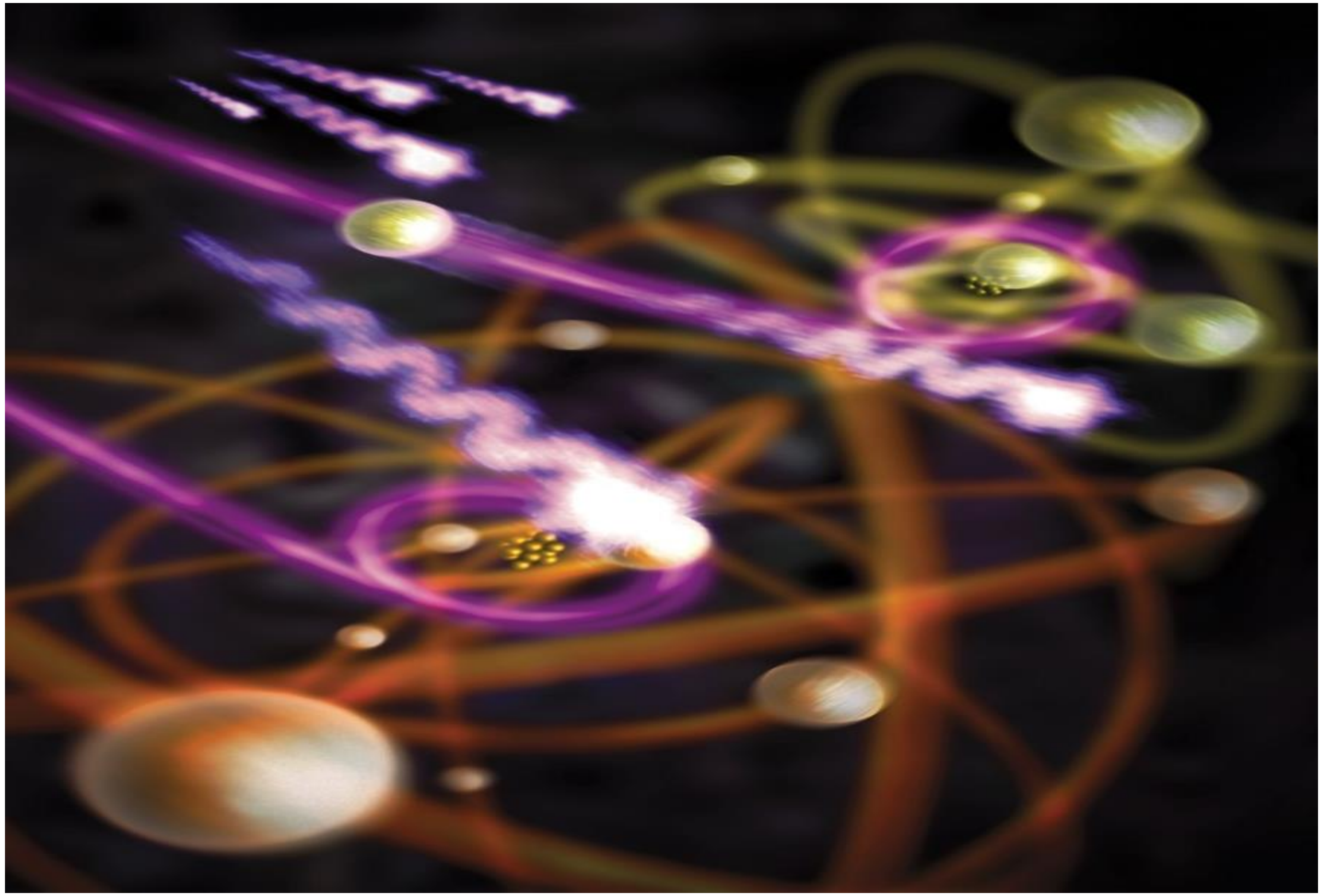
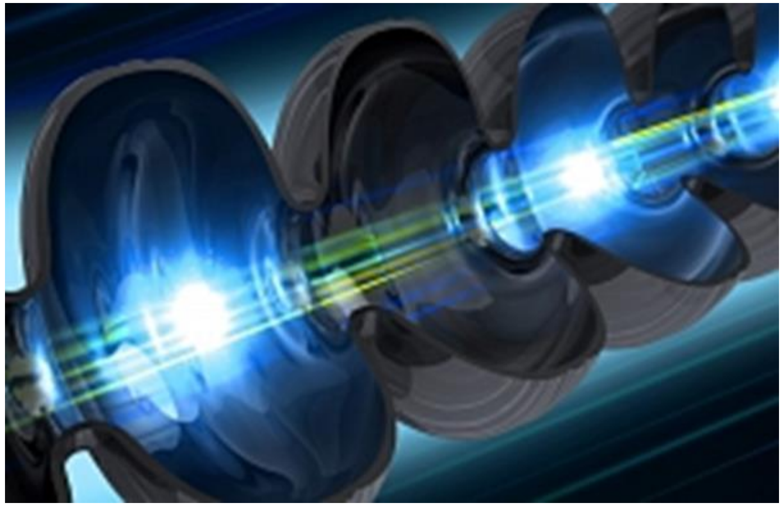
Shaping the future of the European XFEL:Options for SASE4/5 tunnels

Optimized Soft X-Ray beamline to Investigate Dynamics

Nora Berrah, Physics Department, University of Connecticut





European XFEL

LCLS-II

Work Funded by DoE-BES

Outline: Making Molecular Movies of:

- Non-Linear Physics in large systems
- Time-resolved charge transfer dynamics in molecule driven by x-ray induced nuclear motion
- Time-resolved resonant Auger-driven Interatomic Coulombic Decay (ICD)
- Electronic Dynamics: Attosecond Electron migration in Molecules using Non-Linear Multidimensional Methodologies

What is needed and Why?

Required Capabilities for a Soft X-ray (SXR) Beamline

- Chemistry, Atto-chemistry and AMO physics need a Soft X-ray Beamline **optimized** to access the C, N and O K-edges; starting at 270 eV.

- **Attosecond** Capability will Push Further Ultrafast Science.

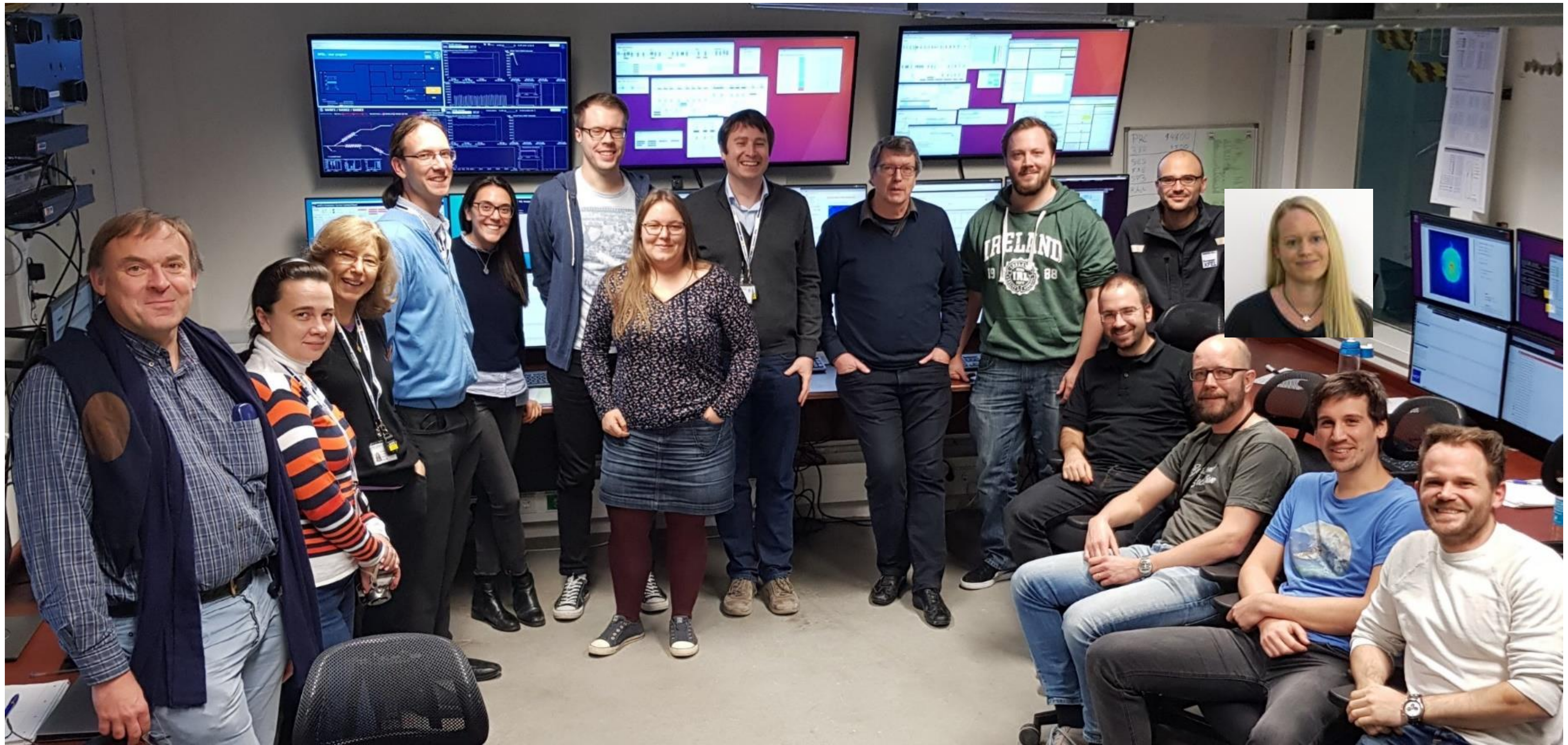
- **Self-seeding** or **Laser-seeding (***a la* FERMI) will allow for resonance-based experiments & time-resolved high resolution spectroscopy.

