AMO from 10 eV to 100 keV: Molecular movie and beyond

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Many more…..

AMO from 10 eV to 100 keV: Molecular movie and beyond

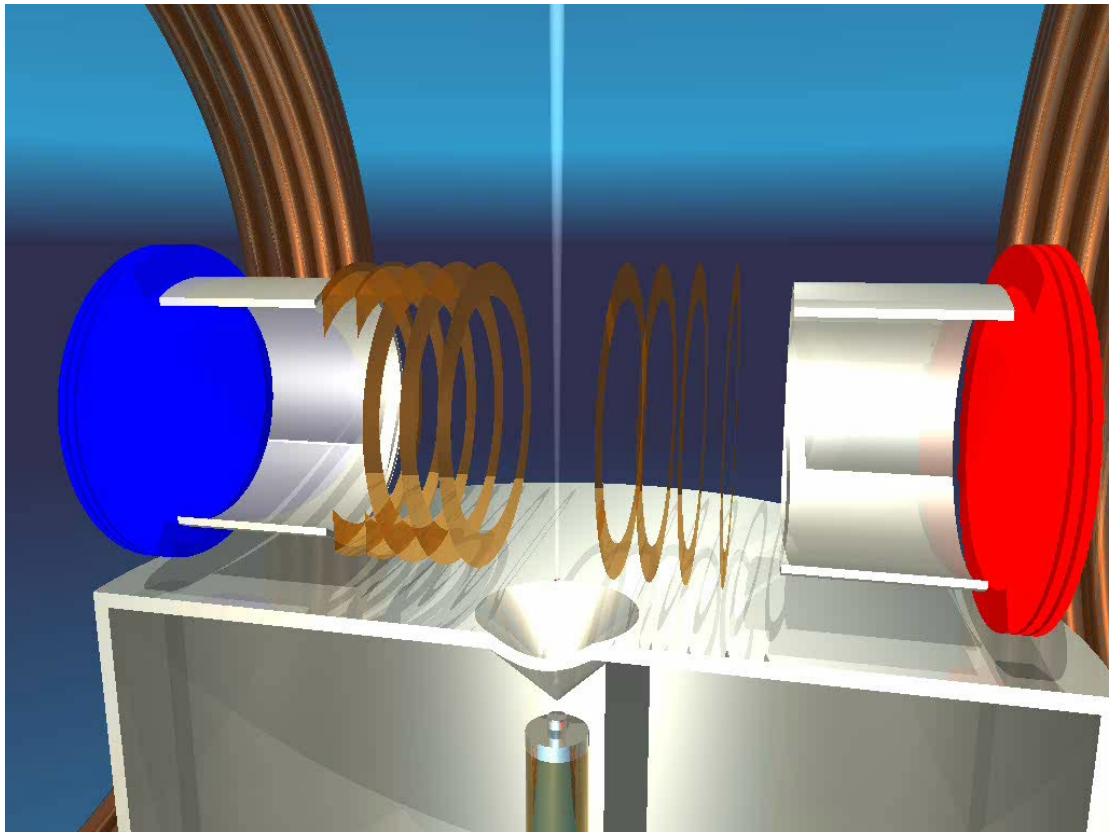
I: **200 (100?)** eV - 3 keV
Molecular movie with two-color SXFEL pump-probe

II: 3 - 25 keV
HX dynamic imaging of nanoparticles

III: 25 - **100 keV**
Photoelectron recoil (imaging and controlling nuclear packet) and Compton (imaging the electron orbital)

IV: **100(30?) eV-** 1 keV (fully coherent seeded FEL, EEHG)
Watching and stirring attosecond electron dynamics

I: Molecular movie with two-color SXFEL pump-probe



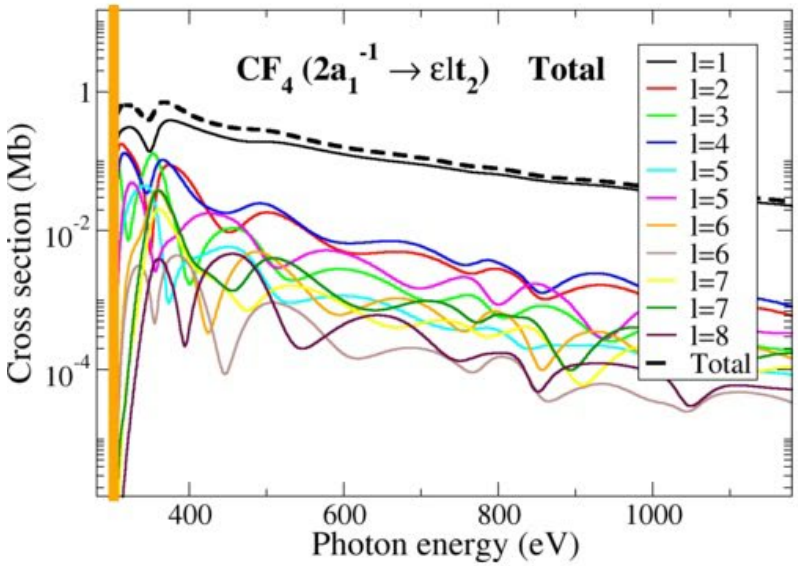
Two approaches to shoot the molecular movie:

