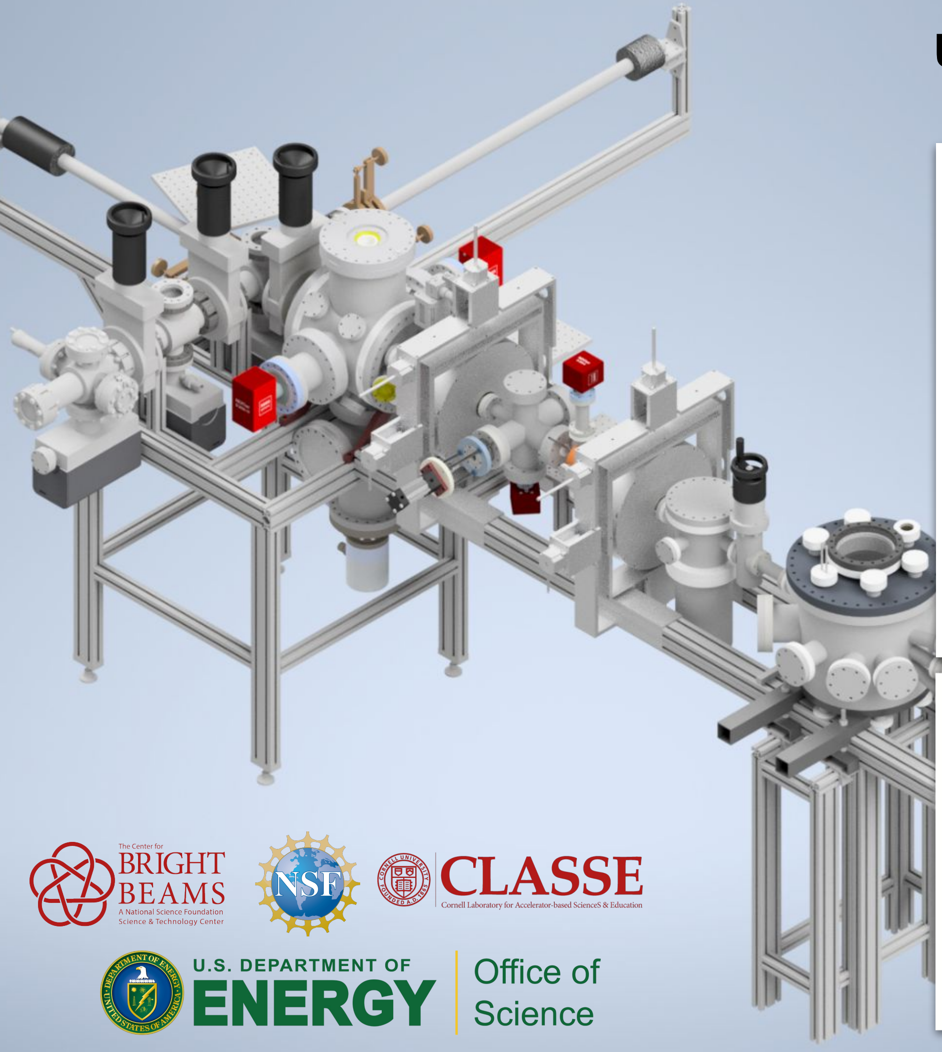
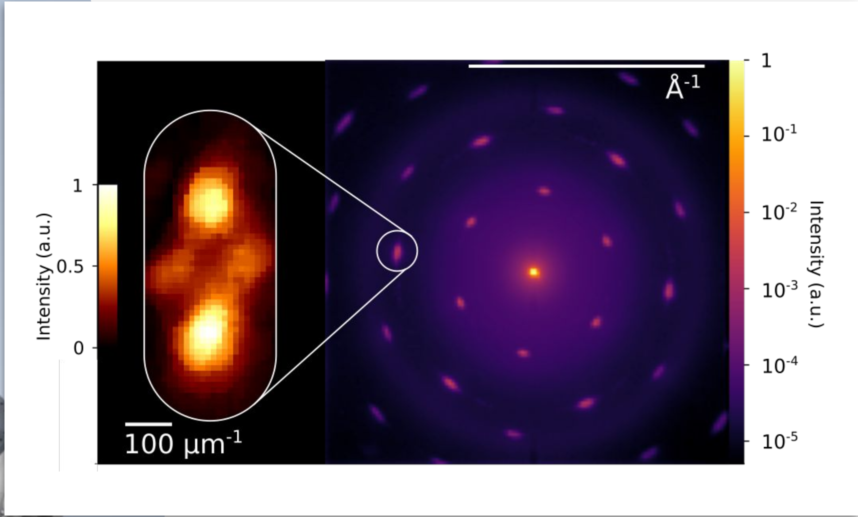
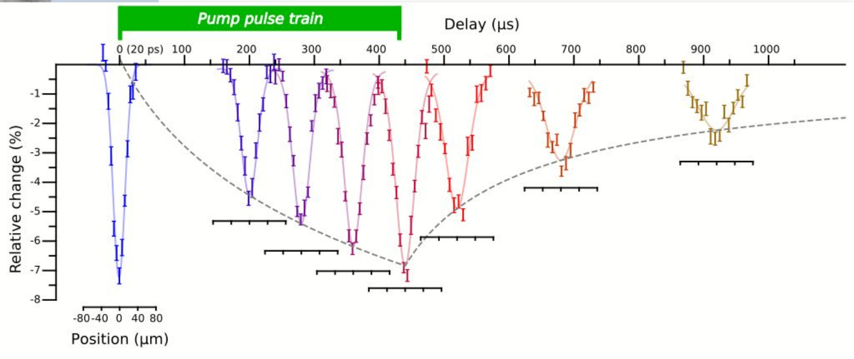
Ultrafast Electron Diffraction (UED) @Cornell







The Center for BRIGHT BEAMS
A National Science Foundation Science & Technology Center

