# Ultrafast multi-edge X-ray spectroscopy with 100 kHz OPCPA-driven HHG source

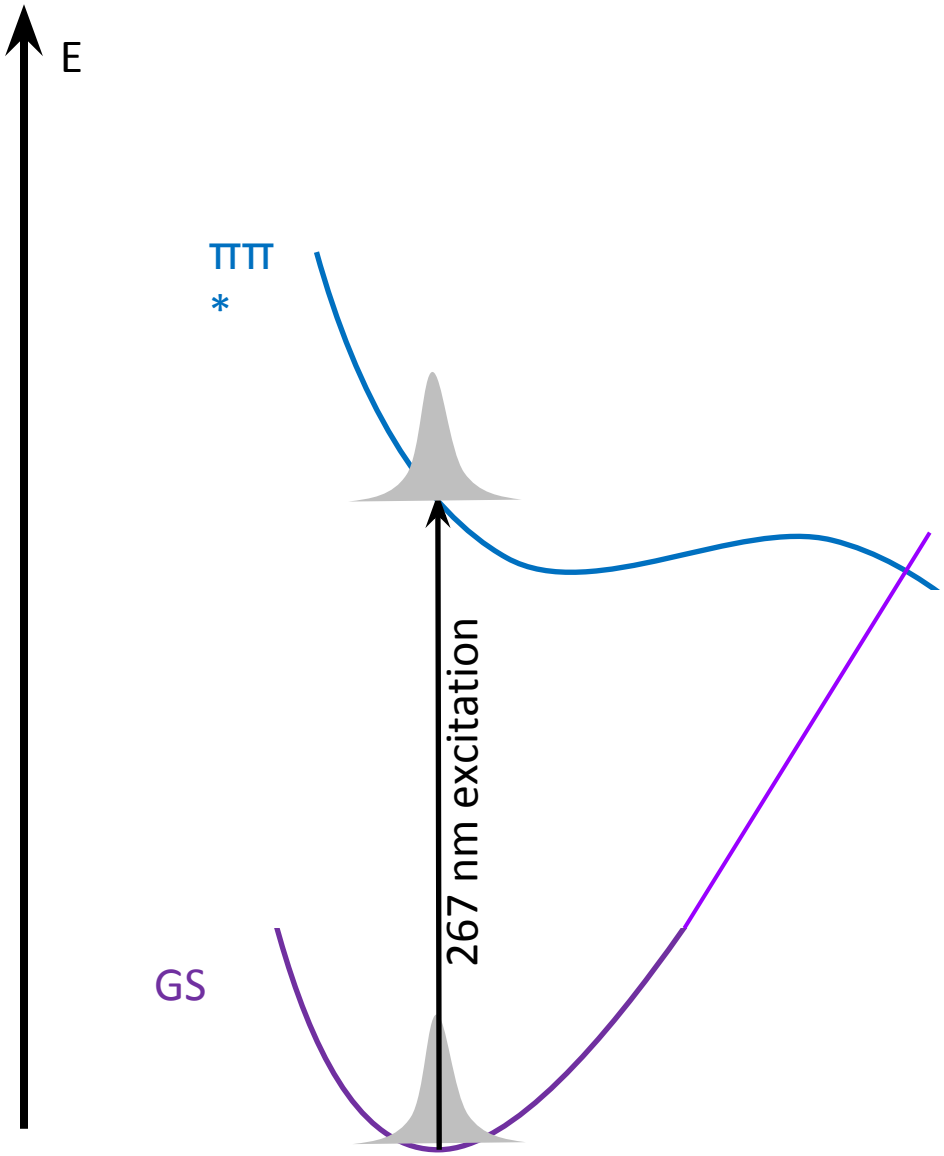
Elio Champenois

ISMS 2019

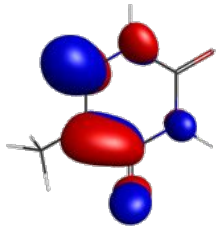
U.S. DEPARTMENT OF ENERGY
Office of Science

PULSE
SLAC NATIONAL ACCELERATOR LABORATORY

# Motivation: excited state dynamics in organic chromophores




π*-MO, delocalized



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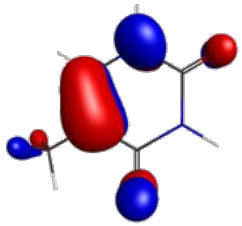

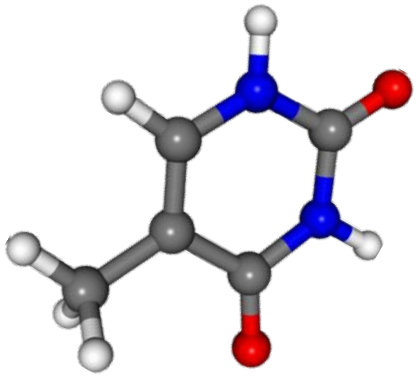

Thymine

TJA Wolf et al. "Photochemical pathways in nucleobases measured with an X-ray FEL." Phil. Trans. A 2019

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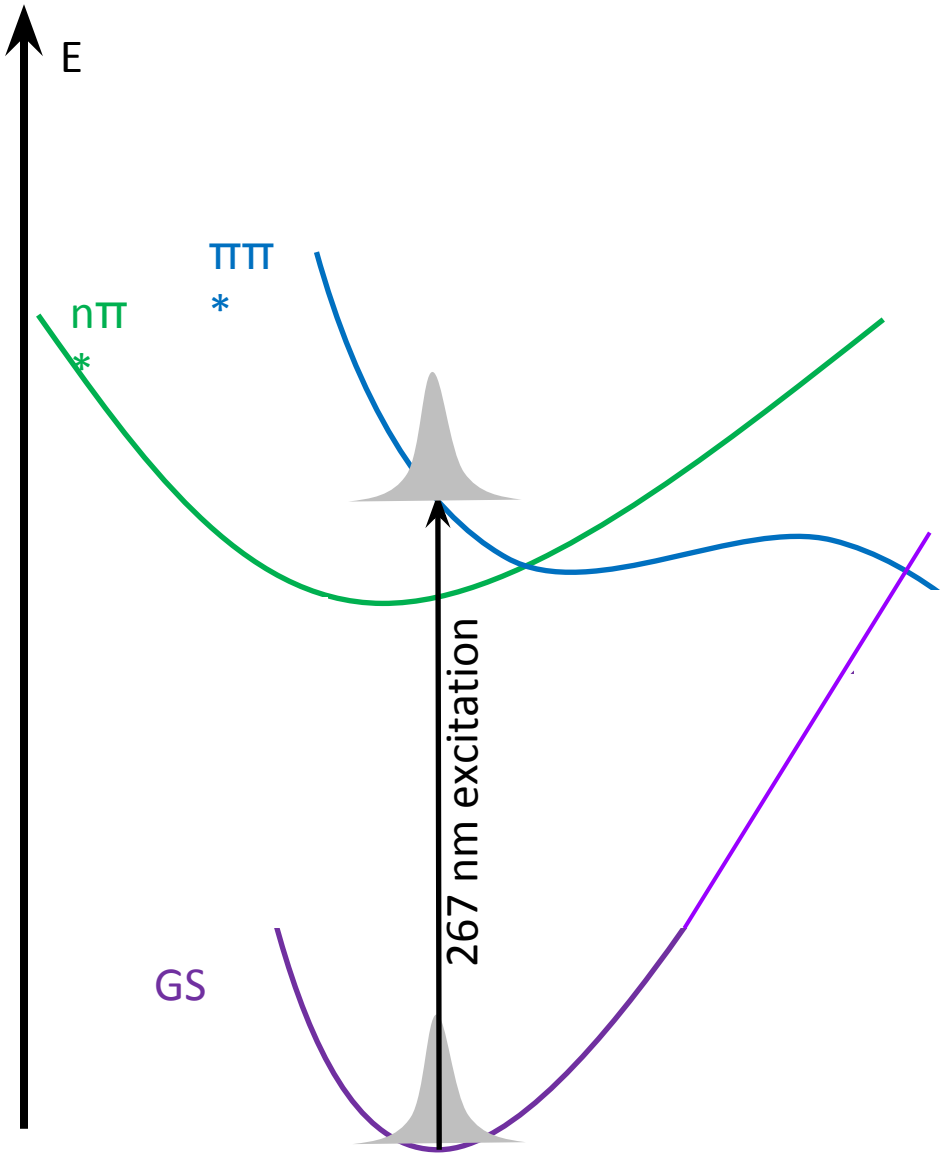


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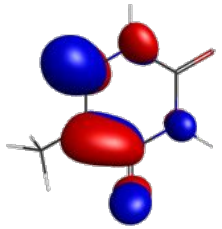

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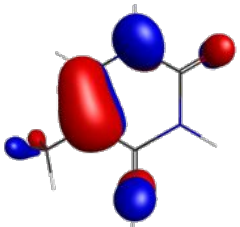
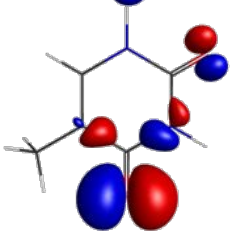
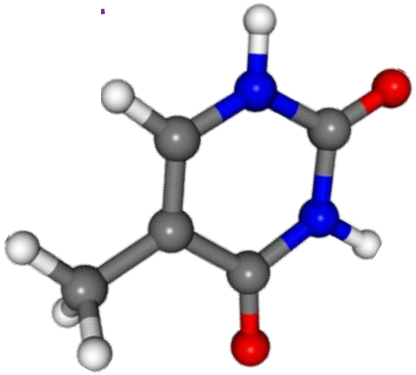


