Possible novel attosecond technology for ELI

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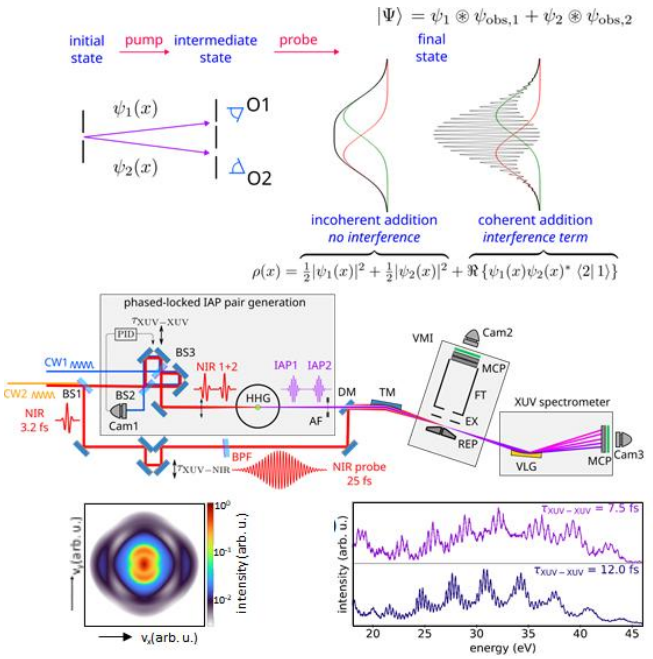
Attosecond Technology @ MBI

- Attosecond XUV+NIR beamline @ 1 kHz
  - APT & IAP capability, including generation of APT & IAP pulse pairs
  - Velocity map imaging (VMI) and/or XUV spectrometer detection
- Attosecond XUV+NIR beamline @ 100 kHz
  - APT & IAP capability
  - VMI or Coincident electron-ion detection using a reaction microscope
- Attosecond XUV+XUV beamline @ 1 kHz
  - Near-IAP capability
  - XUV spectrometer detection (mostly)



L.M. Koll et al., Nature (in press)

- Attosecond XUV+XUV beamline @ 40 kHz (under development)
  - Currently APT capability, IAP under development
  - Charged particle detection (under development)

A few years ago MBI developed a Velocity map imaging (VMI) apparatus for ELI

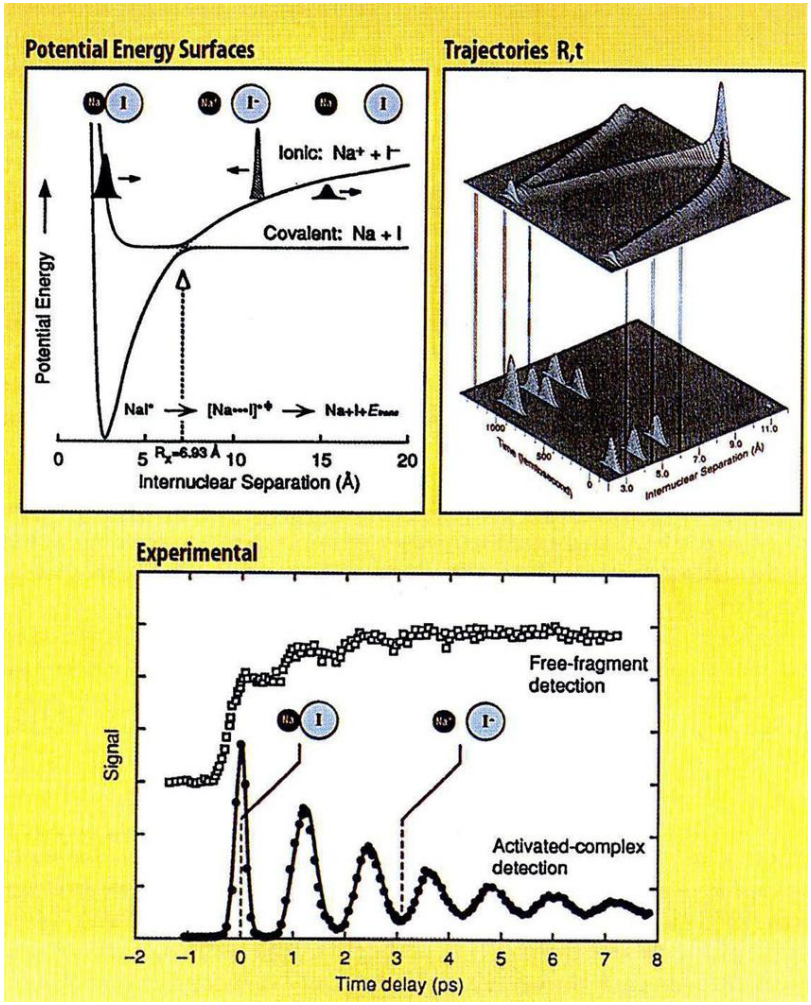
2023 Physics Nobel Prize

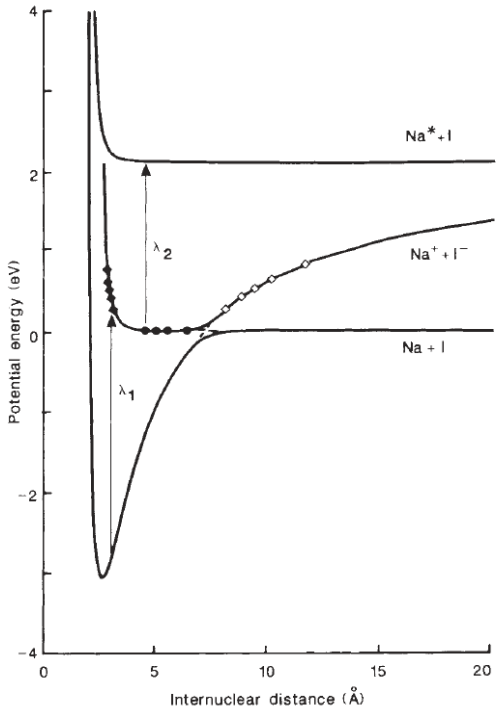
The Nobel Prize in Physics 2023 was awarded jointly to Pierre Agostini, Anne L'Huillier and Ferenc Krausz "for experimental methods that generate attosecond pulses of light for the study of electron dynamics in matter"

From Femtochemistry...

Ahmed Zewail (1946-2016)

Nobel Prize in Chemistry 1999 for the development of Femtochemistry





Femtosecond pump + femtoscond probe → femtosecond time-resolution

A. H. Zewail, Angew. Chem. Int. Ed. 39, 2587–2631 (2000)

to Attosecond Physics ....