NSF

CLASSE
Cornell Laboratory for Accelerator-based ScienceS & Education

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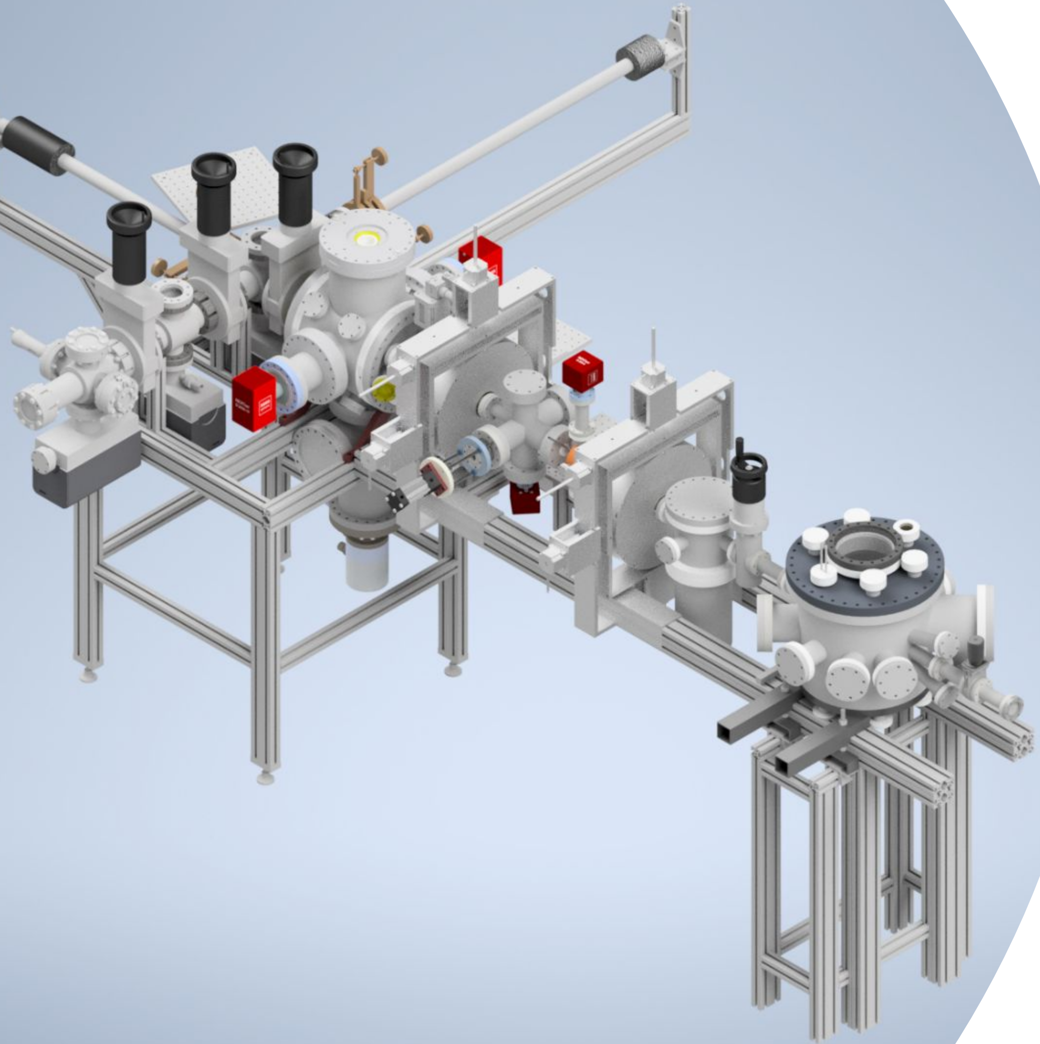
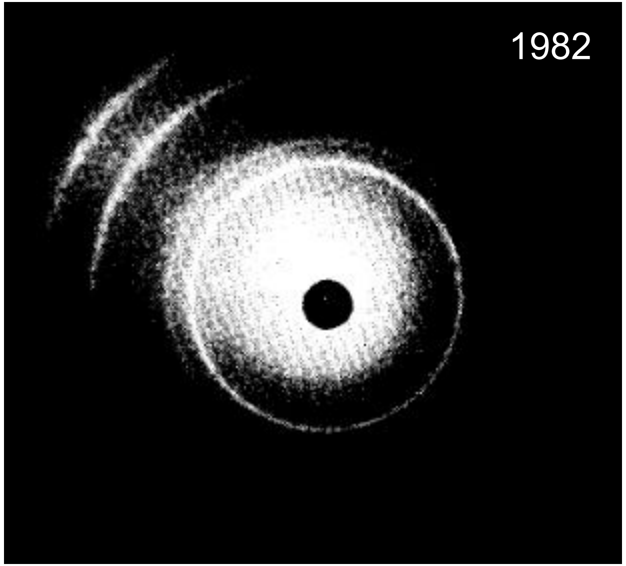


Outline:

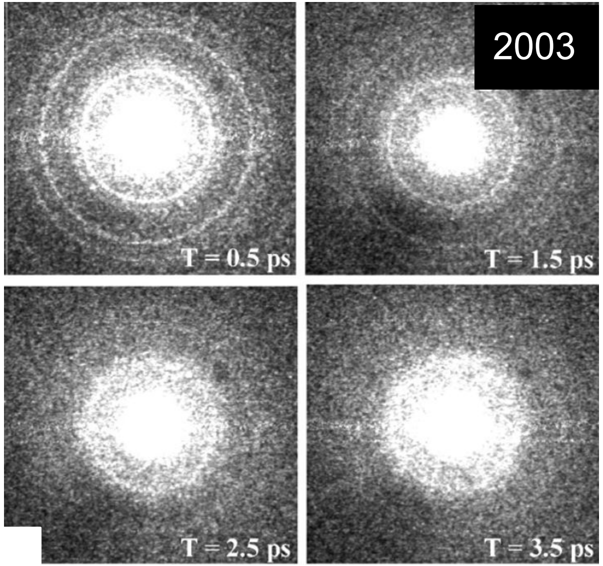
- UED: conceptual introduction
- Experimental setup
  - Beamline
  - Direct electron detector
- First science results
  - 2D Moire material
  - Scanning UED technique
  - Thermal diffusion data
- Summary and Outlook

Forty years of UED



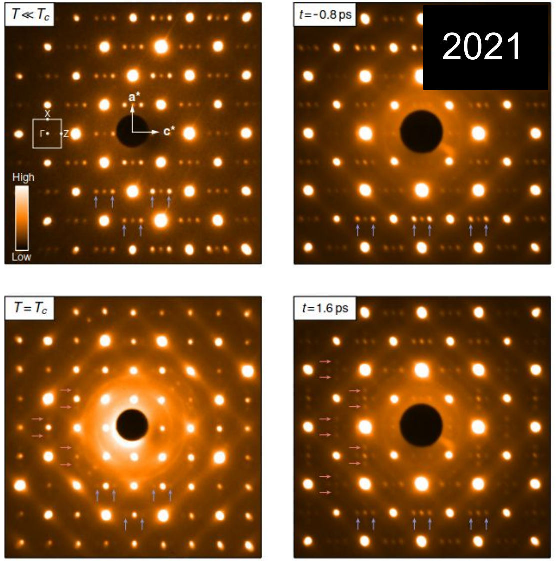
Mourou & Williamson, “Picosecond electron diffraction”, *Appl. Phys. Lett.* **41**(1) 1982