n-MO, localized





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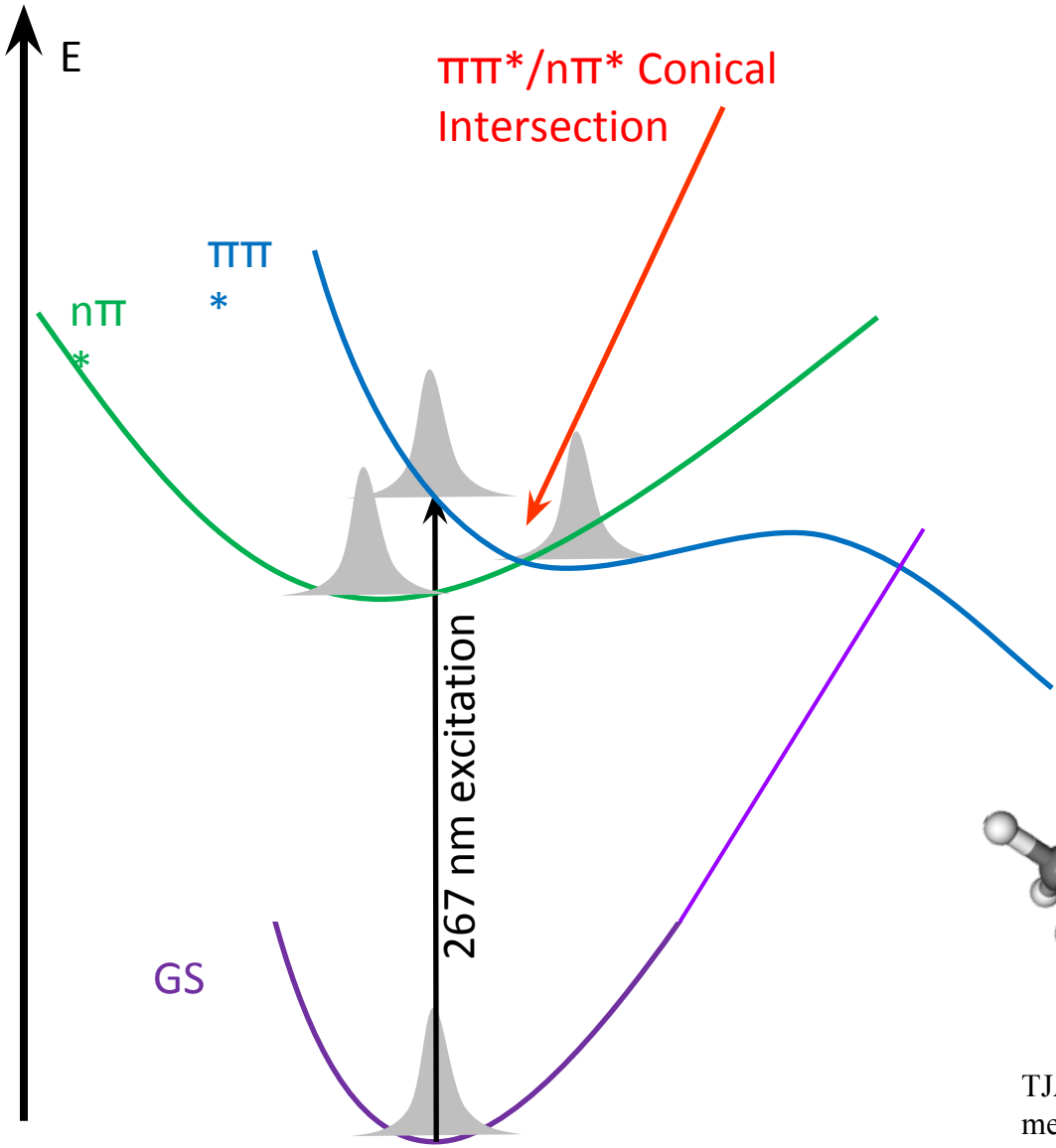
SLAC

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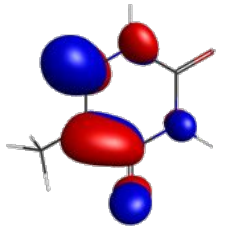

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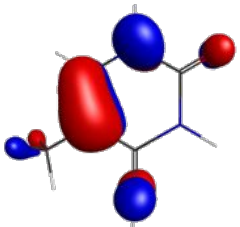

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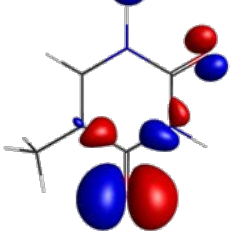

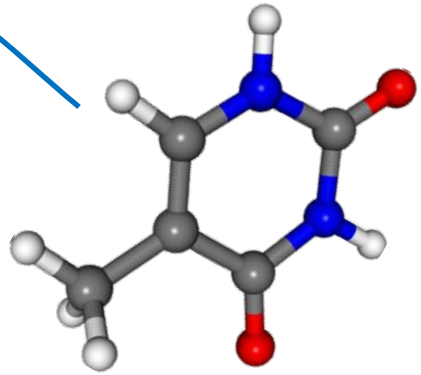

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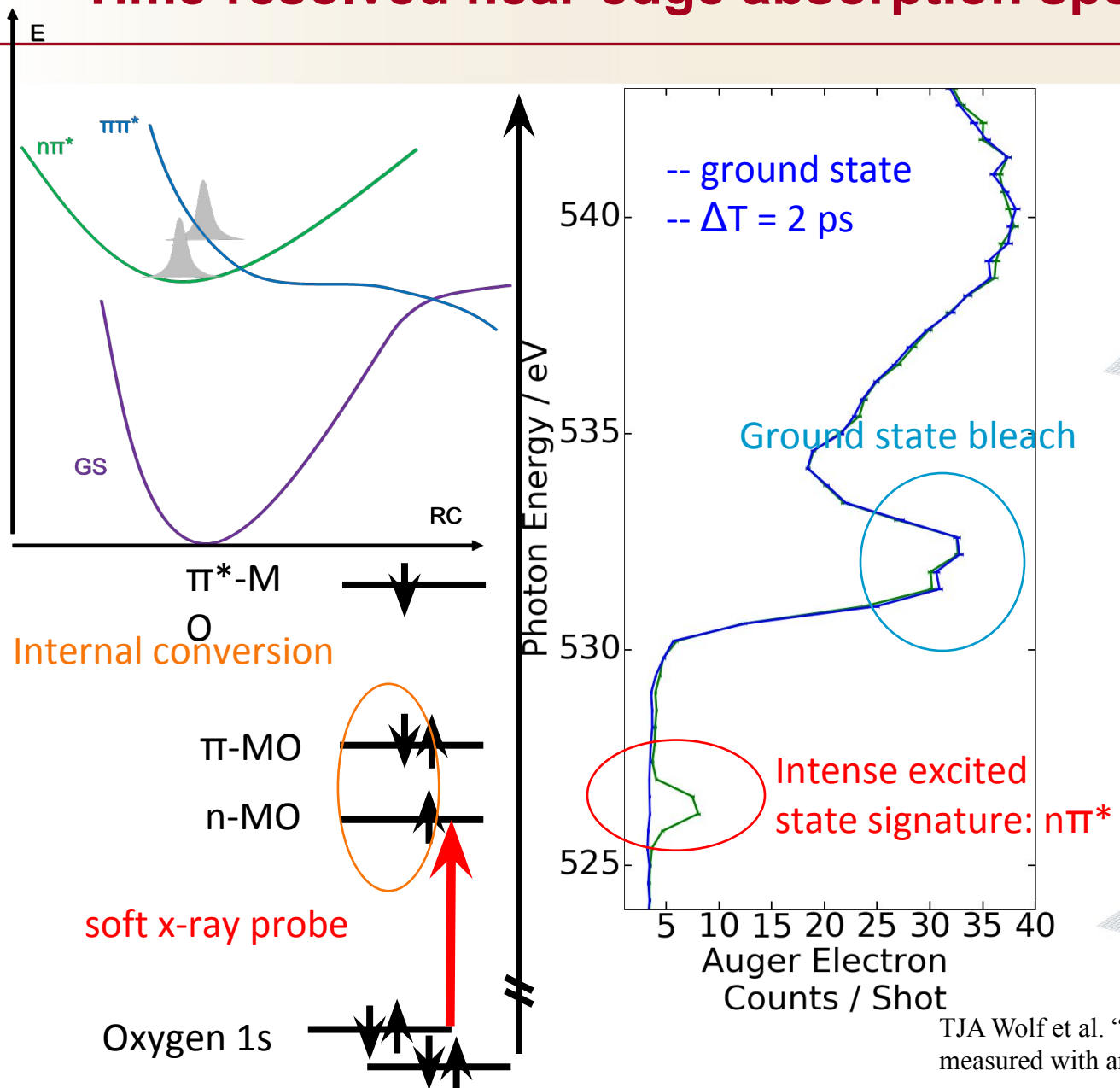
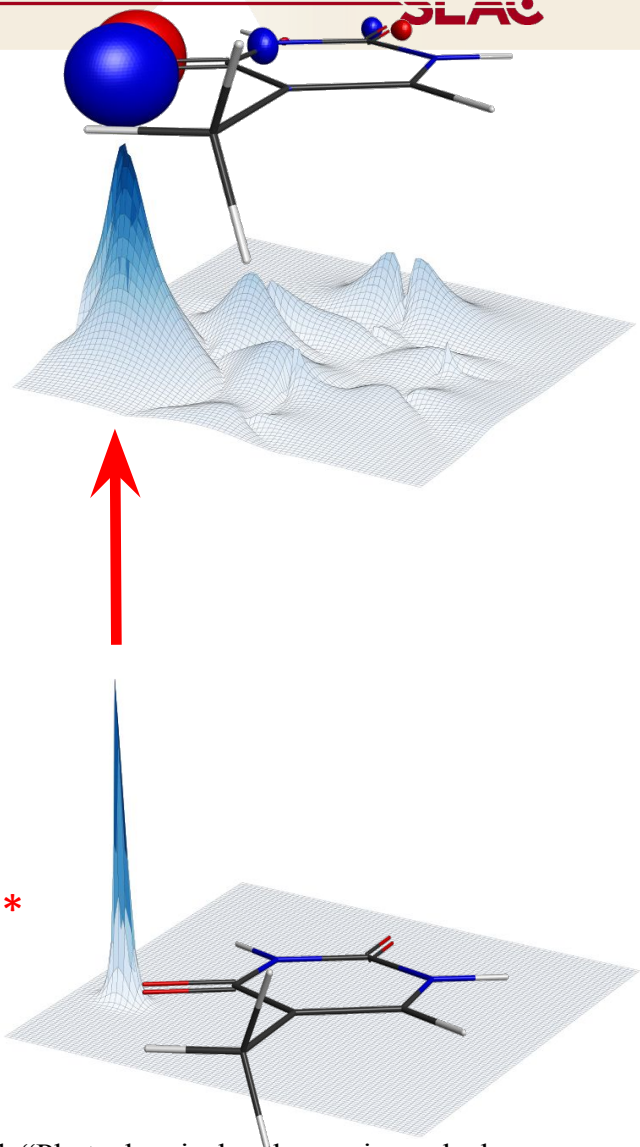
# Time-resolved near-edge absorption spectroscopy







