universität freiburg

High-repetition-rate experiments at ELI-ALPS

Attosecond and Strong-field Physics Group
Institute of Physics University of Freiburg
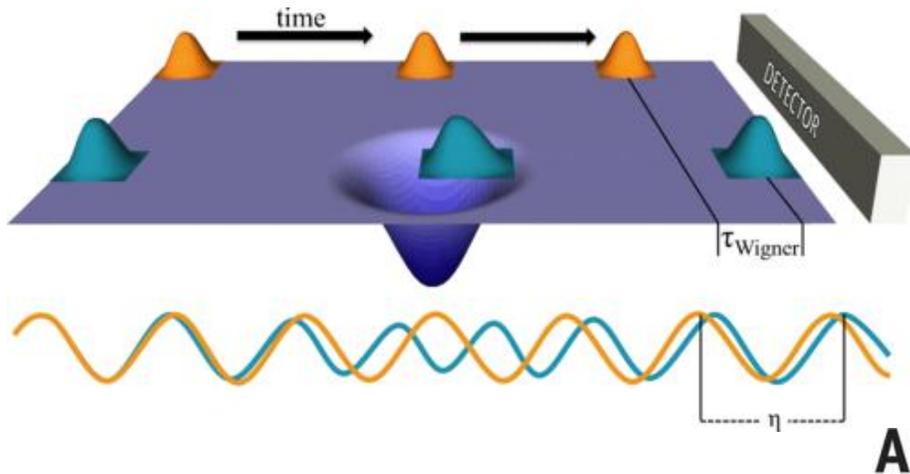Giuseppe Sansone
ELI Ultrafast Science Workshop
12. February 2026
European XFEL

Attosecond photoionization delay

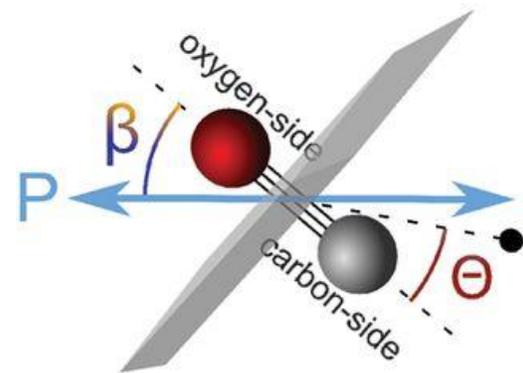Electron passing through a short-range potential accumulates quantum phase

E. Wigner, *Phys Rev* **98,** 145 (1955).
F. Smith, *Phys Rev* **118,** 349 (1960).
L. Argenti *et al., Phys Rev. A* **95** (2017).

Asymptotic phase difference vs free wave propagation

Wigner-Smith delay: $\quad \tau_W = \dfrac{\partial \eta}{\partial E}$


A

Photoemission delays as a sensitive probe

J. Vos *et al.*, Science **360**, 1326 (2018).
M. Huppert *et al.* PRL **117**, 093001 (2016).
S. Biswas *et al.* Nature Physics **16**, 778–783 (2020)
H. Ahmadi *et al.*, Nature Communications **13**, 1242 (2022).

o Influence of the molecular potential
o Presence of electronic resonances (Shape resonances)
o Influence of electronic correlationn effects (shake-up states)
o Coupled nuclear-electronic dynamics