Single shot, 100 ps resolution



Siwick et al., “An atomic view of melting…”, *Science* **302**(5649) 2003

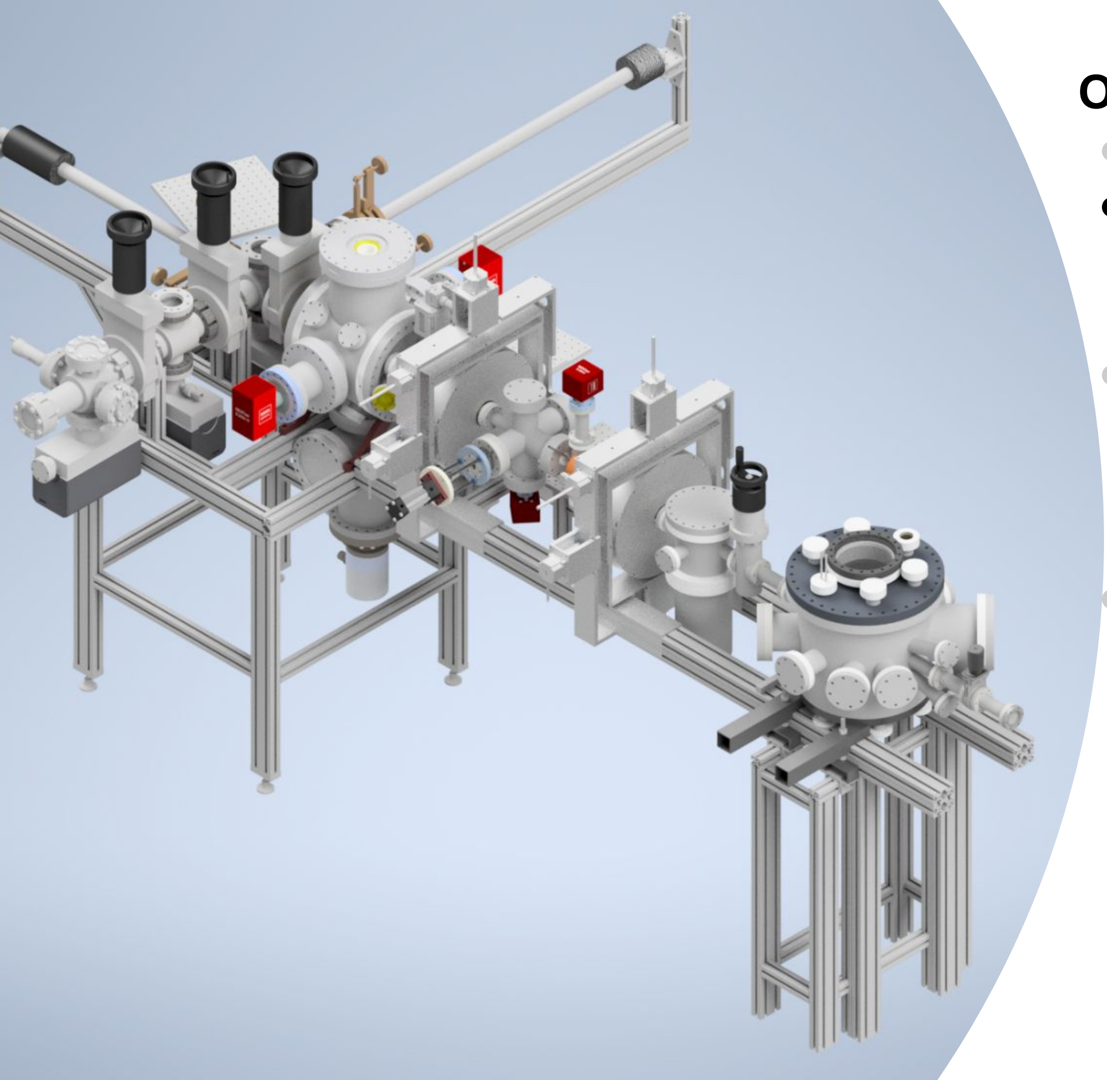
Single shot, 600 fs resolution



Zong et al., “Role of equilibrium fluctuations…”, *P.R.L.* **127** 2021

Stroboscopic, 300 fs resolution

One trend is toward higher sensitivity in stroboscopic measurements, to study diffuse scattering and *superlattice* structure



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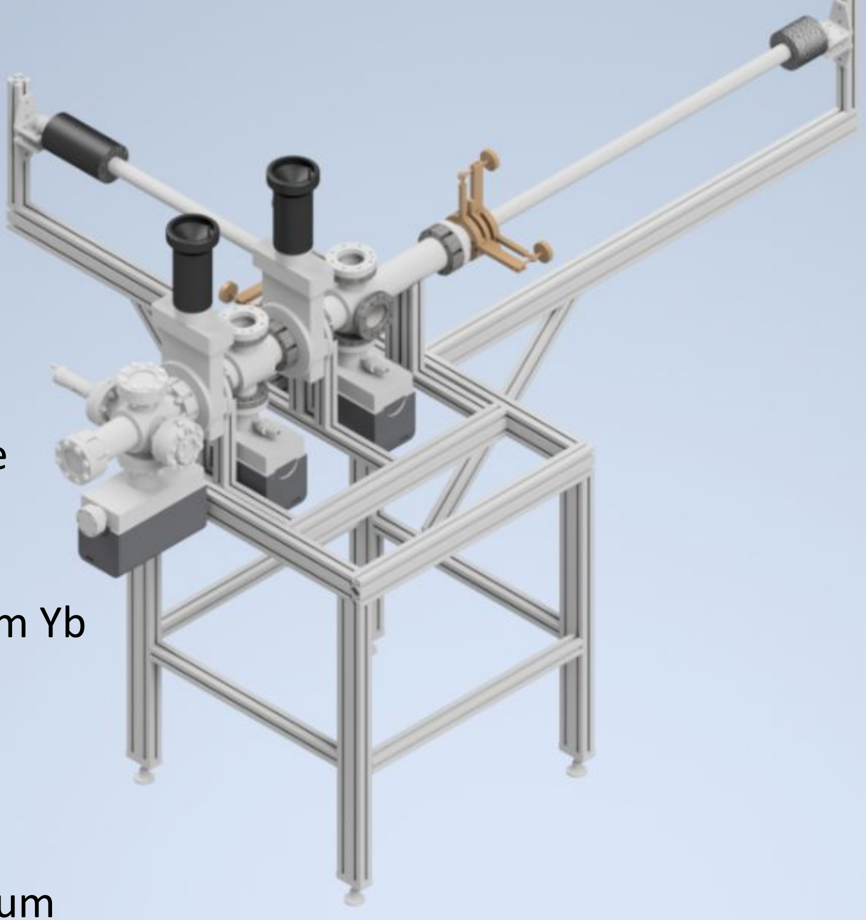
Beamline

High brightness, semiconductor electron source (Na-K-Sb)

650 nm photo-emission wavelengths matches the source bandgap

50 W @ 250 kHz, 1030 nm Yb fiber laser (AS Tangerine) feeds optical parametric amplifier (AS Mango)

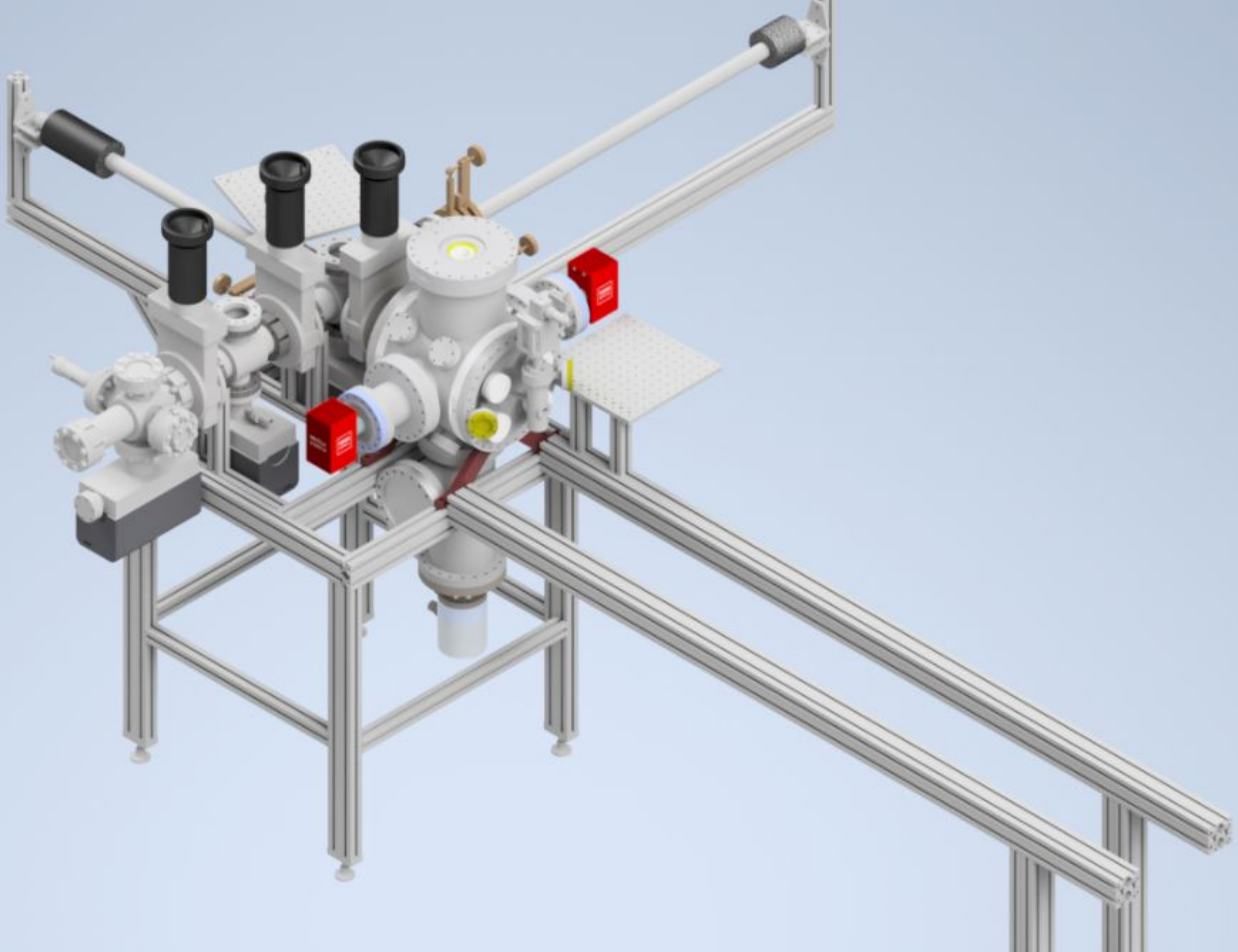
Source is extremely vacuum sensitive (XHV)