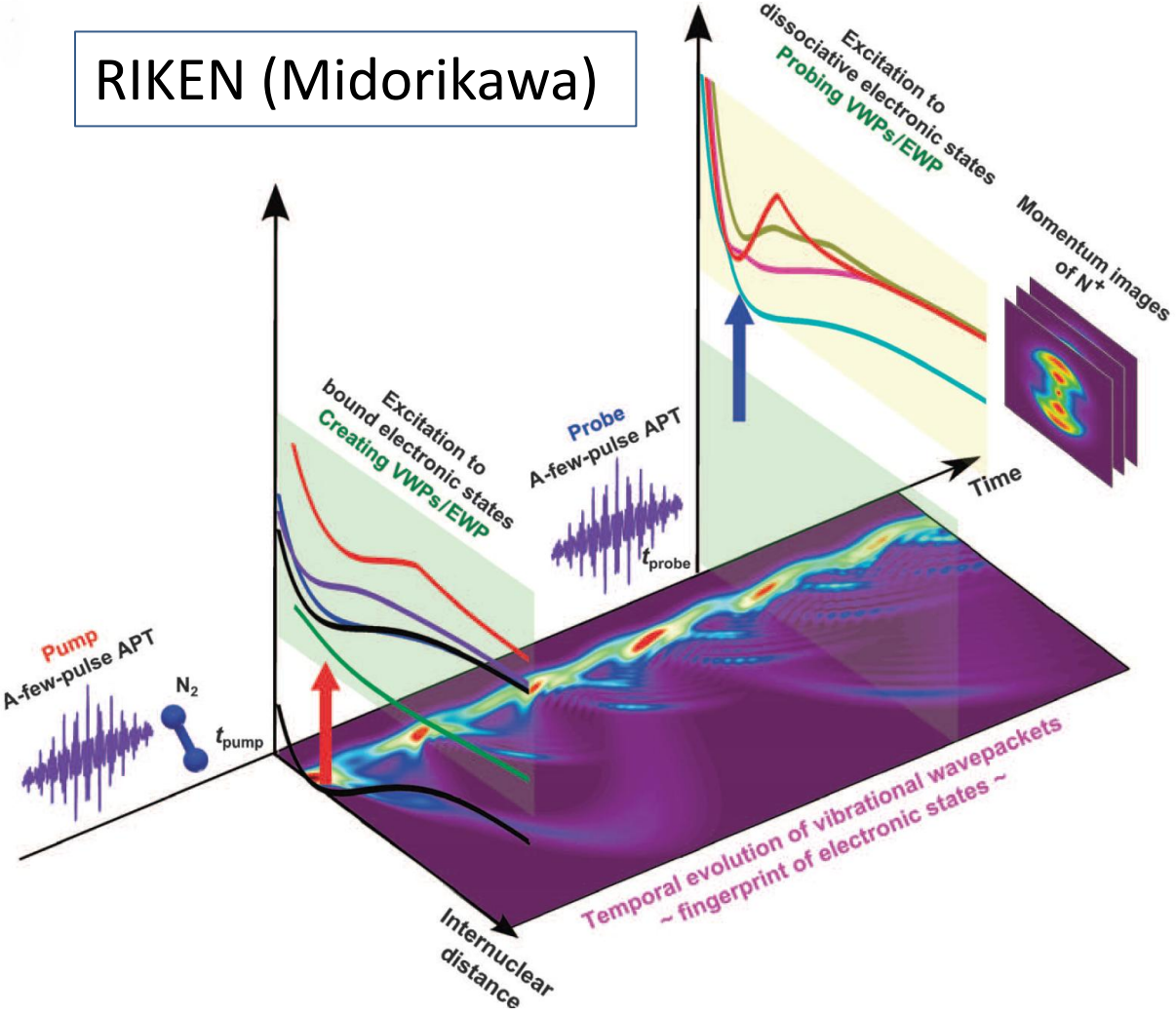
In the spirit of the development of the field of femtochemistry, a logical step following the generation and characterization of attosecond laser pulses would have been their use in attosecond pump-attosecond probe experiments

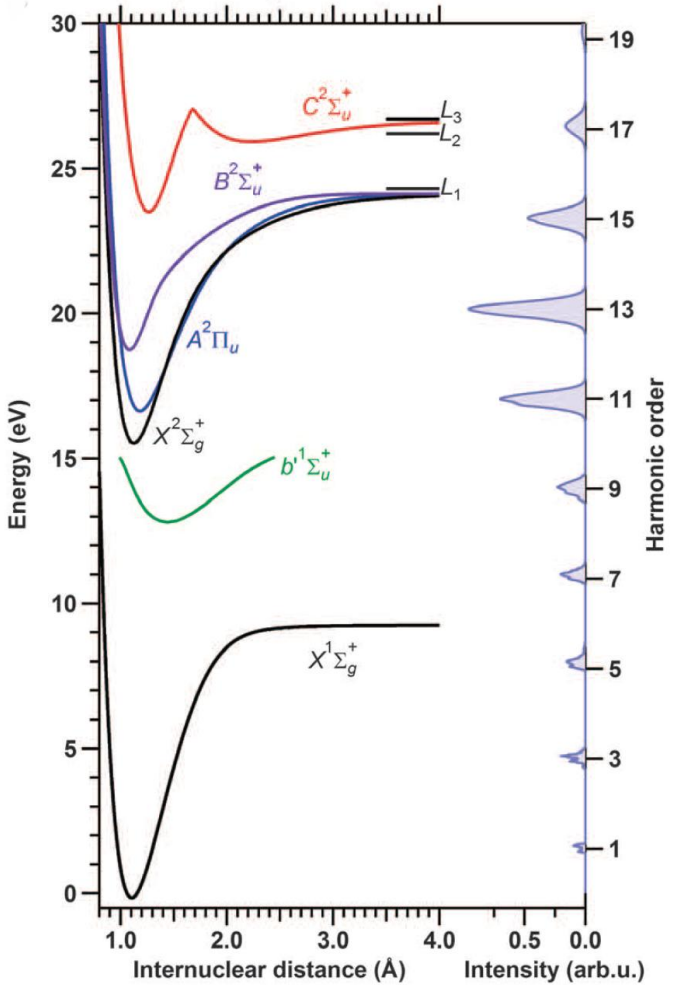
However, such experiments have only been performed on rare occasions up to now

Until the recent emergence of attosecond pulses at free electron lasers the vast majority of attosecond scientists are using variants of the XUV + NIR measurement protocols pioneered by Agostini and Krausz

Attosecond pump-probe

RIKEN (Midorikawa)

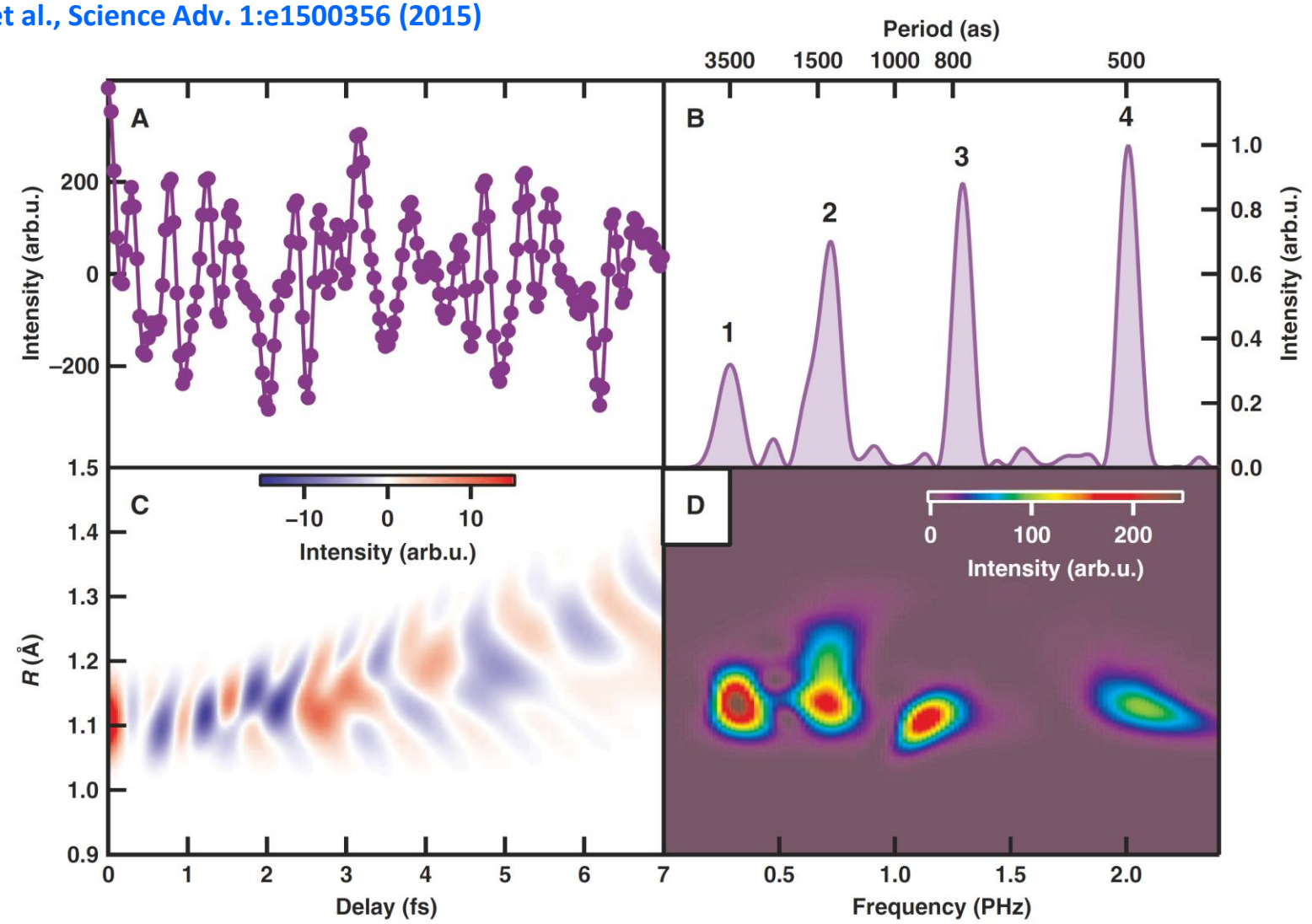




Okino et al., Science Adv. 1:e1500356 (2015)

