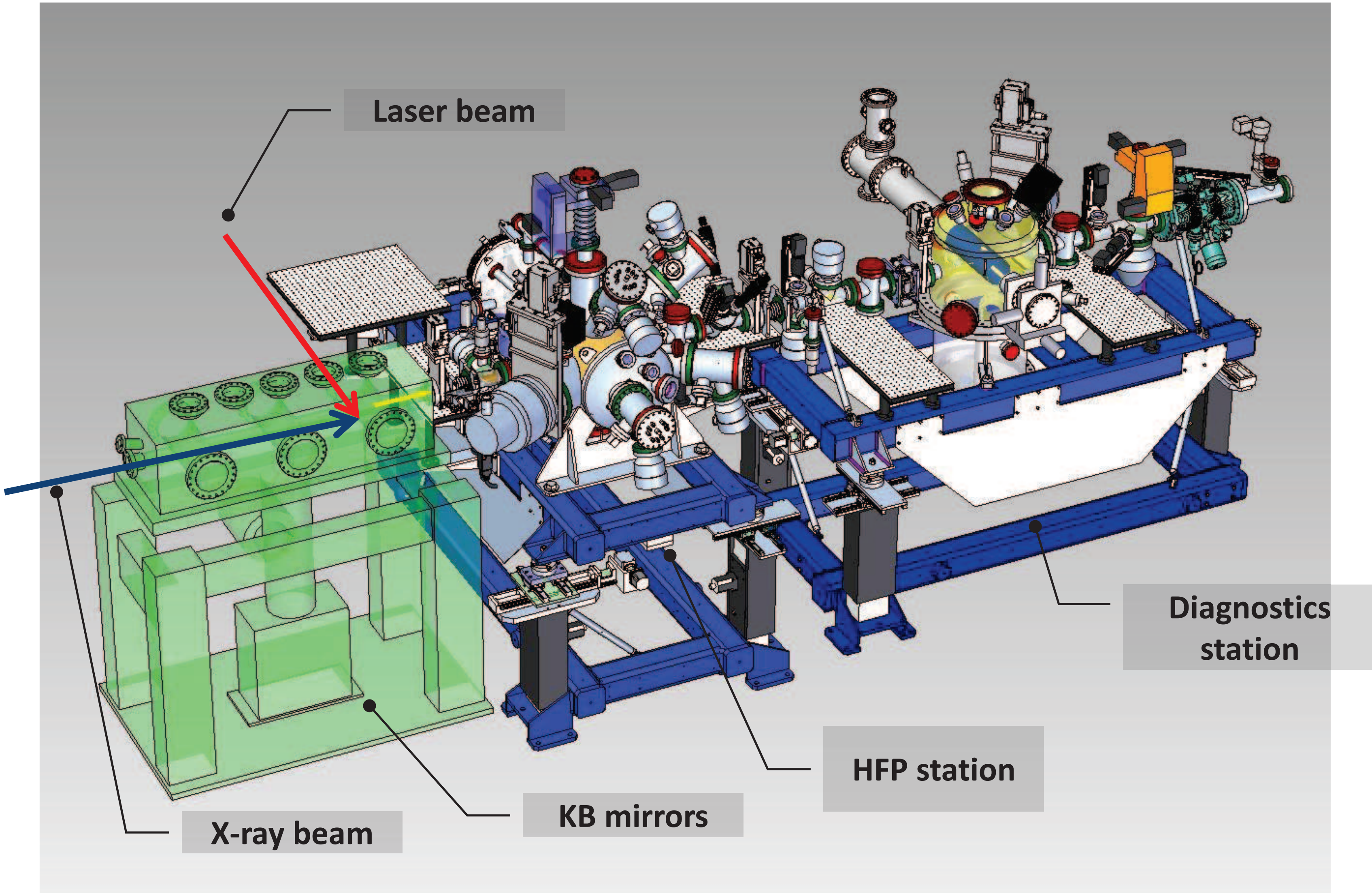
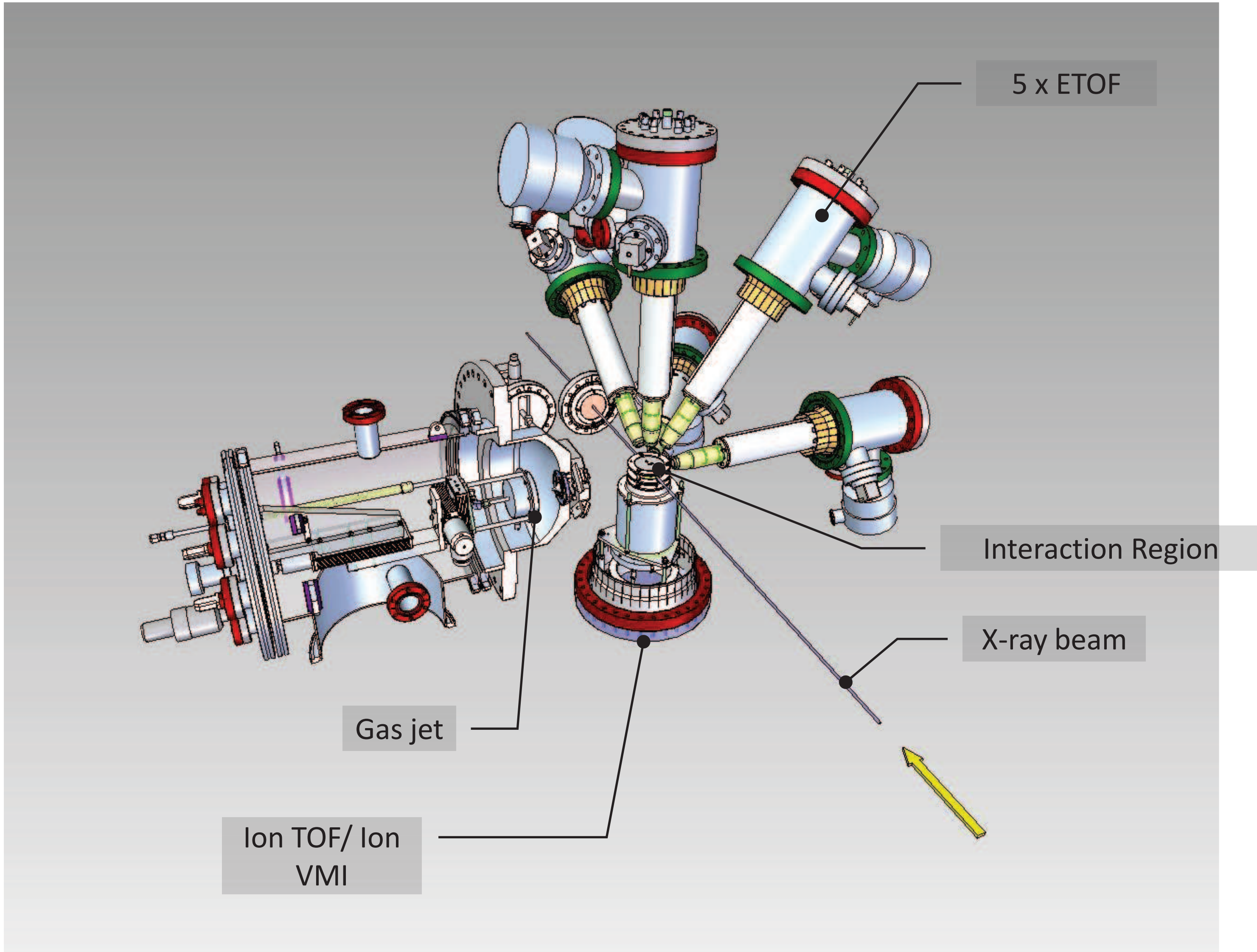
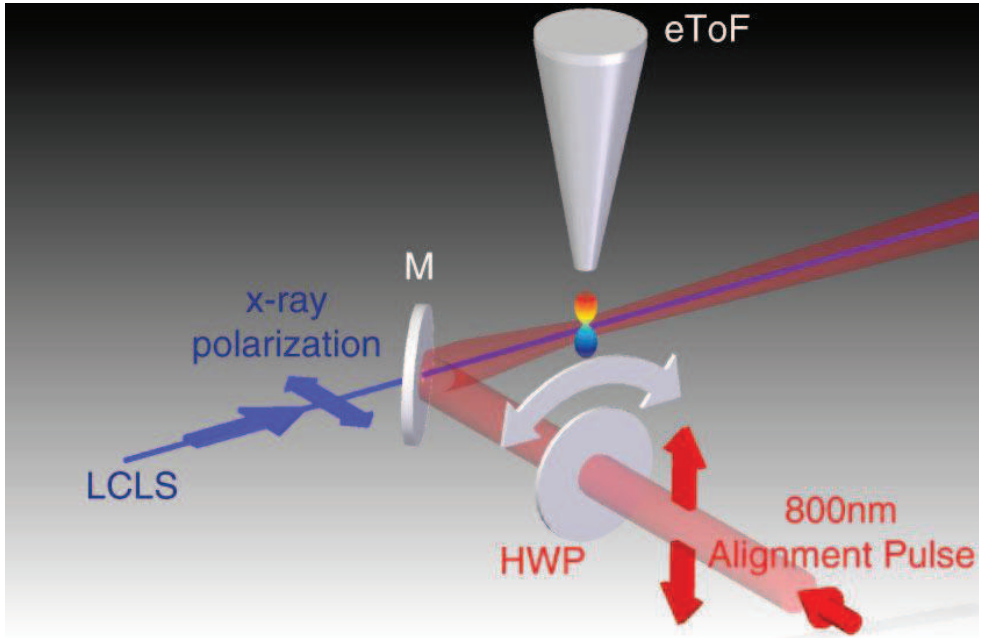
LCLS AMO end-station located in near experimental hall: instruments overview