W. H. Li, et al., (2022) *Structural Dynamics*, *9*(2)

Beamline

200 kV DC gun typically operated at 140 kV (for vacuum safety)



W. H. Li, et al., (2022) *Structural Dynamics*, *9*(2)

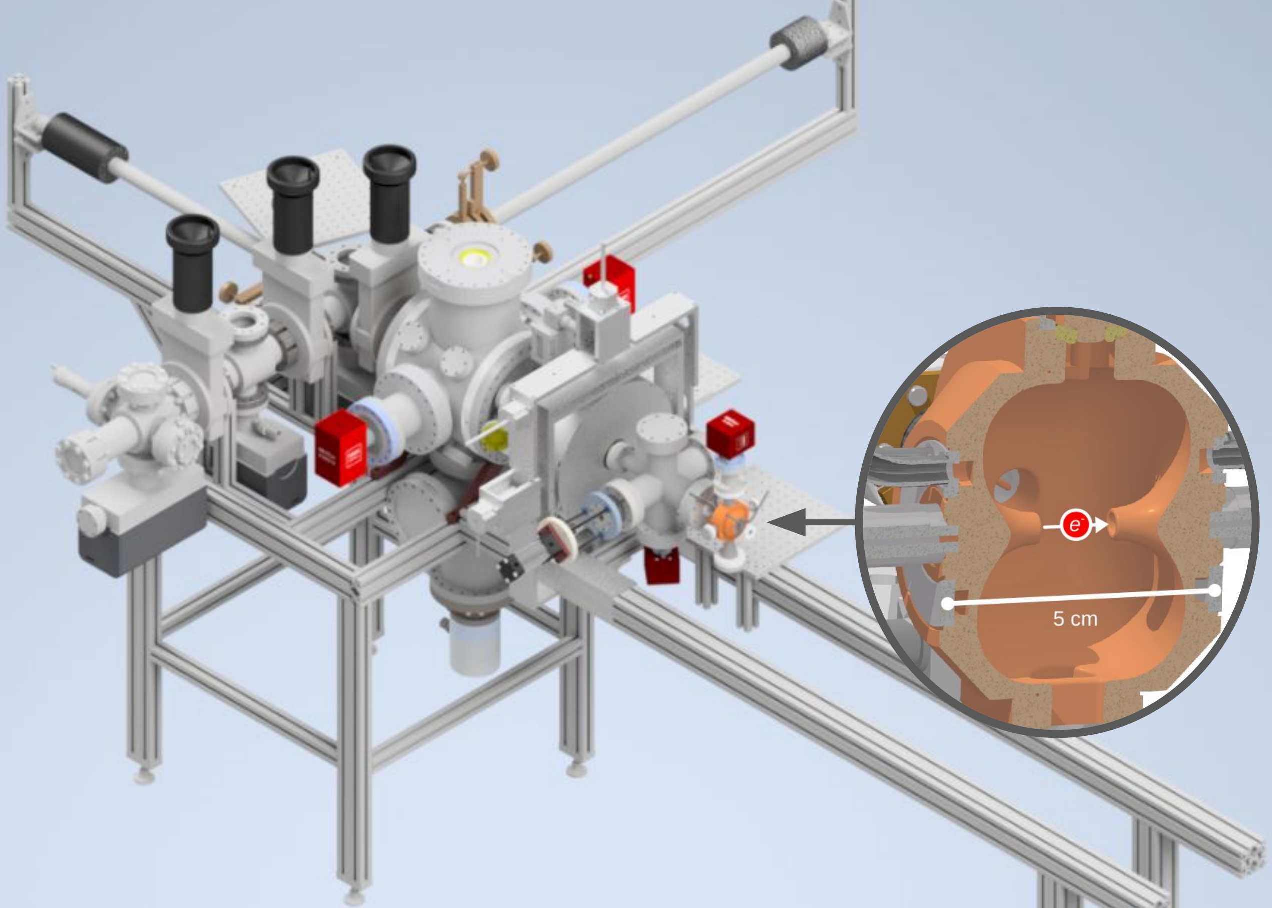
Beamline

Radio-frequency bunching cavity

Long pulse length at cathode: 10 ps

Bunching after acceleration mitigates space-charge

100 fs rms bunch-length at sample



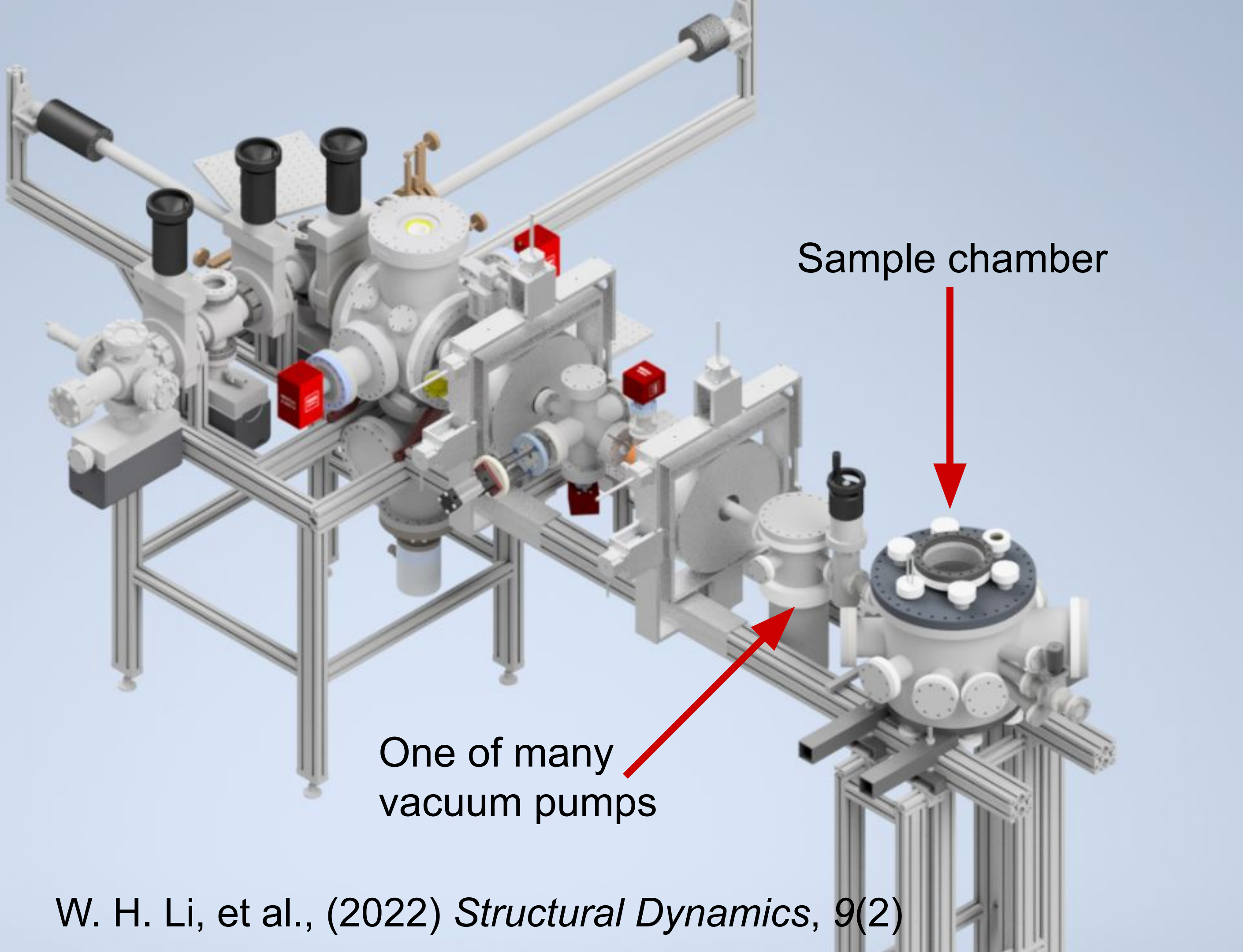
W. H. Li, et al., (2022) *Structural Dynamics*, *9*(2)