universität freiburg

2

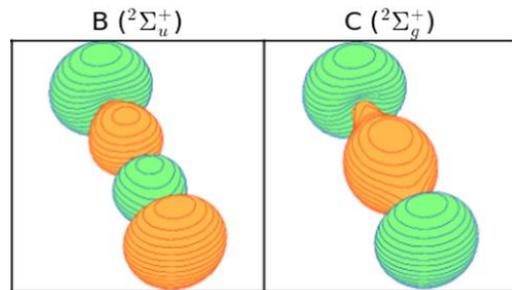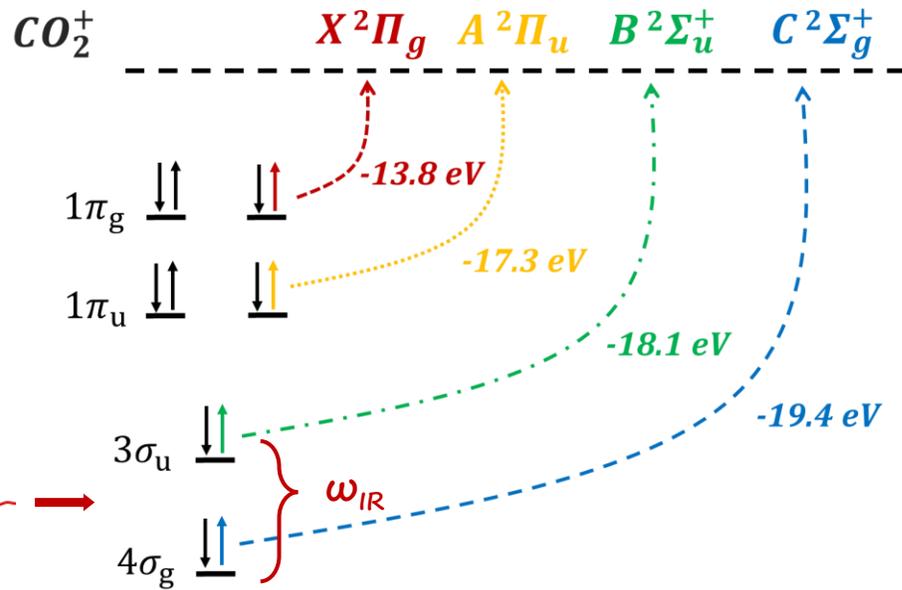# Attosecond photoionization delays using coincidence spectroscopy
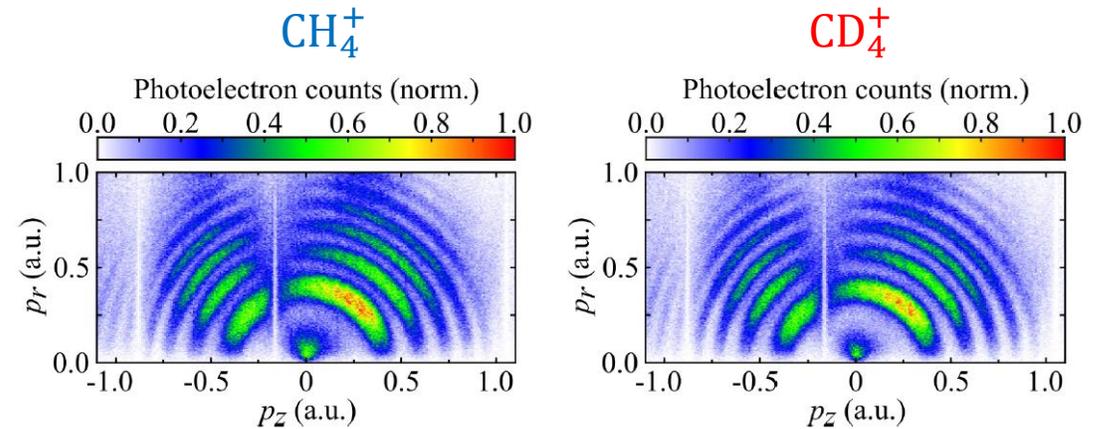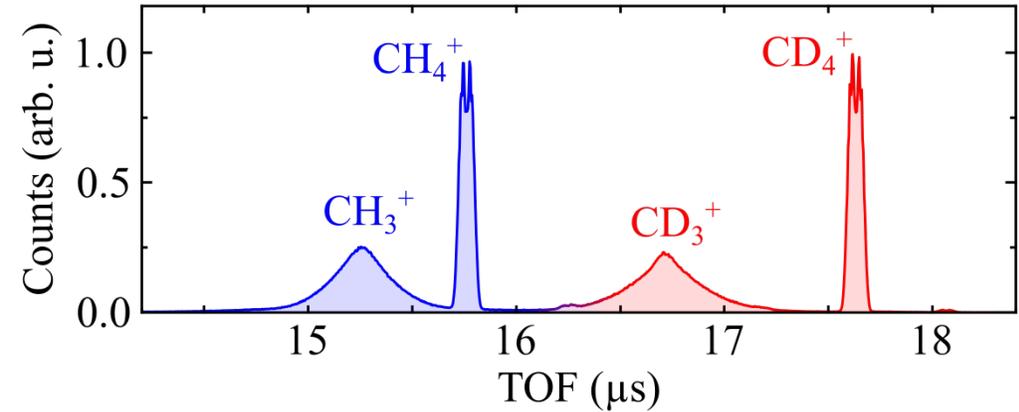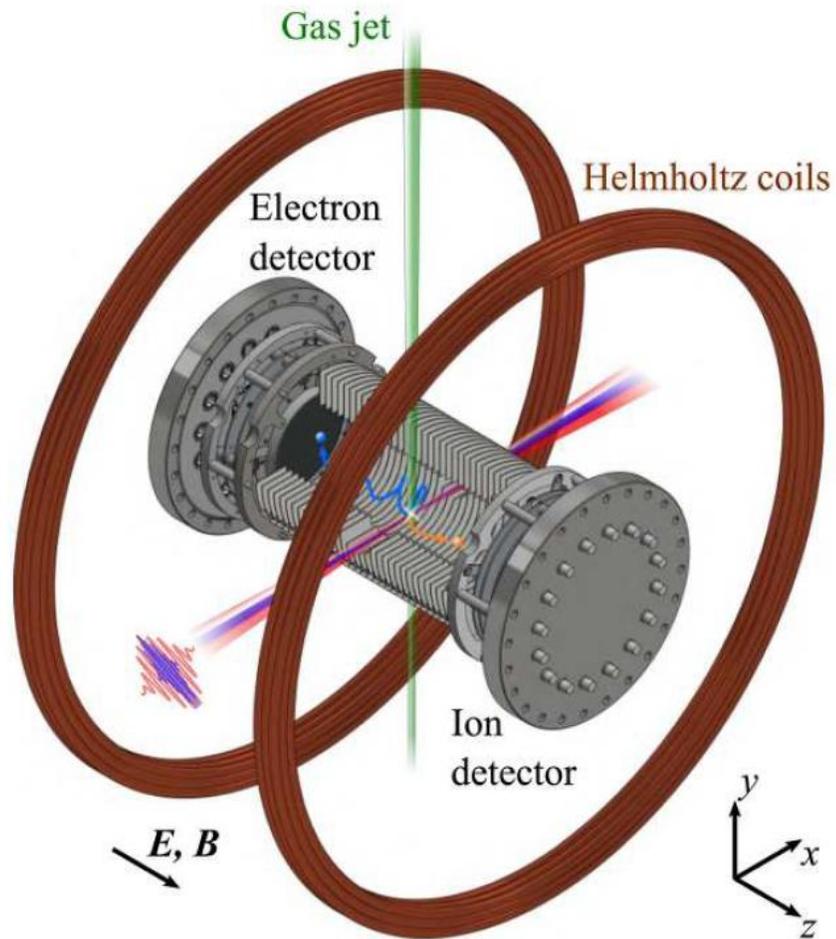
- Measurements under the „same" experimental conditions
- Angular and energy-resolved information
- Information on the different ionization channels
- Attosecond sources operating at 50 kHz
- Acquisition time (about one week)