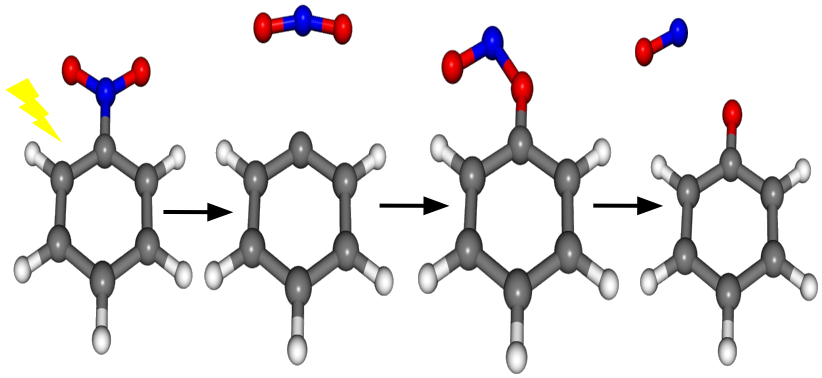


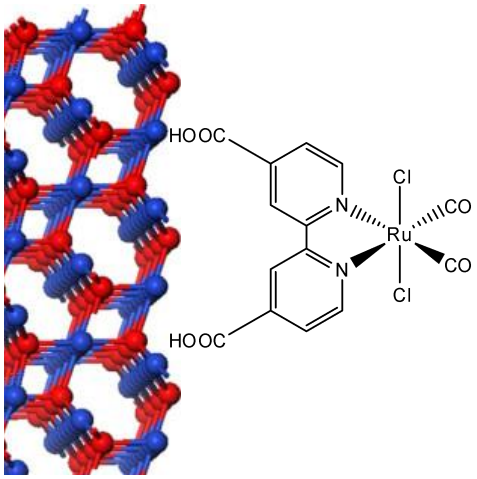
TJA Wolf et al. "Photochemical pathways in nucleobases measured with an X-ray FEL." Phil. Trans. A 2019

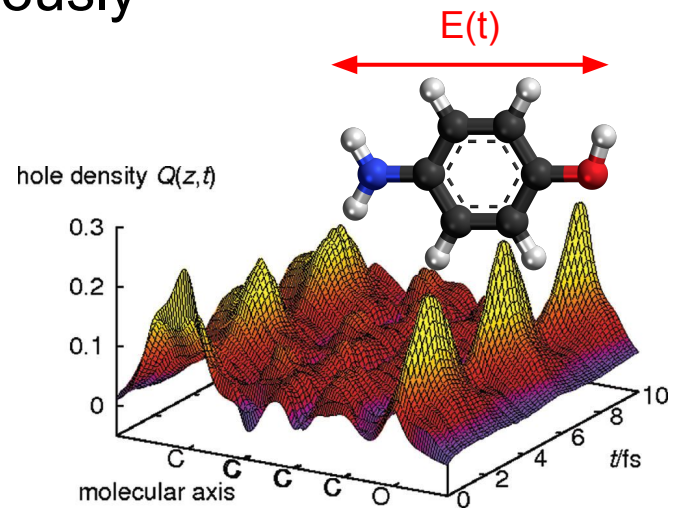
# Time-resolved NEXAFS with an HHG source

SLAC

- Atom-specific local sensitivity to the valence electronic character
- Oscillator sum rule:
  - wide range of applications
  - unlikely to go blind
- Multiple edges probed simultaneously






Kuleff et al. JPC A **114** 8676 (2010)

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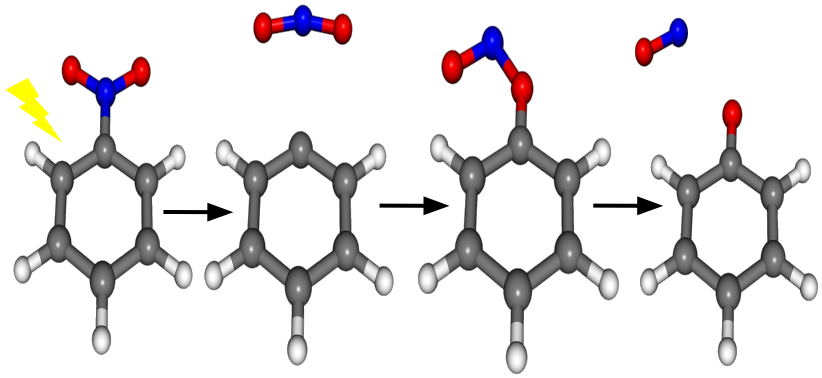
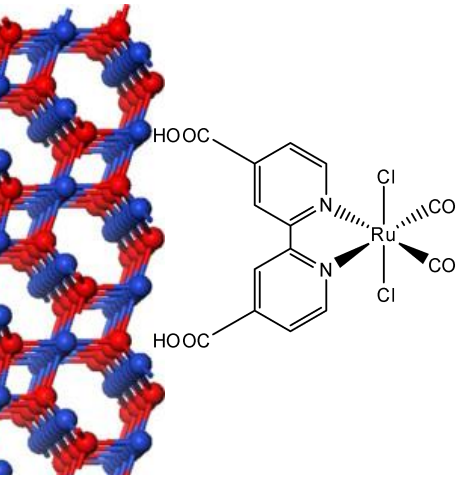
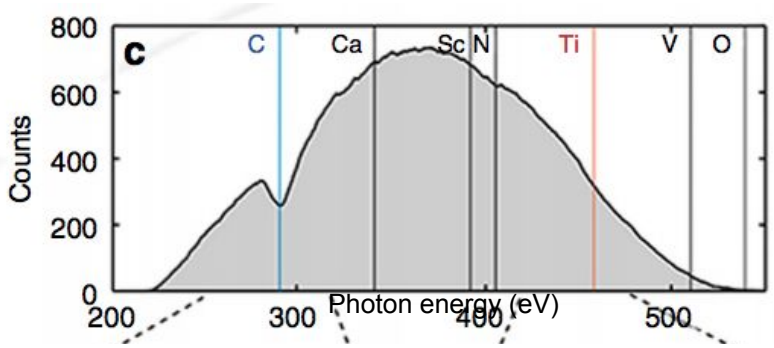
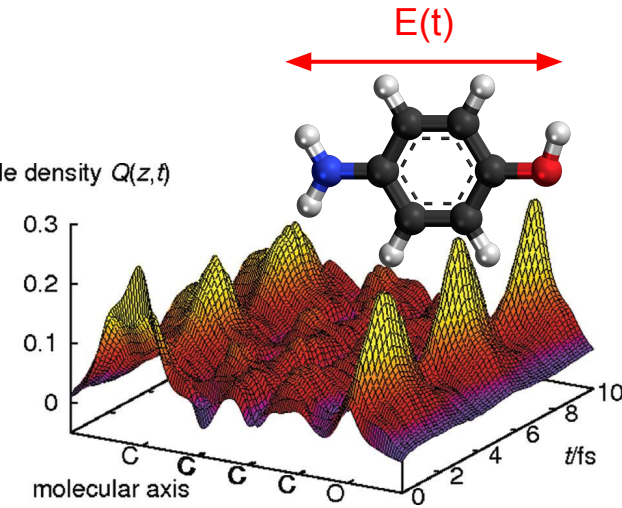