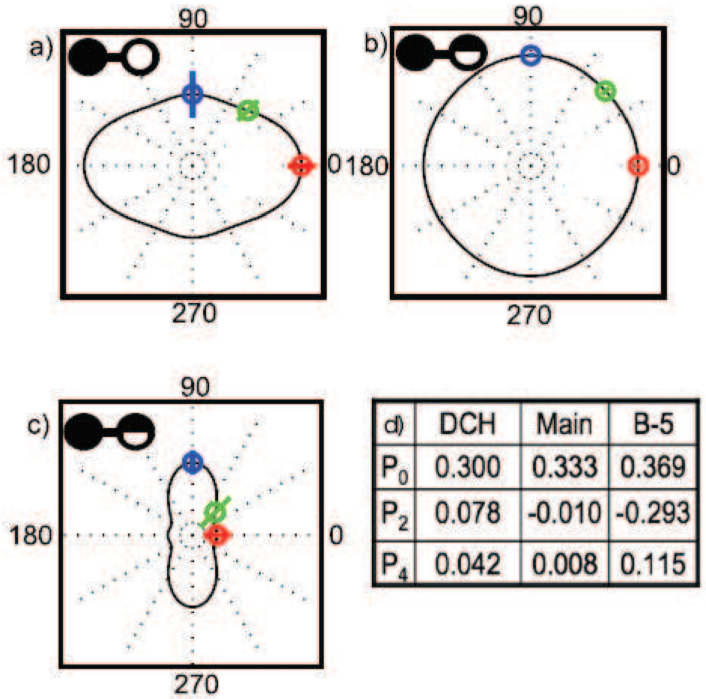


Experimental station – Chamber X-Section



Double core holes in N_2 were studied by probing a time-dependent rotational wavepacket to fix molecules in space





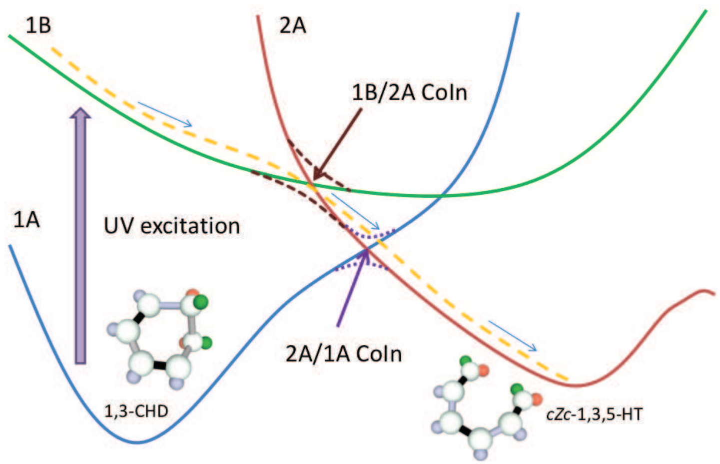
d)	DCH	Main	B-5
P_0	0.300	0.333	0.369
P_2	0.078	-0.010	-0.293
P_4	0.042	0.008	0.115

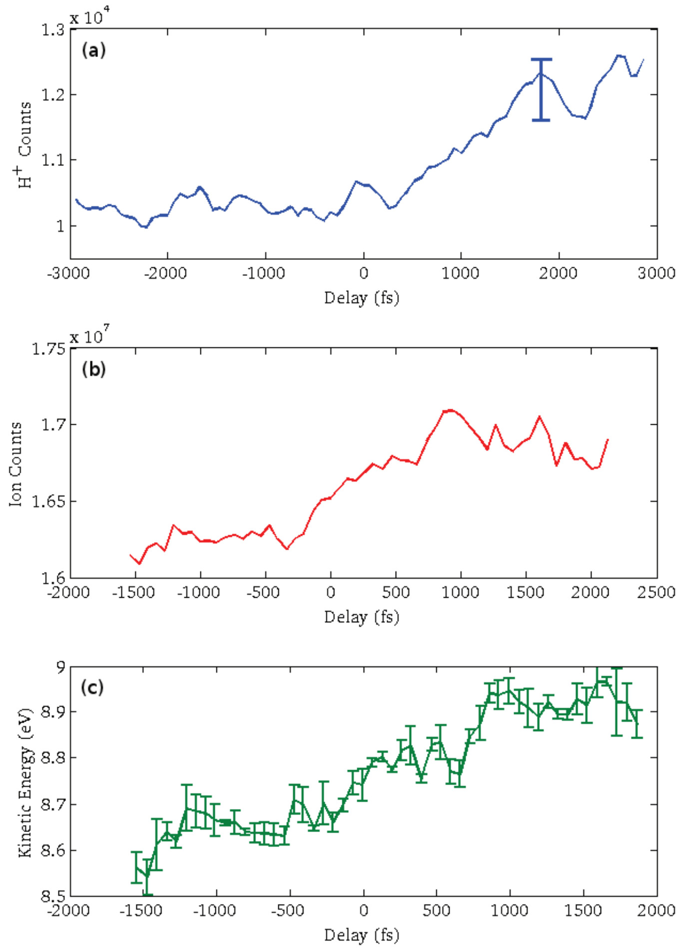
Short intense X-ray pulses enable single (SCH) and double core hole (DCH) formation in molecules. Impulsive alignment allowed us to measure the angular distribution of the Auger electrons from these states for the first time