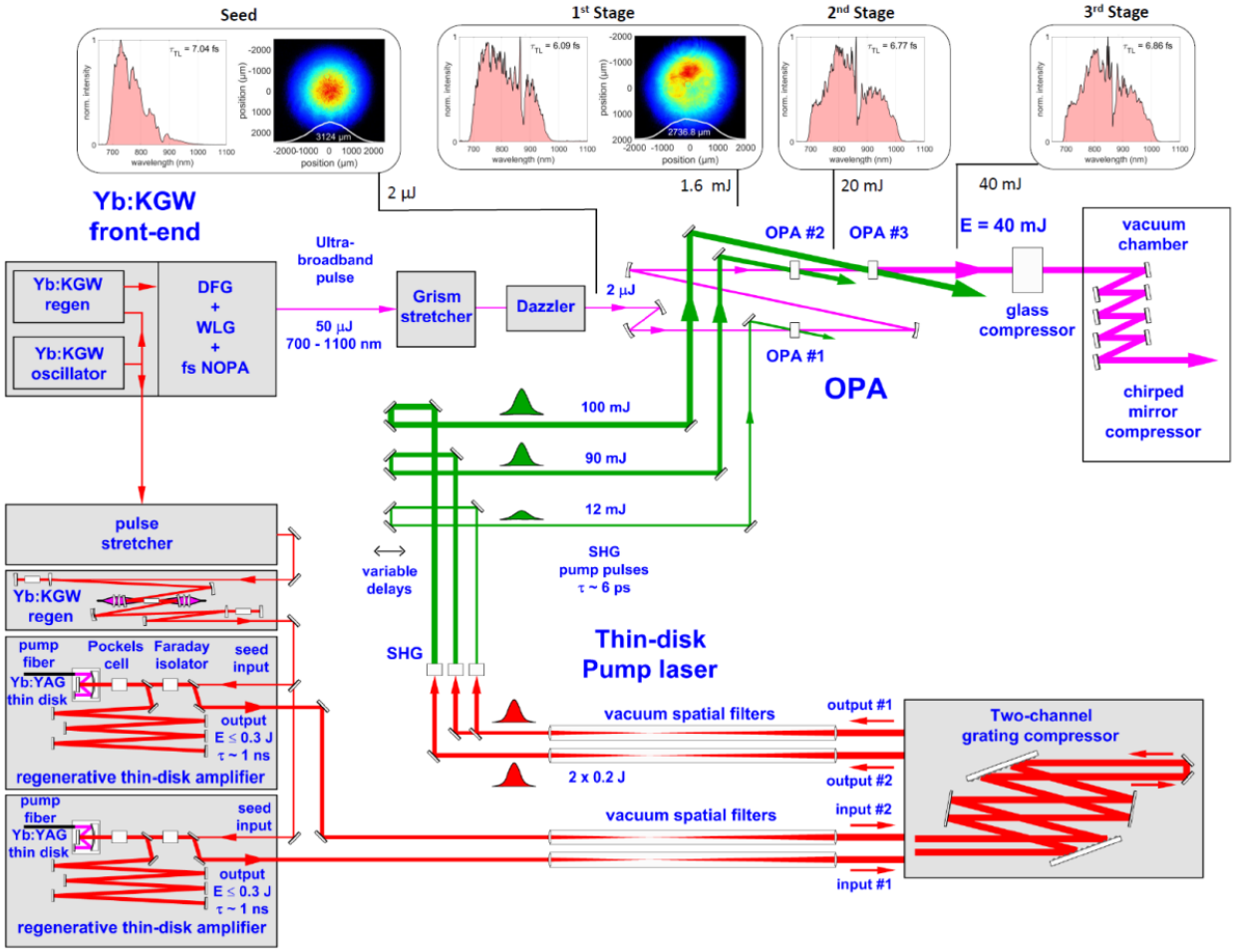
Attosecond pump-probe

Okino et al., Science Adv. 1:e1500356 (2015)



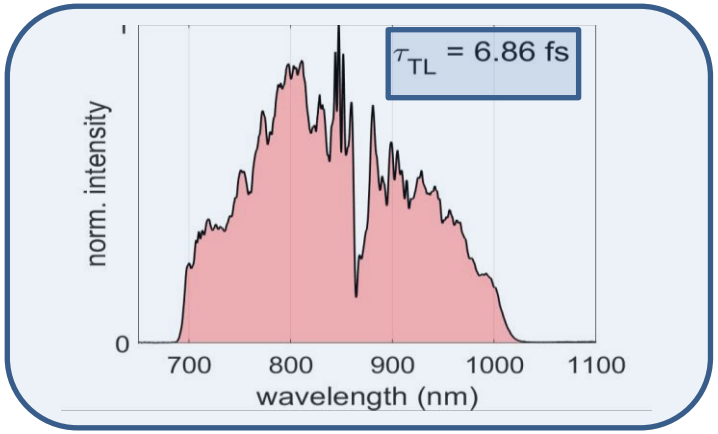
Attosecond pump-probe @ MBI

MBI (Schütte, Vrakking)

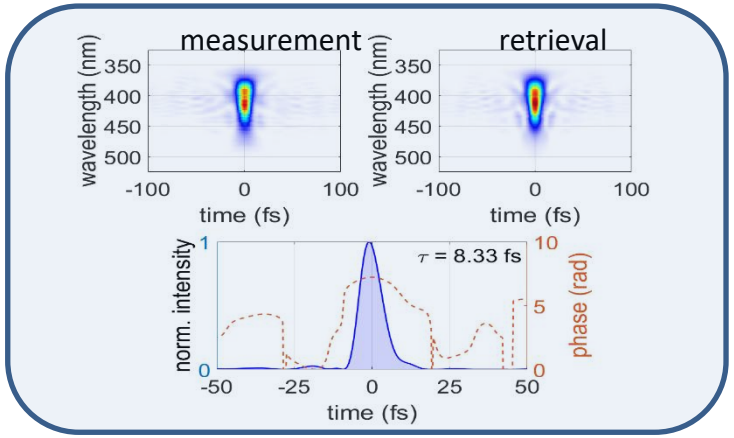


Xtreme Photonics Lab MBI (100 Hz)

Spectrum supporting 6.9 fs duration at 40 mJ pulse energy



FROG measurement confirming 8.3 fs duration

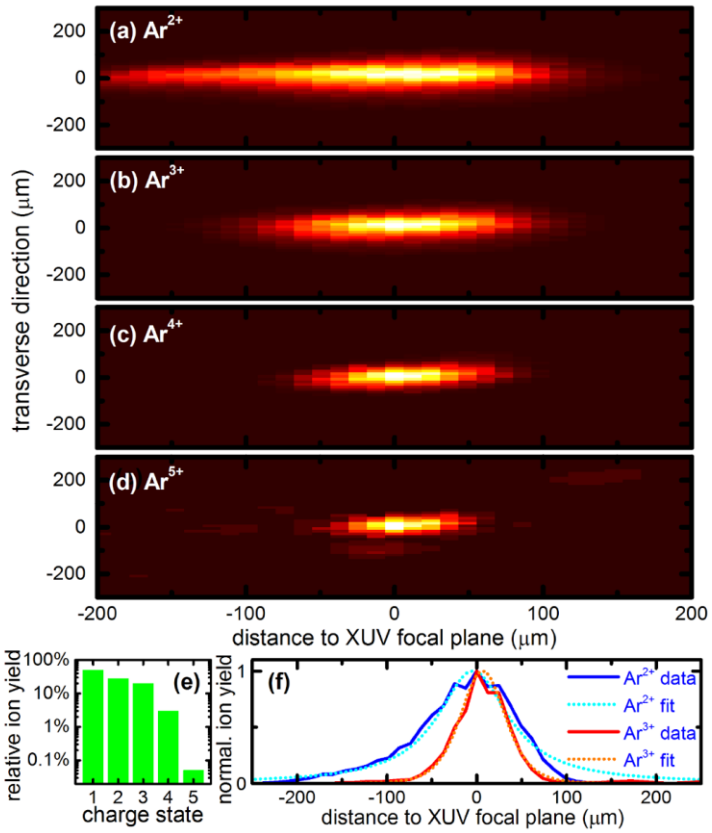


MBI Xtreme Photonics Laboratory in 2021

- Vacuum compressor and vacuum HHG beam line
- CEP detection and stabilization
- Timing jitter compensation for further improvement of stability and reproducibility