S. M. Teichmann et al. Nat. Comms. (2016)

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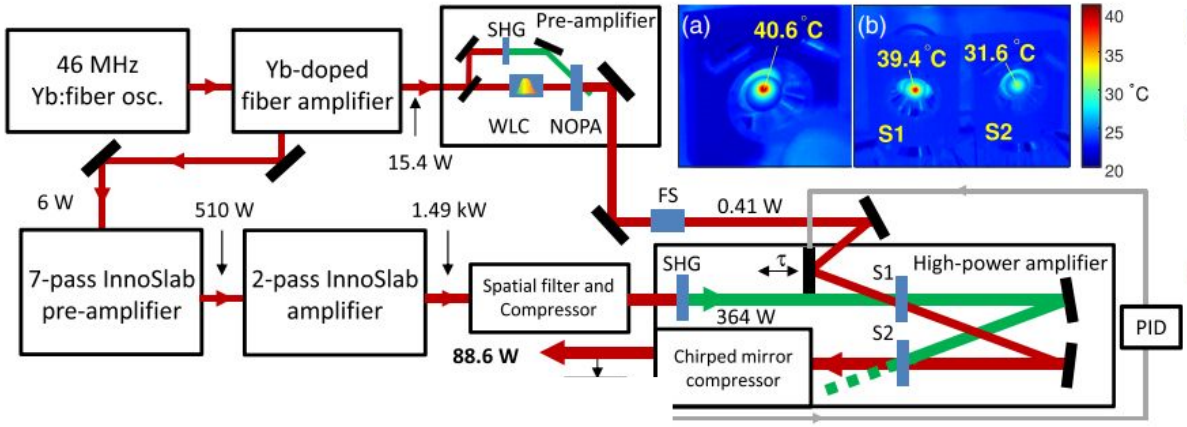
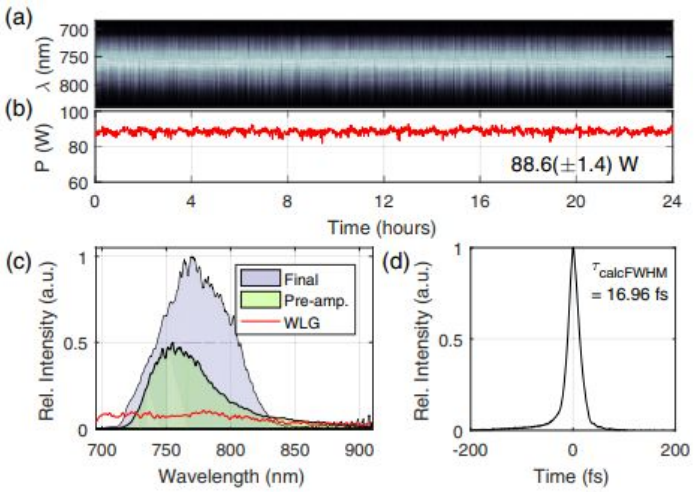
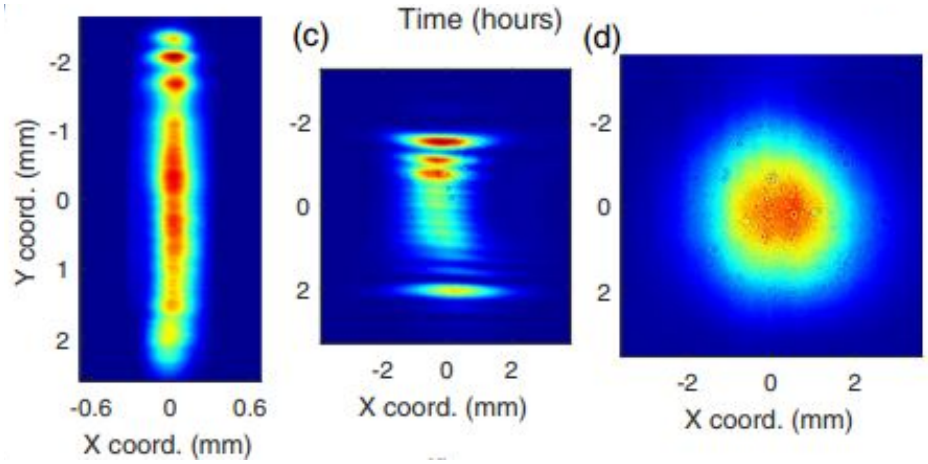
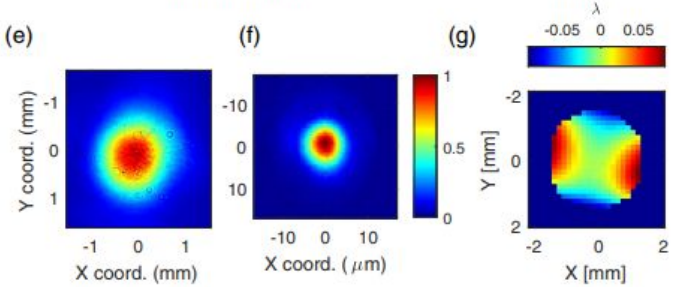
# High average power, high repetition rate OPCPA



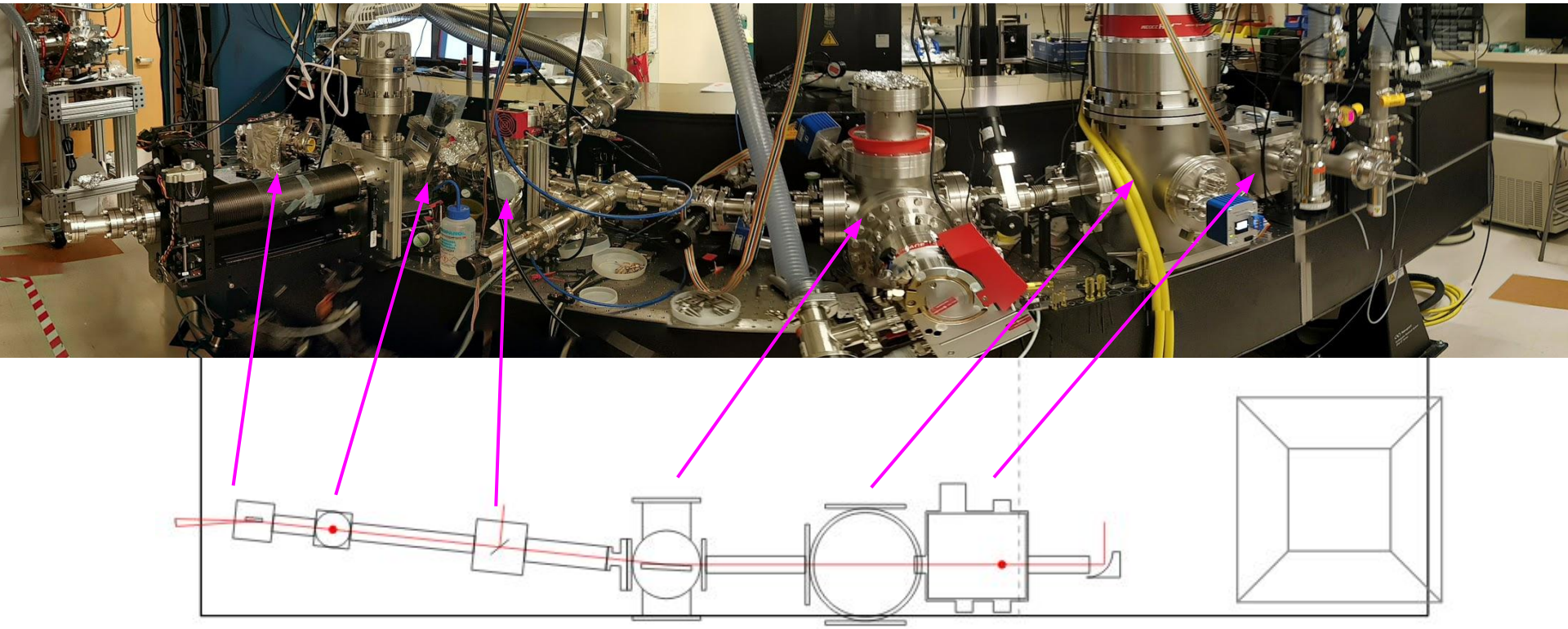

Mecseki et al. “High average power 88 W OPCPA system for high-repetition-rate experiments at the LCLS x-ray free-electron laser.” Optics Letters 2019

Windeler et al. “High Average Power 106W, 1.75 μm, 100 kHz Optical Parametric Chirped Pulse Amplifier.” CLEO 2019

# HHG and transient absorption beamline

SLAC

(6) XUV / X-ray spectrometer | (5) Target delivery: gas, solid, liquid?? | (4) Optical/UV pump recombination | (3) IR rejection and X-ray re-imaging | (2) Two stages of differential pumping | (1) High pressure gas cell for HHG



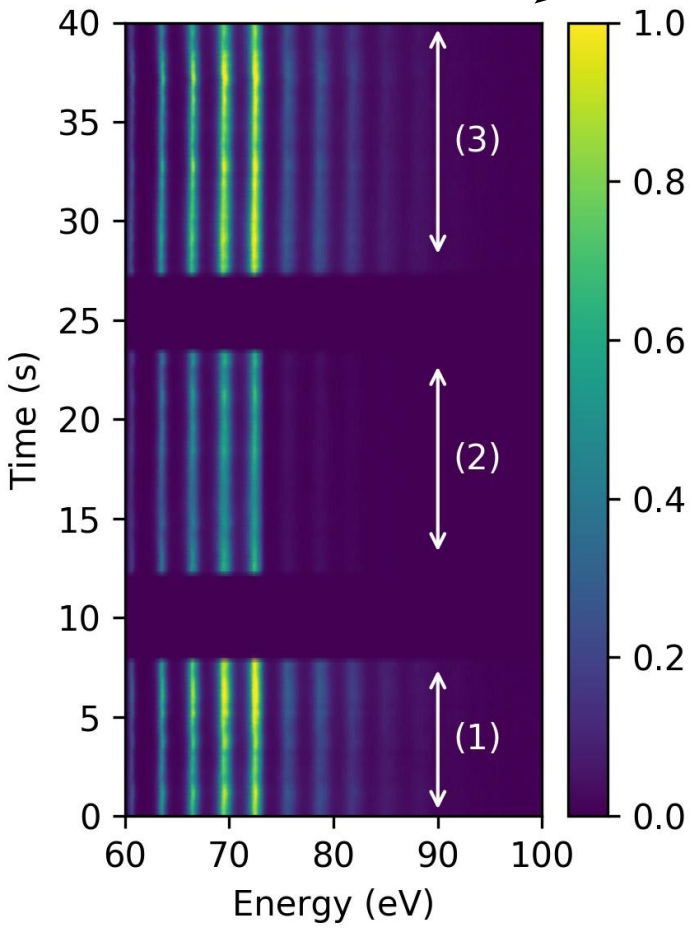
Currently: 800 nm, 21 fs, 400 uJ, 100 kHz (80% for HHG)

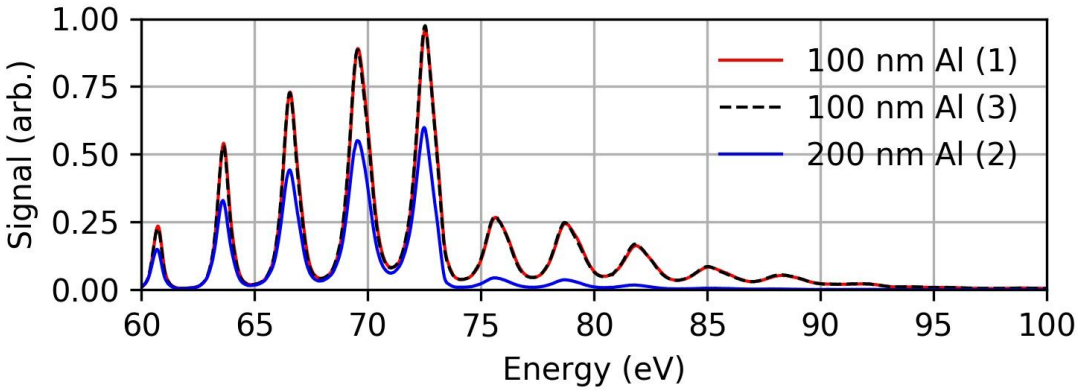
# Static absorption measurements

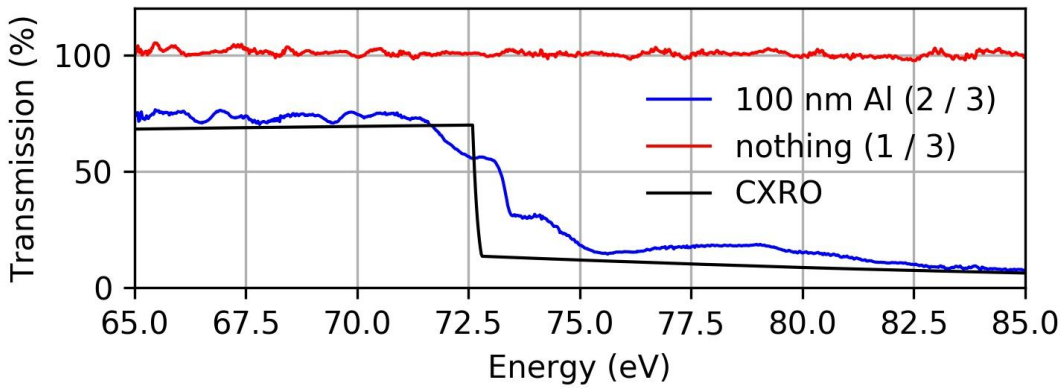
SLAC

25 ms per readout using small vertical ROI, full vertical binning, and 0 ms exposure