Cryan et al, PRL, 105, 083004 (2010)

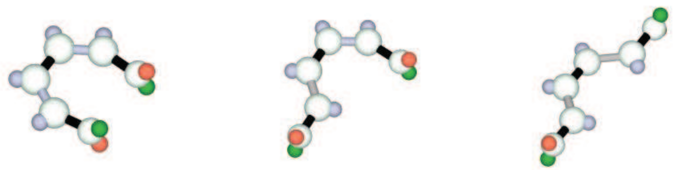
Cryan et al, J.Phys.B, 45, 055601 (2012)

Ring Opening of 1,3-Cyclohexadiene



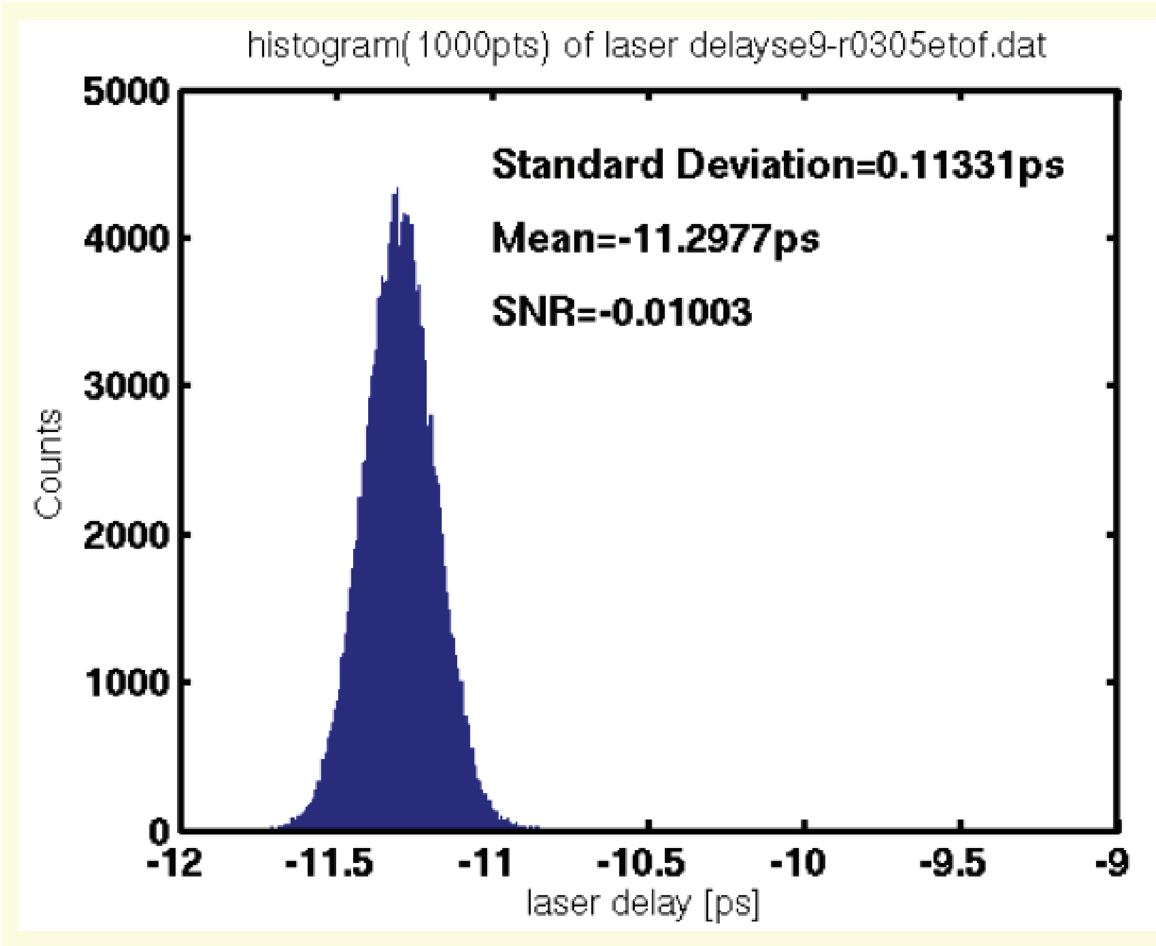


- UV-pump X-Ray probe
- Time jitter hides ring opening …
- … but resolution is enough to see signature of further conformational changes
- X-Ray fragmentation is sensitive to molecular structure.



V. Petrovic, M. Siano et. al., PRL 108, 25, 253006 (2012)

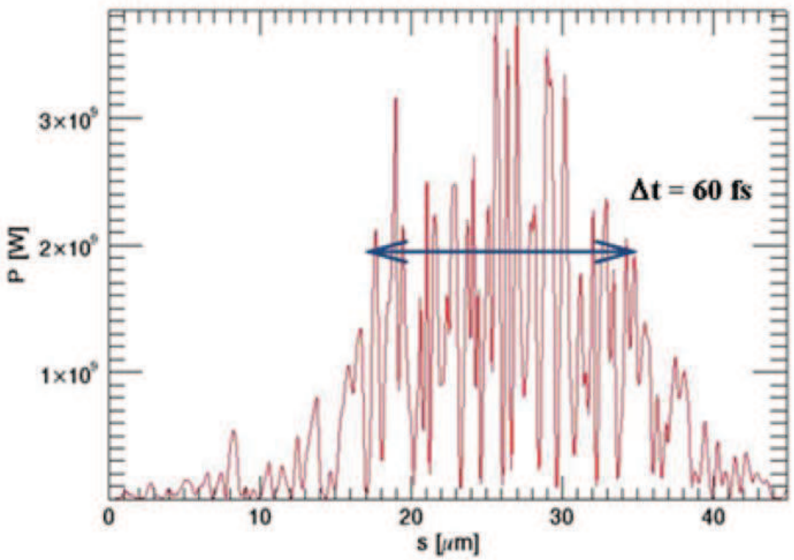
In the course of these measurements X-ray to laser jitter was measured