J. Ullrich *et al.* J. Phys. B At. Mol. Opt. Phys. 30, 2917–2974 (1997).
J. Ullrich *et al.* Prog. Phys. 66, 1463–1545 (2003).
R. Dörner *et al.* Phys. Rep. 330, 95–192 (2000).

D. Ertel, *et al.,* The Journal of Physical Chemistry A **128**, 1685 (2024).

universität freiburg

Attosecond interferometry in CO₂

CO_2^+

$X\,^2\Pi_g$ $A\,^2\Pi_u$ $B\,^2\Sigma_u^+$ $C\,^2\Sigma_g^+$
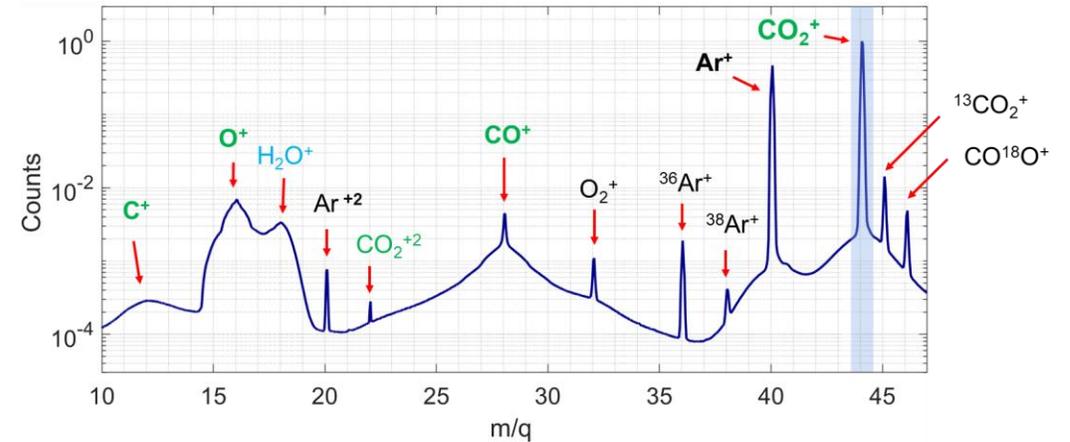
-13.8 eV

-17.3 eV

-18.1 eV

-19.4 eV

$1\pi_g$

$1\pi_u$

$3\sigma_u$

$4\sigma_g$

ω_{IR}

$B\,(^2\Sigma_u^+)$ $C\,(^2\Sigma_g^+)$

A. Kamalov et al. PRA **102** (2020).

Ion time of flight CO₂ /Ar mixture

Counts

CO_2^+
Ar⁺
O^+ H_2O^+
CO^+
Ar⁺²
CO_2^{+2}
O_2^+
³⁶Ar⁺
³⁸Ar⁺
C⁺
¹³CO₂⁺
CO¹⁸O⁺

m/q

RABBIT spectrogram in CO₂⁺

Delay Δt (fs)

Photoelectron energy (eV)