120 Torr Ne,
1x or 2x 100 nm Al



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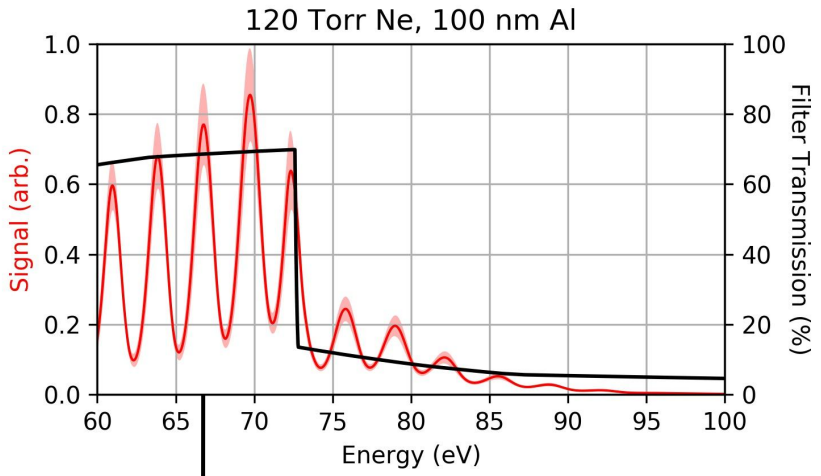
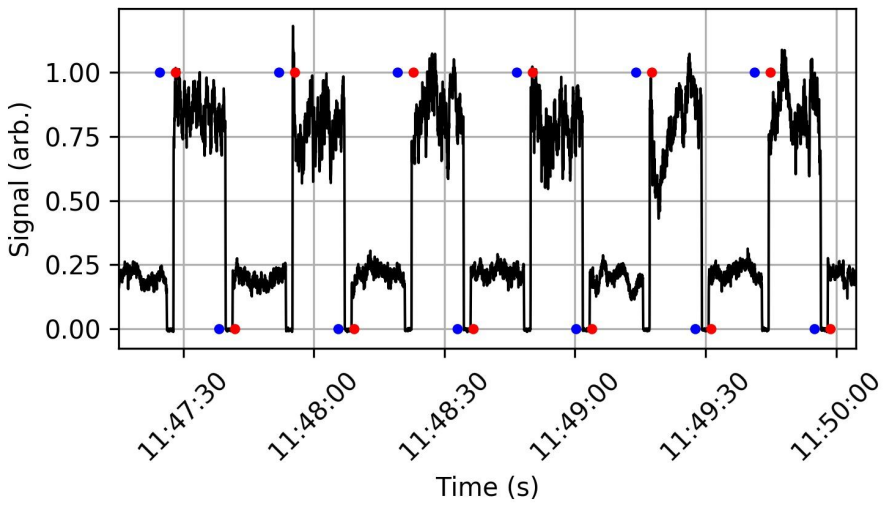
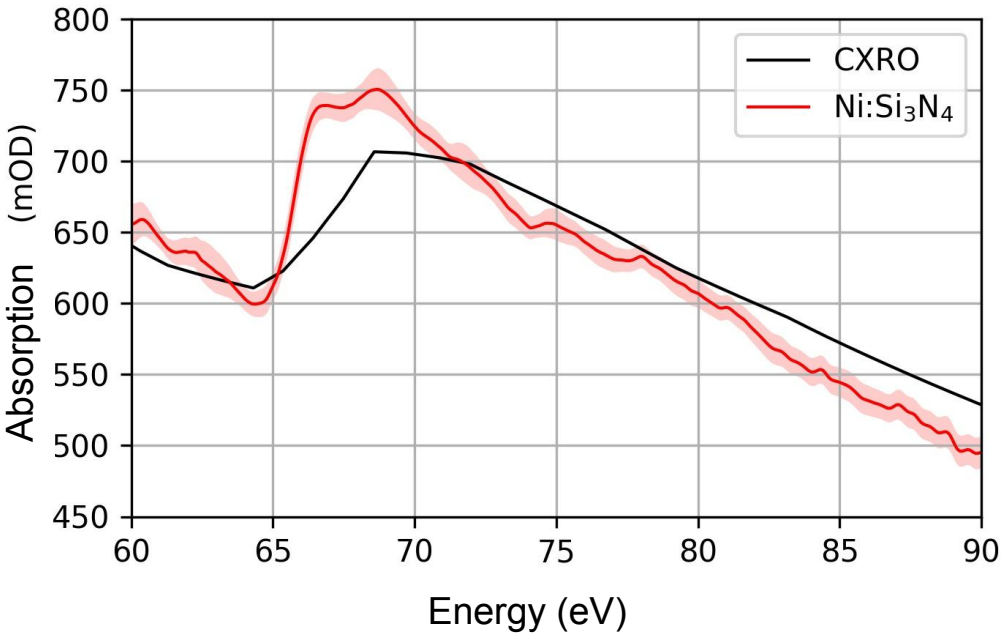


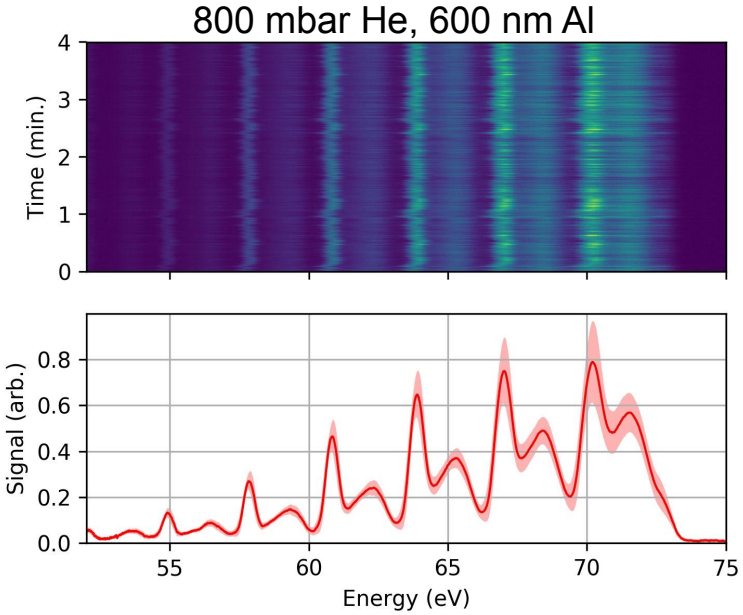

Single pixel,
25 ms exposure




- Limited by the spectral instability on the referencing time-scale (> 1 s)
- Ideally: reference shot-to-shot
- Instead: reference on shots closer in lab time

# Locking in at higher frequencies



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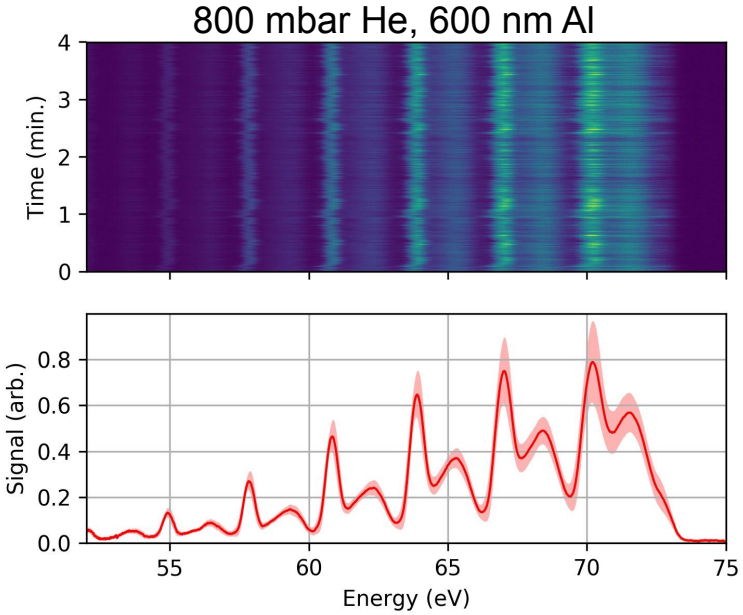