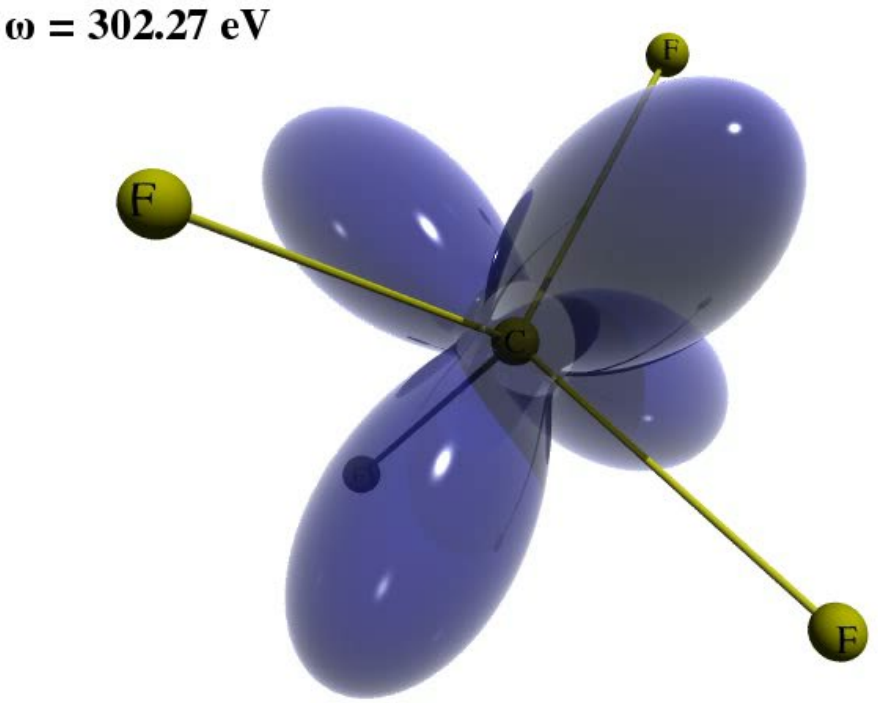
Time-resolved coulomb explosion imaging

Time-resolved molecular frame photoelectron angular distribution: Photoelectron diffraction

Polarization averaged MFPADs of CF_4

Etienne Plesiat,[1] Piero Decleva,[2] and Fernando Martin, Phys. Rev. A 88, 063409 (2013)





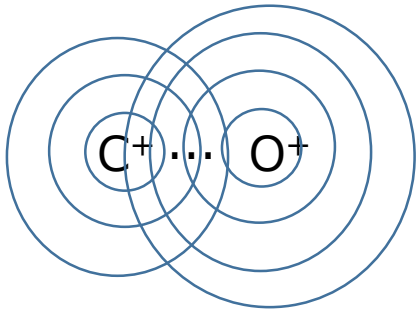
Proposed molecular movie with two-color SXFEL pump-probe I

Pump by 330 eV C 1s ionization and Probe by 660 eV O 1s ionization

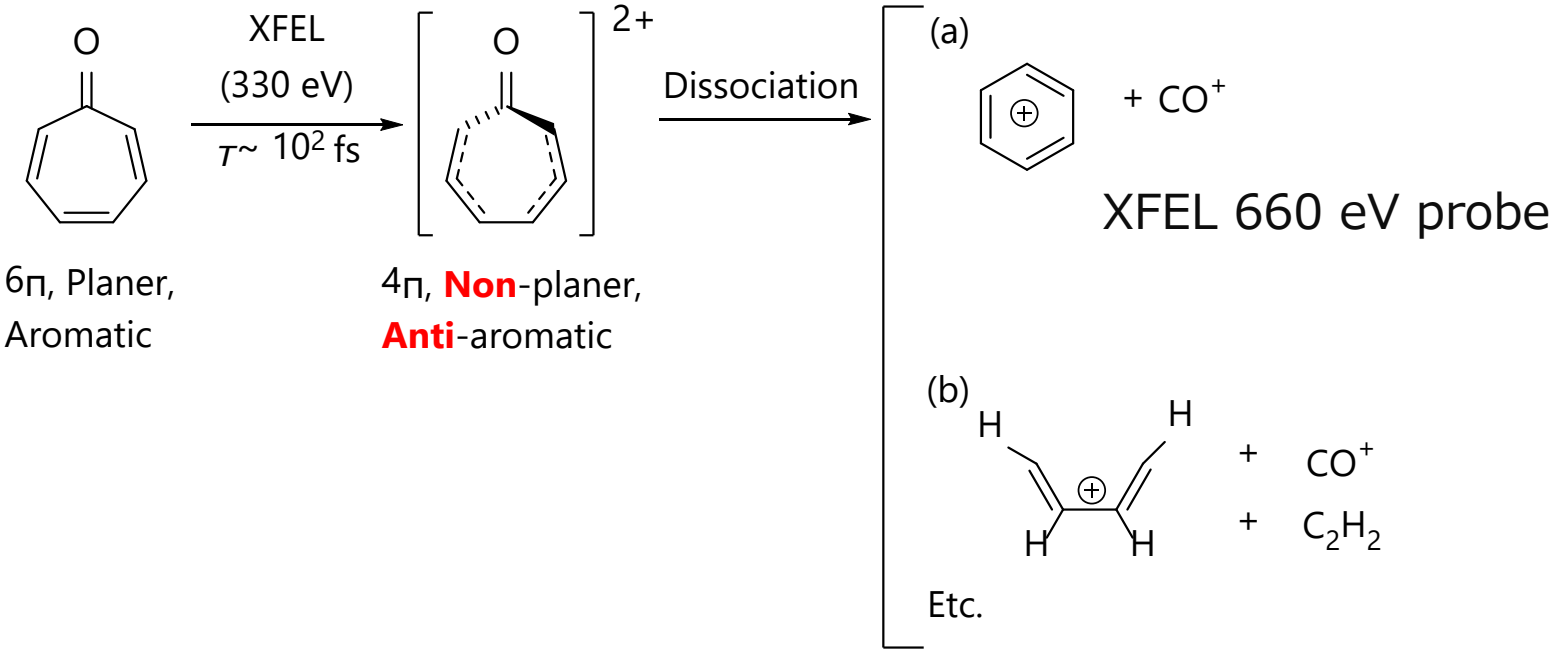
or vice versa

What kind of molecular movie we can shoot:

1. CO^{2+}: C^{+}-O^{+} Coulombic dissociation
(F. Ohta, K. Hatada et al.)



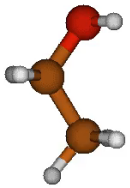
2. $C_7H_6O^{2+}$ (Non-planer sequential dis.)
(K. Yamazaki, F. Ohta, et al)



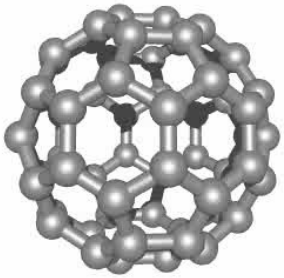
Proposed molecular movie with two-color SXFEL pump-probe II

3. CH_3CH_2OH: H migration (F. Martin, S. Díaz-Tendero F. Ohta et al. initiated by N.Berrah and N. Kling)

000.0 fs

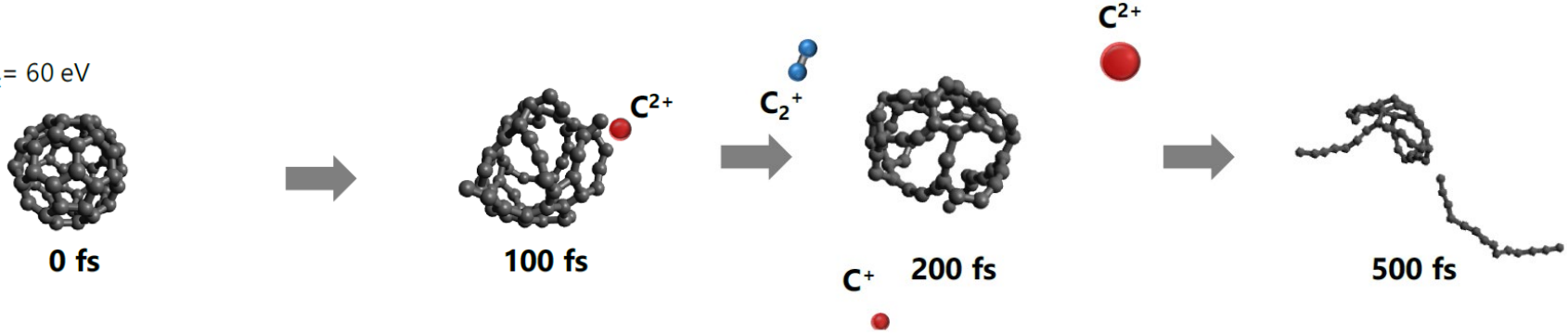


4. C60 (H. Kono, R. Santra, et al.) (Optiical pump SXSFEL probe)



Put Li in it and use it as an electron source (200 eV)

- Z = 12, E_{ex}= 60 eV



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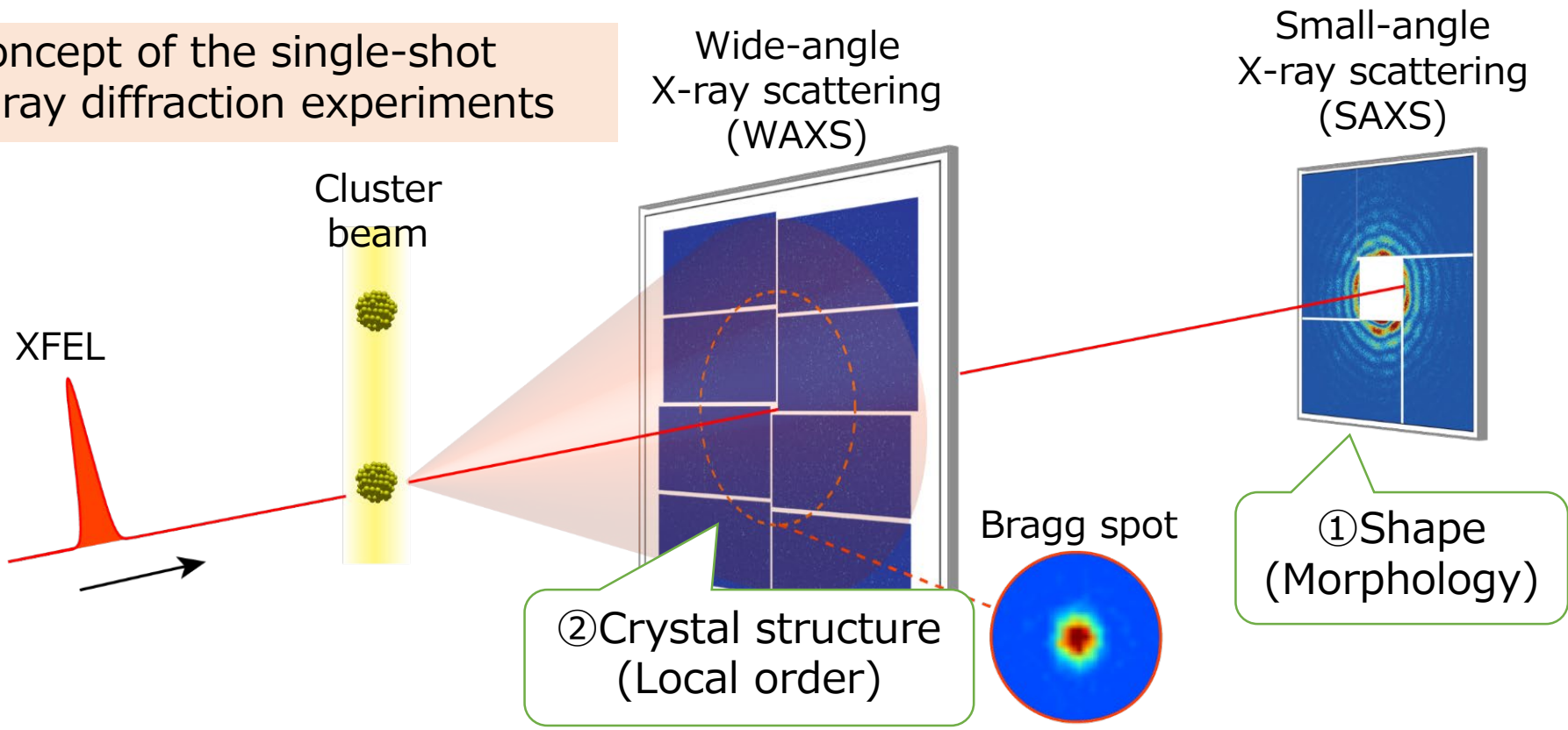
II: HX imaging of nanoparticles