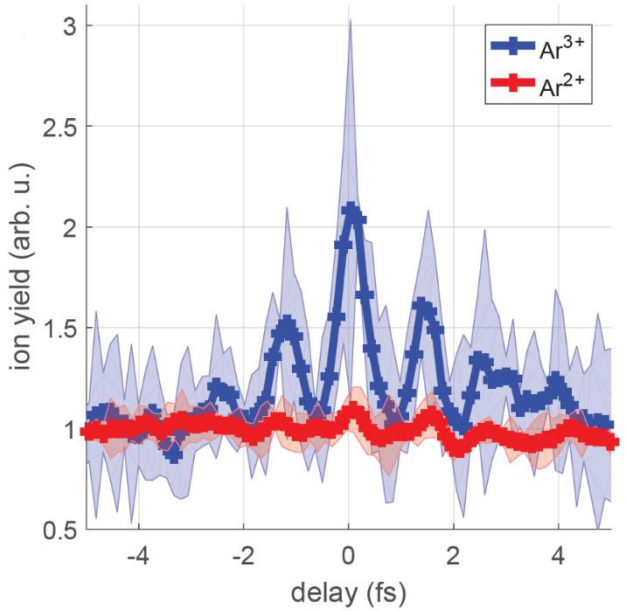
Extreme Photonics Lab MBI

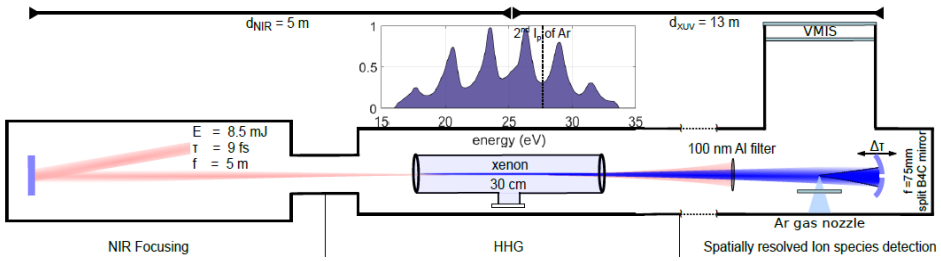
XUV multiphoton ionization using harmonics generation in Kr (appr. 700 nJ)



B. Senftleben et al., J. Phys. Phot. 2, 034001 (2020)

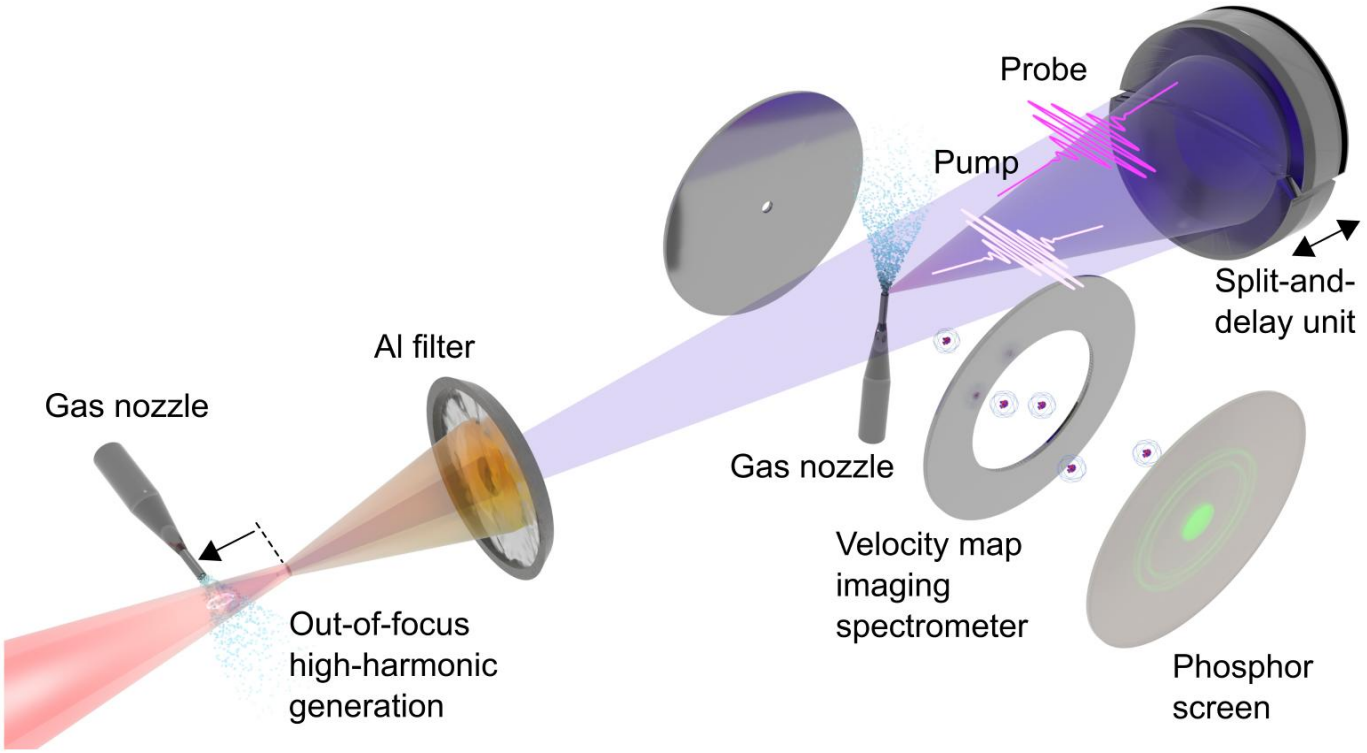
Attosecond pulse train AC measurement





M. Kretschmar et al., Optica 6, 639 (2022)

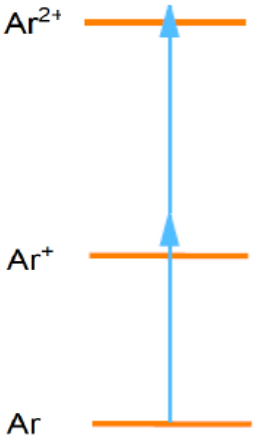
All-attosecond pump-probe spectroscopy



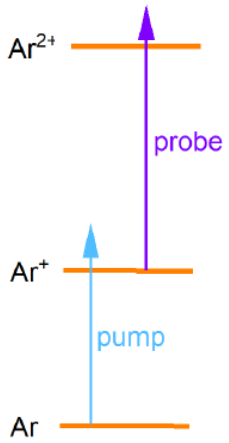
- Post-compressed kHz Ti:Sa laser (3.8 fs, 1 mJ)
- Tight focusing to ~6×10^{15} W/cm^2
- *Out of focus* HHG in a high density gas jet producing divergent XUV with a few-μm virtual source size
- 5-10 fold demagnification to waist radius ~1μm

Optica 8, 960 (2021)
Science Advances 10, eadk9605 (2024)

