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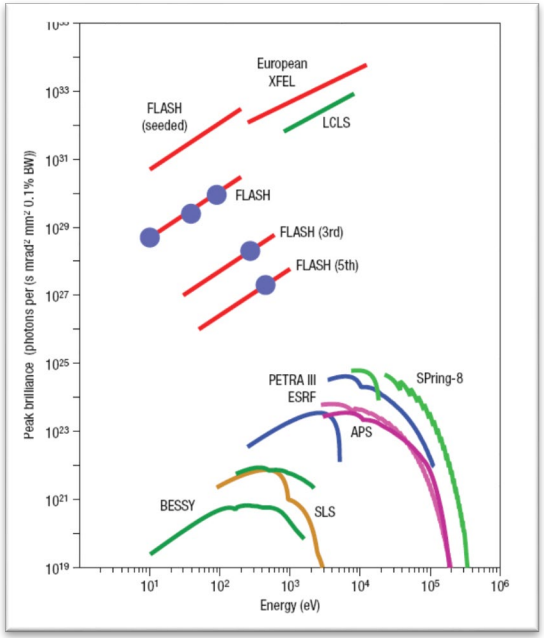
Short Free-electron Laser Pulses: The User Perspective

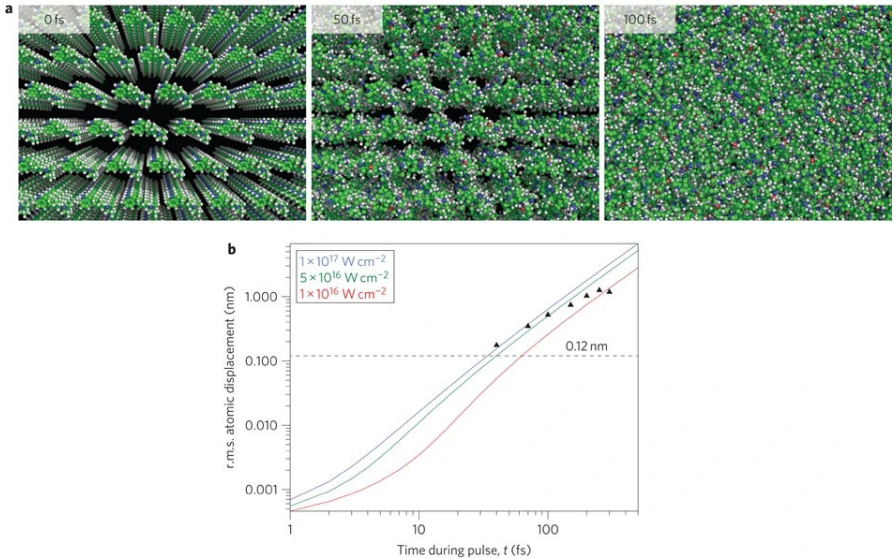
Future Light Source Workshop, Lucerne, 31.08.2023

The figure of merrit in large parts of the XFEL community is pulse energy



Brian WJ McNeil and Neil R Thompson "X-ray free-electron lasers" Nature photonics, 4(12):814-821, 2010.