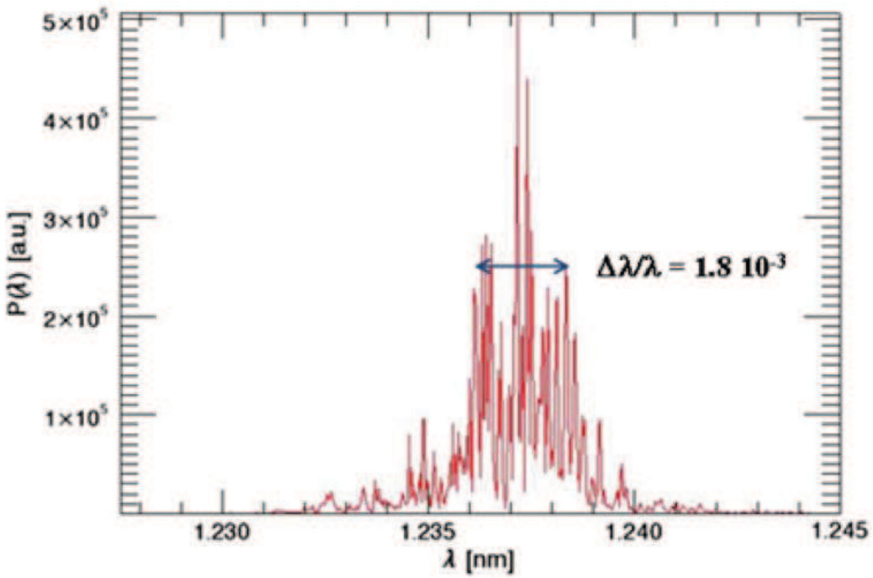


Glownia et al, OE 18, 17620 (2010)

To a large extent this jitter is an inherent limit of a SASE FEL

SASE: Wavelength Fluctuation and Temporal Jitter



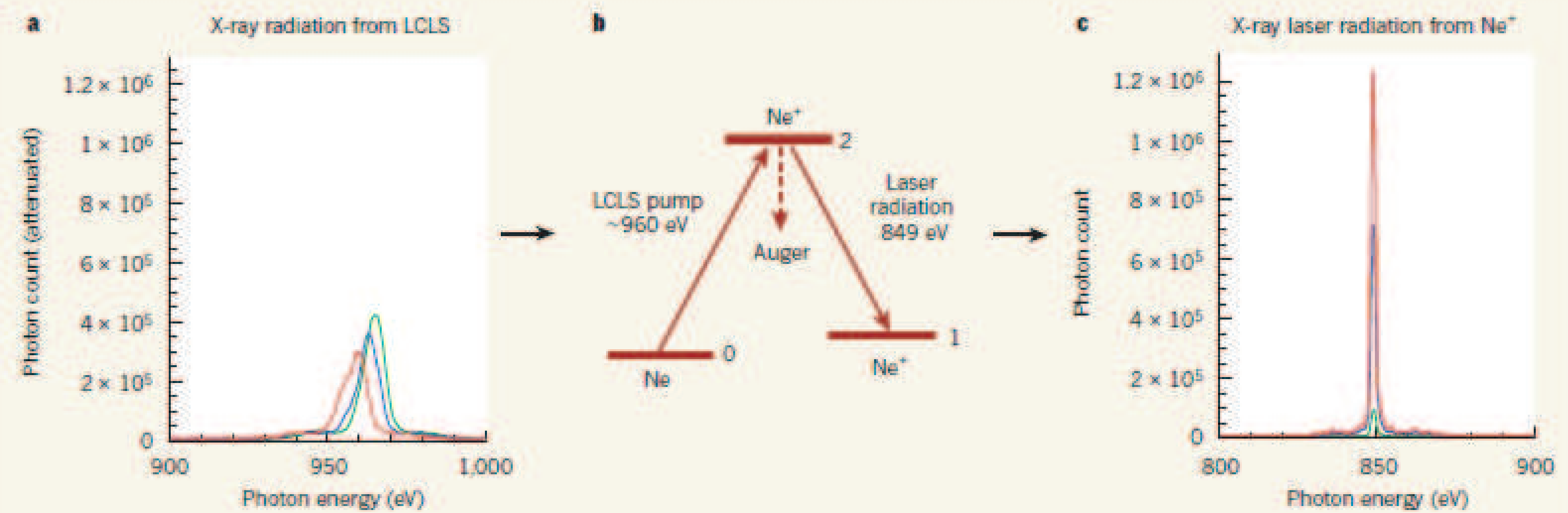


Temporal (+/- 100 fs) jitter inhibits:
- **Synchronisation with external sources**
- **High temporal resolution measurements**
- **Quantitative non-linear interaction studies**

Wavelength fluctuation inhibits:
- **X-ray spectroscopy**
- **Inelastic scattering**
- **Chemically sensitive CDI**

Ultrafast structural dynamic measurements at the femtosecond timescale need to overcome these limitations

Atomic Inversion Laser



Rohringer, N. *et al. Nature* **481,** 488–491 (2012)

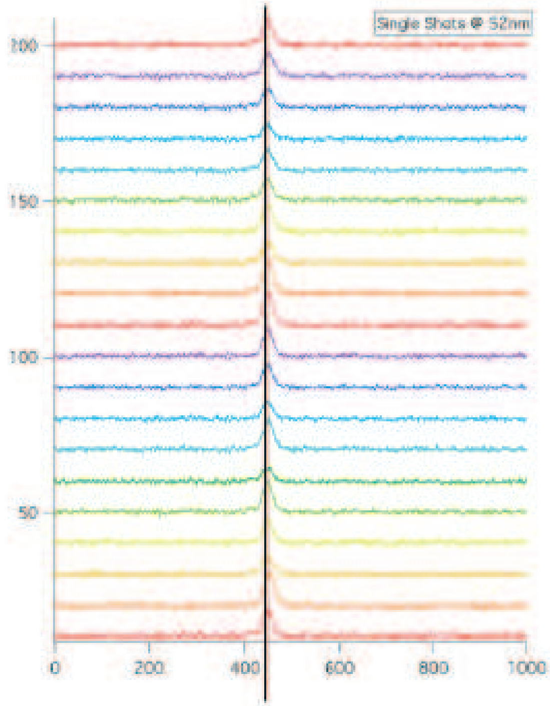
Figure 1 | Atomic X-ray lasing. Rohringer *et al.*[1] have demonstrated X-ray lasing at a photon energy of 849 electronvolts by creating atomic population inversion in a sample of neon gas using the Linac Coherent Light Source (LCLS), which is an ***[OK? to be consistent with main text?]*** X-ray free-electron laser. **a**, The LCLS X-ray radiation has a large spread in photon energy and considerable fluctuation in the average photon energy (about 960 eV) obtained from sequential laser pulses (shown in different colours). Photon count is here measured after transmission through the neon sample and is strongly attenuated. ***[OK?]*** **b**, The LCLS 'pumps' many of the neon atoms from the ground state (state 0) into a higher-energy state of ionized neon (state 2). Most of the ions in this excited state decay through an Auger process, but some will make the radiative transition to a state (state 1) that has a lower energy than state 2. This transition is accompanied by the emission of laser radiation that has a precise average photon energy (849 eV). **c**, The laser radiation has a smaller energy spread than that of the LCLS.

Results in a fixed wavelength but hard to do……..