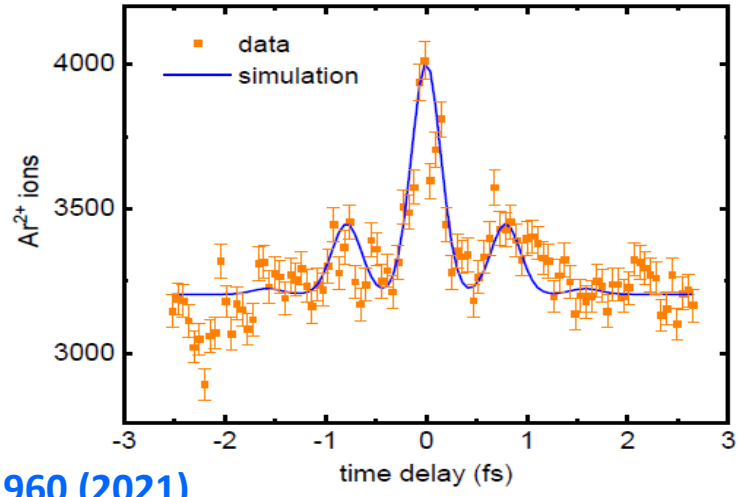
All-attosecond pump-probe spectroscopy

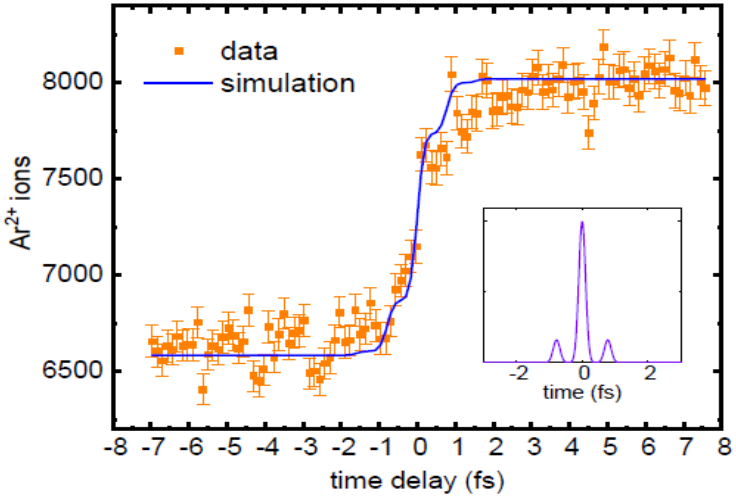


Single-color two-photon ionization: sequential and direct ionization



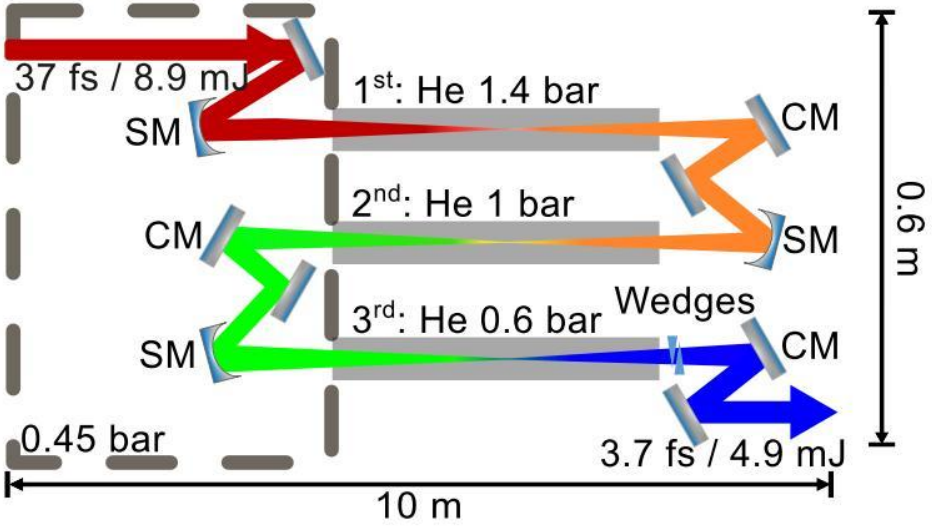
Two-color two-photon ionization: sequential and direct ionization

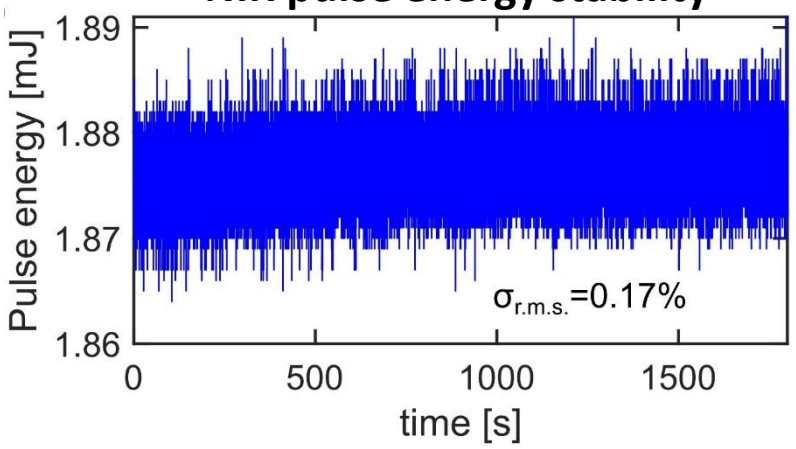


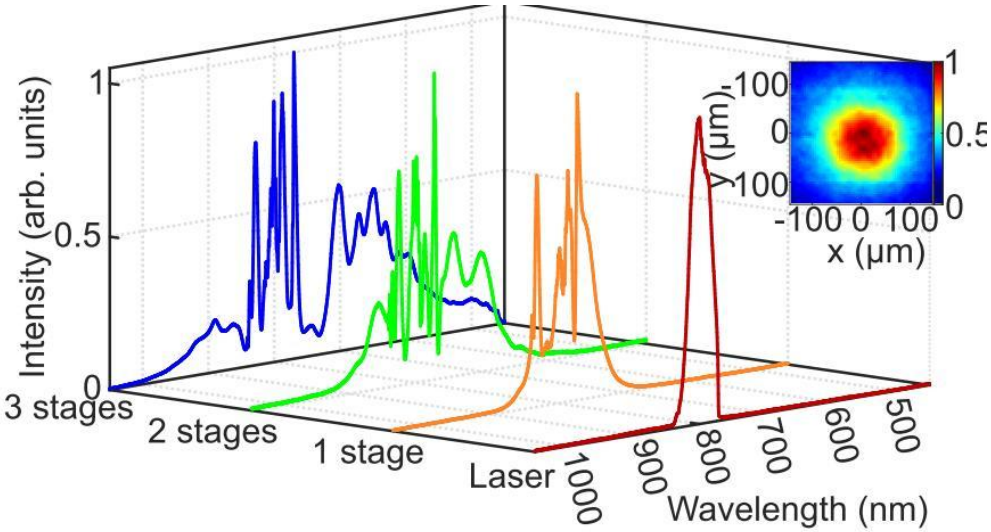


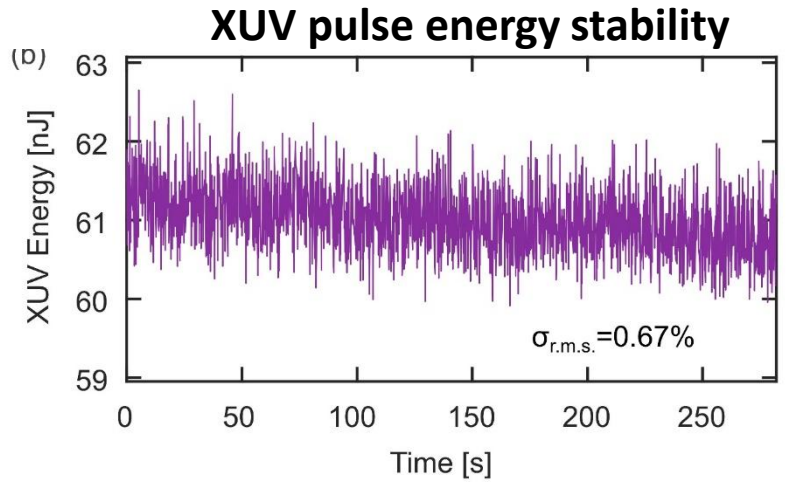
Optica 8, 960 (2021)
Science Advances 10, eadk9605 (2024)