universität freiburg

Resonant coupling in the cation

CO_2^+

$X\,^2\Pi_g$ $A\,^2\Pi_u$ $B\,^2\Sigma_u^+$ $C\,^2\Sigma_g^+$

$1\pi_g$

−13.8 eV

$1\pi_u$

−17.3 eV

−18.1 eV

−19.4 eV

$3\sigma_u$

ω_{IR}

$4\sigma_g$
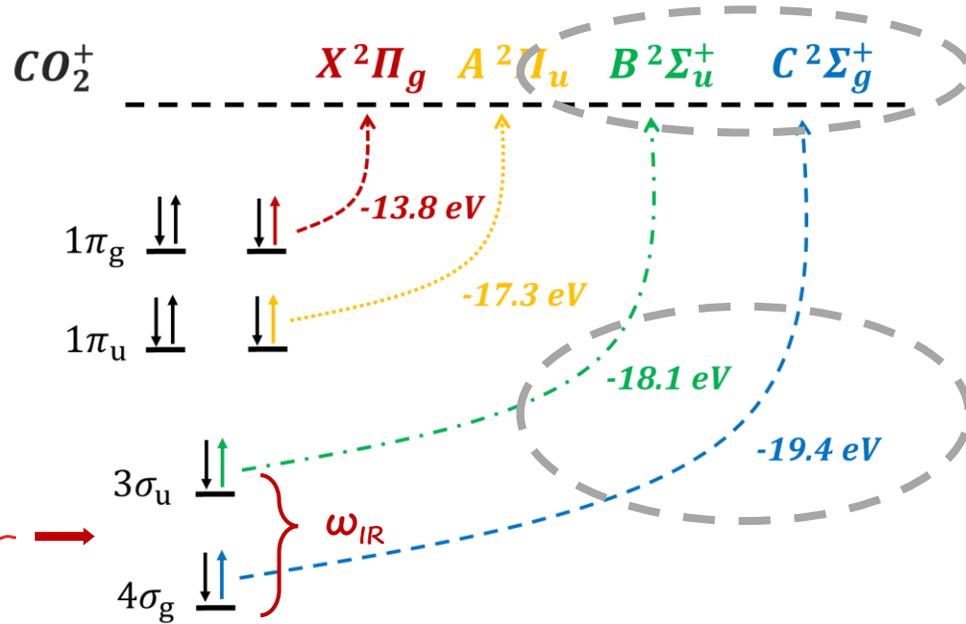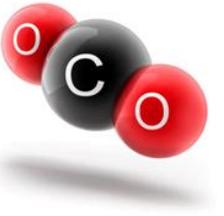
$$\Delta E = E(B) - E(C) \simeq 1.3\ eV$$
$$\hbar\omega \simeq 1.22\ eV\ (@1012\ nm)$$

Ion time of flight CO₂ /Ar mixture
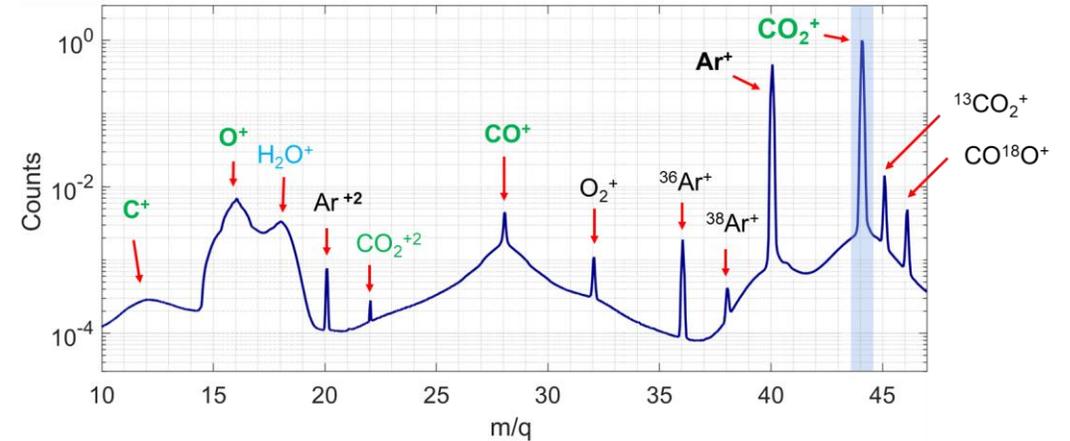
RABBIT spectrogram in CO₂⁺ B state

Z. Mašín *et al.*, *J. Phys. B: At. Mol. Opt. Phys.* **51,** 134006 (2018)
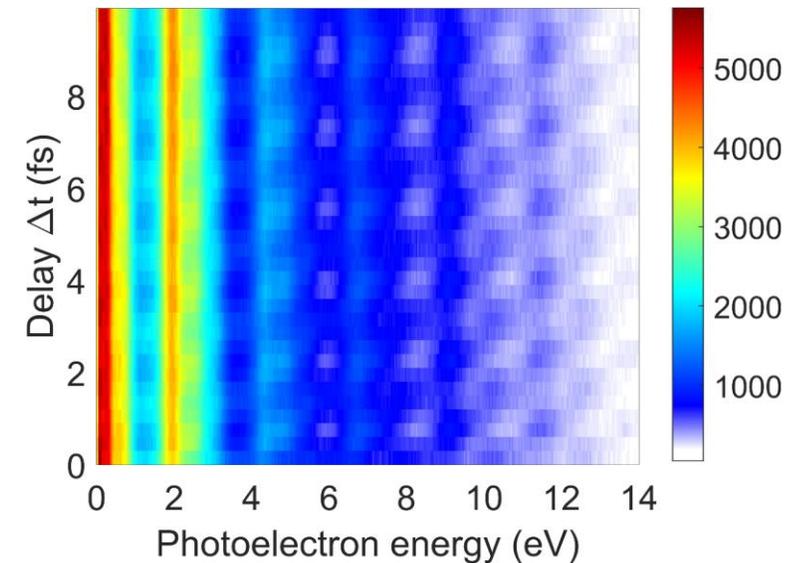
RABBIT: two-pathway photoelectron interferometer

$$\left| \begin{array}{c} e^- \end{array} \right\rangle \otimes \left| \begin{array}{c} ion^+ \end{array} \right\rangle$$
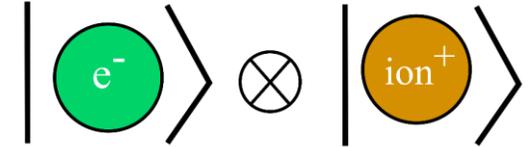
Photoionization leaves the target in an entangled state