- **Pump-probe,** 2 or more colors, is indispensable.

Required Capabilities for a Soft X-ray (SXR) Beamline

- **Full polarization control:** linear, circular, elliptical

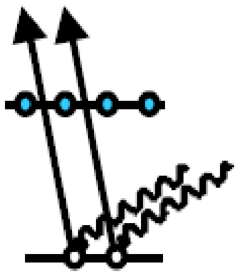
- All AQS instruments in a switch-yard configuration



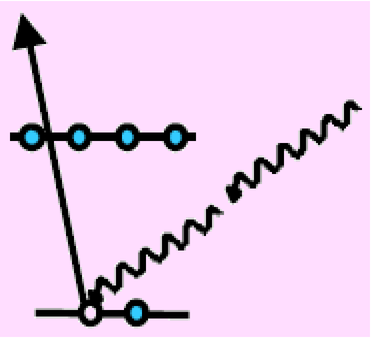
Exellent In-House Team

Some Example of Research Underway, Planned and the DREAMS……..

- Non-Linear Physics With AQS



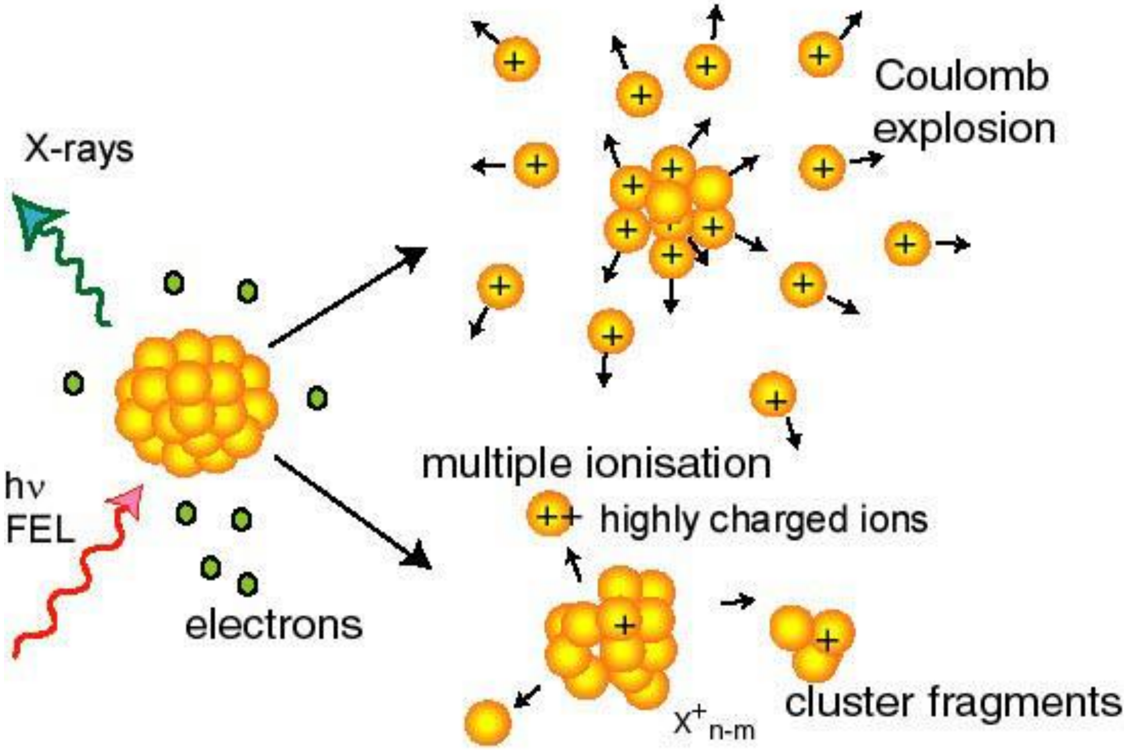
2-photons, 2-electrons



Doumy, PRL**106**, '11

- Time-resolved charge transfer dynamics in molecule driven by x-ray induced nuclear motion

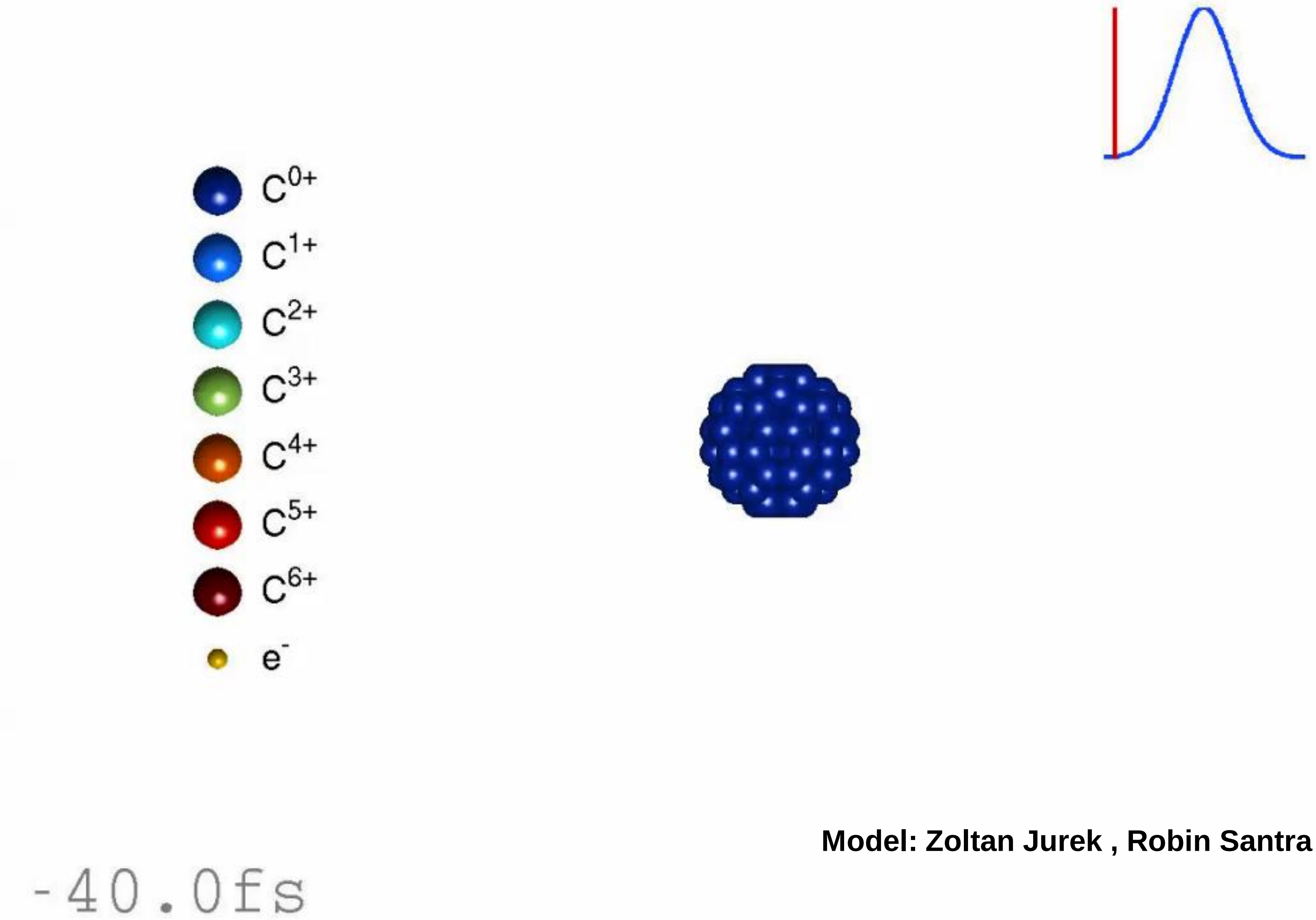
Molecular Movie:
Time-Resolved FEL-Induced Fullerene Dynamics
Complex Model System for Large bonded Molecules



Driving Fundamental Questions:

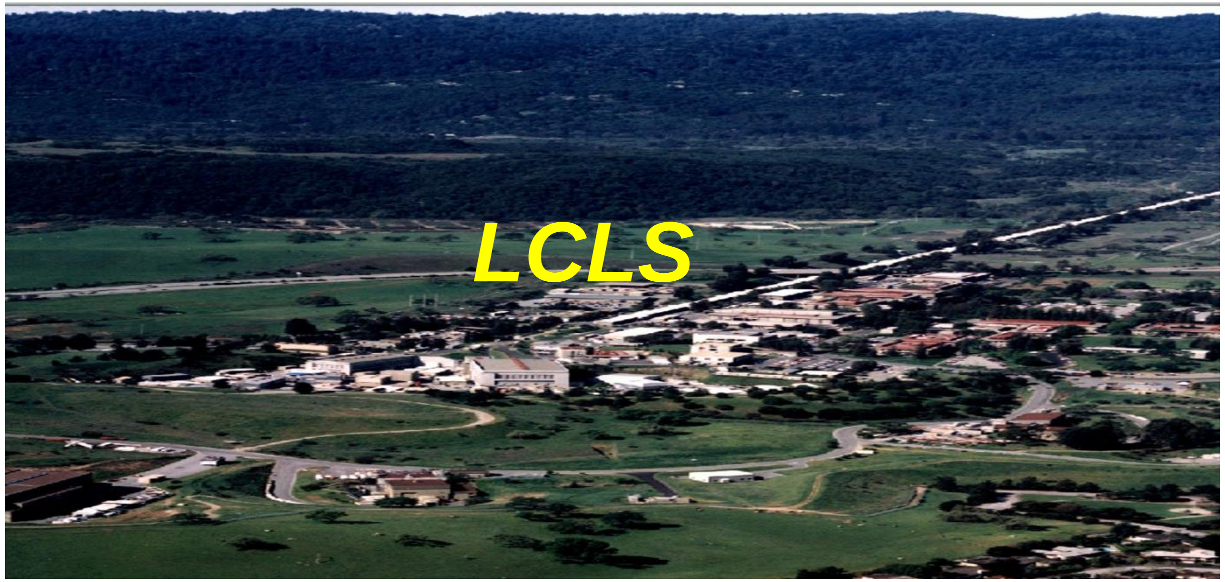
- **Can We, Quantitatively, Understand the Time-Resolved Molecular Dynamics Induced by Intense X-ray Exposure?**
- **Can We Test & Improve Time-Dependent Molecular Dynamics Models?**

Our Findings Impact: **Electronic and Radiation Damage of Bio-Molecules during Imaging with FEL X-ray Exposure.**



Model: Zoltan Jurek , Robin Santra

AMO Hutch @ LCLS

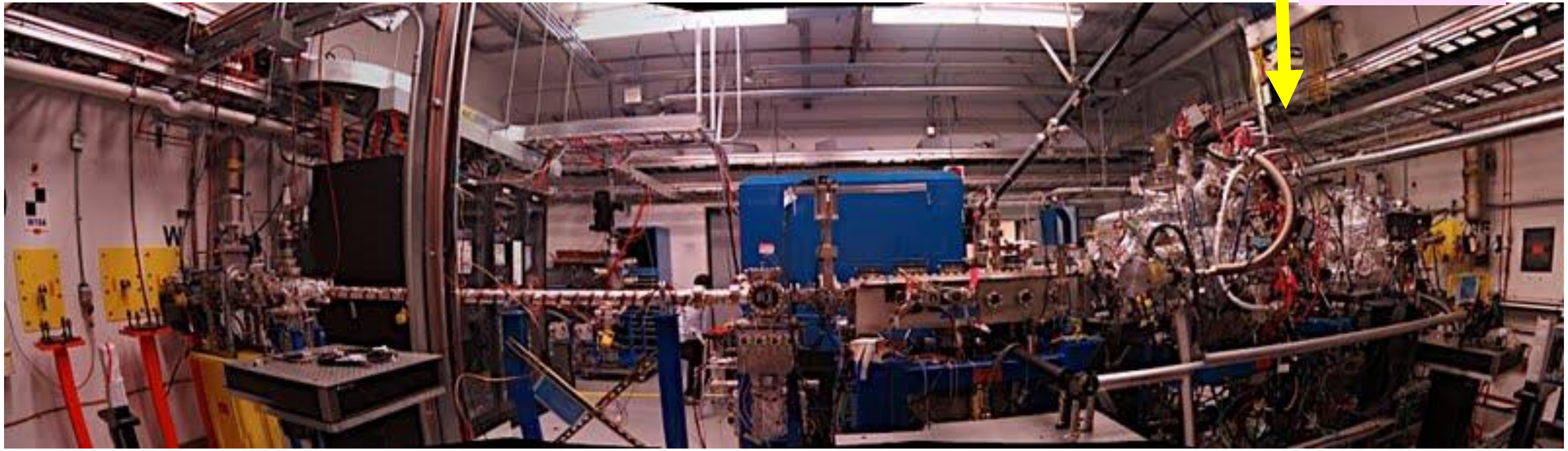


Berrah & Bucksbaum
Scientific American,
310,64,(2014);54,2015.

Bucksbaum & Berrah,
Physics Today, 68, 2015.

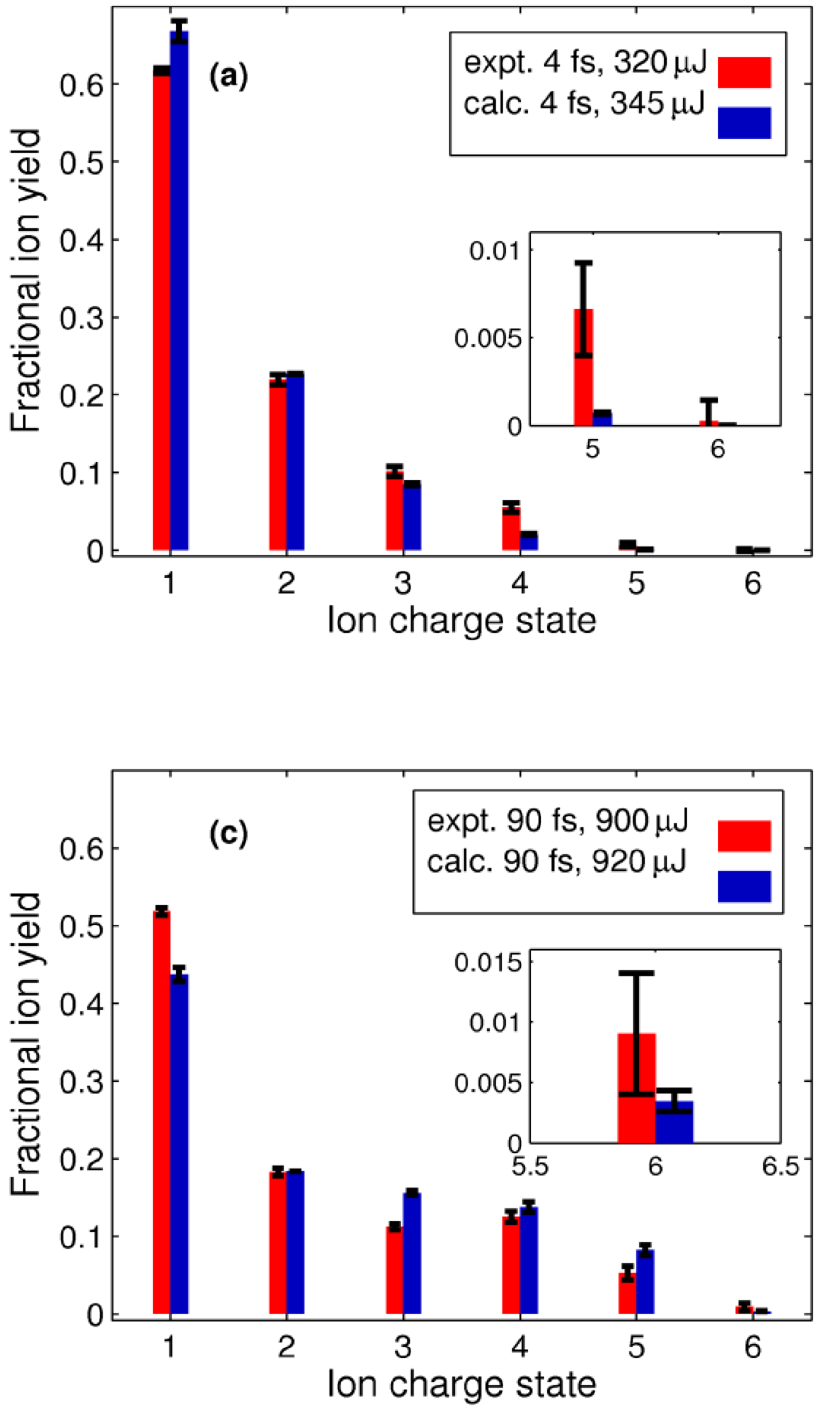
LAMP Instrument: Rev. of Sci. Instr., 89(3), 035112 (2018)

LAMP



High fluence: Comparison of C^{n+} states with model.