New, optimized laboratory (1 kHz)



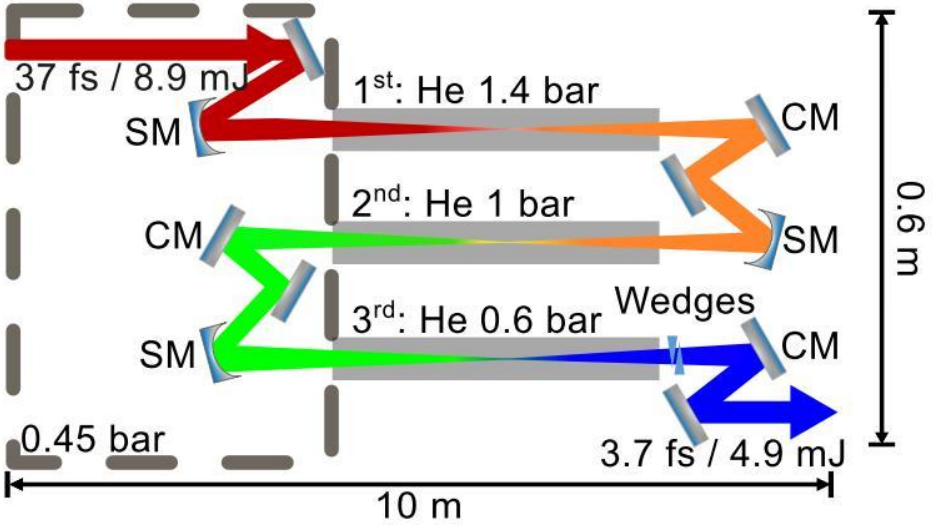


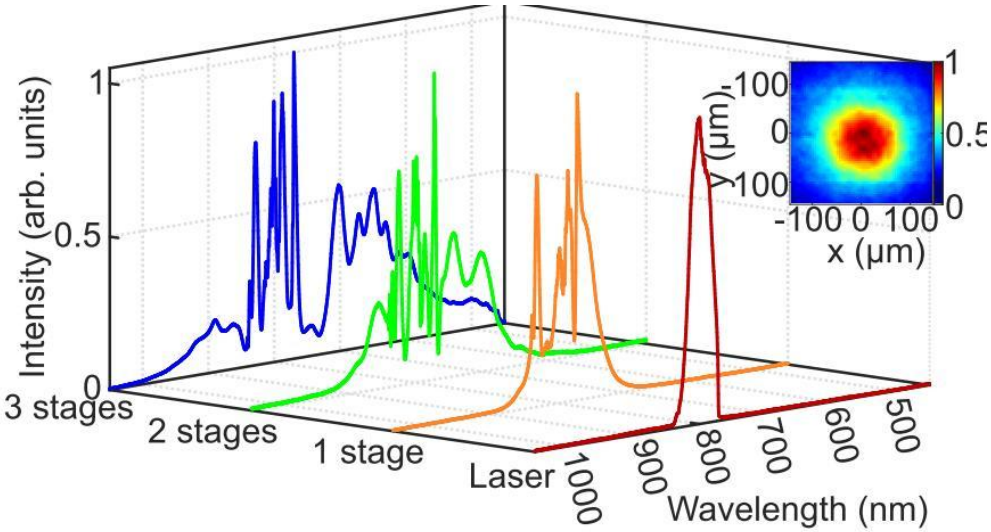


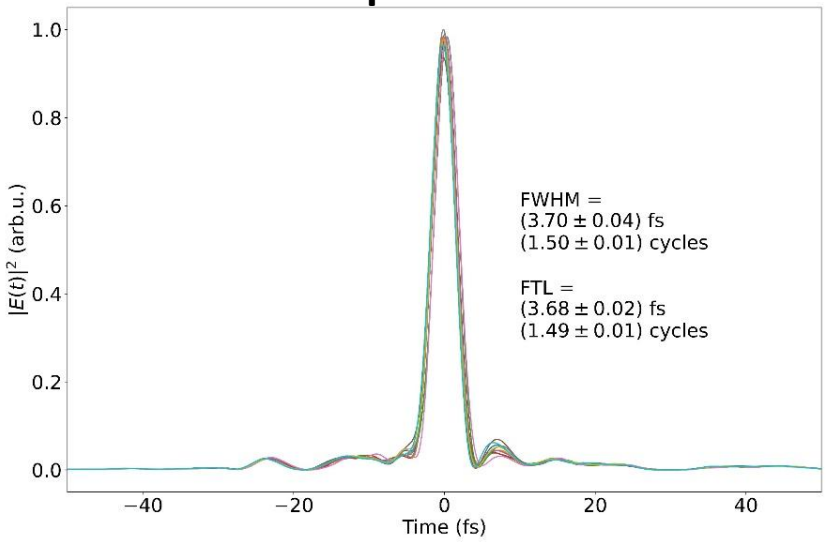


Sobolev *et al.*, Optics Express 32, 46251 (2024)

New, optimized laboratory

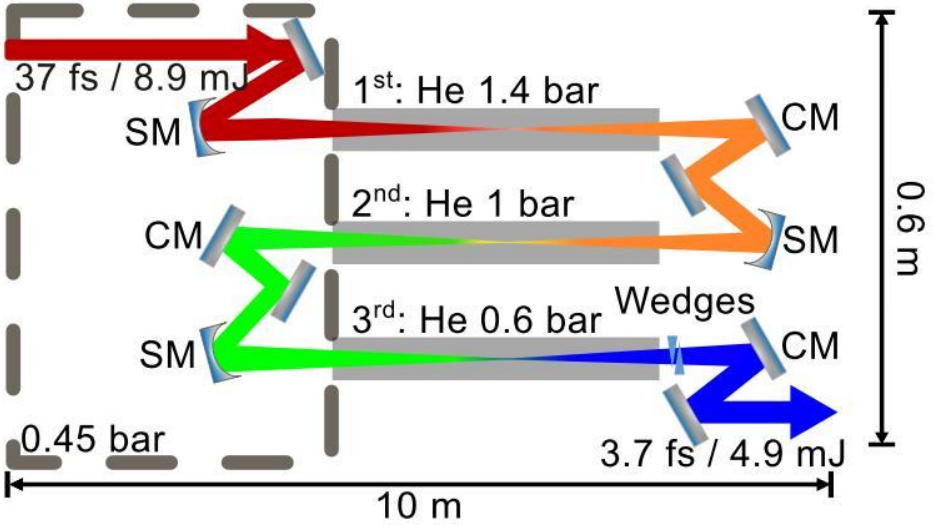


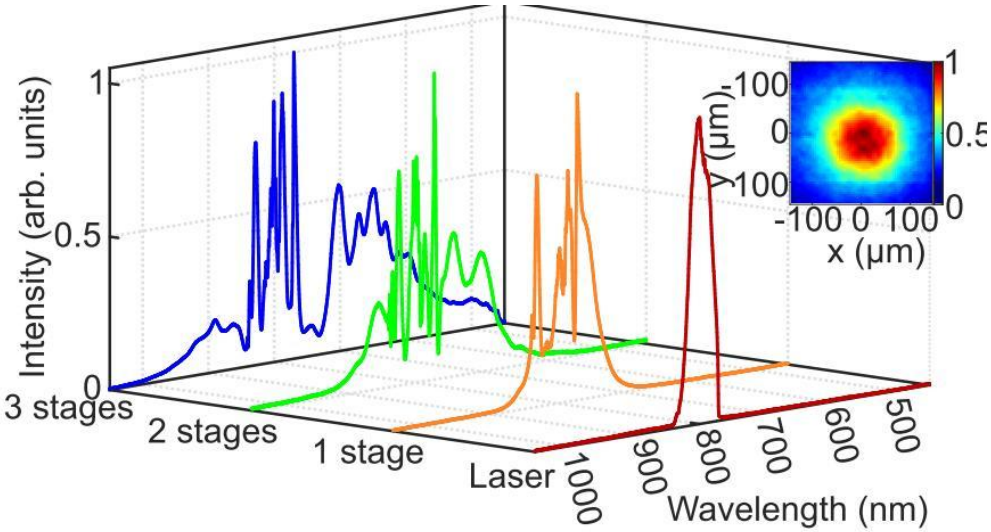




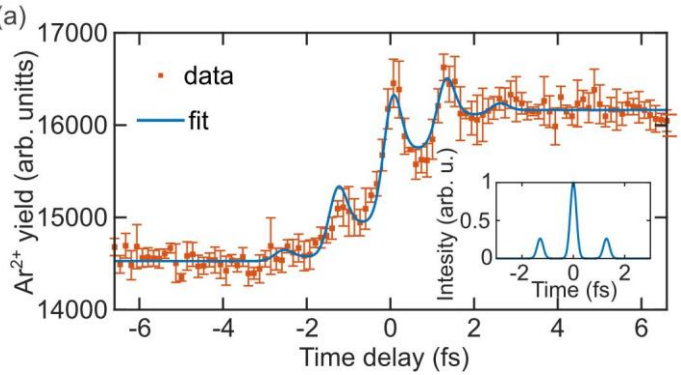
Sobolev *et al.*, Optics Express 32, 46251 (2024)