$$\Psi(t) = \sum_j \psi_j(t)\, \Phi_j(t)$$

$$e_{2q+1} = (2q+1)\, \hbar\omega_{IR} - I_P(B)$$

XUV $(2q+1)\hbar\omega_{IR}$ Path 1

IR $r \cdot F(t)$

$e_{e_{2q+1}}$ Φ_B e $-\hbar\omega_{IR}$

$e_{2q} = 2q\, \hbar\omega_{IR} - I_P(B)$

Ψ_{B2q}

e Φ_B e_{2q}

$B\ ^2\Sigma_u^+$

$C\ ^2\Sigma_g^+$

$e_{2q} = 2q\, \hbar\omega_{IR} - I_P(B)$

H_{2q+1}

H_{2q-1}

Ip

Path 1 – 2

Path 2 $(2q-1)\hbar\omega_{IR}$

e Φ_B e_{2q-1}

$r \cdot F(t)$ e $+\hbar\omega_{IR}$
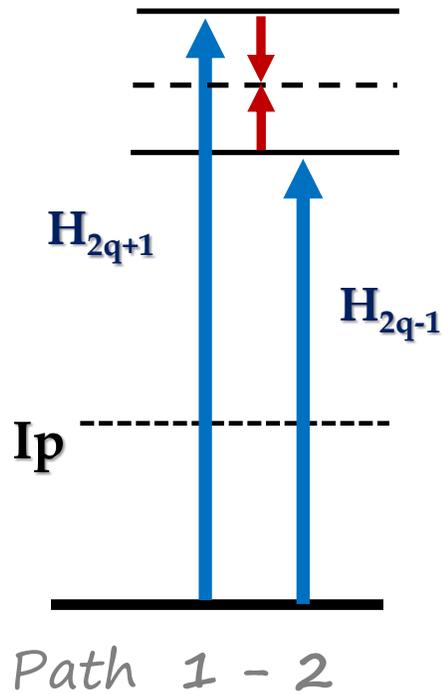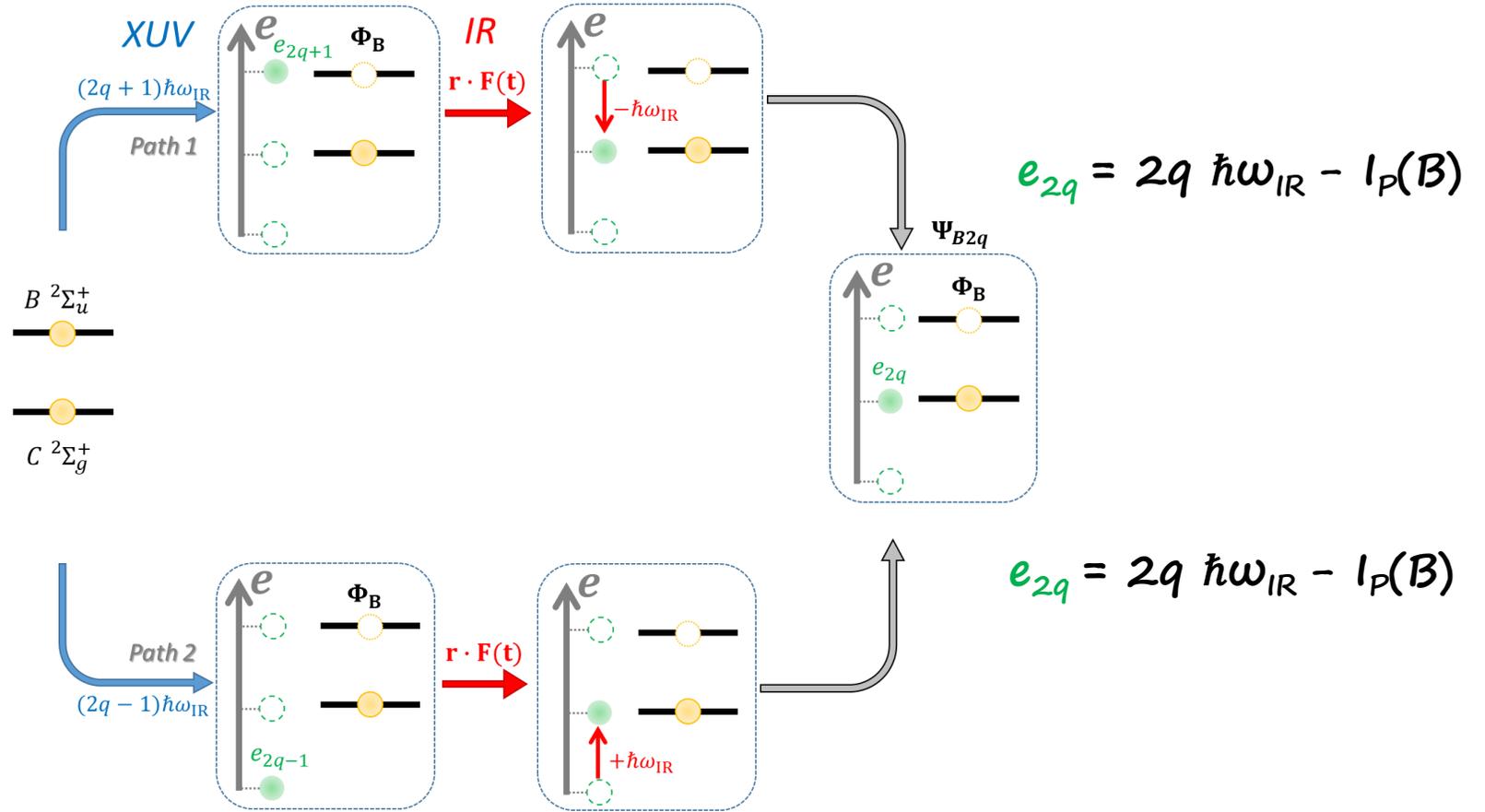
$e_{2q-1} = (2q-1)\hbar\omega_{IR} - I_P(B)$

