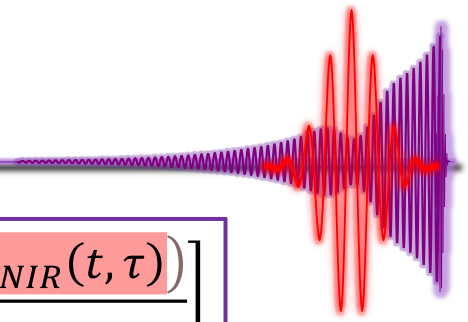
in literature: ranging from picoseconds to nanoseconds

Conclusion



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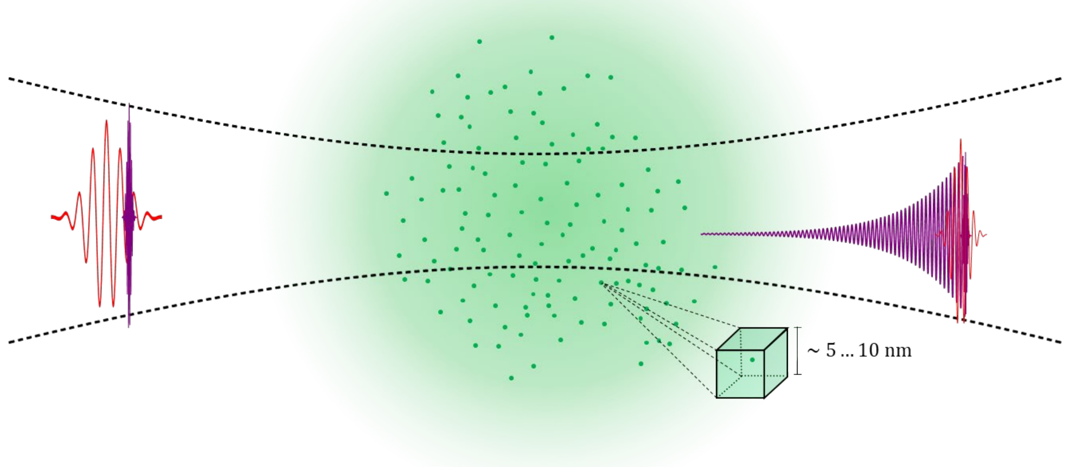
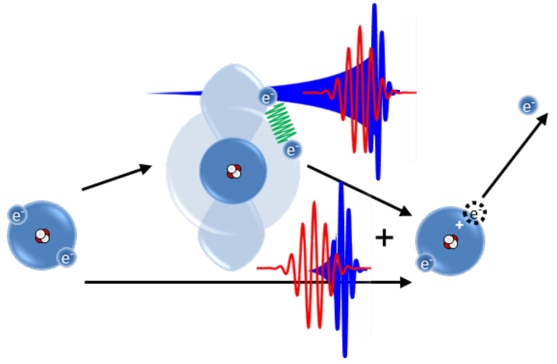
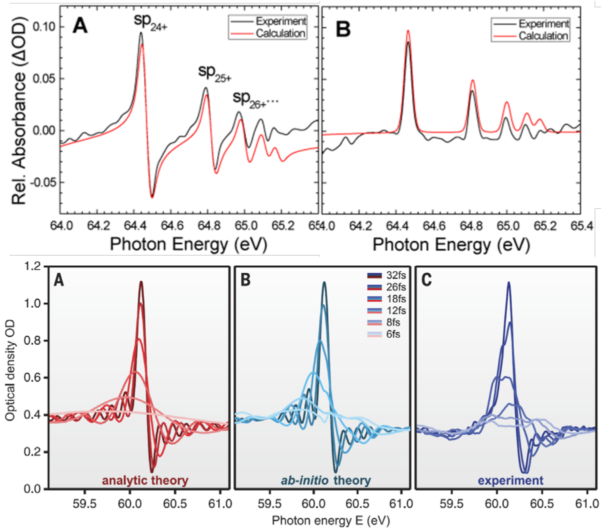
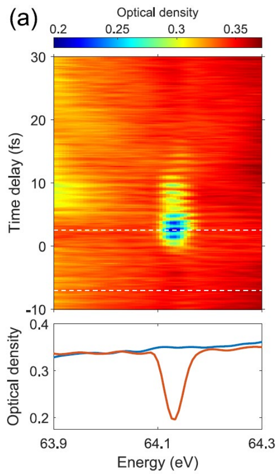