Simultaneous measurement of small-angle vs wide-angle X-ray scattering

Concept of the single-shot
X-ray diffraction experiments



Three pulse-sequence with 2 or 3 color X-ray pulses + optical pump laser

1st HX pulse at for characterization of the nanoparticle

Plan 1: 1st pulse at 5 KeV for SAX
Plan 2: 1st pulse at 20 KeV for WAX
Plan 3: 1st pulse at 5 and 20 KeV for SAX and WAX

2nd pulse by intense optical laser for pump

3rd HX pulse for WAX for probe

Plan 1: 3rd pulse at 10 KeV for WAX
Plan 2: 3rd pulse at 10 KeV for WAX
Plan 3: 3rd pulse at 10 KeV for WAX

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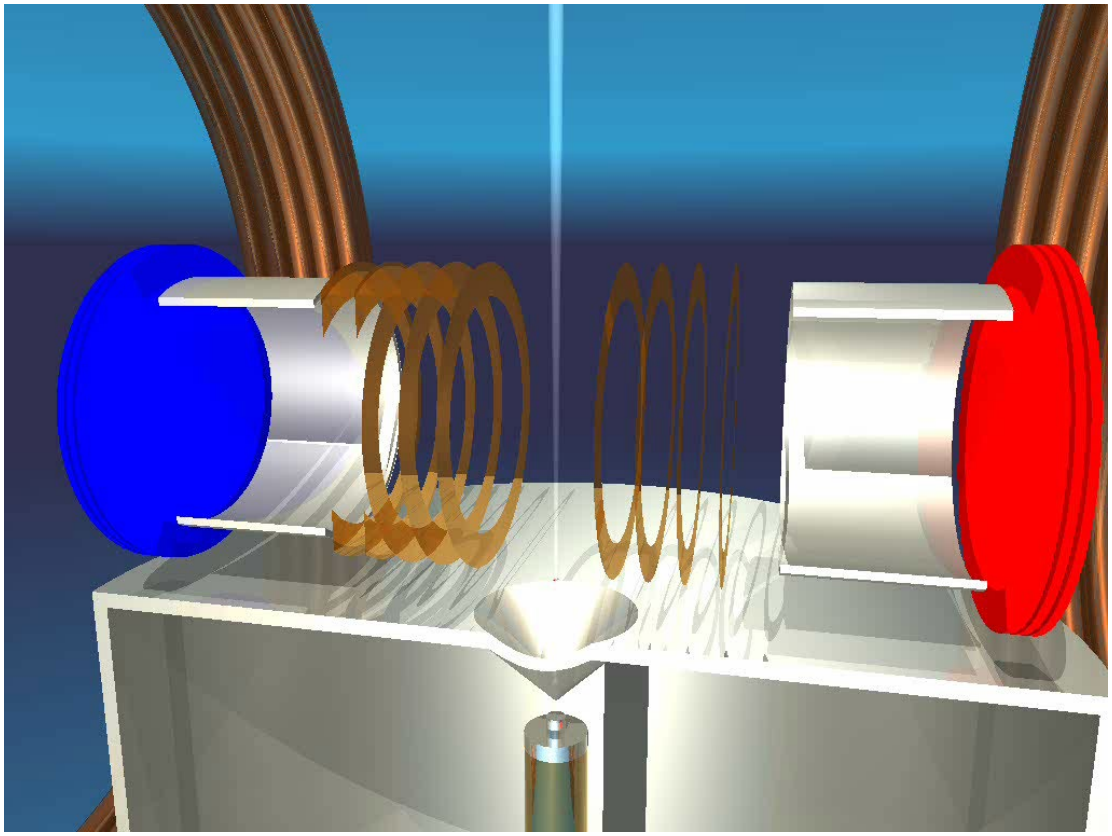
III: Photoelectron recoil and Compton above 40 keV with COLTRIMS



Photoelectron recoil for imaging and controlling nuclear packet

Compton scatting for imaging the electron orbital:
beyond the molecular movie

Photoelectron Recoil with SX (SR)

[1] W. Domcke and L. S. Cederbaum, J. Electron Spectrosc.Relat. Phenom. 13, 161 (1978). For homonuclear diatoms, photoelectron recoil energy goes to vibrational (1), rotational (2), and translational (3) eneries (degree of freedoms).

[2] E. Kukk et al., Phys. Rev. Lett. 95, 133001 (2005).
First observation of C1s photoelectron-recoi-induced vibratoinal excitation in CH4

[3] T. D. Thomas et al., J. Chem. Phys. 128, 144311 (2008)
C1s photoelectron-recoil-induced vibratoinal excitation in CF4

[4] T. D. Thomas et al., Phys. Rev. A 79, 022506 (2009).
Photoelectron-recoil-induced rotational excitation in N2