3rd - pathway in RABBIT interferometer

- due to the interaction of the IR with the residual ion

⮕ 3rd pathway arises

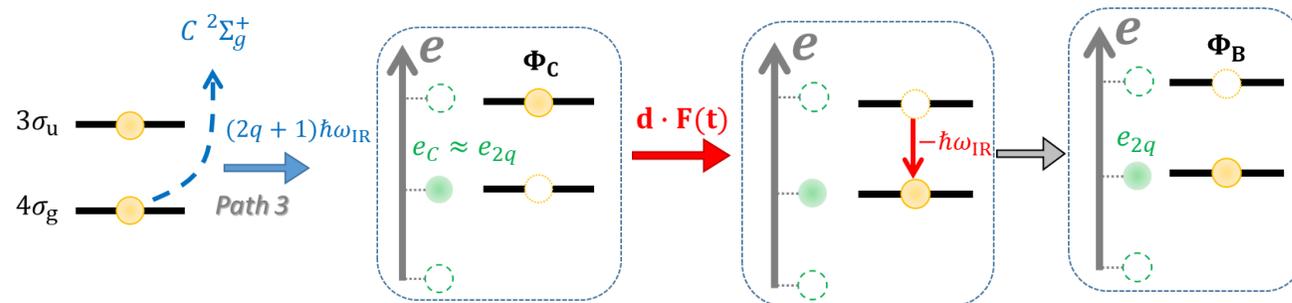$$e_C = (2q+1)\, \hbar\omega_{IR} - I_P(C) = 2q\, \hbar\omega_{IR} - I_P(B) \approx e_{2q}$$

$$I_P(B) - I_P(C) = \hbar\omega_{IR}$$

$$e_{2q} = 2q\, \hbar\omega_{IR} - I_P(B)$$

- IR couples the cationic states without modifying the kinetic energy of the photoelectron