«Self-terminating diffraction» during Coulomb explosion solves the problem

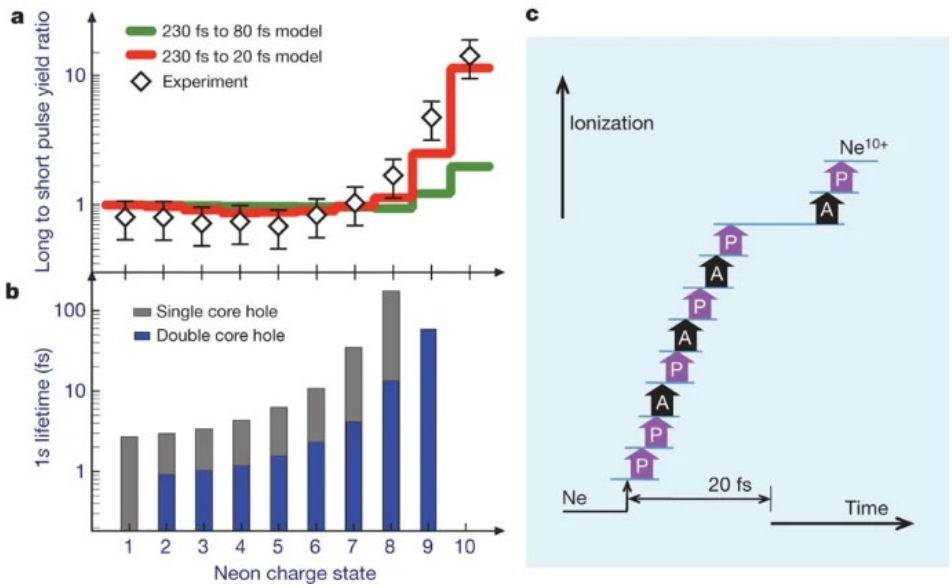


A. Barty et al., "Self-terminating diffraction gates femtosecond X-ray nanocrystallography measurements" Nature photonics, 6:35, 2012.

But I do believe that pulse length is an at least equally important parameter!

Argument 1: Core vacancy decay time

- Already first experiments confirmed strong dependence of ionization dynamics on pulse lengths
- Creation of (multiple) core vacancies can lead to x-ray induced transparancy
- Pulse length (and energy) determines number of ionisation/decay cycles
- Core vacancy lifetimes few femto- to attoseconds

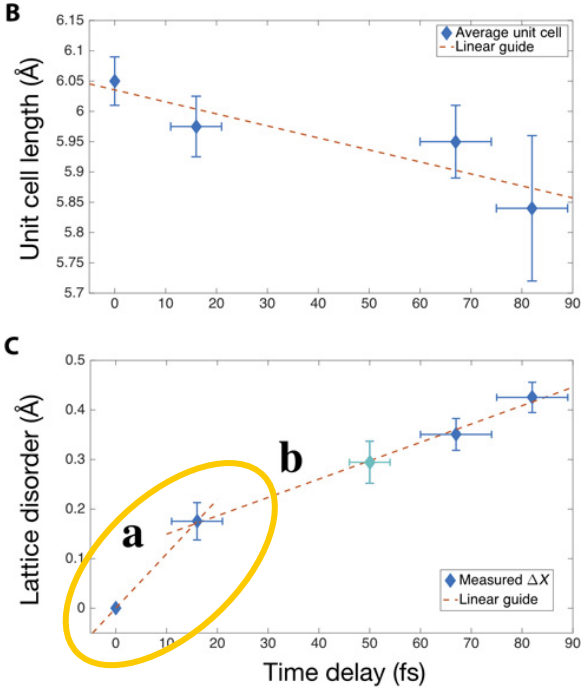


Young et al., Femtosecond electronic response of atoms to ultra-intense X-rays, Nature 466, 56 (2010)

Natural clock for X-ray induced processes is on the few femto to attosecond scale

Argument 2: Electron delocalization

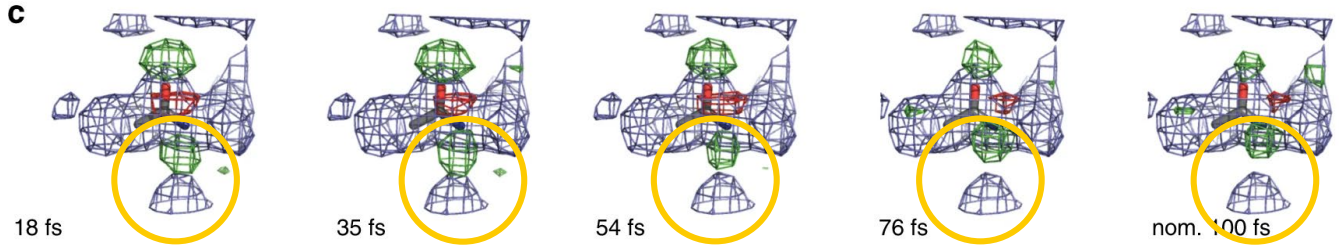
- This is from an X-ray pump / X-ray probe experiment
- Diffraction system on simple xenon nanocrystals
- Overall interpretation within the Debye-Scherrer formalism, relating Bragg intensities to lattice order
- Particularly strong change within first femtoseconds, interpretation as electron excitation and delocalization, leading to immediate reduction of Bragg intensities



K. Ferguson et al., "Transient lattice contraction in the solid to plasma transition" Sci. Advances 2, e1500837(2016) .

X-ray induced electron delocalization has immediate impact on scattering response; increasing for increasing number of decay cycles

Argument 3: Real damage in SFX (bio exps.)