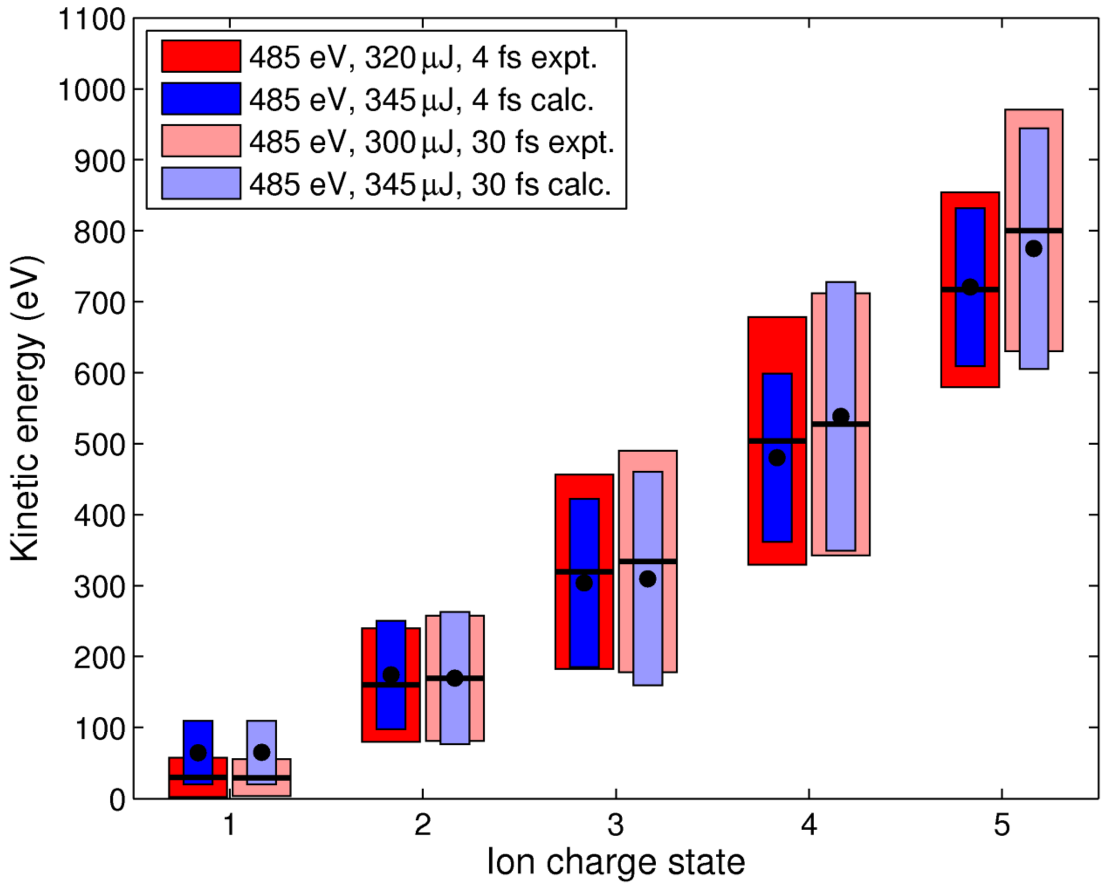
(hv=485 eV)



- ✓ **Yield for higher charge states increases when we increase the pulse duration.**
- ✓ **Model predicted initially more abundant charge states ⇨ strong recombination of electrons with ions after the pulse ends.**

Murphy et al., Nature Comm. 5, 4281, (2014)
Berrah et al., Faraday Disc., 171 (1),(2014)

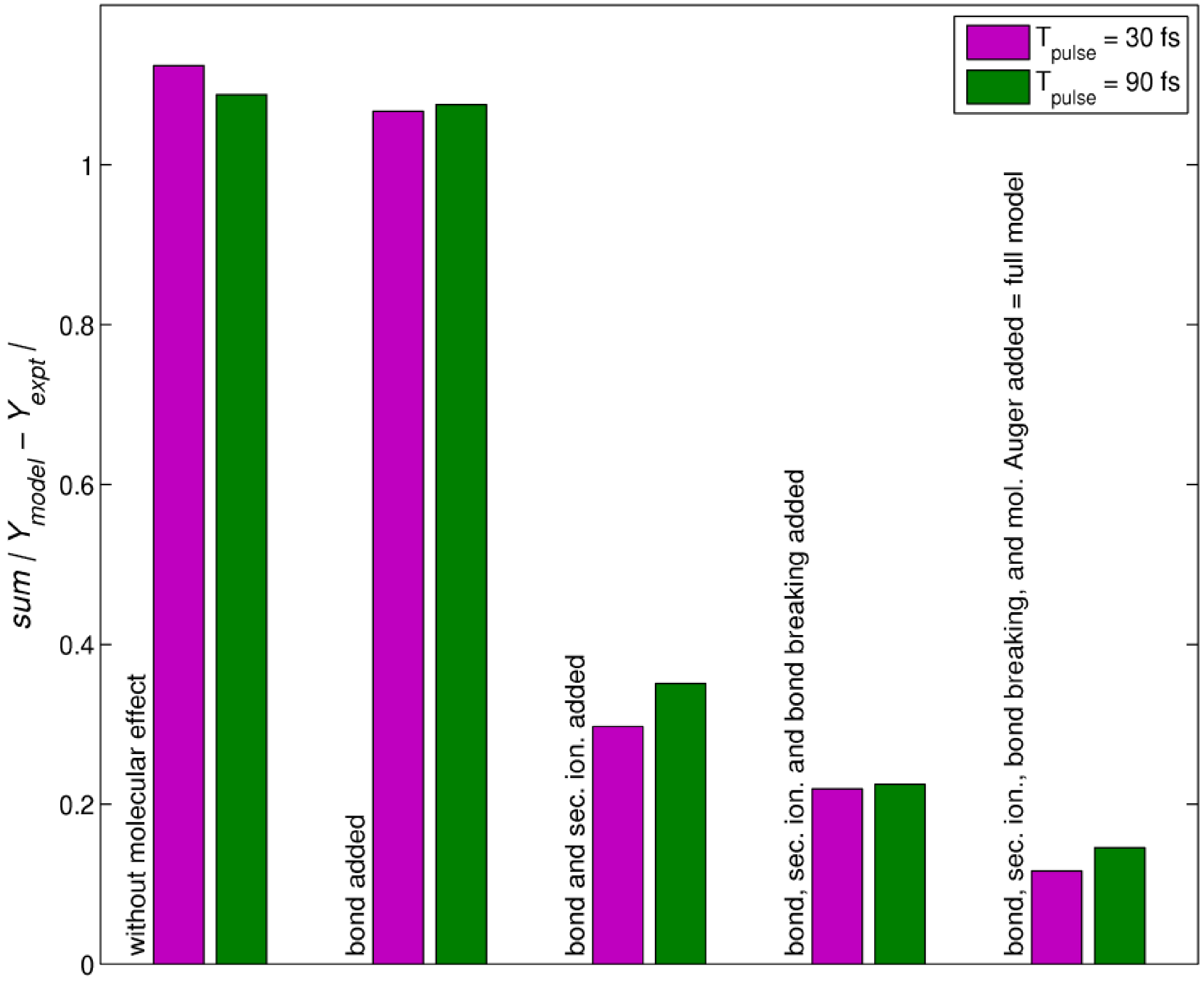
Fragment Atomic C Ion Kinetic Energy: Central high-fluence region



Quadratic Scaling of Mean Ion KE with C^{n+} Expansion is Coulombic.

Mean ion kinetic energy: **circles (experiment)** and **lines (simulation).**
RMS kinetic energy width: Height of each rectangle.

Comparison of Experimental data with Molecular Dynamic model



Validated a fundamental assumption: Charged particles behave as if they were classical particles

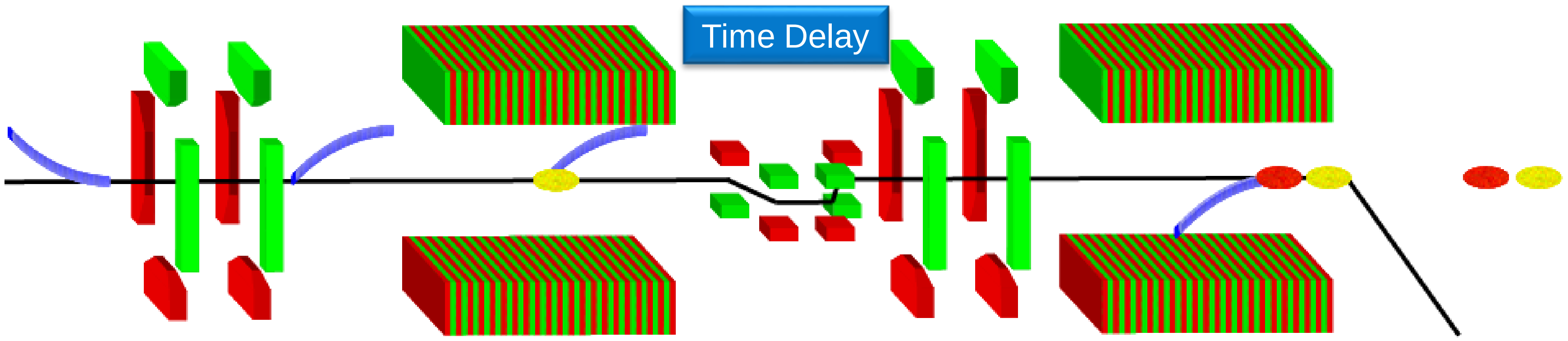
* **Significant secondary ionization by P and A e-** in C_{60} compared to Van der Waals clust.

* **Molecular influence are also strong** compared to VdW clust.