Injection Seeding

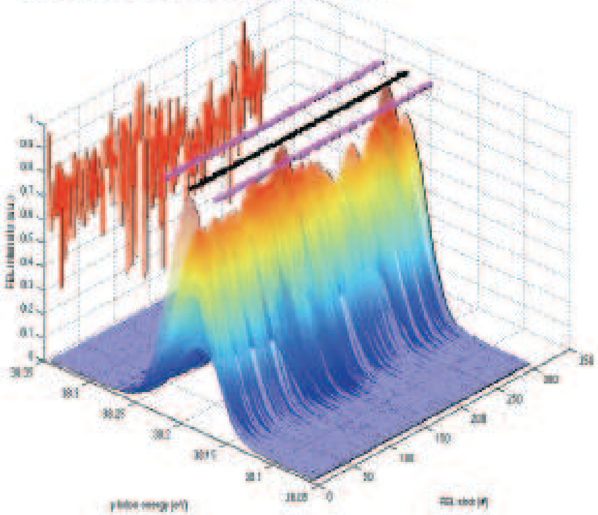
SASE AND SEEDED BANDWIDTH AND SPECTRAL STABILITY

In addition to the very narrow spectrum FERMI is characterized by a very good spectral stability. Both short and long terms measurements show that the spectral peak move less than 10^{-4}.



D. Cocco, C. Svetina, M. Zangrando

Reported data refer to an electron beam of 350 pC at 1.24 GeV compressed about a factor 3. The 6 radiators are tuned at 32.5 nm.



FEL photon energy	~ 38.19eV
Photon energy fluctuations	= 1.1meV (RMS)
FEL bandwidth	= 22.5meV (RMS)
	= $5.9e^{-4}$ (RMS)
FEL bandwidth fluctuations	= 3% (RMS)

E. Allaria, W. Fawley

courtesy of Fulvio Parmigiani

Nature Photonics 6, 699 (2012)

Highly suitable for valence shell studies, but not viable for hard X-rays…..

Self Seeding: Eliminates Wavelength Jitter

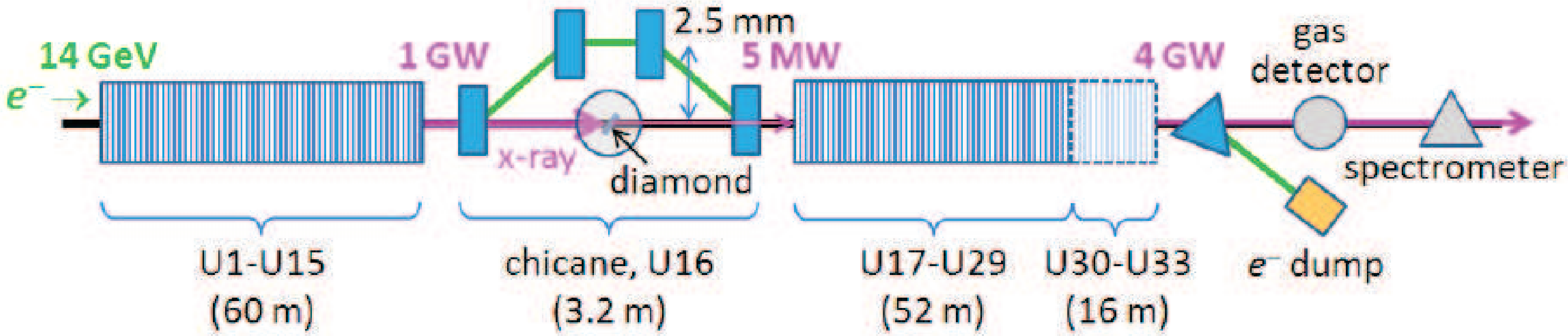
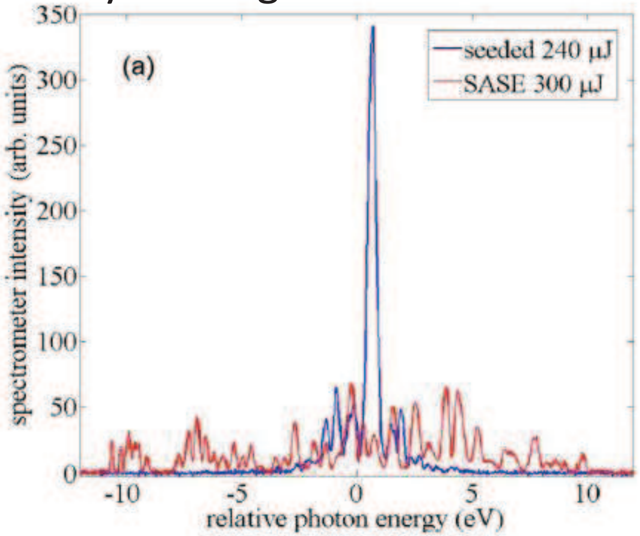
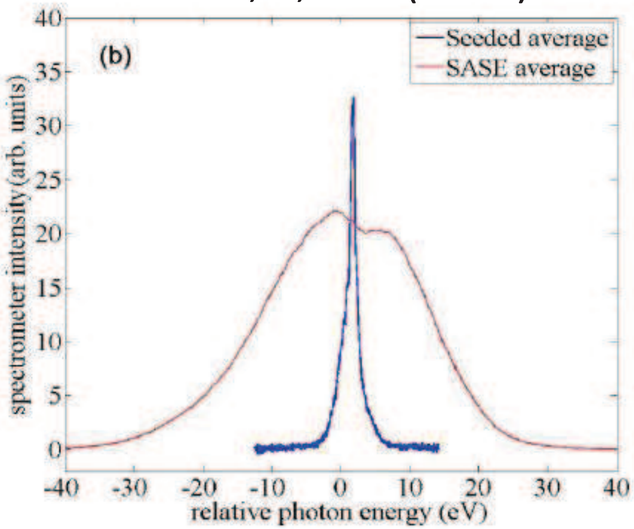


Figure 1. Layout of LCLS undulator with self-seeding chicane, diamond monochromator, gas detector, and hard x-ray spectrometer. The chicane is greatly exaggerated in scale here.

courtesy of Jerry Hastings

Nature Photonics, 6, 693 (2012)

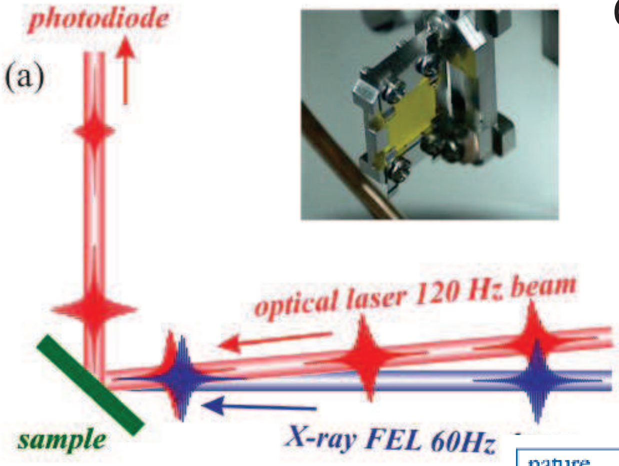




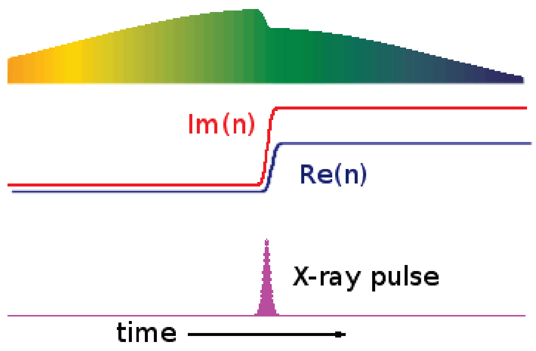
Promising and fixes wavelength jitter, but not temporal jitter…….