$$\Phi_C \quad ⮕ \quad \Phi_B$$

3rd path introduces an additional time delay
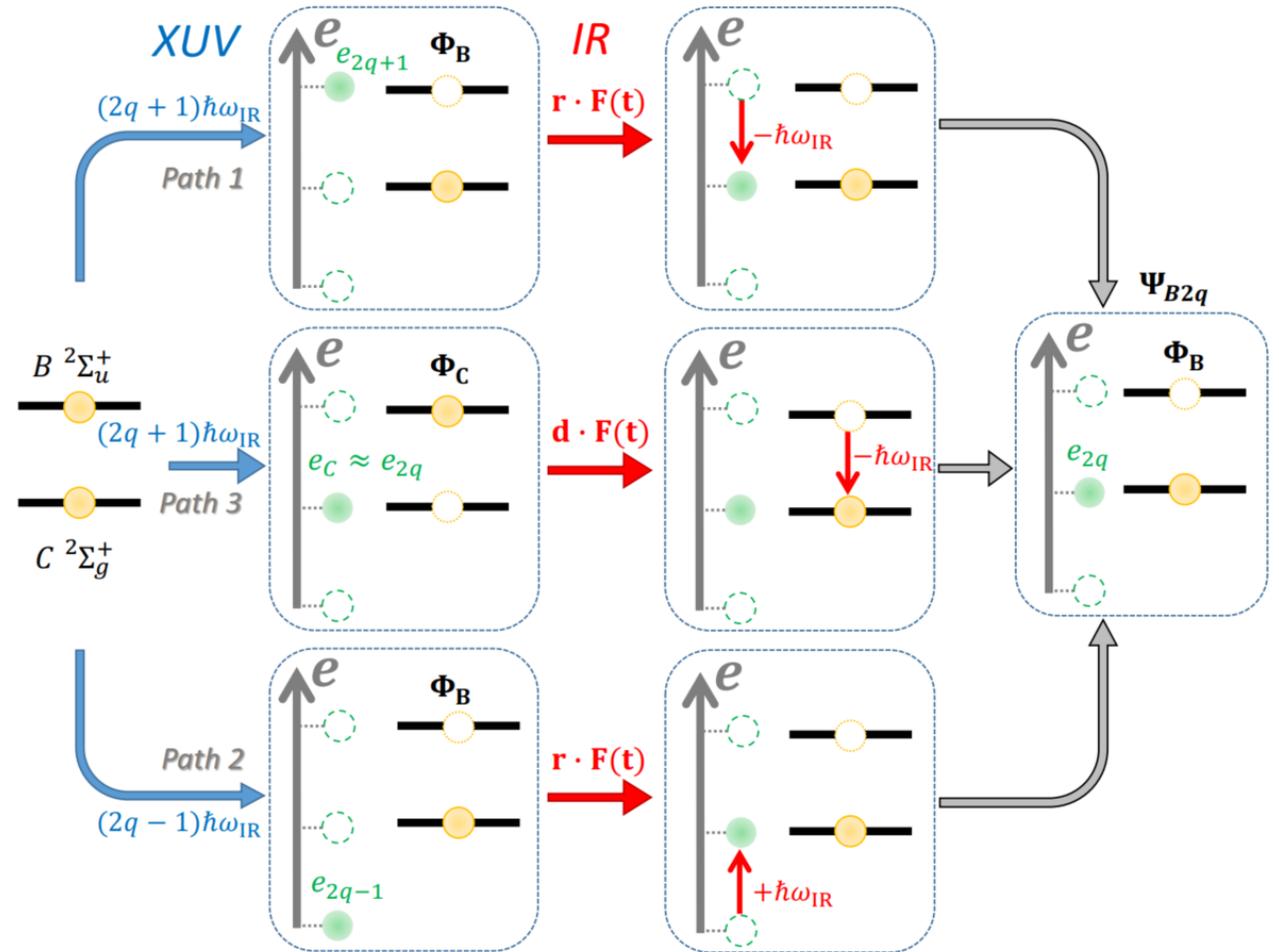
Total wave function: superposition of photoelectron coupled to **B** cationic state & photoelectron coupled to **C** cationic state.
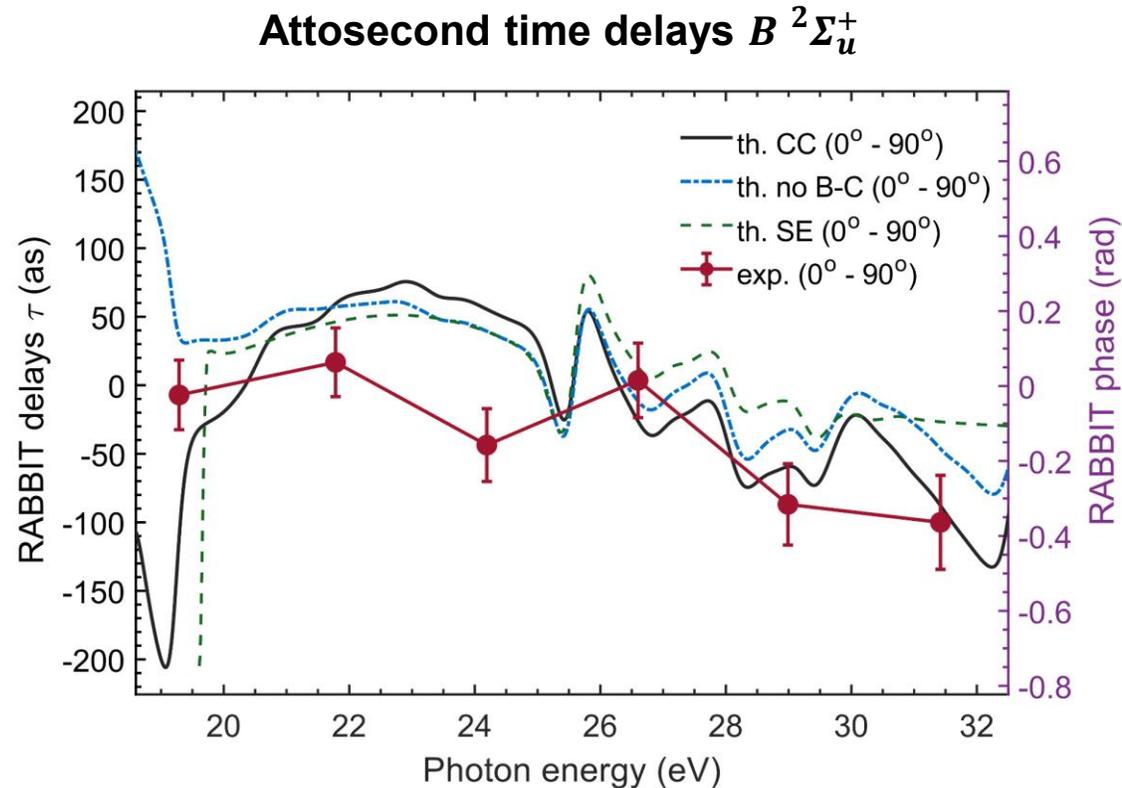
$$\Psi_n(t) = \psi_{Bn}(t)\,\Phi_B(t) + \psi_{Cn}(t)\,\Phi_C(t)$$

- Laser induced dipole coupling in the residual ion introduces an additional time delay:

$$\tau_B = \tau_e + \tau_{ion}$$

J. Delgado *et al.* PRA **111**, 063107 (2025)

Effect of the ionic coupling onto the time delays

Attosecond time delays $B\ ^2\Sigma_u^+$

- Close coupling model (*CC*) with or without B-C coupling, considers dipole transitions in the residual ion

- Static exchange (*SE*) : single channel picture

➡ Stationary multiphoton R-matrix approach for the equilibrium geometry of the neutral molecule

J. Benda *et al.*, *Phys. Rev. A* **105**, 053101 (2022).   I. Makos *et al. Nature Communications* **16**, 8554 (2025)

Limitations of current setup

o Photon energy range 30-40 eV (mainly limited by driving laser intensity)

o Multi-cycle optical pulses

o Long acquisition time (at 50 kHz)

universität freiburg

HR1 system at ELI-ALPS

HR1: The High Repetition rate laser system provide millijoule level, few-cycle laser pulses at 100 kHz repetition rate, ideally suited for experiments relying on high statistics. The laser systems work in the near infrared, at 1030 nm wavelength.