Beamline

Sample chamber

Pressure 10^4 × higher than gun (UHV)

515 nm pump pulses, second-harmonic of Tangerine



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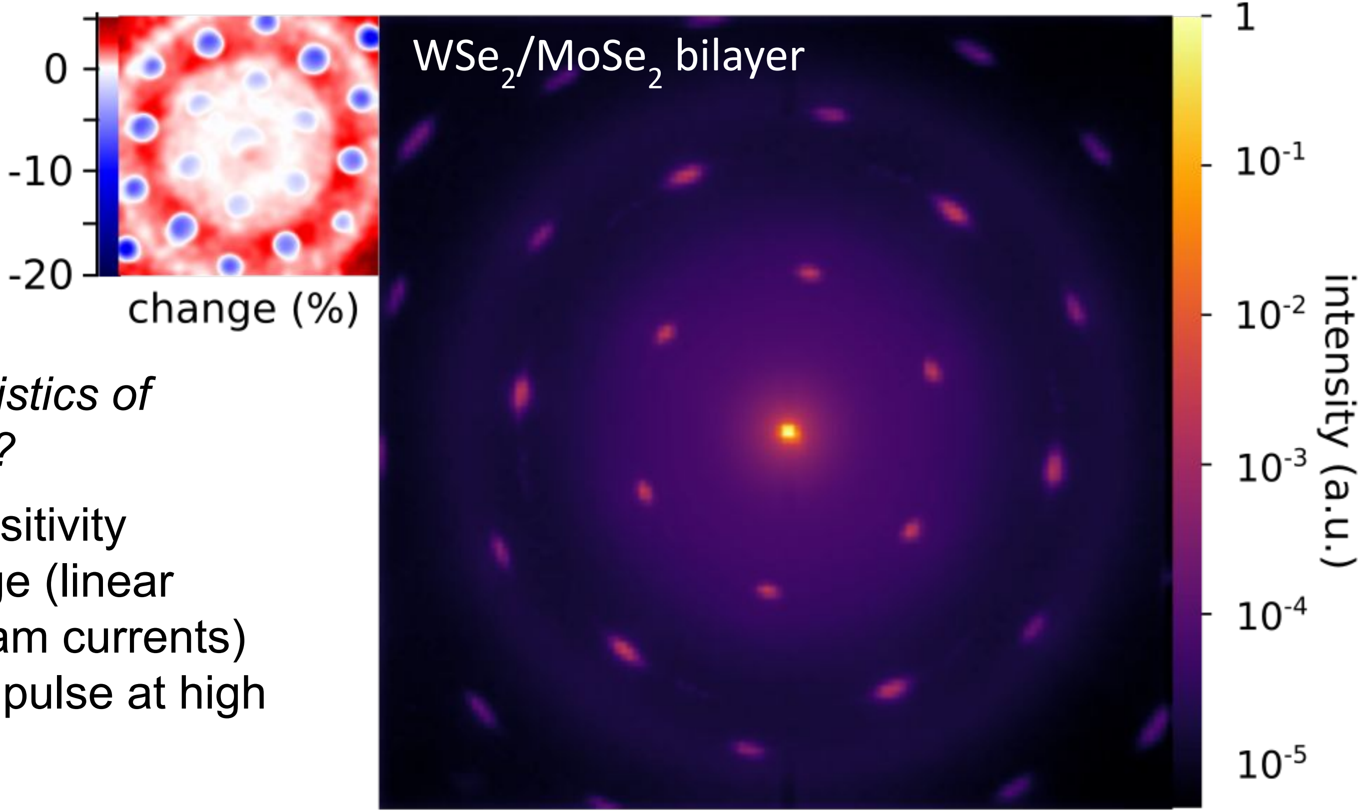
Direct electron detector



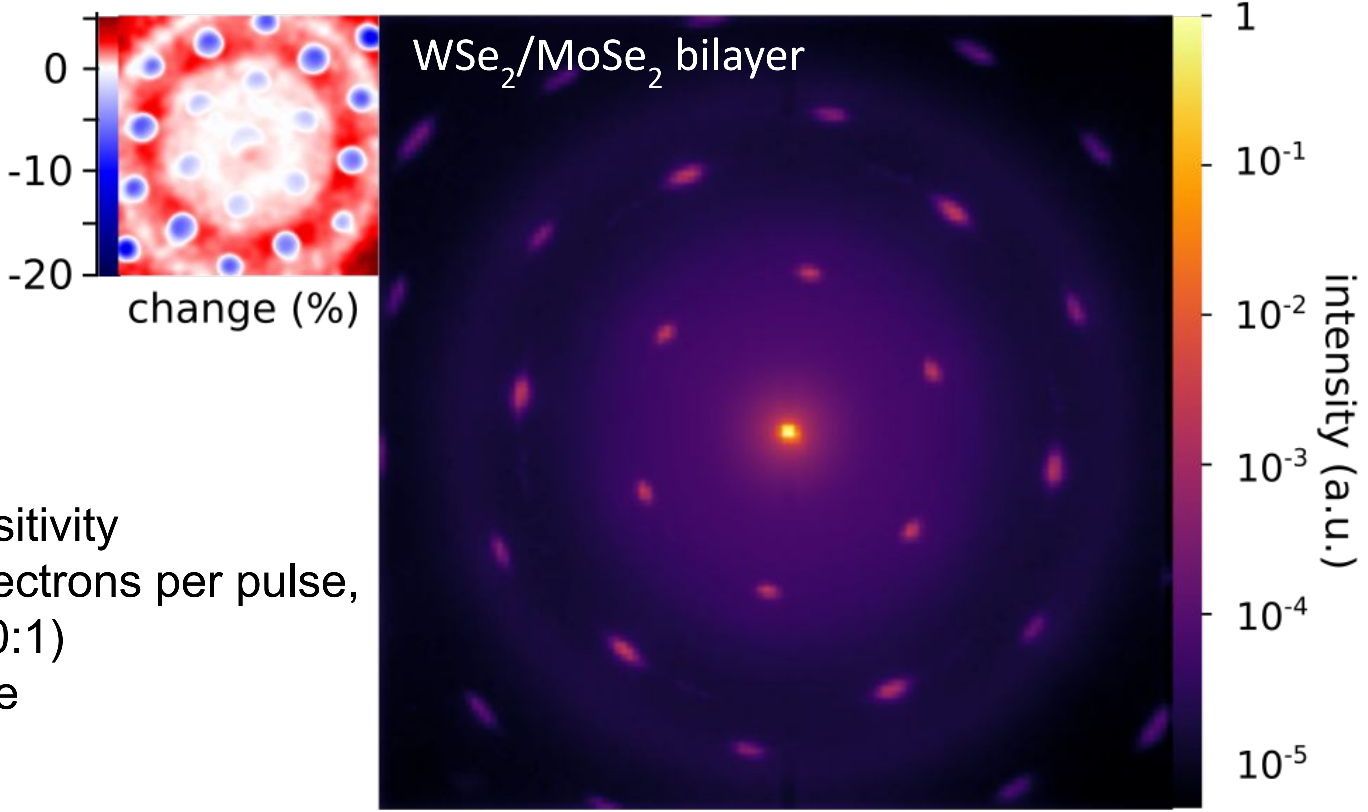
What are the characteristics of the ideal UED detector?