* C-C short bond length → **strong Coulomb repulsion**

Zoltan Jurek
Sang-Kil Son
Robin Santra

Pump-probe Scheme: ***Fresh-temporal slices of electron bunch***-- Orbit Selective Spoiling; Delay:25fs-925 fs

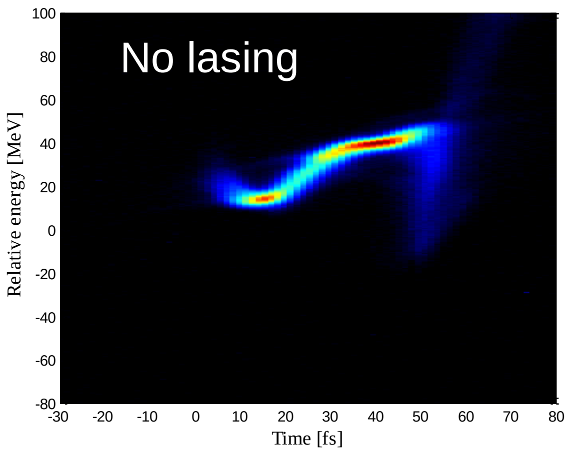


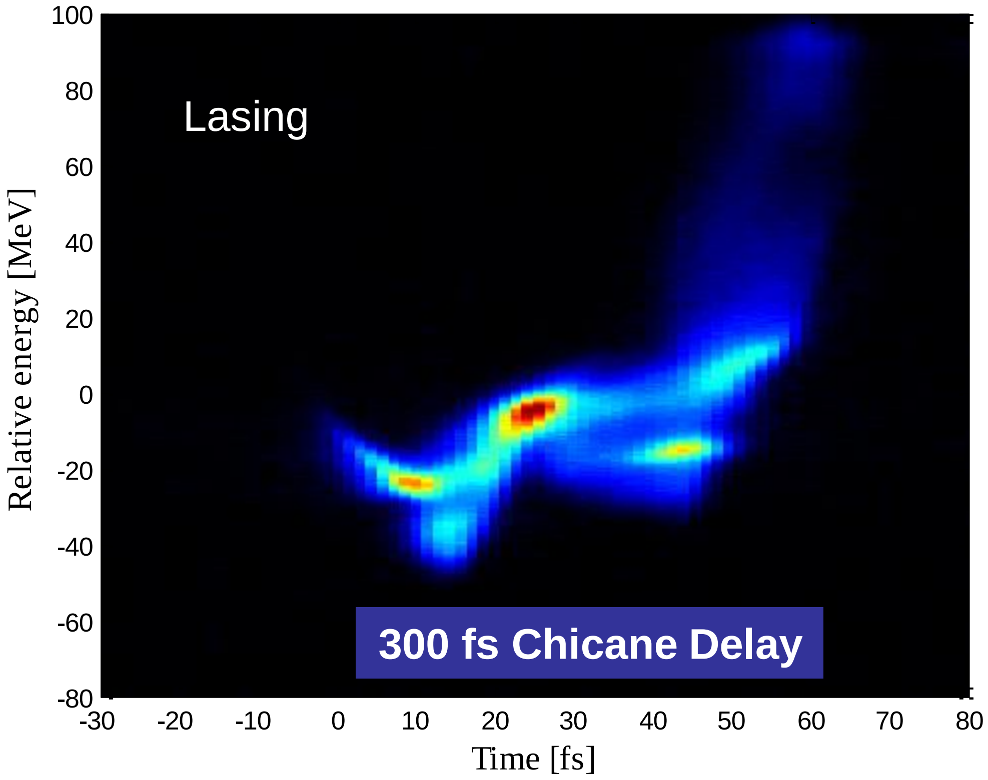
- X-Y dipoles correct orbit for the **tail** before 1st undulator to prepare it to lase
- 1st undulator section tuned at K1 **makes FEL** on **the tail** of the beam
- Chicane can **delay** the electrons with respect to the first beam
- X-Y dipoles after 1st undulator section correct the orbit for **the head**
- 2nd undulator section tuned at K2 **makes FEL** on **the fresh head** of the beam

✓ Allows true zero delay
✓ Simple delay scans (chicane only)
✓ Simple color scans (undulator K)

A. Lutman , T. Maxwell, J.MacArthur, M. Guetg, N. Berrah, R. Coffee Y. Ding, Z. Huang, A. Marinelli, S. Moeller, and J. Zemella, Nature Photonics 10, 745 (2016)

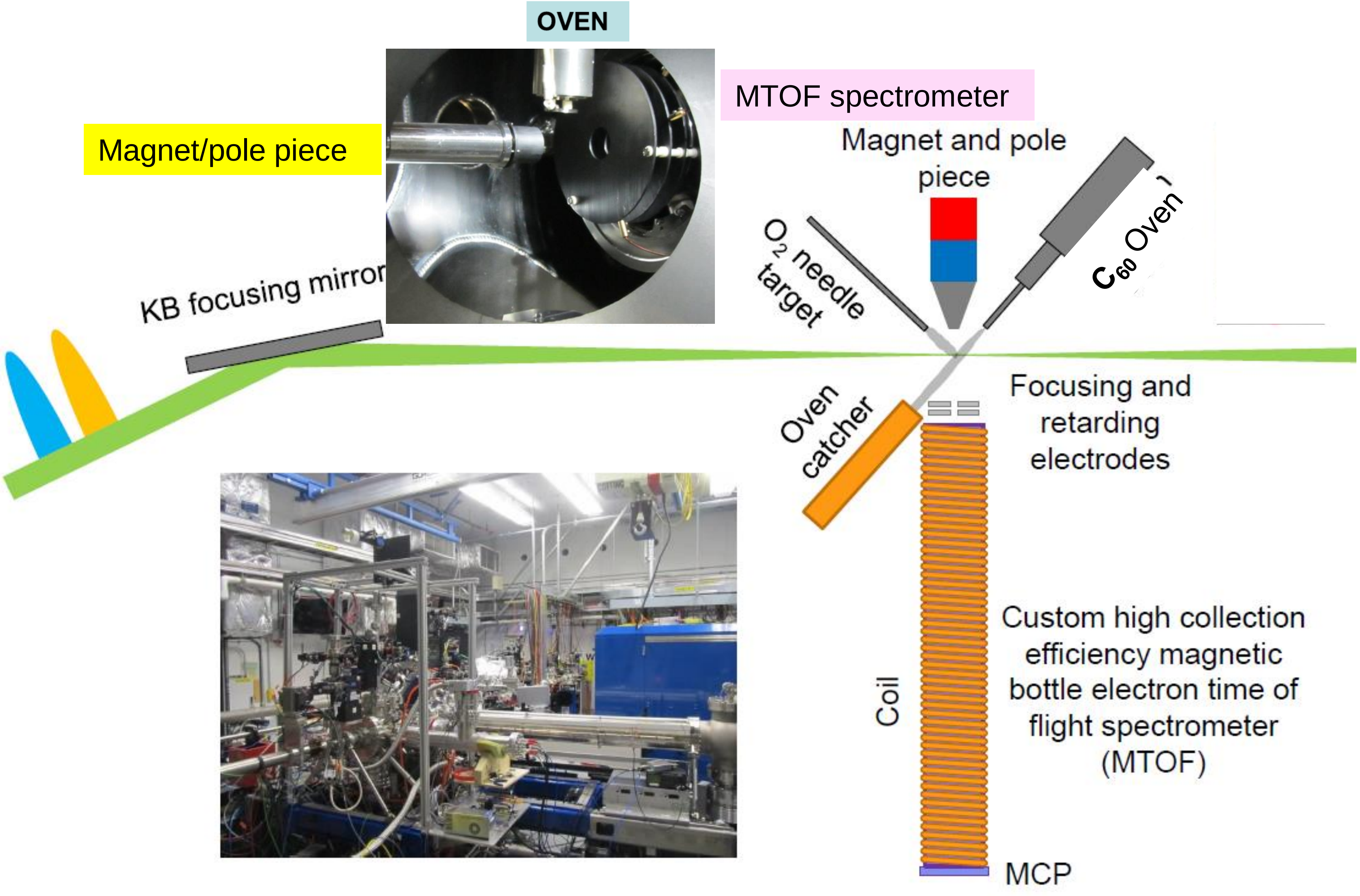
Fresh-Slice Mode: Electron-Bunch Head-Tail Lasing for X-ray Pump-Probe Technique