New, optimized laboratory

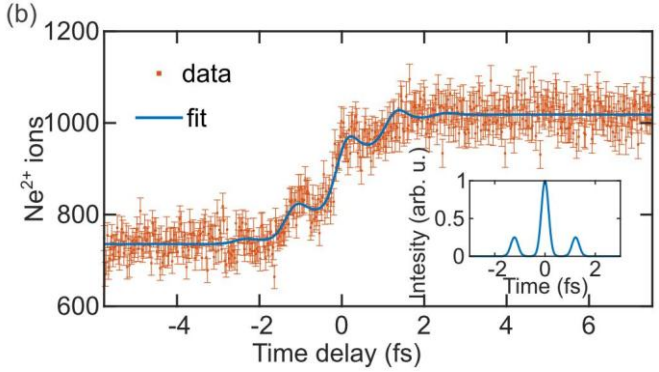




Attosecond pump pulse at 23 eV generates Ar^{+} ions
Attosecond probe pulse at 33.5 eV generates Ar^{2+} ions

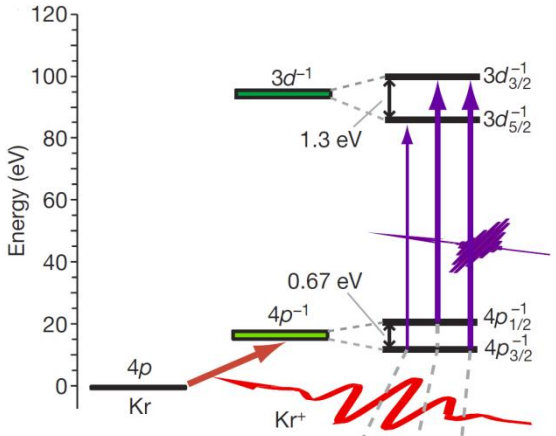


Attosecond pump pulse at 27 eV generates Ne^{+} ions
Attosecond probe pulse at 45 eV generates Ne^{2+} ions



Sobolev *et al.*, Optics Express 32, 46251 (2024)

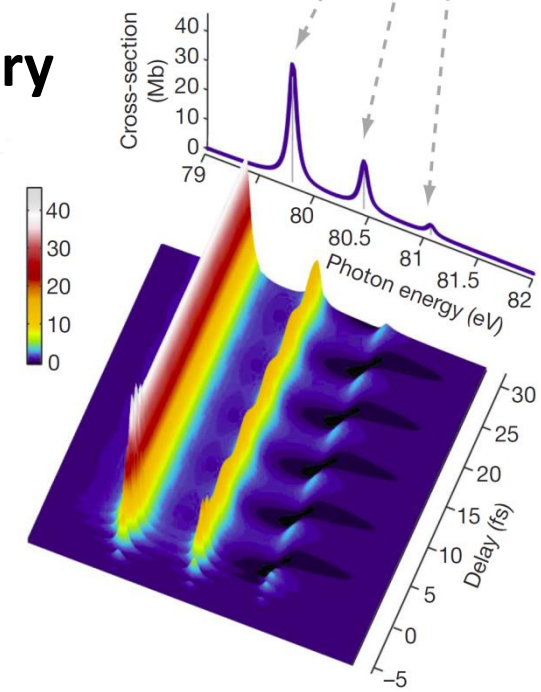
Observation of electronic coherence



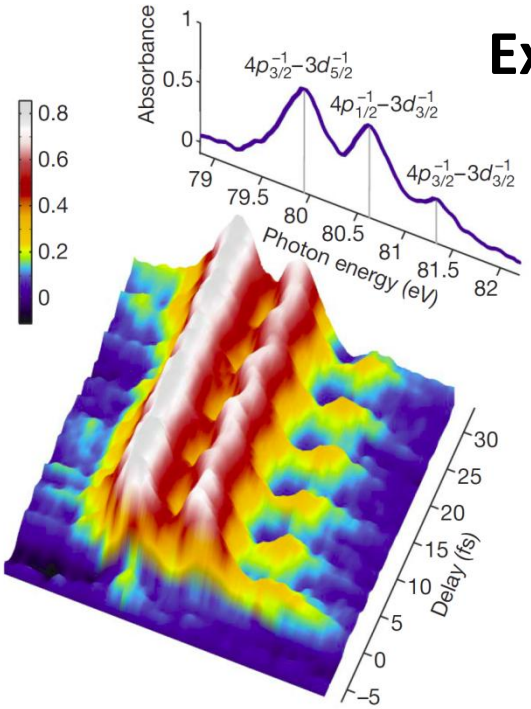
After ionization the Kr^+ ion is in a $4p_{1/2}$ or $4p_{3/2}$ state

Both configuration can be excited to a $3d_{3/2}$ state of the ion → interference