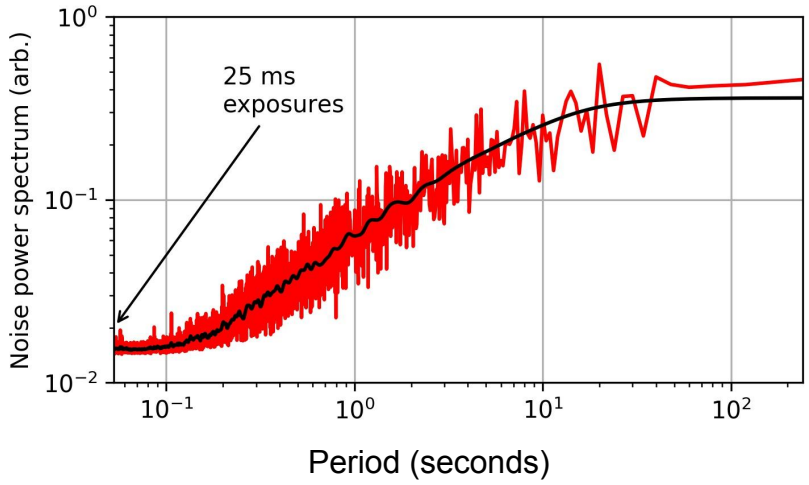

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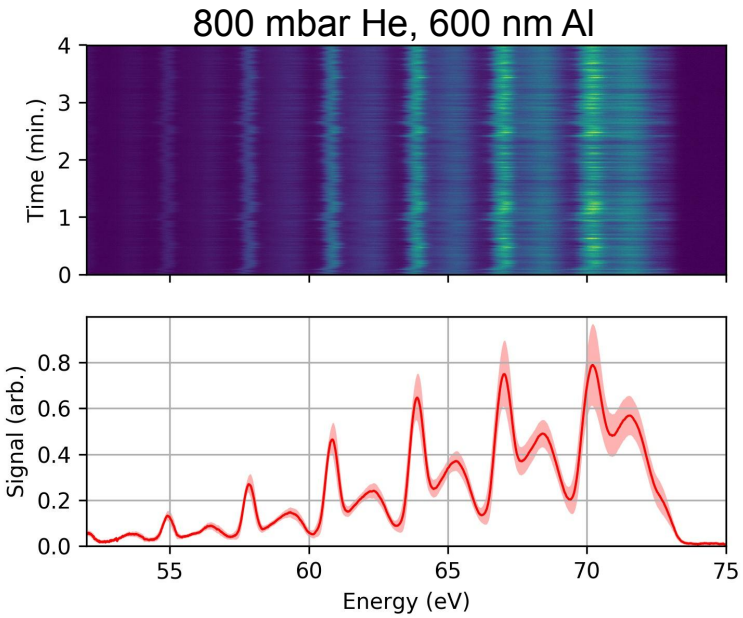


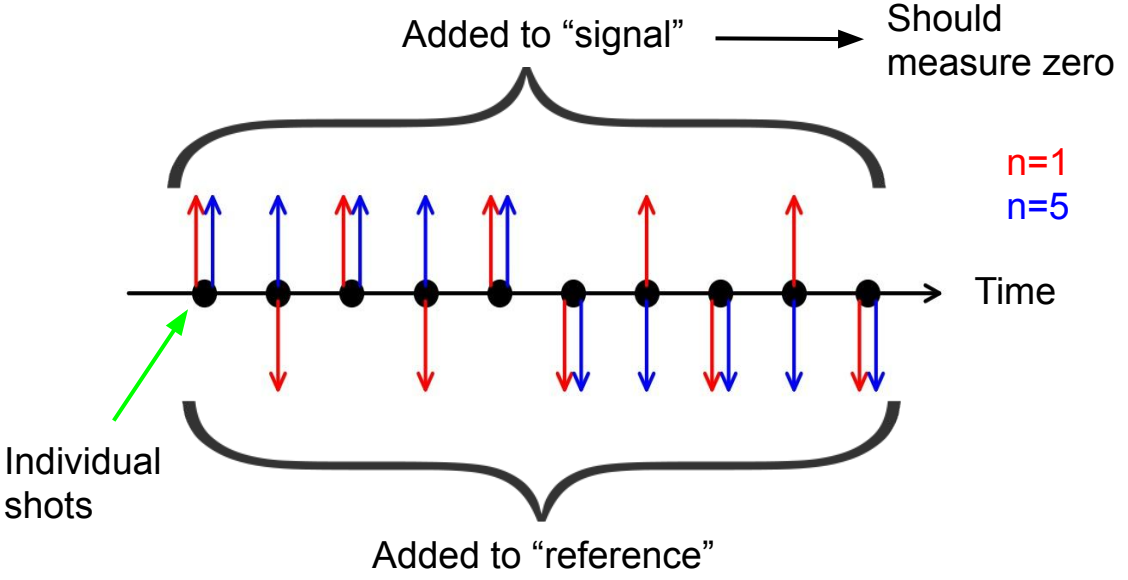


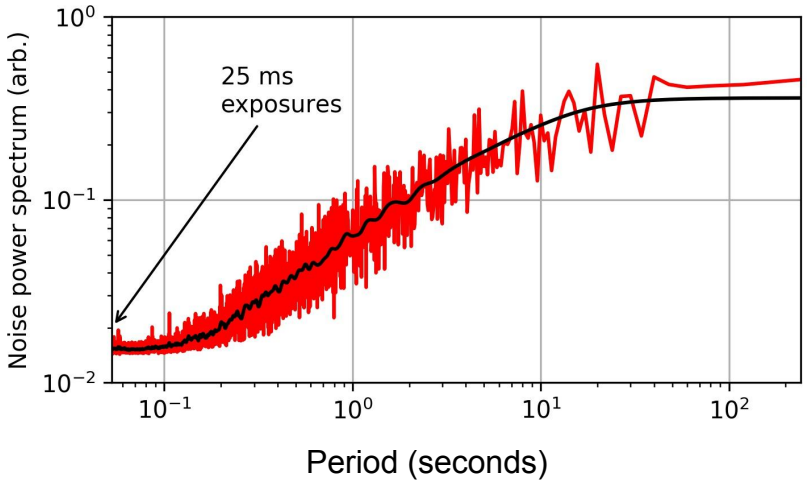

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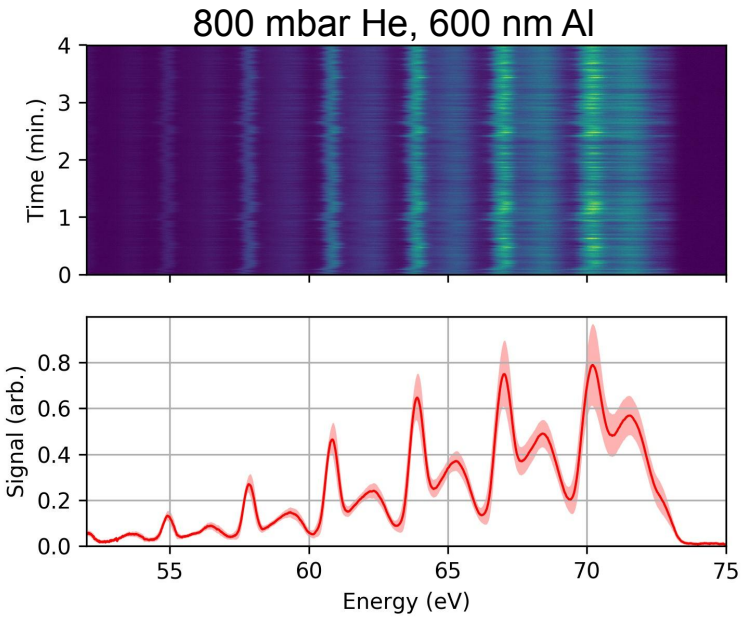
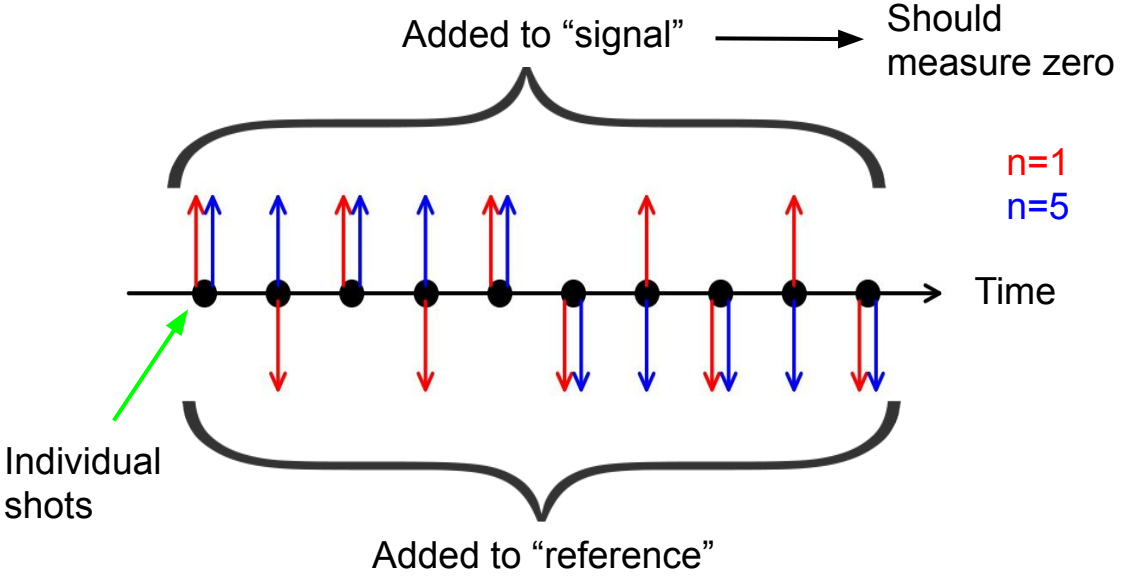
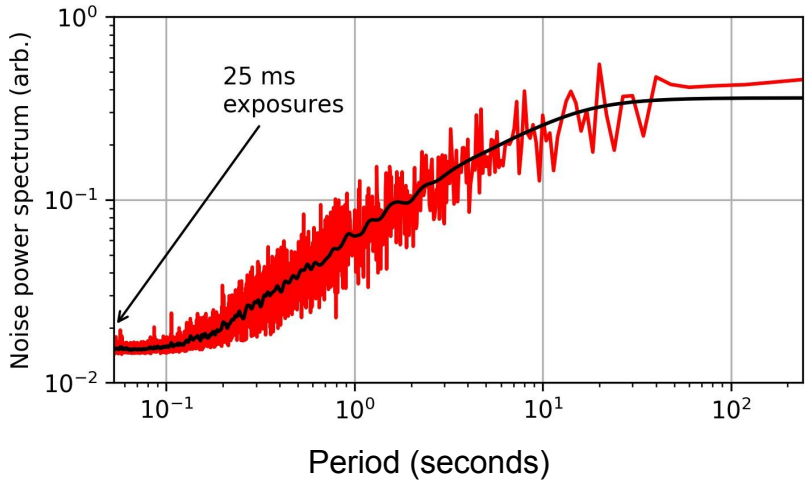
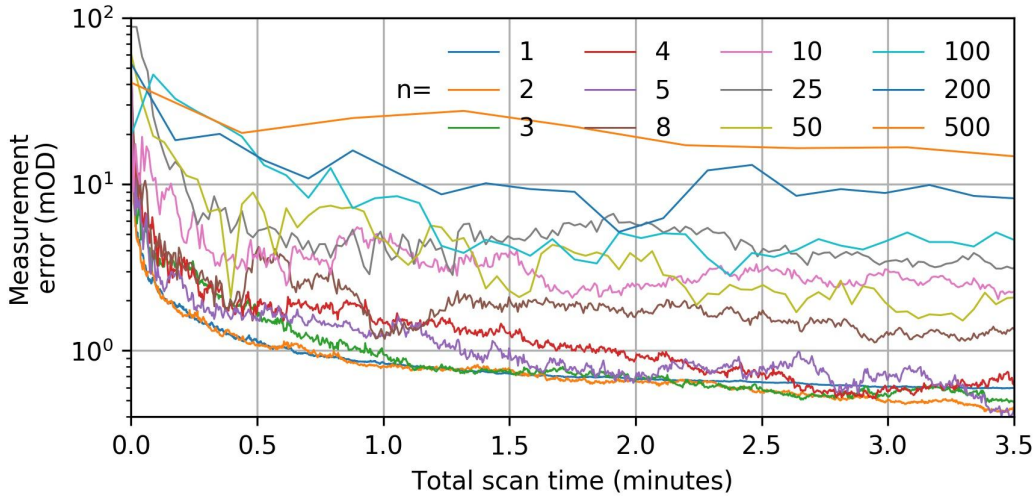