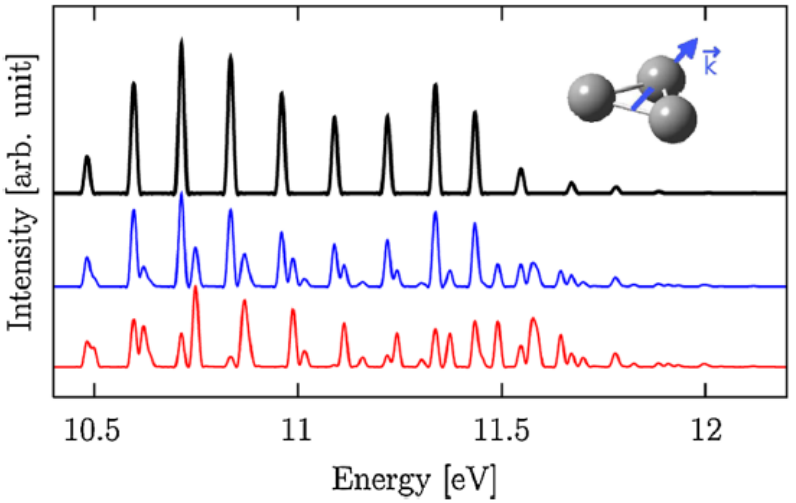
[5] K. Kreidi et al., Phys. Rev. Lett. 103, 033001 (2009)
Recoil-induced dynamics of ICD – COLRIMS

More experiments in Tender X-rays.

….

[6] L. S. Cederbaum and M. Basler,
Phys. Rev. Lett. 103, 133001 (2009)
Nonadiabatic Effects by Photoelectron Recoil

No observation so far…



Proposed experiment above 40 keV

Exploring Nonadiabatic Effects by Recoil of ~50 keV Photoelectrons probing them by ~25 keV Photoelectrons

Evolution of molecular orbitals by Compton scattering during laser-induced reactions:
beyond the molecular movie

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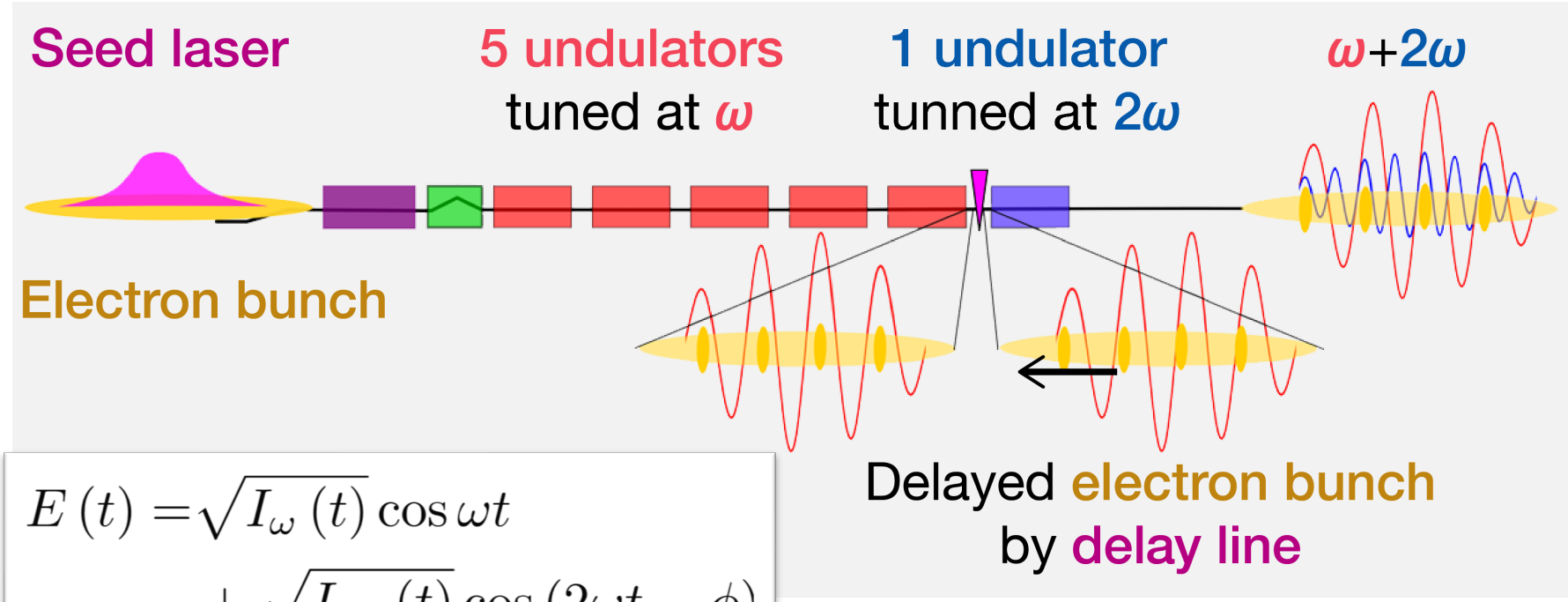
IV. Fully coherent seeded SX FEL

Phase-coherent two-color pulses

Delaying the electrons' arrival to the last undulator, controls the phase difference between the two FEL pulses with 3 attosecond precision.



$$E(t) = \sqrt{I_\omega(t)}\cos\omega t + \sqrt{I_{2\omega}(t)}\cos(2\omega t - \phi)$$