- Single-particle sensitivity
- High dynamic range (linear response at all beam currents)
- One exposure per pulse at high rep-rate

Direct electron detector

Electron
Microscope
Pixel
Array
Detector
(EMPAD)

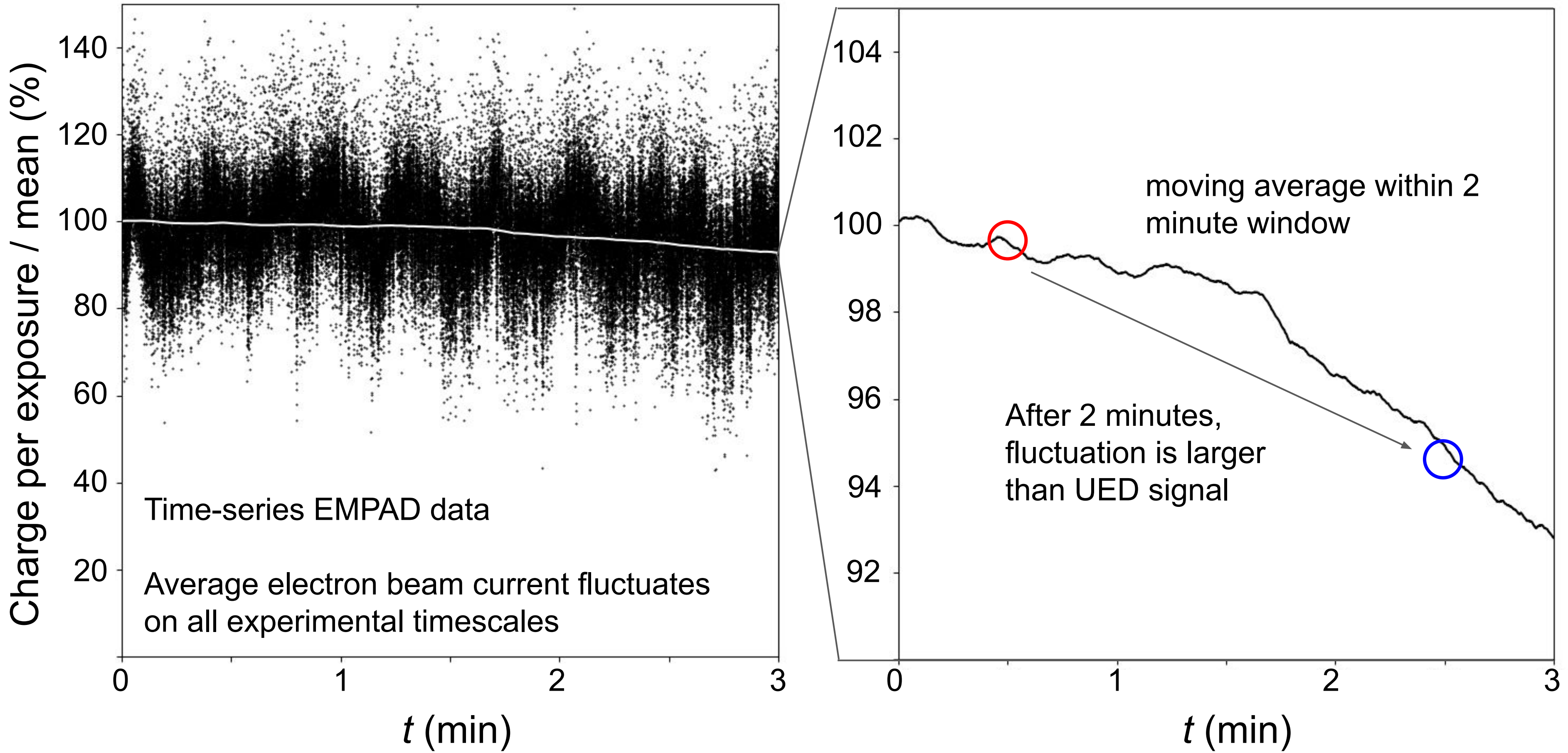
- single-particle sensitivity (saturates at 60 electrons per pulse, signal-to-noise 100:1)
- 10^6 dynamic range
- 1 kHz frame-rate



Thank you to David Muller, Sol Gruner, Julia Thom-Levy and Mark Tate for helping to deploy the EMPAD for UED

Detector performance

Eliminating "flicker-noise" with lock-in detection



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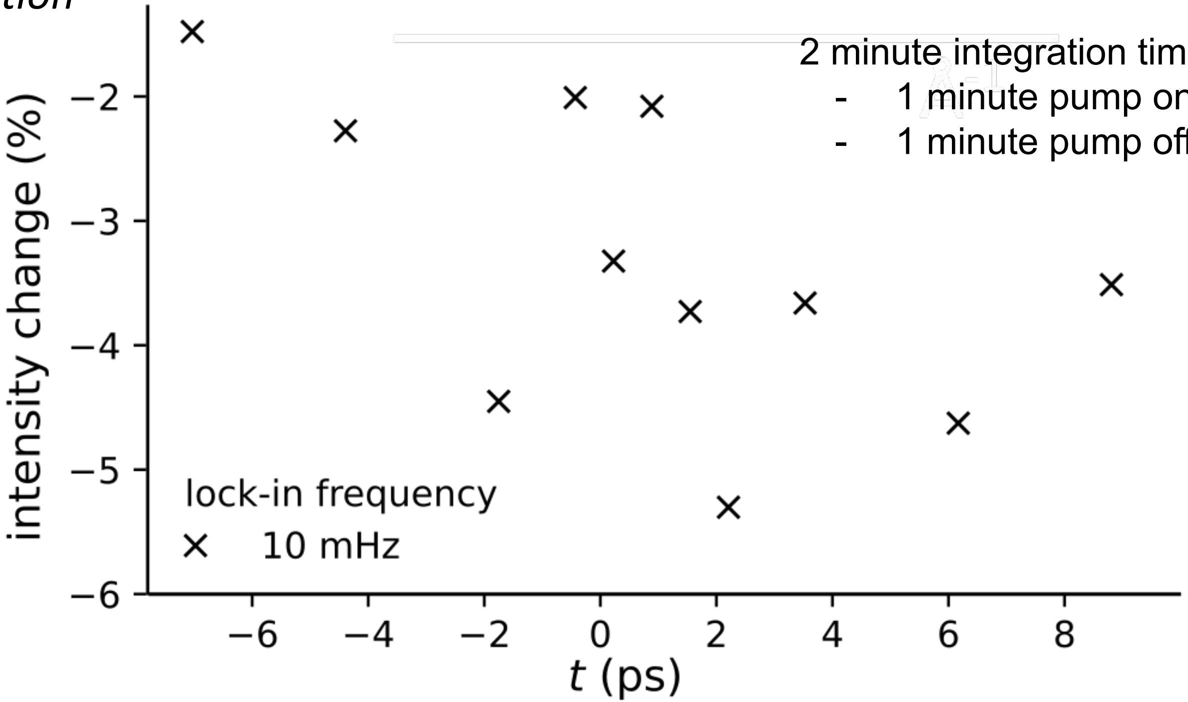