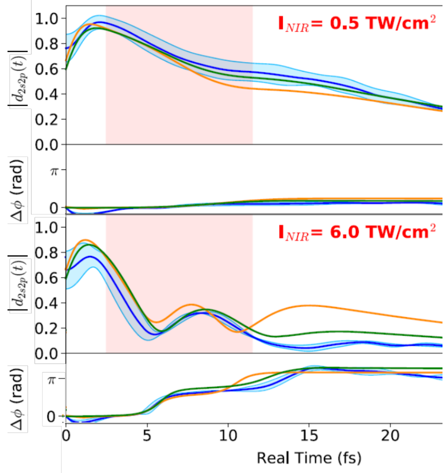
Laser control of the coherent XUV dipole response & phase sensitivity of spectral line shapes enabled by attosecond „delta pulses“

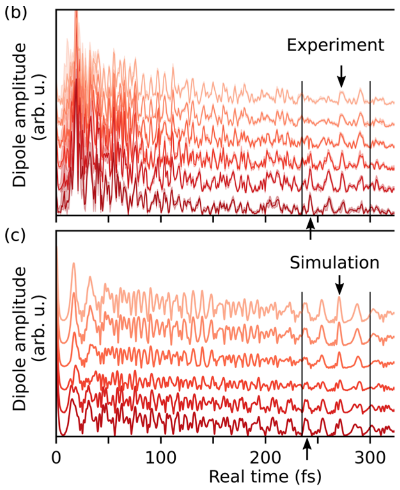


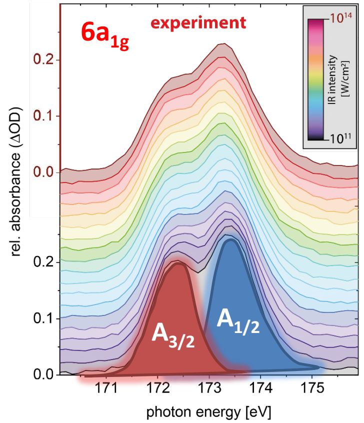
Phase-sensitive and state-resolved measurement and control of quantum dynamics in bound-state few-body systems: from atoms to molecules

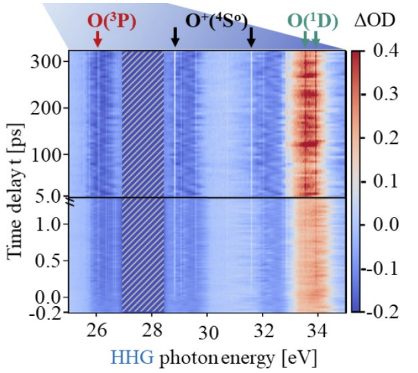












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MAX-PLANCK-INSTITUT FÜR KERNPHYSIK

Division 2
Quantum Dynamics & Control
Prof. Dr. Thomas Pfeifer

Laser-control of excited-state quantum dynamics group:

Gergana D. Borisova
Alexander Magunia
Yu He
Patrick Rupprecht
Carlo Kleine
Lina Hedewig
Harijyoti Mandal
Ana Sofia Morillo Candas
Pankaj Seliya
Noah Wach
Eleni Tzima
Leonard Münchenbach
Felix Wieder
Mohamed Attia

Collaborations:
Greene, Keitel, Evers, Argenti, Martin, Burgdörfer, Lin, Saenz, Haverkort, Kovacev, Calegari, …