Theory

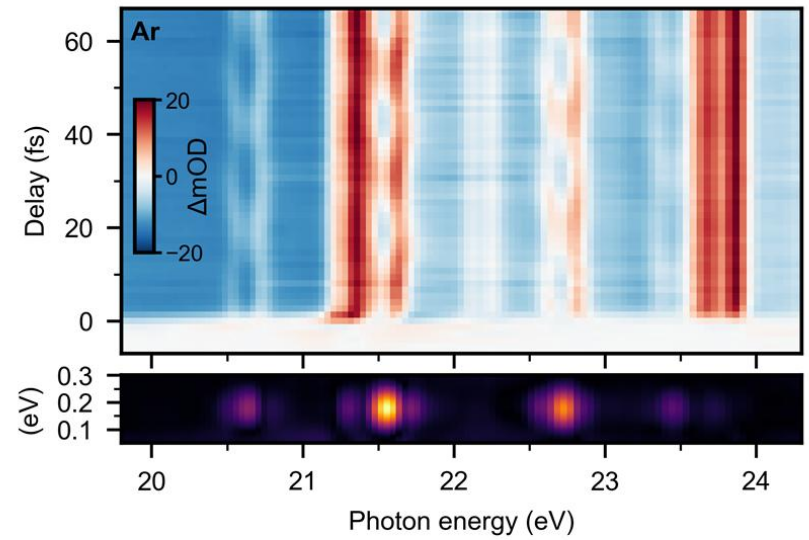


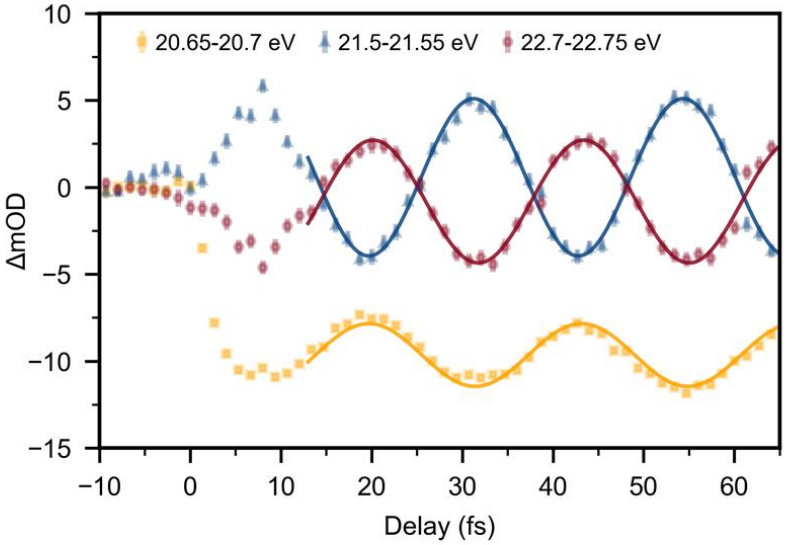
Experiment

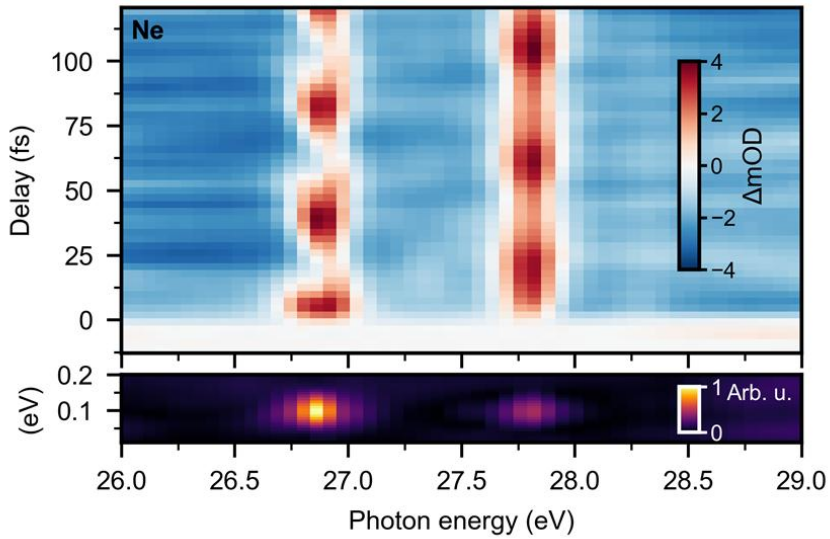


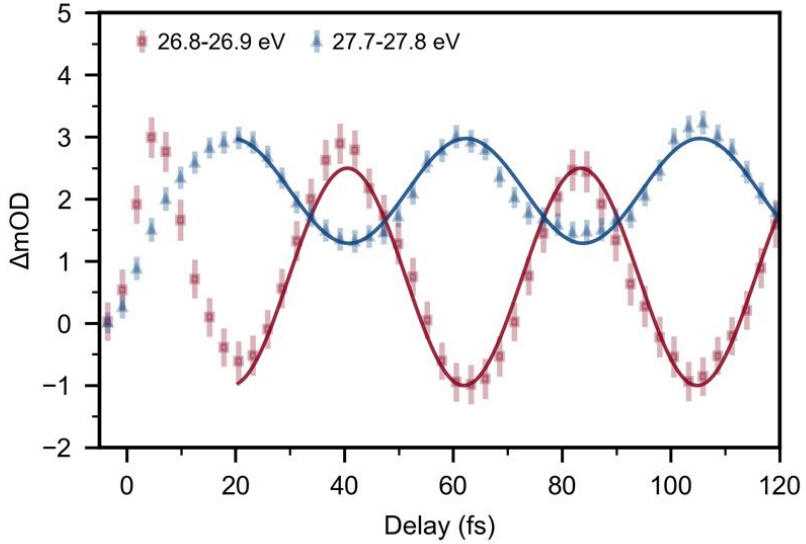
Goulielmakis et al., Nature 466, 739 (2010)

All-attosecond transient absorption



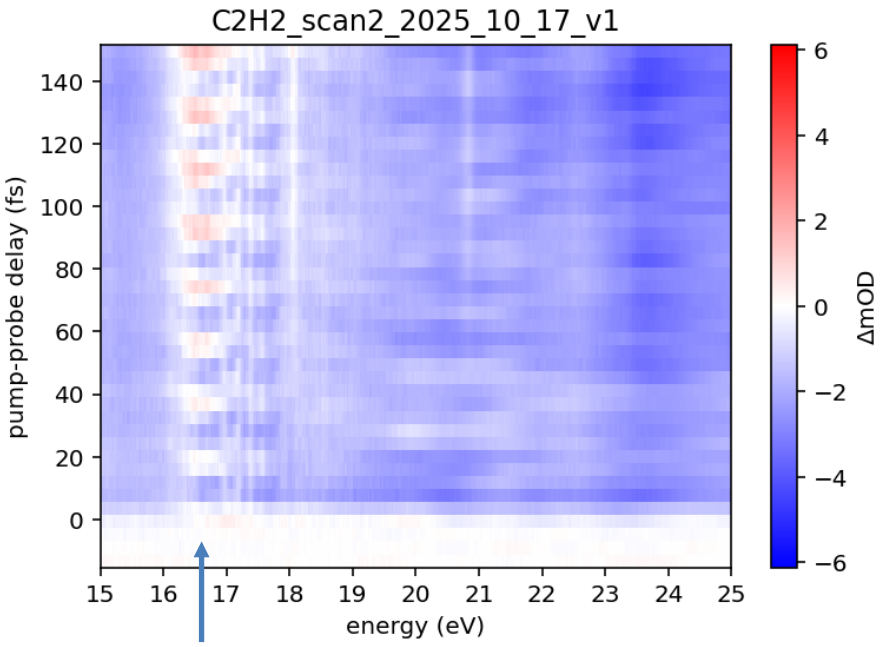




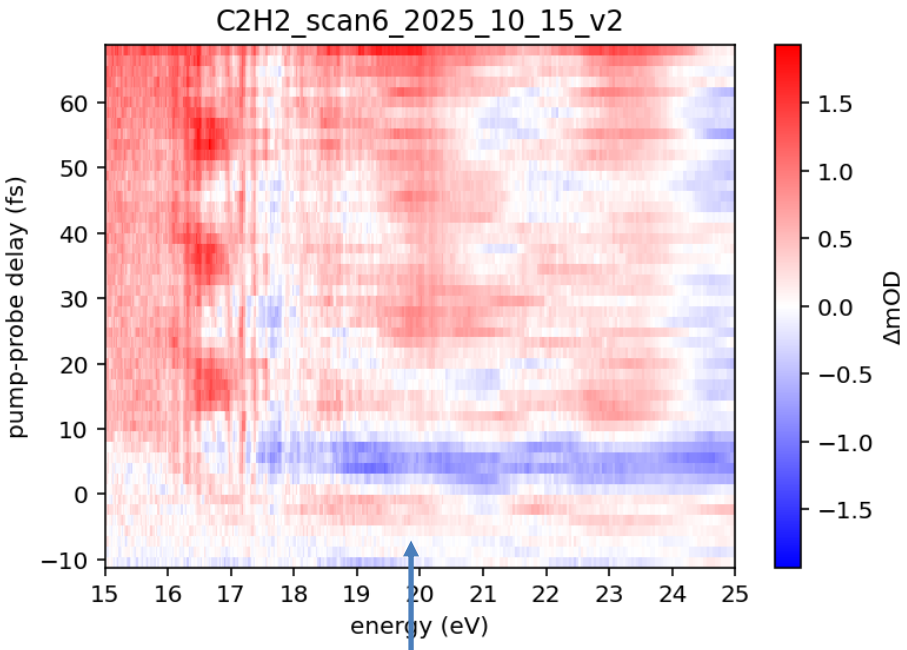


All-attosecond transient absorption

C^+ formation



Vibrational dynamics in cation



Conical intersection?

Conclusions

More than 20 years after the first characterization of attosecond pulse trains and isolated attosecond laser pulses, the execution of attosecond pump-attosecond probe spectroscopy has ***finally*** become readily accessible in the laboratory, including the use of informative differential detection techniques such as transient absorption

Initial experiments have revealed electronic coherences in atoms, as well as ultrafast coherent dynamics in molecules and solids

If this is of interest to ELI, we can help creating this capability at the user facility

Acknowledgements

Team leader: Bernd Schütte
Postdocs: Michael Volkov, Gabriel Emperauger et al.
PhD students: Evaldas Svirplys and Eli Sobolev