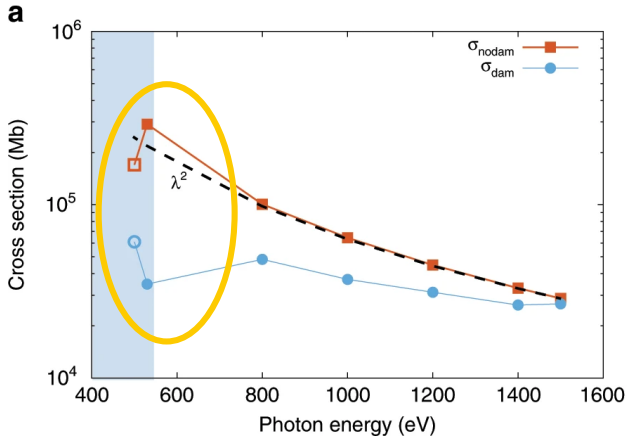
K Nass et al., "Structural dynamics in proteins induced by and probed with X-ray free-electron laser pulses", Nature Comm. 11, 1814 (2020).

- Also hier X-ray pump / X-ray probe study
- Clearly identified changes in reconstructed electron density distribution for longer delays
- Local damage processes specifically relevant around heavy atoms

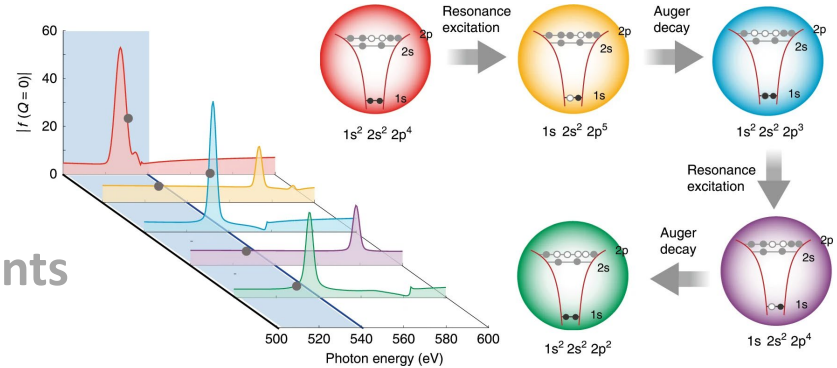
X-ray induced ionization processes («damage») play a real role in real samples

Argument 4: Damage (and opportunities) in imaging

- Combined experimental and theoretical study with full electronic structure calculations
- Initial experimental design: max pulse energy
- Strong loss of signal (cross section) towards resonances due to electron excitation and dynamics
- For short pulses: opportunites for resonant imaging with transient states



P.J. Ho et al., "The role of transient resonances for ultrafast imaging", Nature Comm. 11, 167 (2020).

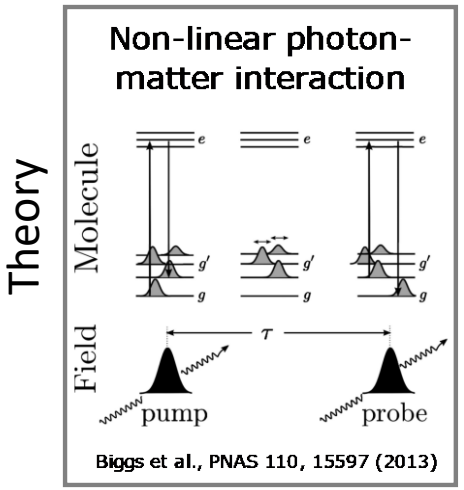


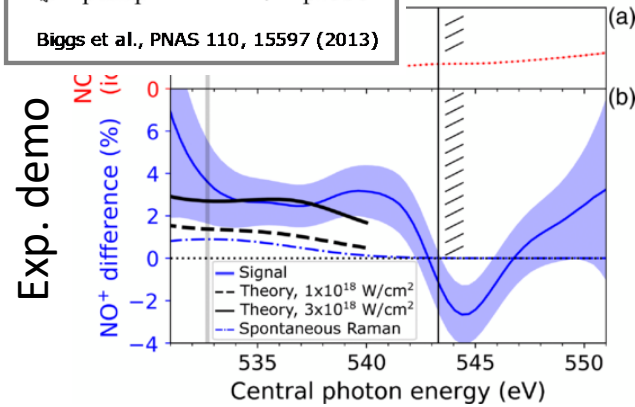
X-ray induced electron dynamics also fundamentally important in imaging experiments