2 minute integration time per point:

- 1 minute pump on
- 1 minute pump off

"Lock-in": cycling between pumped and unpumped exposures

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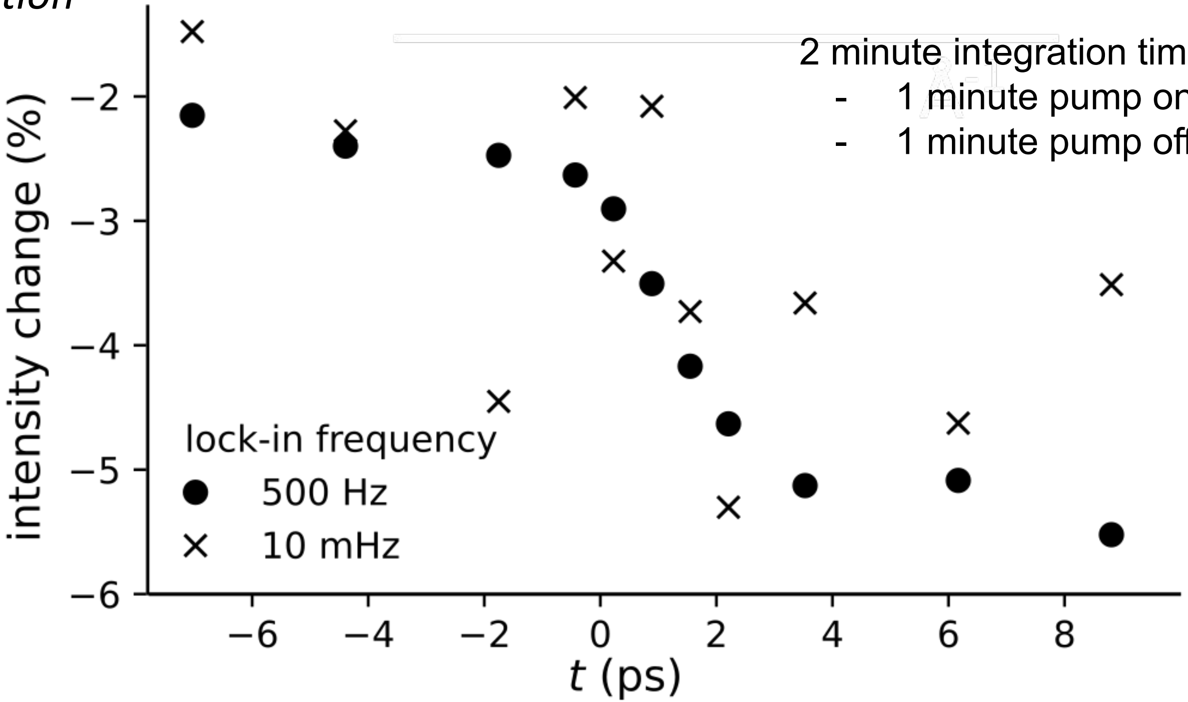

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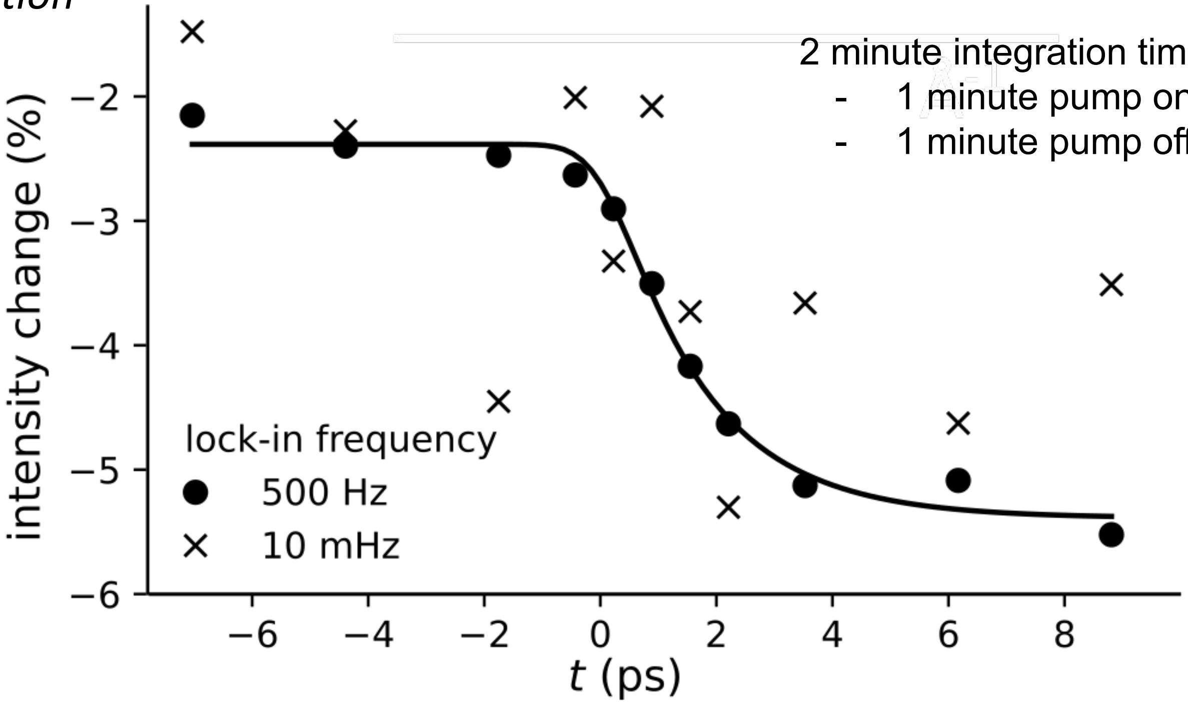