Time Stamping

Courtesy of Ryan Coffee

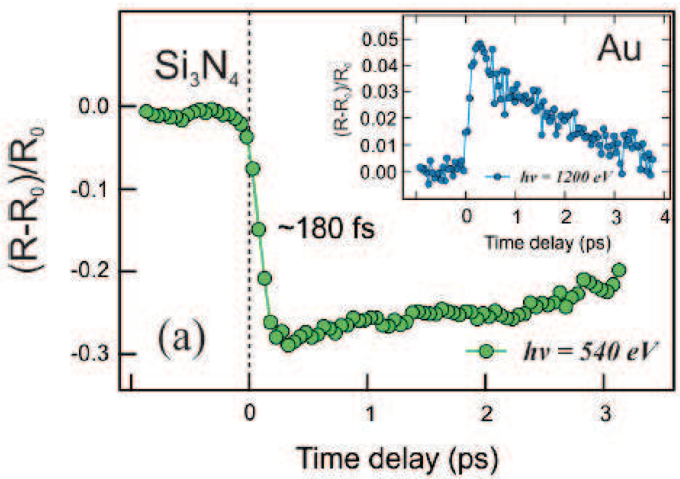


X-ray gated reflectivity

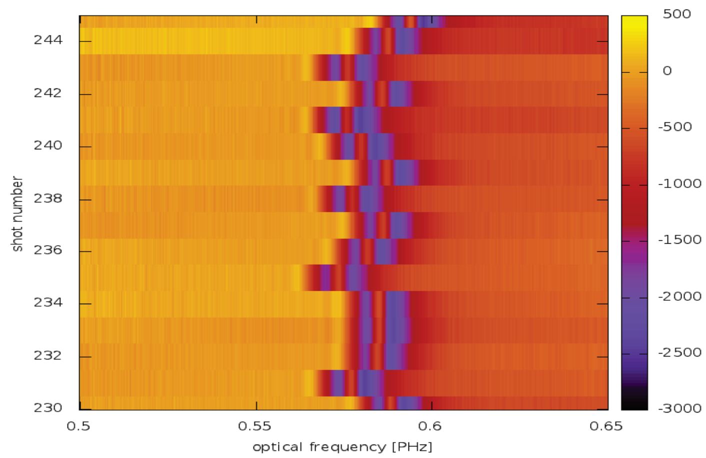


nature photonics | LETTERS
PUBLISHED ONLINE: 17 FEBRUARY 2013 | DOI: 10.1038/NPHOTON.2013.11

Achieving few-femtosecond time-sorting at hard X-ray free-electron lasers



Using chirped pulse for single shot timing measurement – Sub 20fs resolution demonstrated



Combined with self seeding this might just do, but still need higher rep-rate to overcome fluctuations (in self seeding specral fluctuations transferred to intensity fluctuation)….

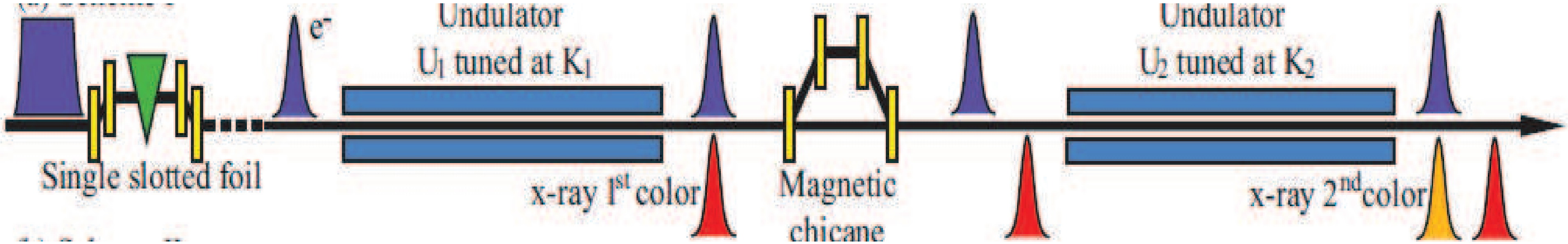
To eliminate temporal jitter X-ray pump- X-ray probe methods are now being developed

Several options are currently used at LCLS

- Split and delay (recently commissioned on AMO)
- Two-pulse generation at single frequency
- Two-pulse / two-colour generation

All offer pulses as short as 2 fs and jitter free delay down to 0fs

To eliminate temporal jitter X-ray pump- X-ray probe methods are now being developed



Two-colour two-pulse scheme using an intra-undulator chicane and tuning the two undulator sections slightly differently:

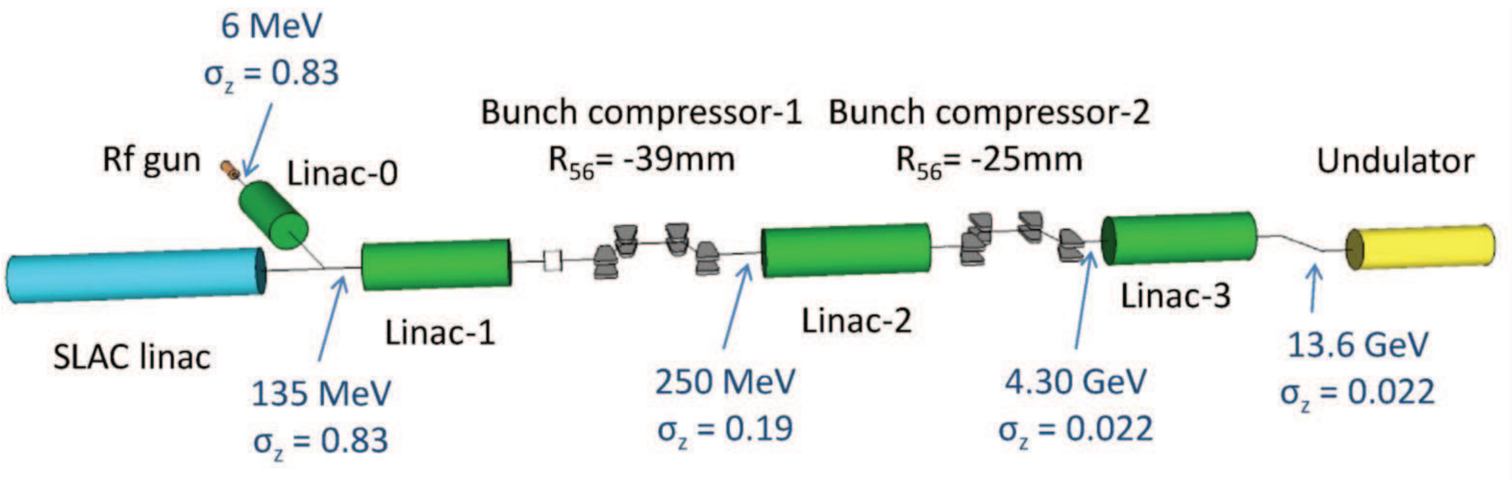
Two pulses each of <3fs

Variable delay 0 to 20 fs

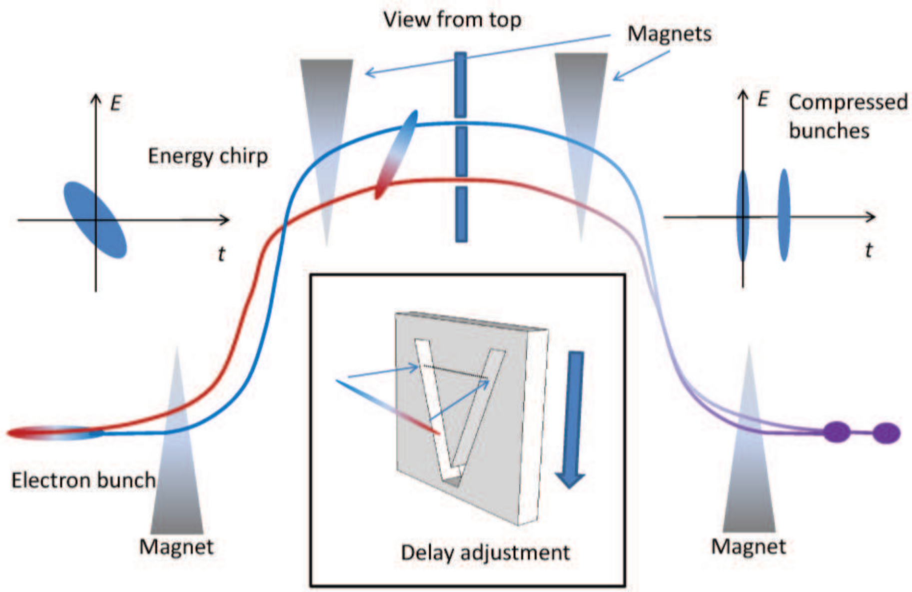
Photon energy difference in pulses of a few percent possible (e.g at 500eV +/- 10 eV)

A. A. Lutman, R. Coffee et al, PRL, 110, 134801 (2013)

FELS: Pump-Probe



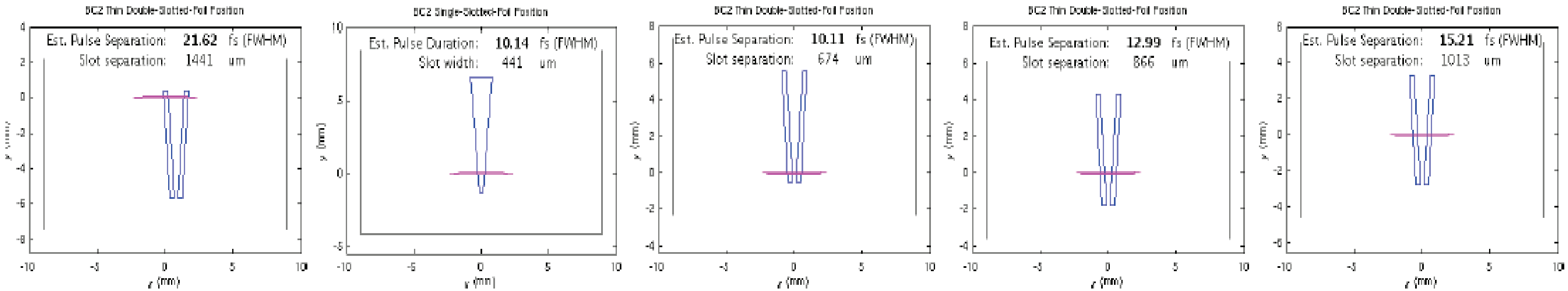
Generation of two few fs short pulses with variable delay



PRL 109, 254802

X-ray pump-X-ray probe

- Split and delay of X-rays is now being implemented at FLASH and LCLS
- A promising method for high temporal resolution is use of a slotted emittance spoiler placed in the electron beam in a dispersed section:



Creates single or double pulses ~ 3 fs duration with variable delay up to ~20fs

This has recently been used to study ultrafast dynamics triggered by X-ray core exctitation in O_2

Group of Ryan Coffee LCLS, SLAC Stanford

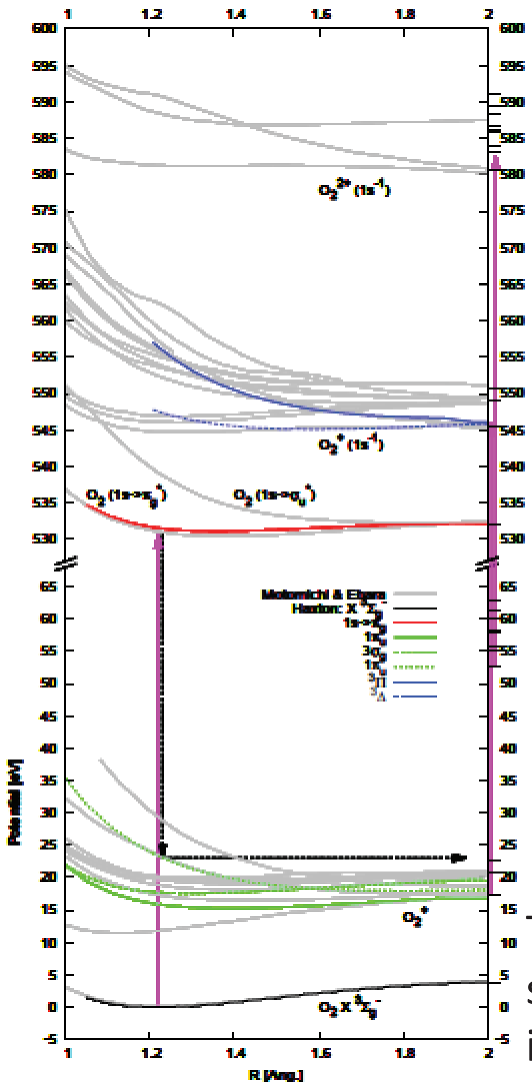
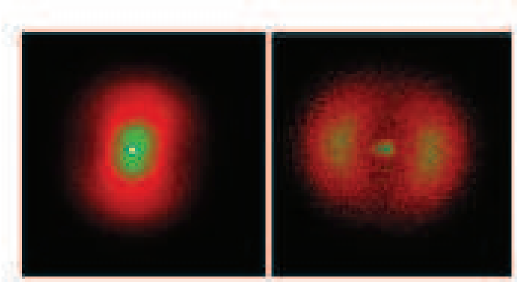
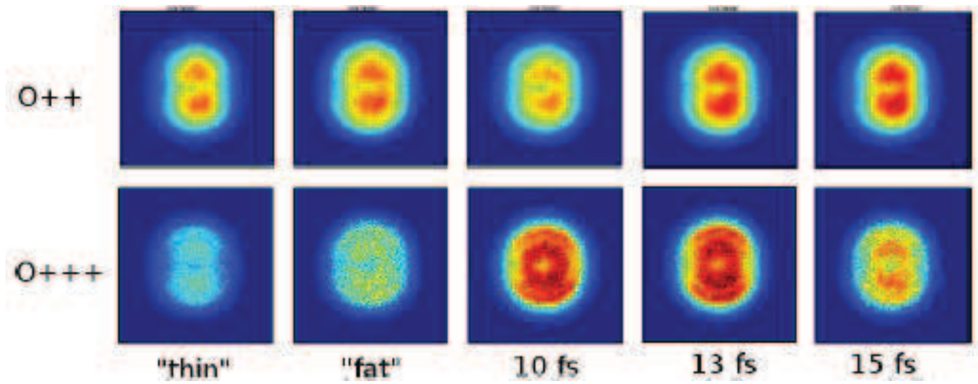


Figure 1: Various potential energy curves. Tashiro Motomichi and Masahiro Ehara produced the gray curves while Dan Haxton produced the symmetry labeled curves.