New frontier: x-ray attosecond pulses

- Impulsive stimulated X-Ray Raman (Electronic Raman) processes allow element specific probing of local electronic density of states.
- Promising route to study charge motion in molecules.
- Need broad bandwidth short pulses: bandwidth needs to cover electronic states, pulse needs to be shorter than Auger lifetime
- First demonstrated with XLEAP pulses at LCLS

Coherent attosecond pulses open door for truly non-linear spectroscopy with core electrons.
Note: we need short (below Auger lifetime) and large bandwidth (covering valence dos)

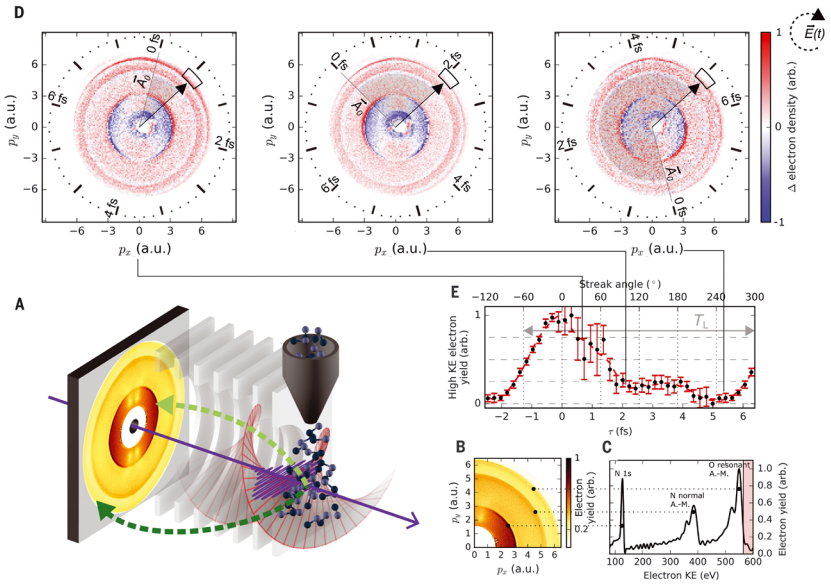




J.T. O'Neil et al., "Electronic Population Transfer via Impulsive Stimulated X-Ray Raman Scattering with Attosecond Soft-X-Ray Pulses", Phys. Rev. Lett. 125, 073203 (2020).

New frontier: x-ray attosecond pulses

- Angular streaking experiment to probe electronic coherences
- Use attosecond soft x-ray pulses to excite coherent superposition of core-excited states
- Map time dependent current of the Auger Meitner decay
- Demonstrates the ability to use XFEL pulses for exploring electronic coherences during charge transfer processes



Experimental techniques have matured to apply attosecond pulses for current questions in (chemical) research

S. Liet al., “Attosecond coherent electron motion in Auger-Meitner decay”, Science 375, 285 (2022).

Notes and Conclusion

- Notes
  - The presented selection is a personal view and there are many more facets to the discussions.
  - A short pulse is always to be considered with respect to the required bandwidth (resolution). High-resolution spectroscopy will require longer short pulses

- Conclusions
  - Pulse length is a very important parameter and the natural reference clock is the core vacancy (in SXR mostly Auger-Meitner) lifetime.
  - Important parameter for many established applications should be «photons per femtosecond» and pulse length should be few to few tens of femtoseconds.
  - Attosecond generation and application provide unique opportunities at the time-resolution frontier and for truly non-linear X-ray spectroscopy.

- Famous last words
  - Two X-ray pulses are always better than one – for pump/probe applications.

Questions

I acknowledge my wonderful colleagues at SwissFEL and in the worldwide XFEL community. I am grateful for many discussions, insights, exchanges, and collaborations around the globe.