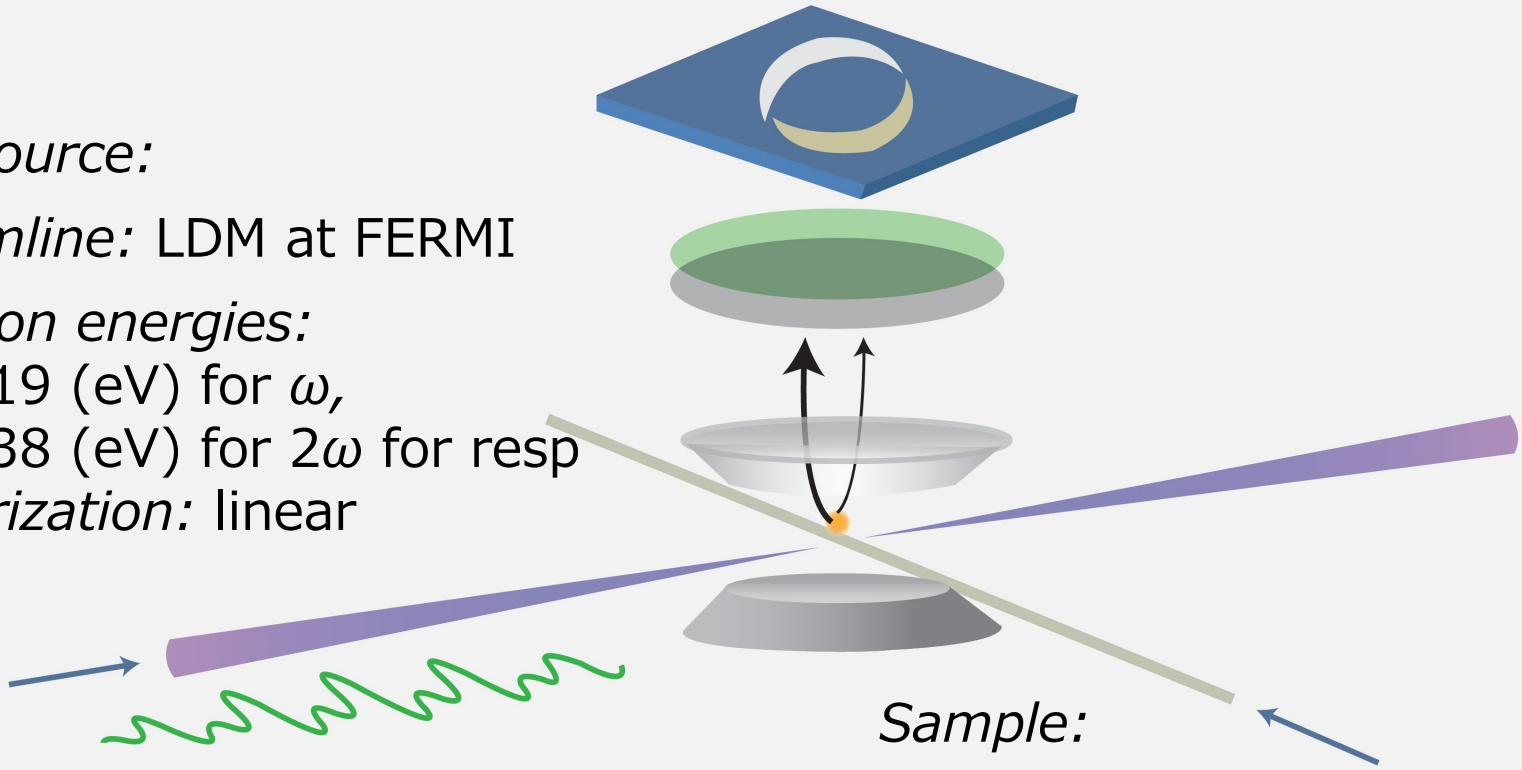
K. C. Prince *et al.* Nat. Photonics 10 176 (2016) and L. Giannessi *et al.* Sci. Rep. 8 7774 (2018)

Experimental setup

Measuring method: Velocity map imaging (VMI) spectrometer

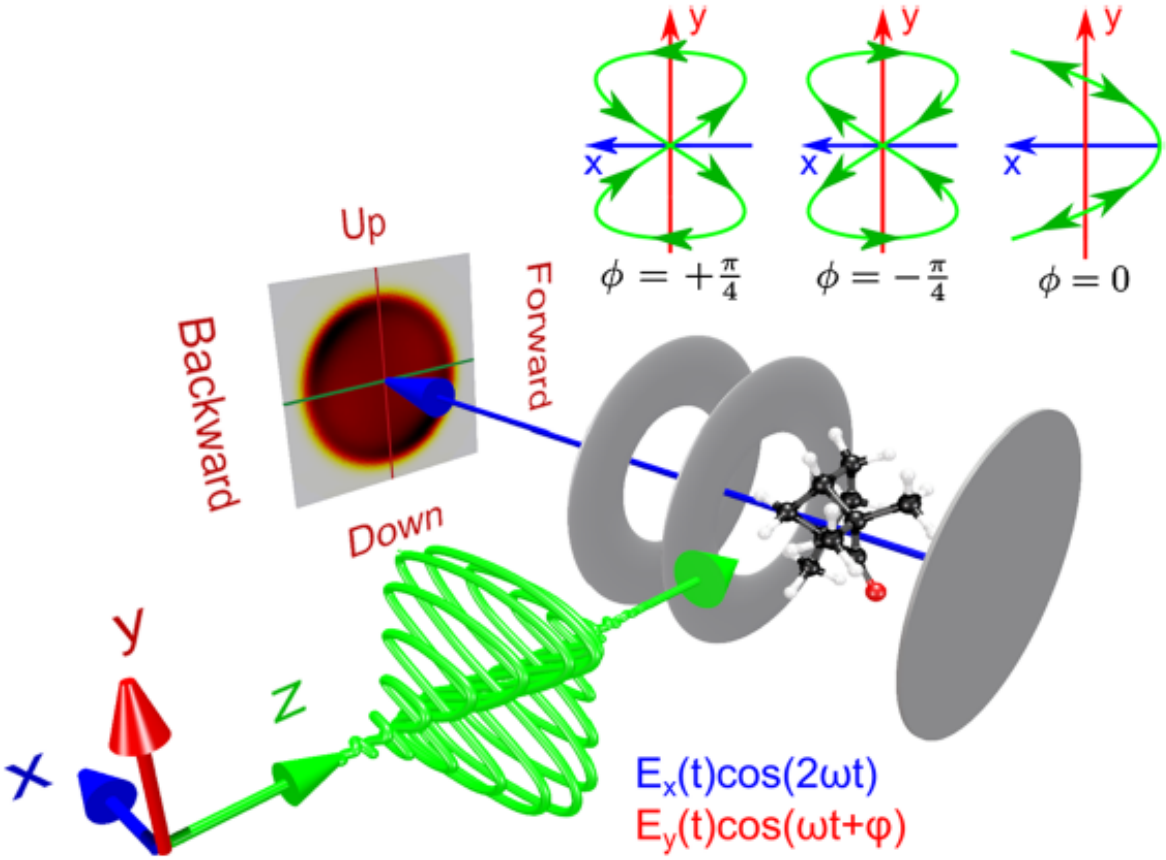
Light source:

- *Beamline:* LDM at FERMI
- *Photon energies:*
  14~19 (eV) for ω,
  29~38 (eV) for 2ω for resp
  Polarization: linear



Original figure from K. C. Prince *et al. Nat. Photonics* **10** 176–179 (2016)

Photoelectron Circular Dichroism with ω+2ω



P.V. Demekhin,* A. N. Artemyev, A. Kastner, and T. Baumert, PRL 121, 253201 (2018)

Proposed Objectives:

Time-resolved chiral dynamics of the photoexcited states

Photoelectron Circular Dichroism with ω+2ω

Proposed Objectives:

Time-resolved chiral dynamics of the photoexcited states

1. Angle-resolved "attosecond time delay" btwn electrons ejected in $1^{st}+3^{rd}$ quadrant versus those in 2^{nd} and 4^{th} quadrant, for different orbitals (enantio-sensitive coefficient $\beta_{2,-1}$ in PAD decomposition)

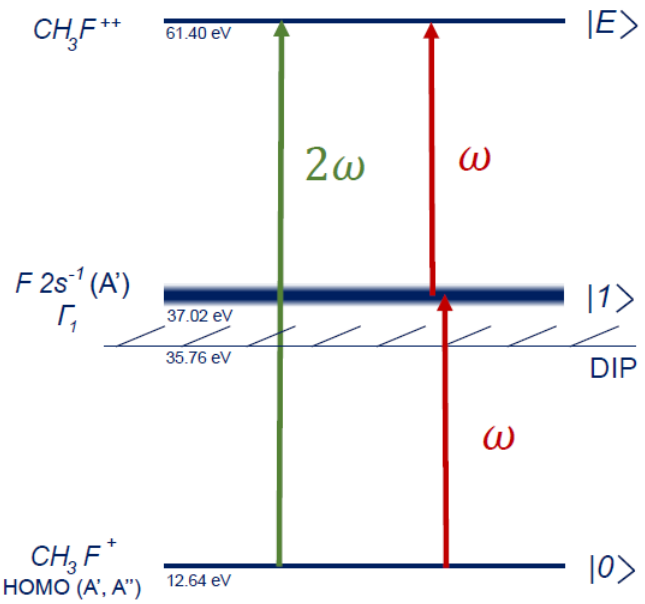
2. Use w-2w interferometry to gain attosecond resolution without as pulses. We benefit from energy-resolved PES to identify different final states. This is the strength of attosecond interferometry – we gain attosecond resolution without having very broad pulses.

Important issue:

Geometry is not cylindrically symmetric, but we want 3D imaging (tomography). Thus we need high-rep. rate FERMI with COLTRIMS!

By Olga Smirnova and Misha Ivanov

Molecular Auger Interferometry



M.A. Khokhlova[1;], B. Cooper[2], K. Ueda[3], K.C. Prince[4;5], P. Kolorenc[6], M.Yu. Ivanov[1;7] and V. Averbukh,, submitted to PRL

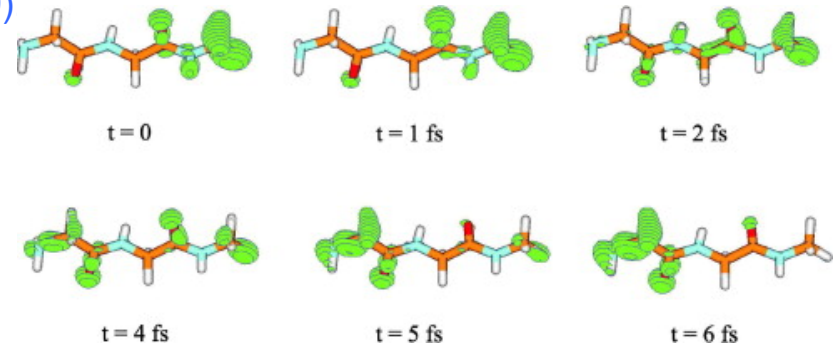
1. One can coherently control of the electron yield in molecules observe variation of the yield for certain molecular symmetries.
2. One can extract the lifetime of the intermediate state.

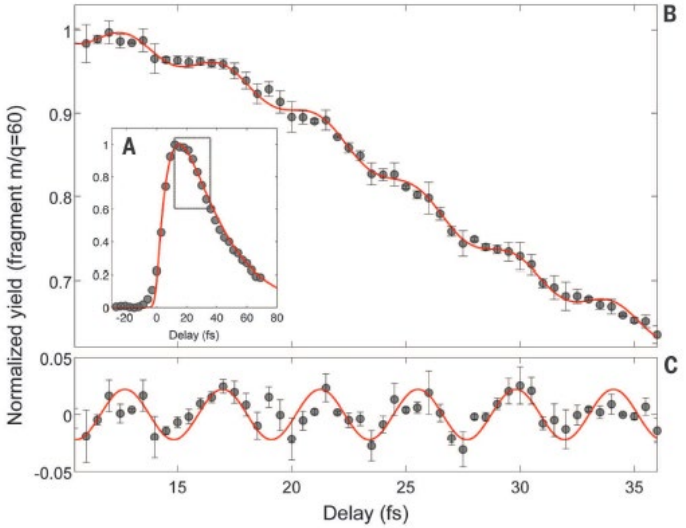
We want to fix the molecule in space and see electron emission in molecular frame.

High-rep. rate fully coherent pulses (EEHG) with COLTRIMS!