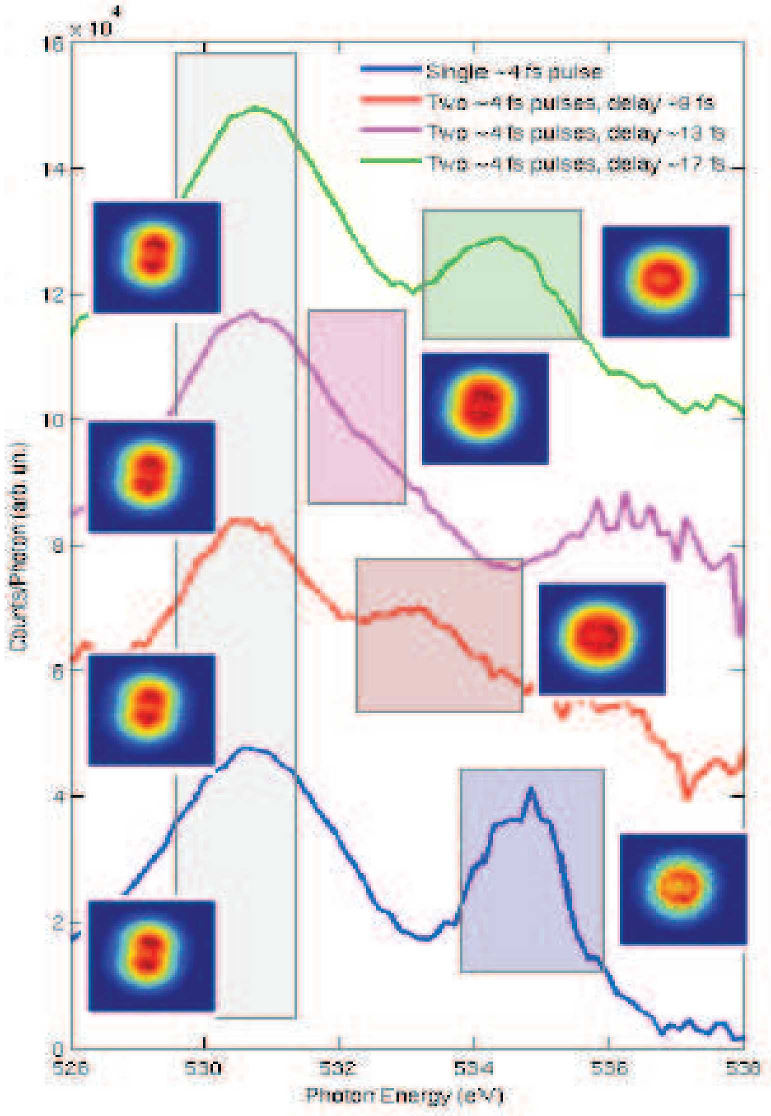


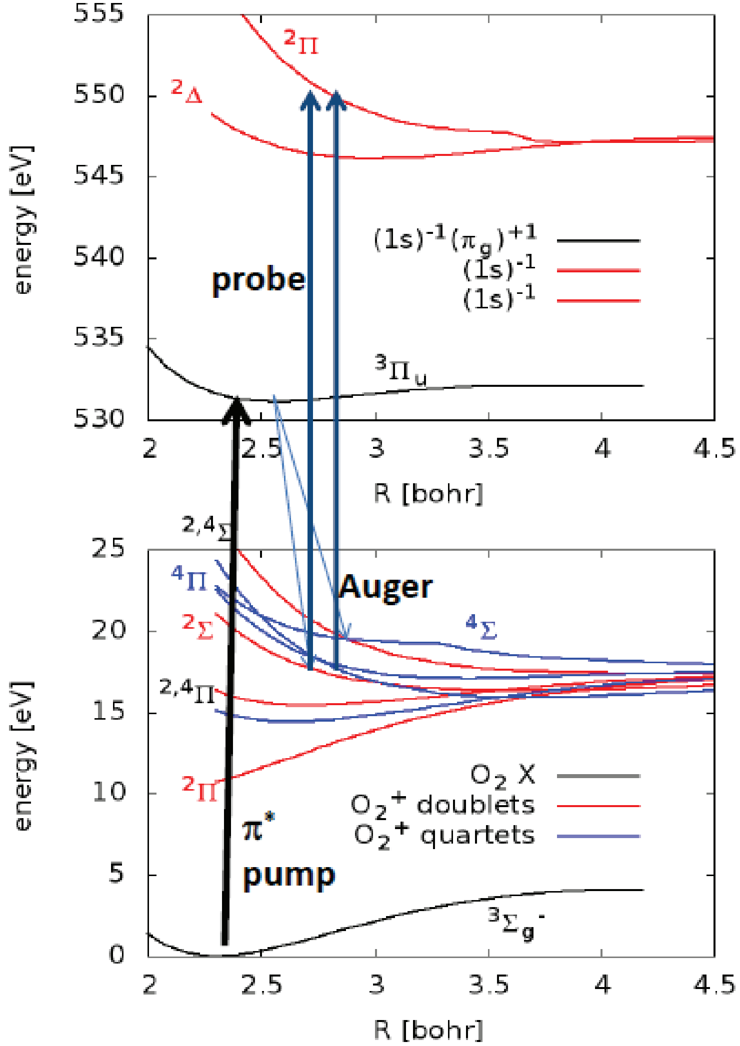
Left fragment pattern from $\pi*$ resonance, right fragment pattern from $\sigma*$ resonance



The symmetry goes form $\pi*$ to convolution of $\pi*$ with $\sigma*$ (indicating second absorption is via $\sigma*$ moment) then recovery of the $\pi*$ by 15 fs. indicates that the second absorption has become atomic like rather than contributing as a molecular symmetry.

Few-fs time resolved measurements with X-ray FEL





X-ray FEL Capabilities for Ultrafast Measurements

- IR/Vis/UV pump – X-ray probe to <20fs resolution (with time stamping)
- X-ray pulses to ~2 fs
- X-ray pump-X-ray probe with ~2fs resolution over delays >30 fs
- Future prospects for (a) two-colour two-pulse and (b) sub-fs measurements being considered.

Acknowledgements

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Ryan Coffee

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Christoph Bostedt

et al

Pulse – Stanford

Phil Bucksbaum

Vladimir Petrovic

James Cryan

Mike Glownia

et al

UCL

Jonathan Underwood

Russell Minns

And many more ....