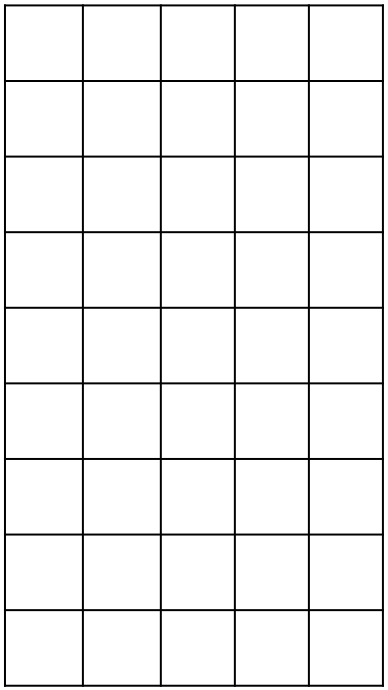



# Locking in at even higher frequencies

SLAC

- Expose n rows for Δt (using a slit)
- Shift them up (24 μs per row)
- When full, read out the full frame
- Become readout noise limited at Δt=1 ms due to slit (throwing away >90% of photons)



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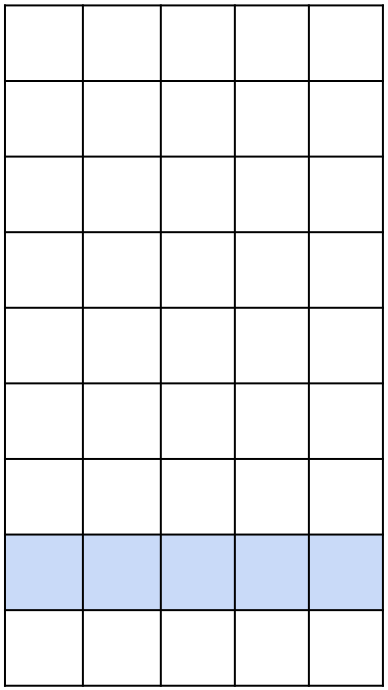

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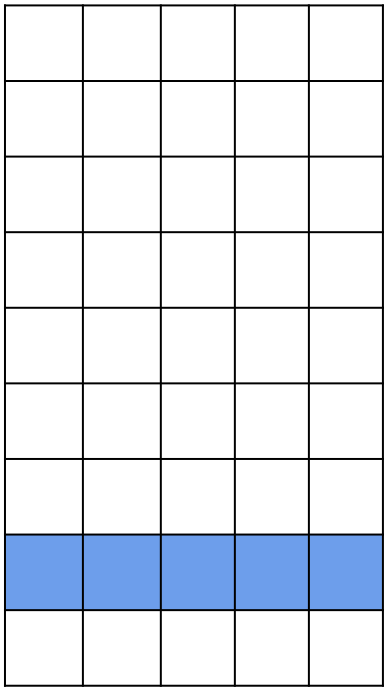

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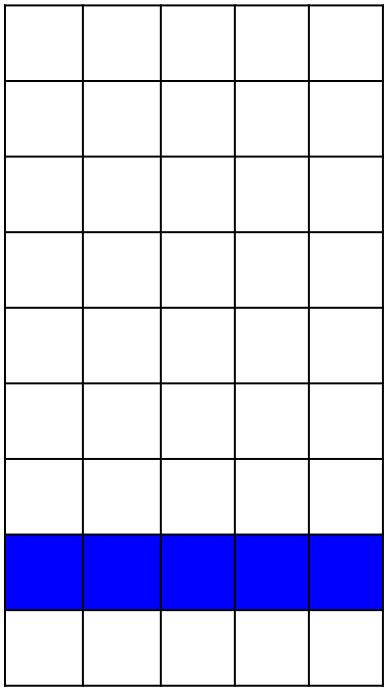

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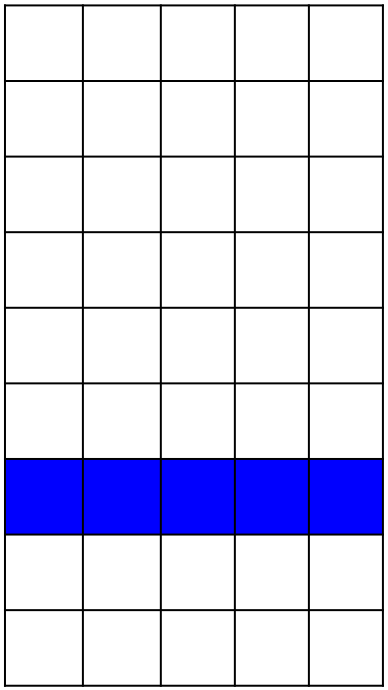

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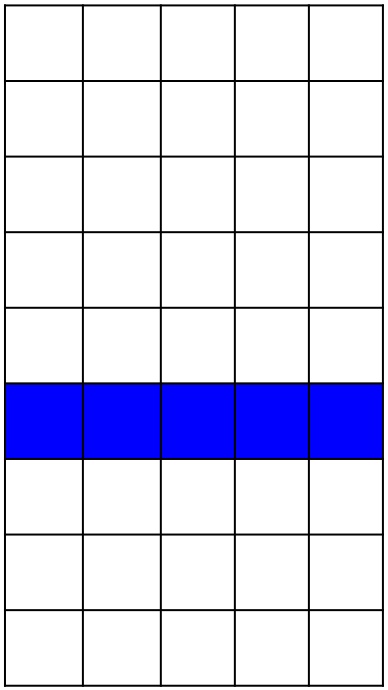

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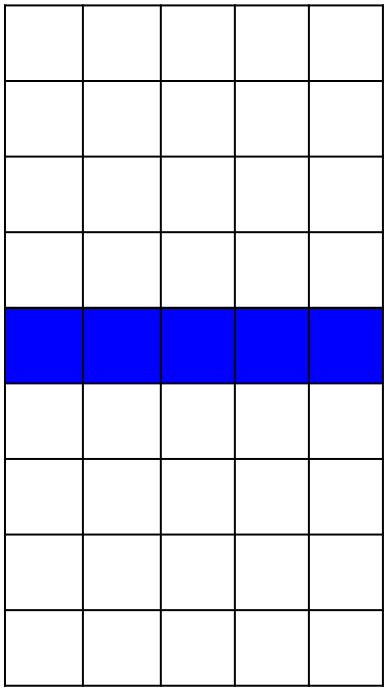

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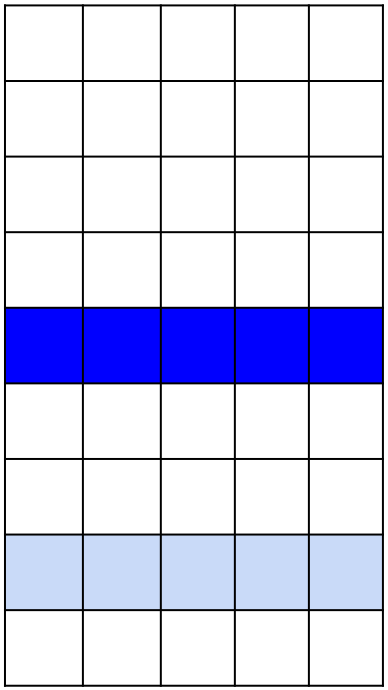
- Become readout noise limited at Δt=1 ms due to slit (throwing away >90% of photons)



# Locking in at even higher frequencies

- Expose n rows for Δt (using a slit)
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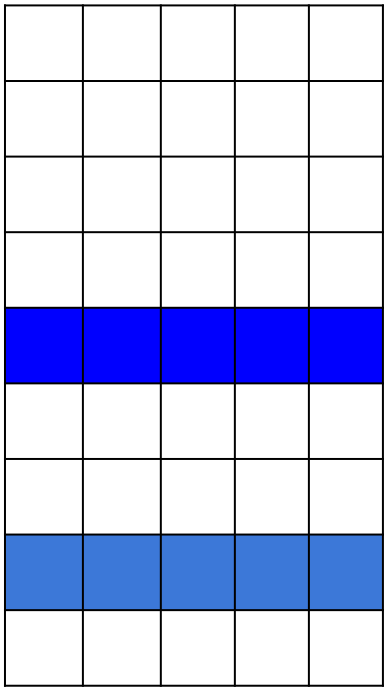