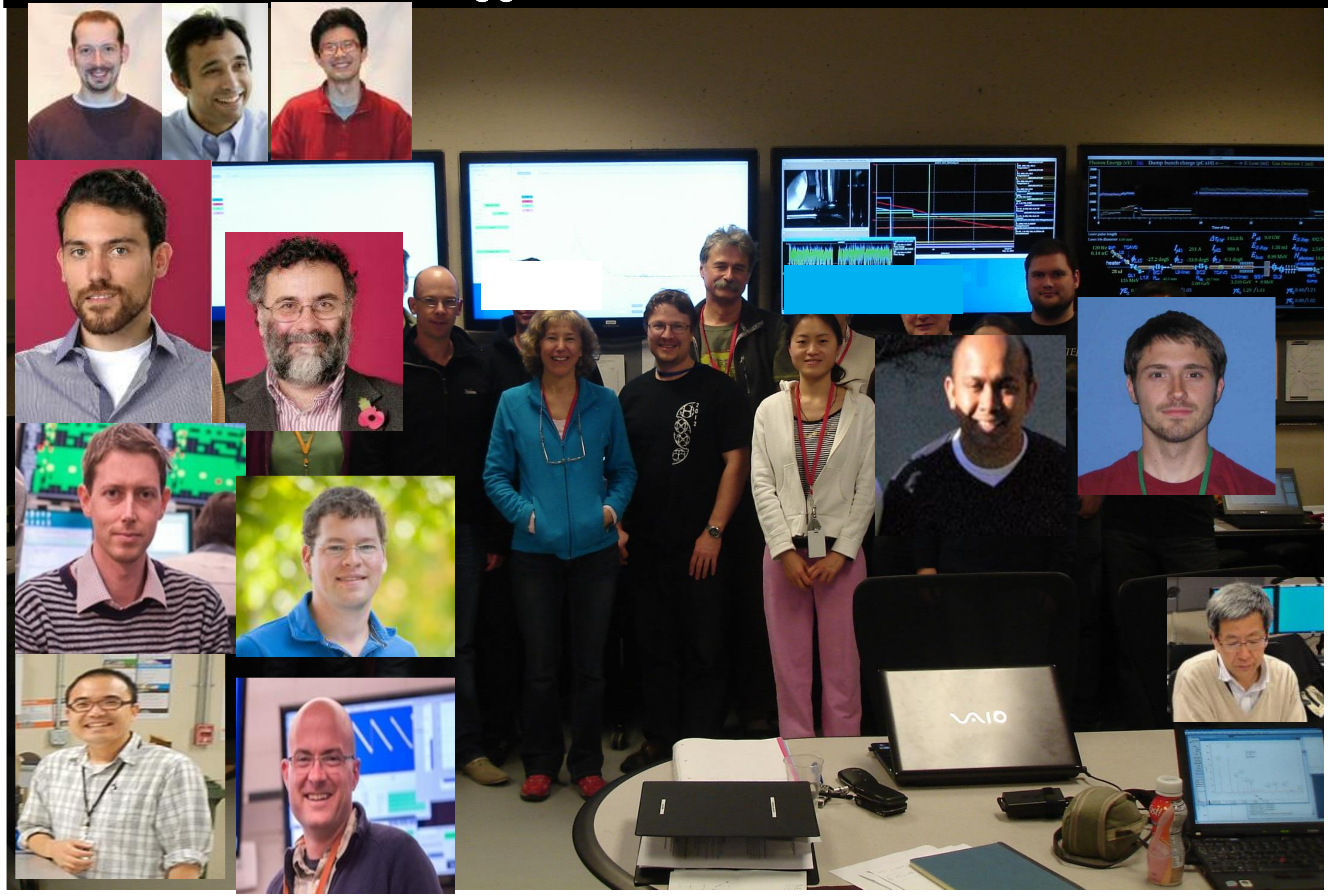
Electron bunch of 30fs generates two x-ray pulses of 10-20 fs

A. Lutman , T. Maxwell, J.MacArthur, M. Guetg, N. Berrah, R. Coffee Y. Ding, Z. Huang, A. Marinelli, S. Moeller, and J. Zemella, Nature Photonics 10, 745 (2016)

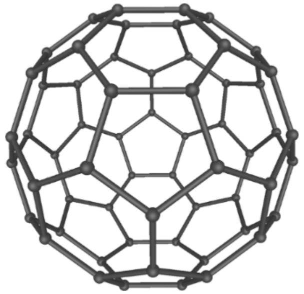


Osipov, Bostedt, Ferguson,Obaid,Rolles, Rudenko, Bozek, Berrah, Rev. Sci. Instr, **89**, 035112 (2018)

Collaboration: C_{60} Time-Resolved Experiment



Collaboration: Time-Resolved C_{60} Investigation



UConn, USA: Razib Obaid, Hui Xiong, Nora Berrah

CFEL-Hamburg, Germany: Zoltan Jurek, Sang- Kil Son, Robin Santra

LCLS-SLAC, USA: Timur Osipov, Alberto Lutman, R. Coffee, D. Ray, S. Moeller

Imperial College, London, UK: Alvaro Sanchez Gonzalez, Thomas Barillot, Jon Marangos, Leszek Frasinski.

Goteborg University, Sweden: Richard Squibb, Raimund Feifel

University of Texas, Austin, USA: Li Fang

PULSE, SLAC, USA: Thomas Wolf, James Cryan, Philip Bucksbaum,

SOLEIL, France: John Bozek

Kansas State University, USA: Daniel Rolles, S. Augustin

Sendai University, Japan: Hironobu Fukuzawa, Koji Motomura, Kyoshi Ueda & Kono group
University Potsdam, Berlin, Germany: Mario Niebuhr,

MPI, Heidelberg, Germany: Kirsten Schnorr, Thomas Pfeifer

MBI, Berlin Germany: Claus-Peter Schulz

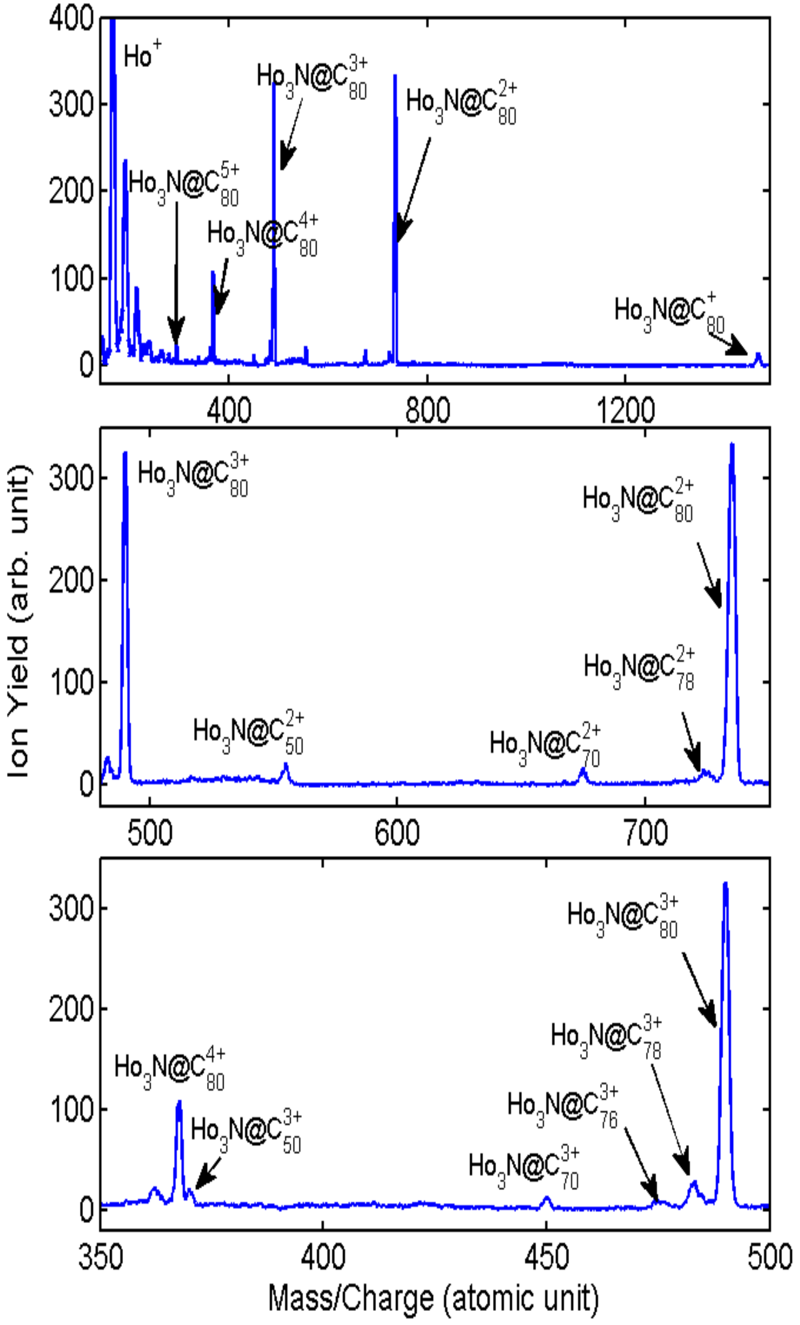
WE Could Do Much More! Using the Ultra High XFEL Pulse Energy

Saturation parameters projected for SASE5 @ E = 17.5 GeV

Radiation wavelength	nm	0.5	1	2	6
Pulse energy	mJ	8.15	11.4	14.0	17.4

We could use beam parameters corresponding to Ultra High Fluence conditions that mimic the degree of ionization encountered in hard-X-ray imaging experiments; SFX. Even for single molecule imaging

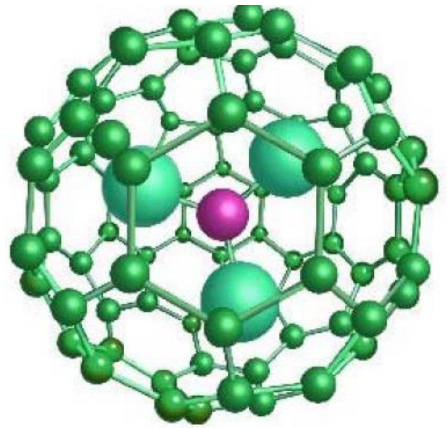
Using X-ray pump-X-ray probe spectroscopy, we could observed new fundamental physical phenomena,



We can also study:

Multiphoton X-Ray Induced Fragmentation of $Ho_3N@C_{80}$

Proof of principle @ LCLS

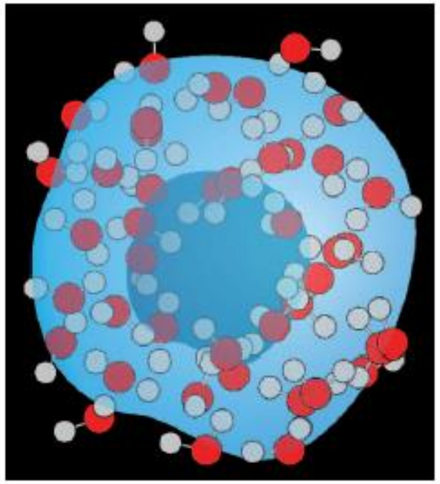


Site Specific Ho 3d ionization hv=1530 eV, 80 fs, 2.2 mJ ~ 6.7 10^{18} ph/cm^2.