Charge migration: electronic wave packet motion

Ultrafast charge migration by electron correlation
L. S. Cederbaum, J. Zobeley, *Chem. Phys. Lett.* **307**, 205 (1999)

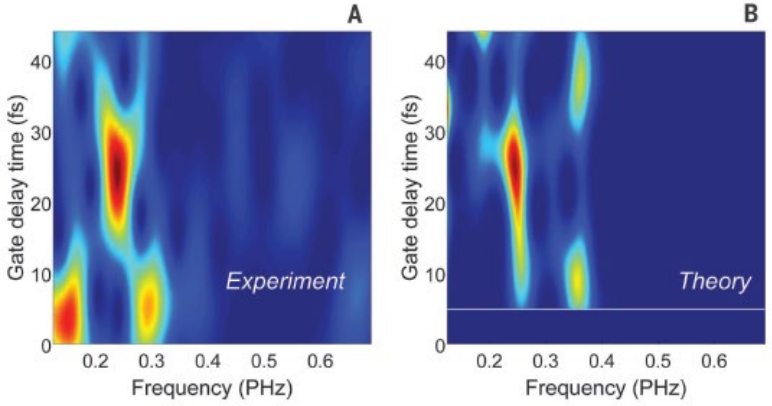




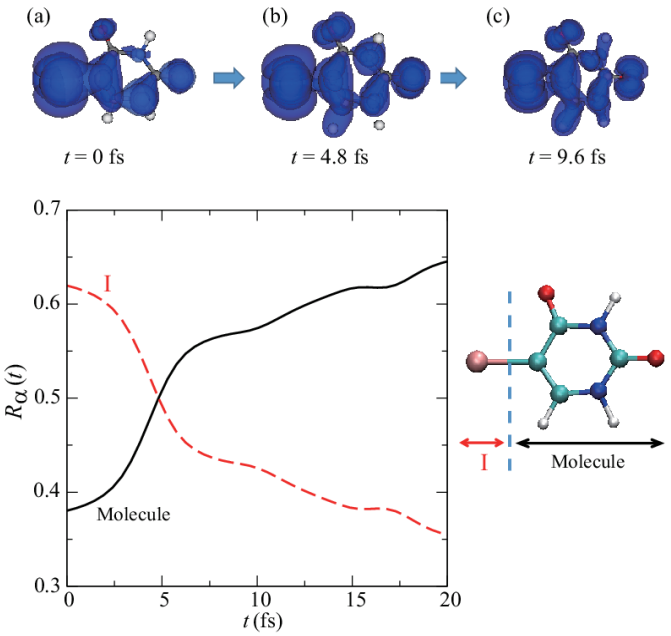
Electron-correlation-driven charge migration in oligopeptides
A. I. Kuleff et al. *Chem. Phys.* **414**, 100 (2013).

実験

Ultrafast electron dynamics in phenylalanine initiated by attosecond pulses
F. Calegari et al. *Science* **346**, 336 (2014).

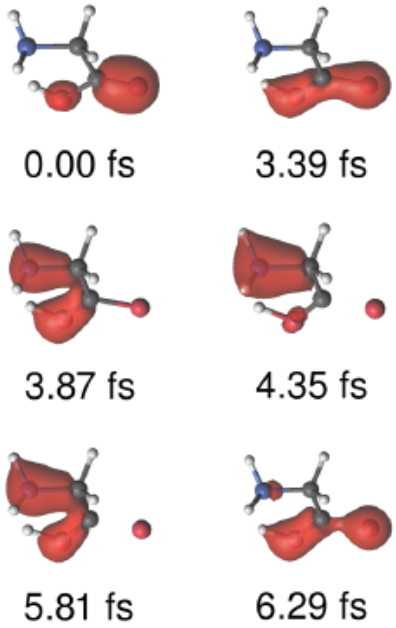


Ultrafast charge transfer of a multiple valence hole in molecules driven exclusively by nuclear motion



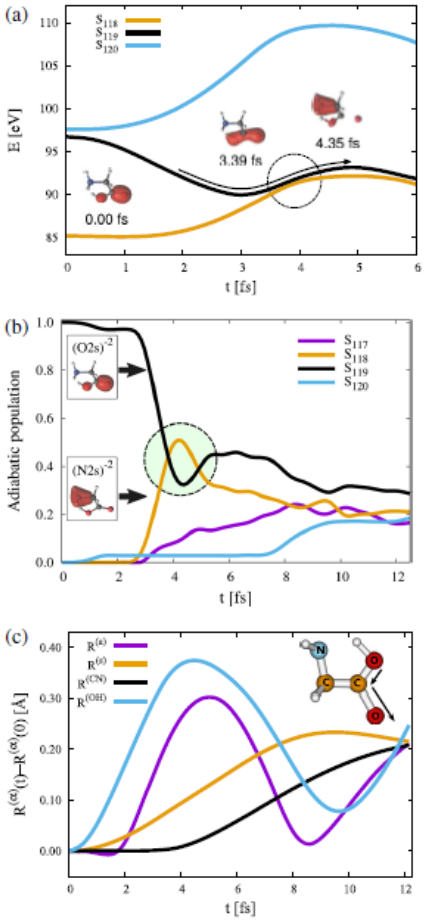
Charge transfer and localization in $5IU^{6+}$

K. Nagaya et al., *Faraday Discussions* **194**, 537(2016).



Charge transfer in Glycine^{2+}

Z. Li, O. Vendrell, and R. Santra
PRL **115**, 143003 (2015).



COLTRIMS can probe the evolution of the electronic state but to see the valence electron we need the second pulse at ~100 eV.

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III: 25-100 keV

Photoelectron recoil, imaging and controlling nuclear packet and Compton, imaging the electron orbital)

IV: 100(30?) eV -1 keV (fully coherent seeded FEL -EEHG)

Watching and steering attosecond electron dynamics