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SLAC

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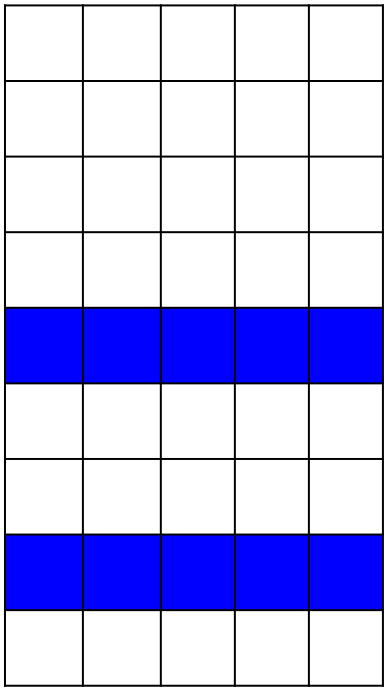

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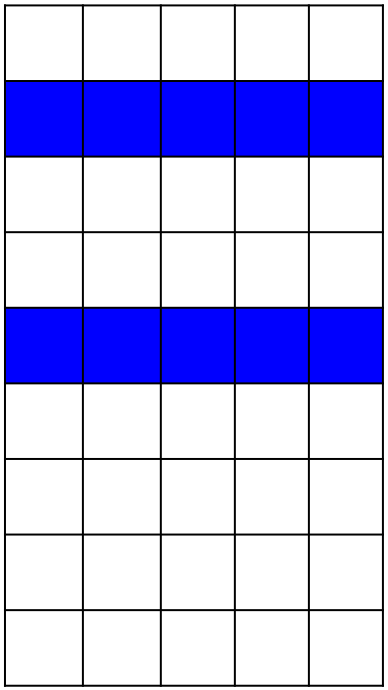

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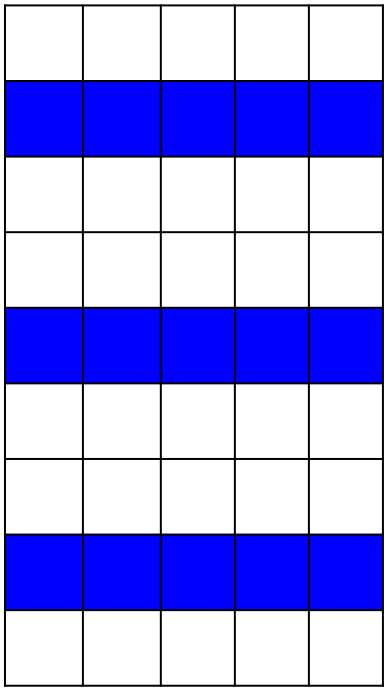

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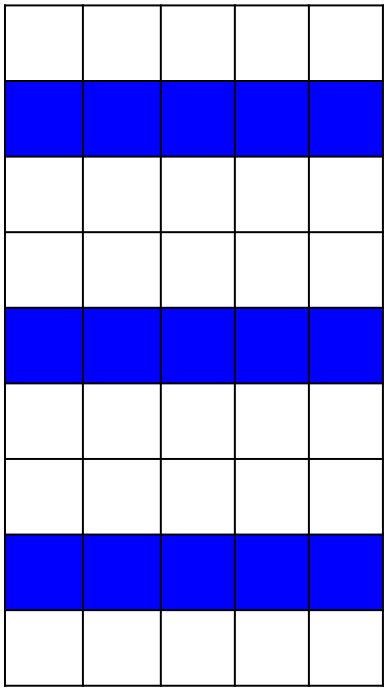

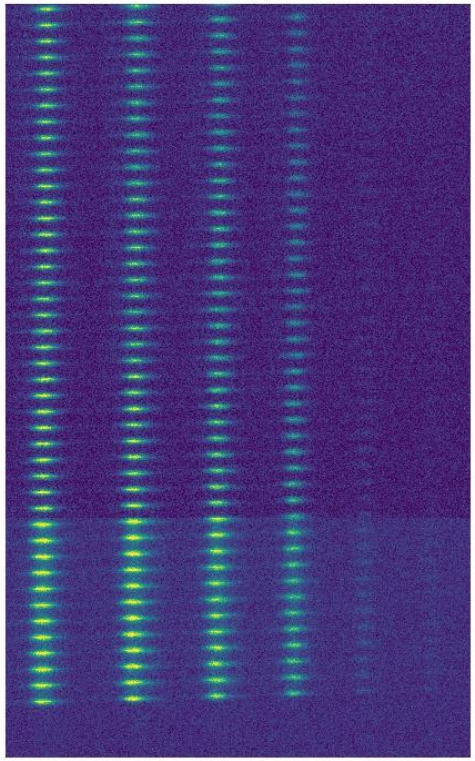

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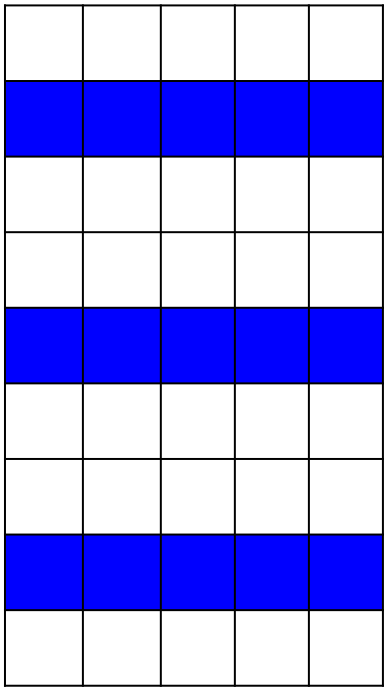
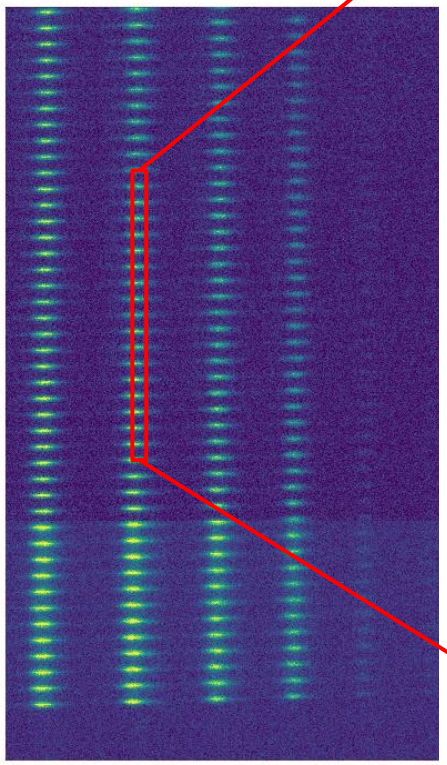
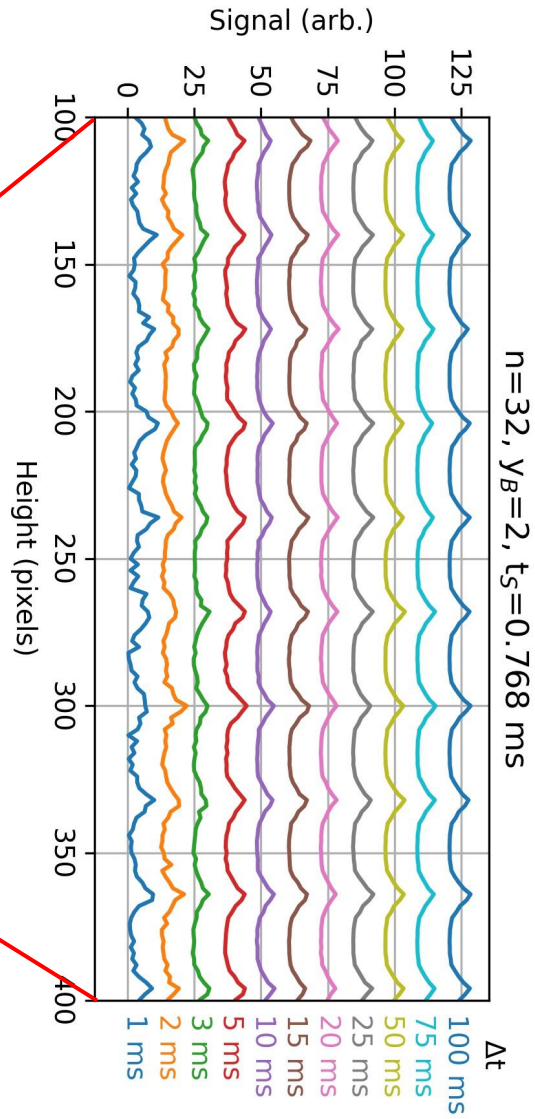


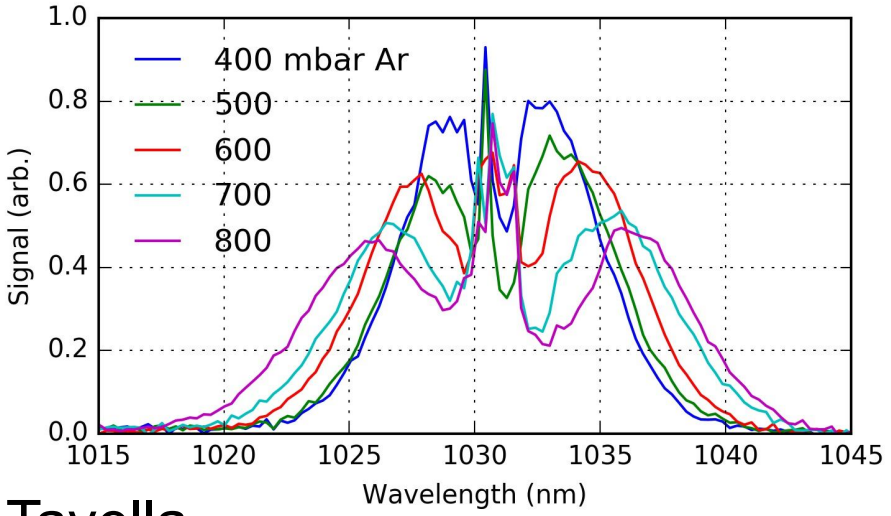




# Next steps and acknowledgements

SLAC

- Back to 1.8 um for the long term: extend HHG to 550+ eV
- First planned experiments in organic molecules at C, N, and O K-edges




PULSE

James Cryan
Amy Cordones-Hahn
Thomas Wolf

**LST Division**

Franz Tavella
Katalin Mecseki
Matt Windeler