Photoionization cross section at 1530 eV: Ho(3d)=1.2Mb; C= 0.013 Mb;

N. Berrah et al., J. of Mod. Opt., 63, 390 (2015)

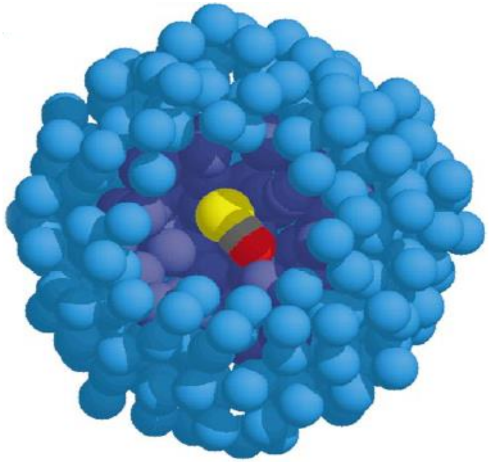
We Can Also Study: Intermolecular ionization dynamics in weakly-bound systems



- Real time electron solvation dynamics in water clusters

- Intermolecular decay mechanisms in doped helium nanodroplets

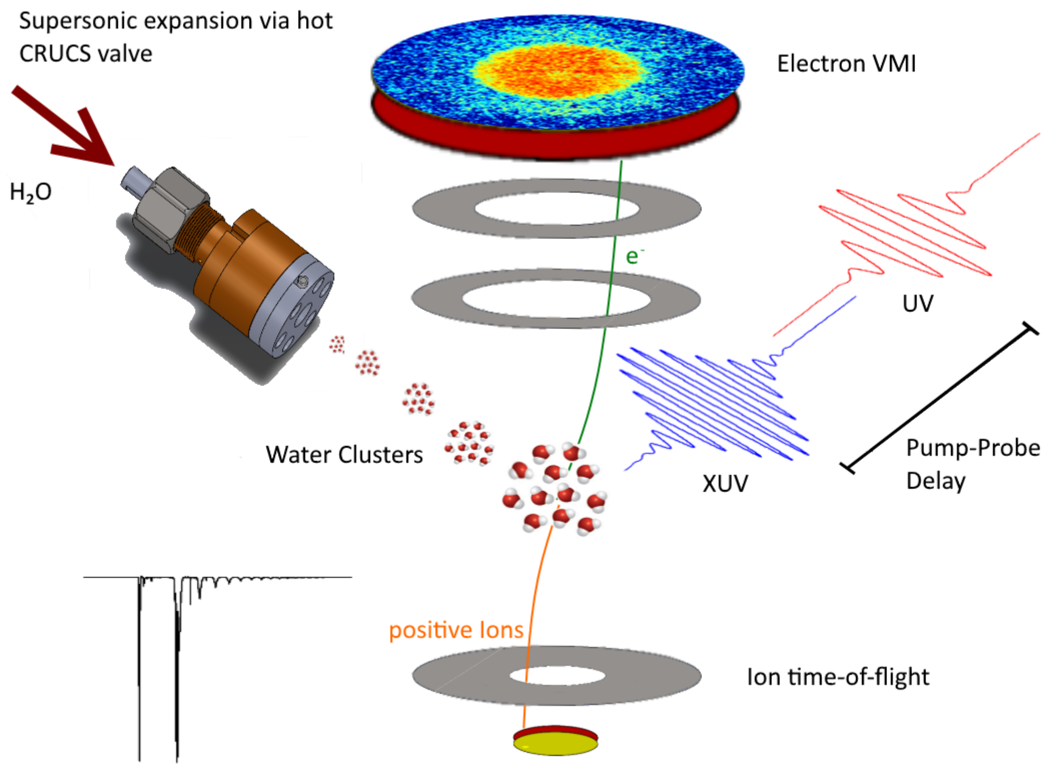
- Doped clusters/liquid water droplets
  - Solvation triggered by excitation/ionization of dopant followed by charge transfer to the solvent



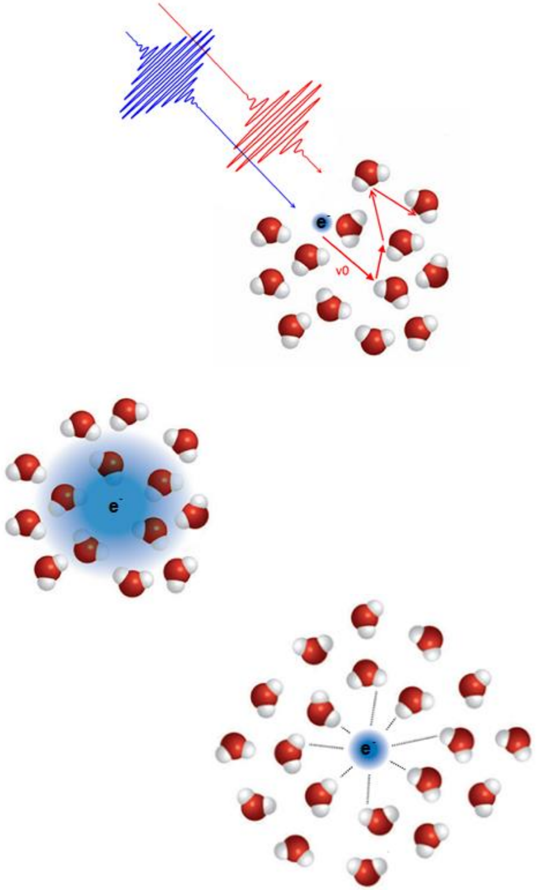
- He nanodroplet spectroscopy

  - Nearly transparent in the x-ray regime (0.019 MB -0.0009 MB)
  - Molecules rotationally/vibrationally cooled to the ground state (0.4 K)
  - molecular complexes difficult to form in the gas phase (specialized donor-acceptor systems)

We Could Study with X-rays: ……….Experiments@ FEMI LDM endstation



- **Pure water vapor expansion**
- **T_{H20}: 100 – 220 °C**
- **Cluster size: 500 – 4000**
- **$T_{cluster}$: ≈ 150K**



Schematic

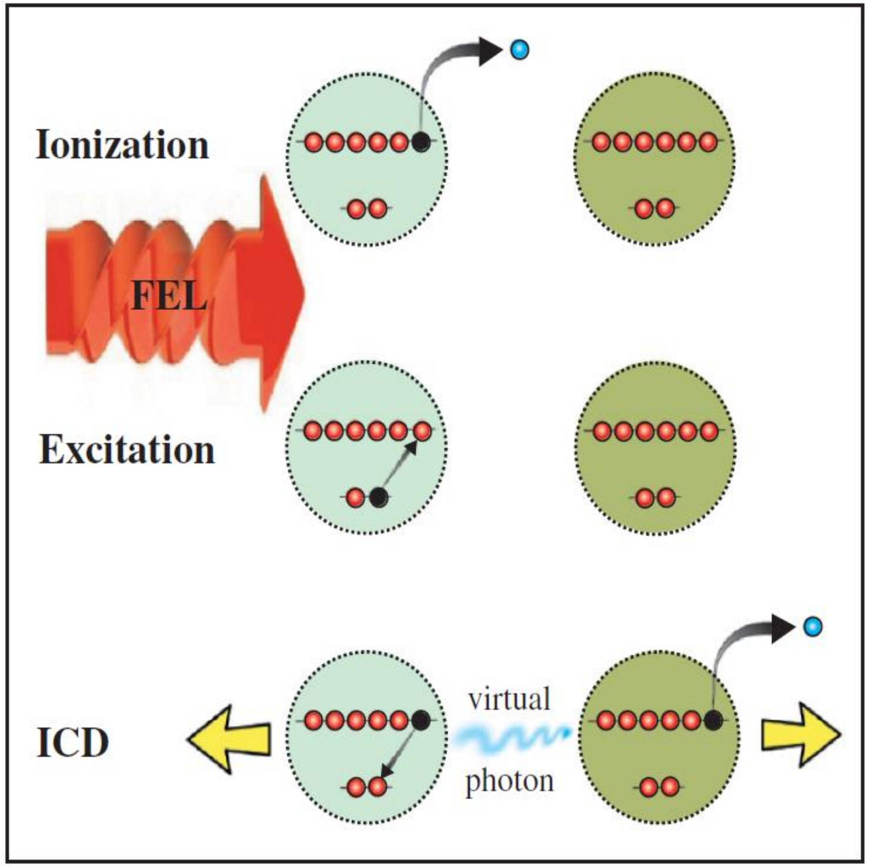
1. **Electron ionized by XUV pulse**
2. **'hot' electron is scattered within the cluster**
3. **Electron is trapped within the conduction band (100s fs)**
4. **Neighboring molecules rearrange forming a solvated state (ps)**

A. C. LaForge et al., Nature Physics 15, 247 (2019)
A. C. LaForge et al., Phys. Rev. Lett. (2019) (in press)
A. C. LaForge et al., Phys. Rev. Lett. 116, 203001 (2016)

Time-Resolved Resonant Auger-Driven Interatomic Coulombic Decay (ICD)

- ICD is an ultrafast decay mechanism that occurs when local electronic decay is energetically forbidden. **It has an ultrafast decay path, fs and sub-fs timescales, where energy is exchanged with a neighboring atom leading to its ionization.**

- Weakly-bound complexes offer an environment in which locally excited electrons can interact with neighboring molecules, leading to new *intermolecular* interaction mechanisms.