✓ Operation at 100 kHz

✓ Few-cycle pulses with CEP tagging

✓ Generation of photons up to 100-150 eV

✓ Electronic relaxation dynamics and molecular fragmentation

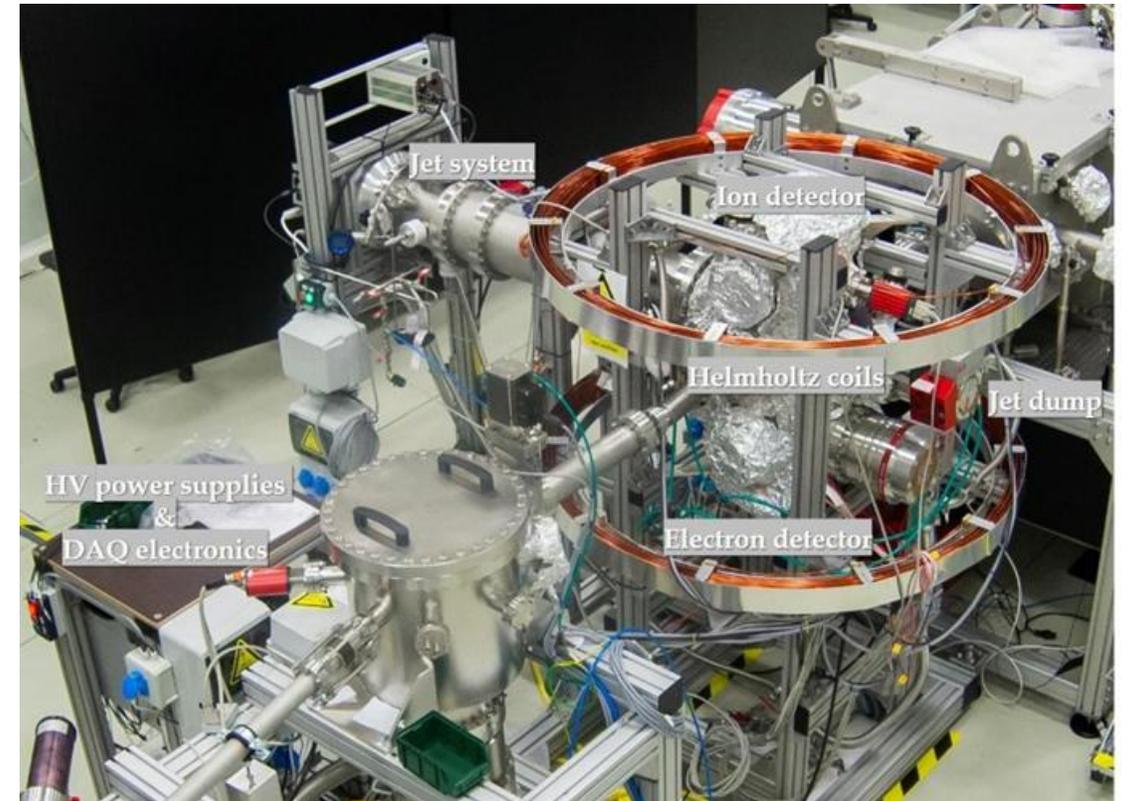✓ Measurement of Auger decay in Krypton (differential photoelectron angular distributions)

Y. Peng *et al J. Phys. B: At. Mol. Opt. Phys.* **53** 154004 (2020)
Y. Peng *et al Ultrafast Sci.* 2022;2022
T. Csizmadia *et al. APL Photonics* **10**, 080803 (2025)

Reaction Microscope End Station -REMI-ES

Active stabilization of the attosecond beamline at ELI-ALPS

T. Csizmadia *et al. APL Photonics* **10**, 080803 (2025)

- ✓ More flexible configurations for the auxiliary pulse

- ✓ Possibility to use a second or third harmonic as auxiliary pulse
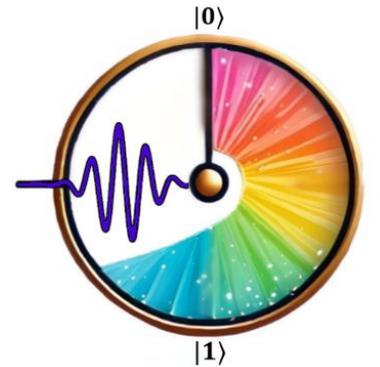
universität freiburg

The team

- **Ioannis Makos**
- David Busto
- Domninik Ertel
- Barbara Merzuk
- Hamed Ahmadi
- Matteo Moioli
- Marvin Schmoll
- Jakub Benda
- Fabio Frassetto
- Luca Poletto
- Claus Dieter Schröter

- Thomas Pfeifer
- Robert Moshammer
- Zdeněk Mašín
- Serguei Patchkovskii

DFG Deutsche Forschungsgemeinschaft

Funded by the European Union

QU-ATTO

universität freiburg

Thank you for your attention!

Giuseppe Sansone

Attosecond and Strong-Field Physics Group

giuseppe.sansone@physik.uni-freiburg.de