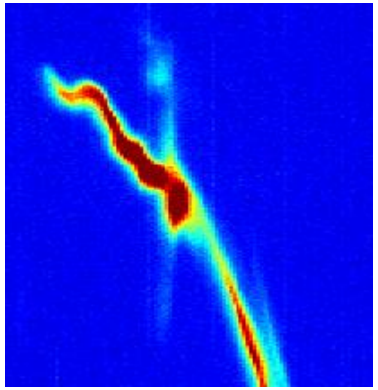


Impact:

- Long-range correlation effects can be obtained via ICD which impacts solution-phase actinide chemistry.

- ICD produces low energy electrons → damage DNA and proteins.

Exciting Future for Attosecond Dynamics with FELs!

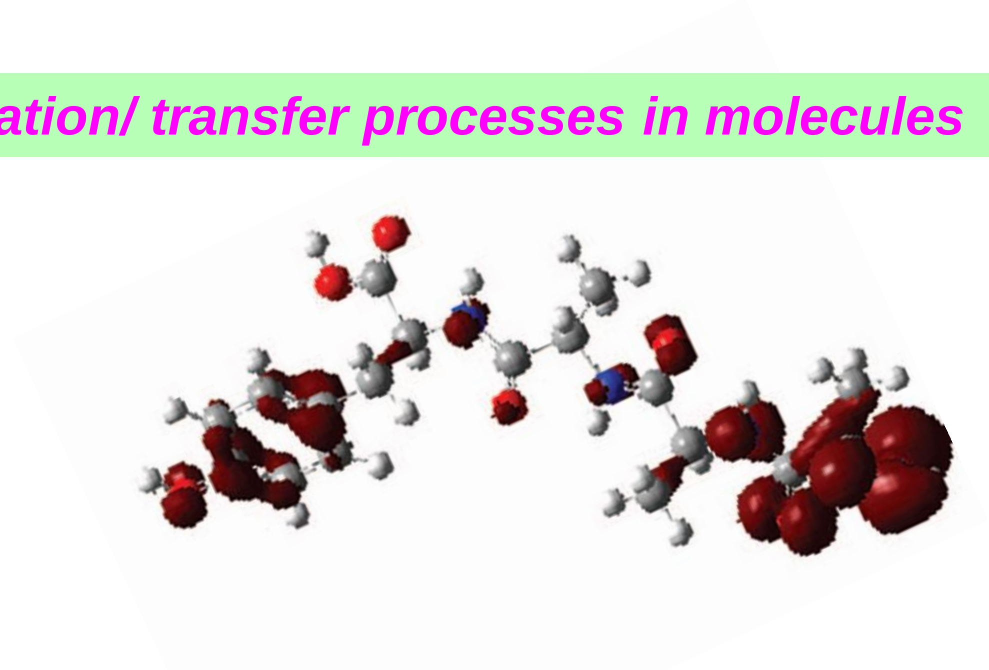


Attosecond Mode

500 as @LCLS in 2018!

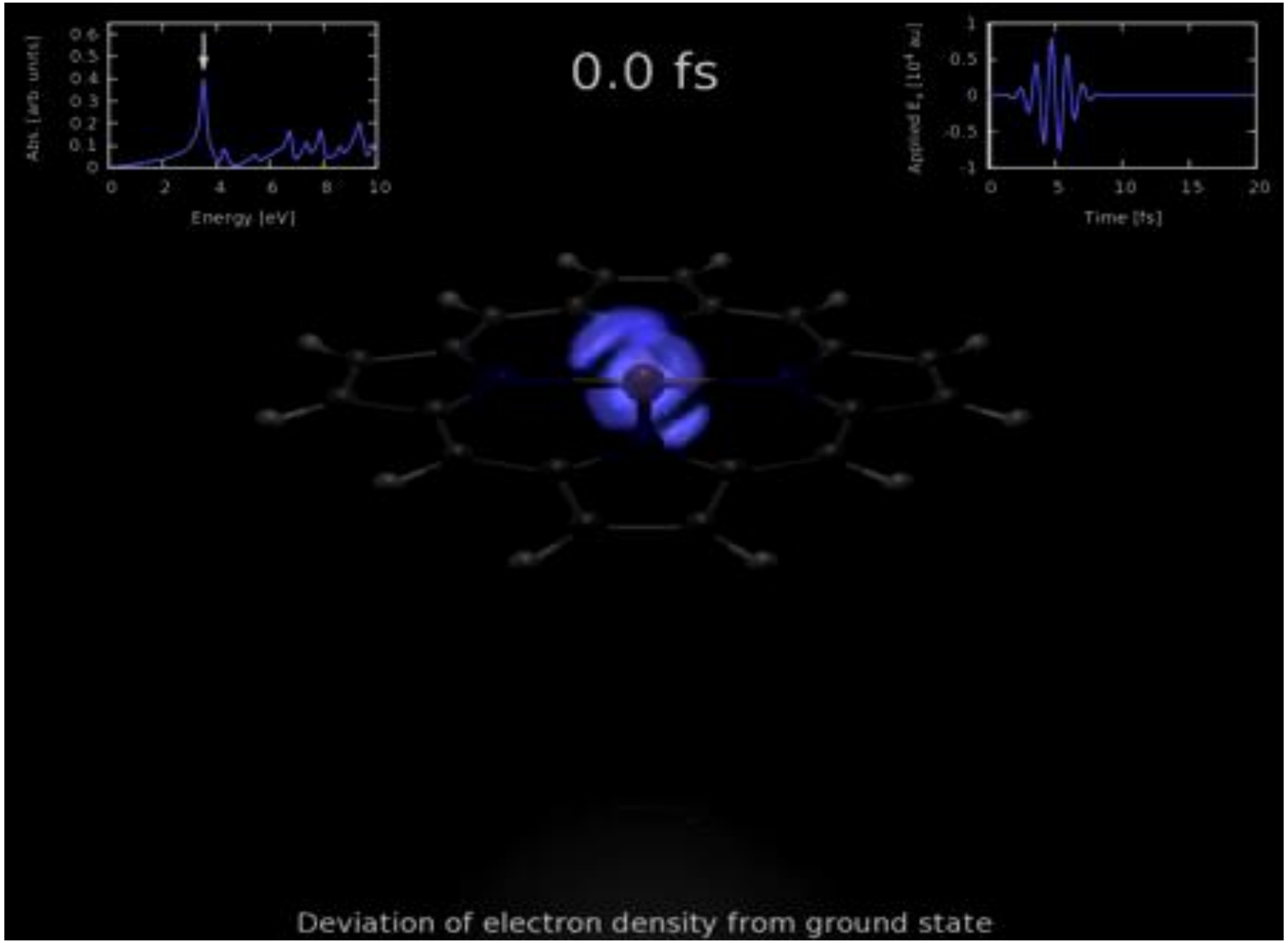
S. Huang et al., "Generating Single-Spike Hard X-Ray Pulses with Nonlinear Bunch Compression in FEL", **PRL119, 154801 (2017)**

Charge migration/ transfer processes in molecules

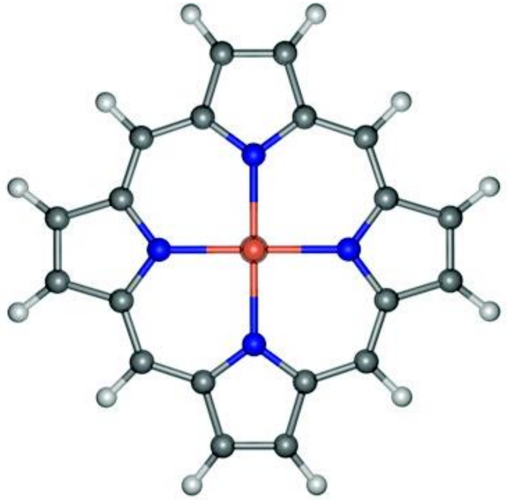


Molecular Movie: Can we track the evolution of electrons on their natural time scales ?

Electrons motion is very important in all fields of physics/ chemistry; their timescale is attosecond



Resonant excitation



Zinc porphyrin

Environmental Molecular Sciences Laboratory (EMSL) @ PNNL: https://www.youtube.com/watch?v=ZYsktRlhMOg J. Chem. Theory Comput. **7**, 1344–1355 (2011)

FIN