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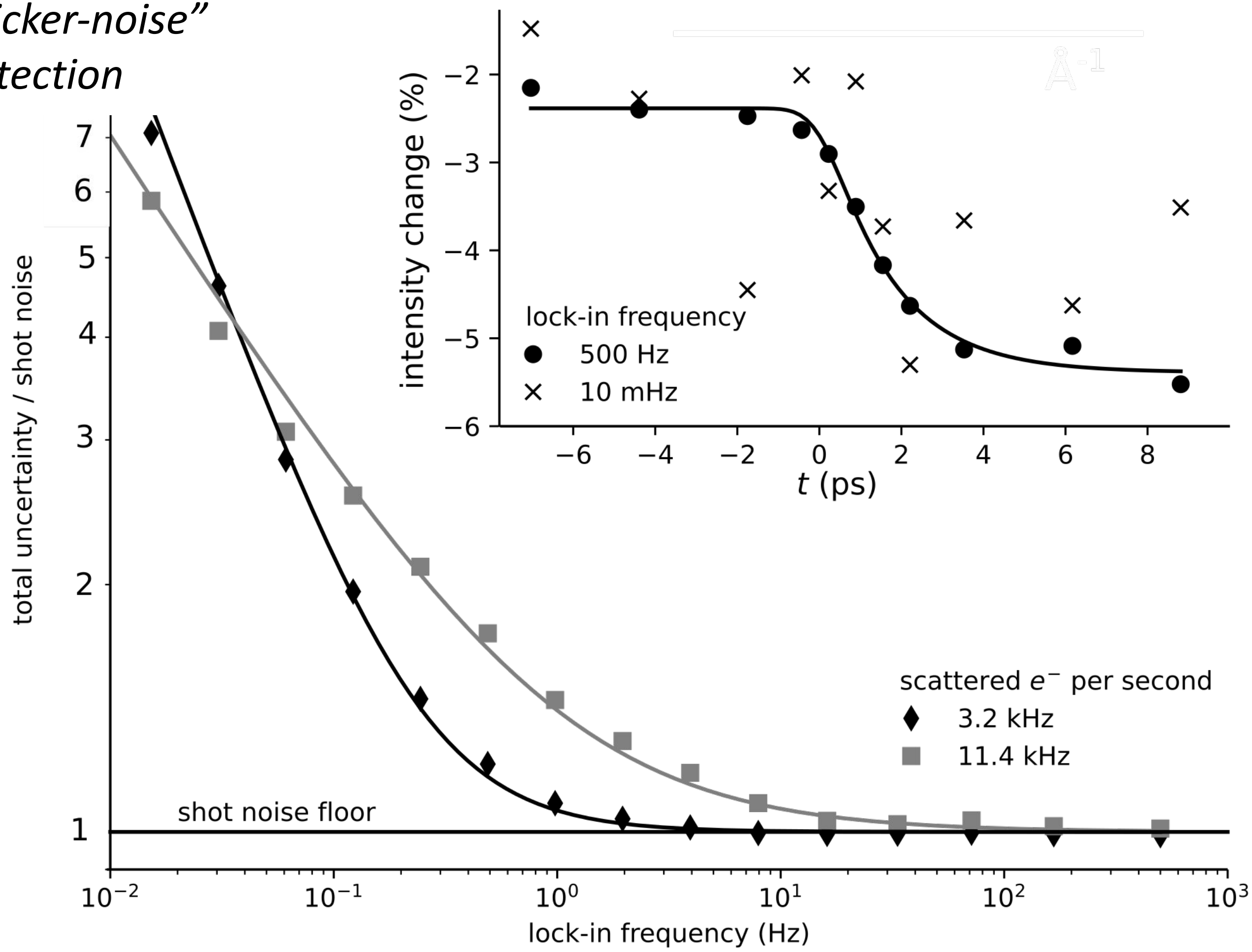
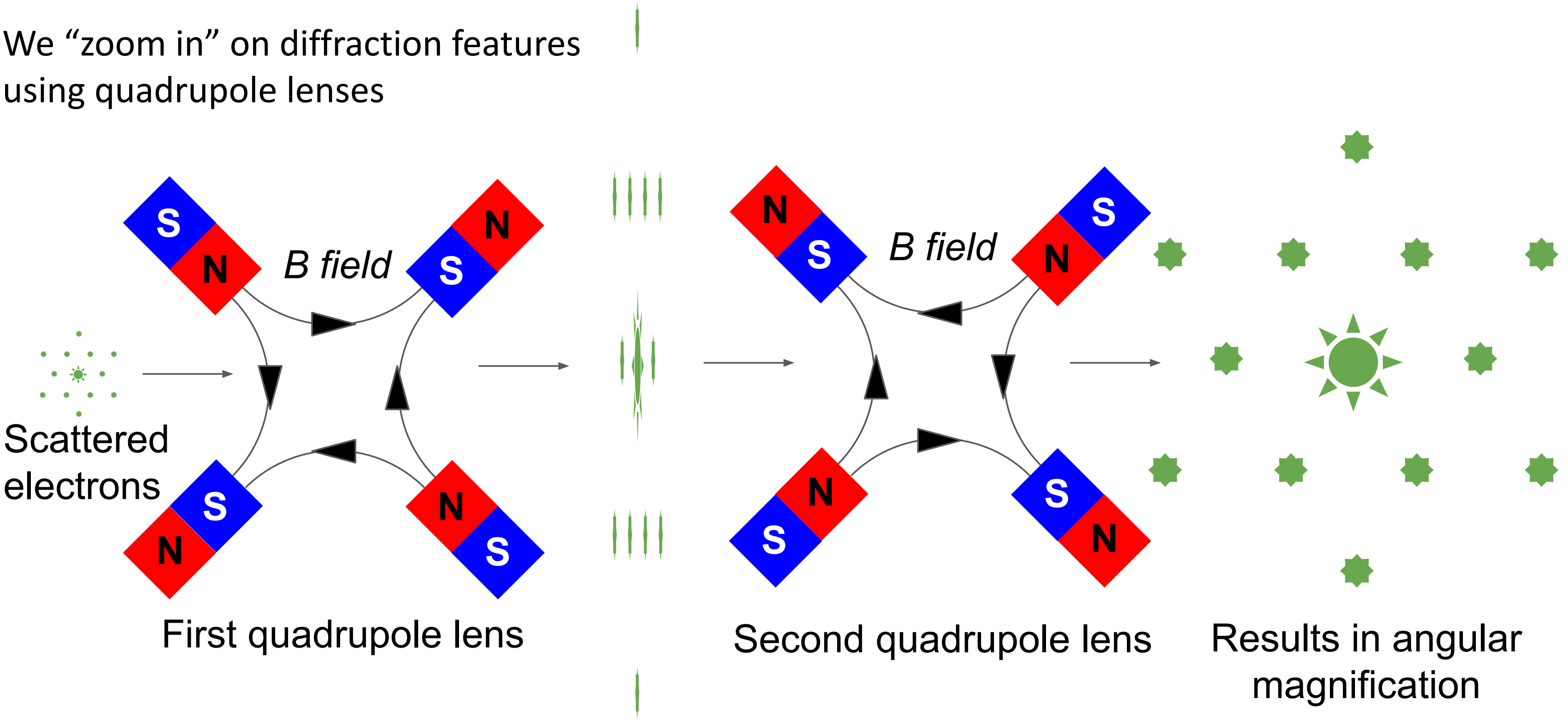


Detector performance

Variable camera length electron optics

We “zoom in” on diffraction features using quadrupole lenses

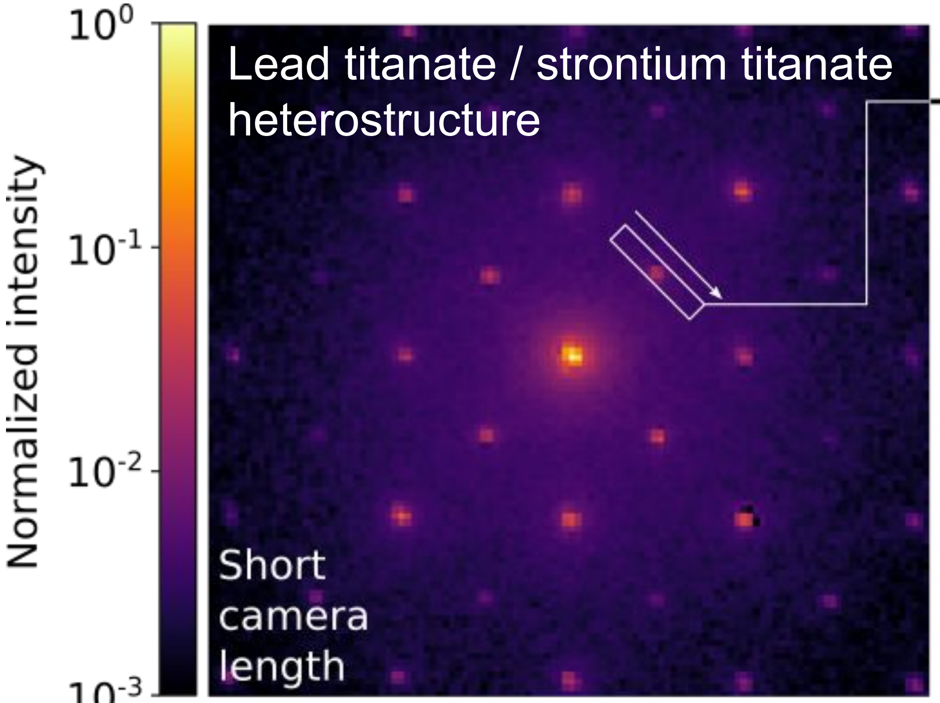


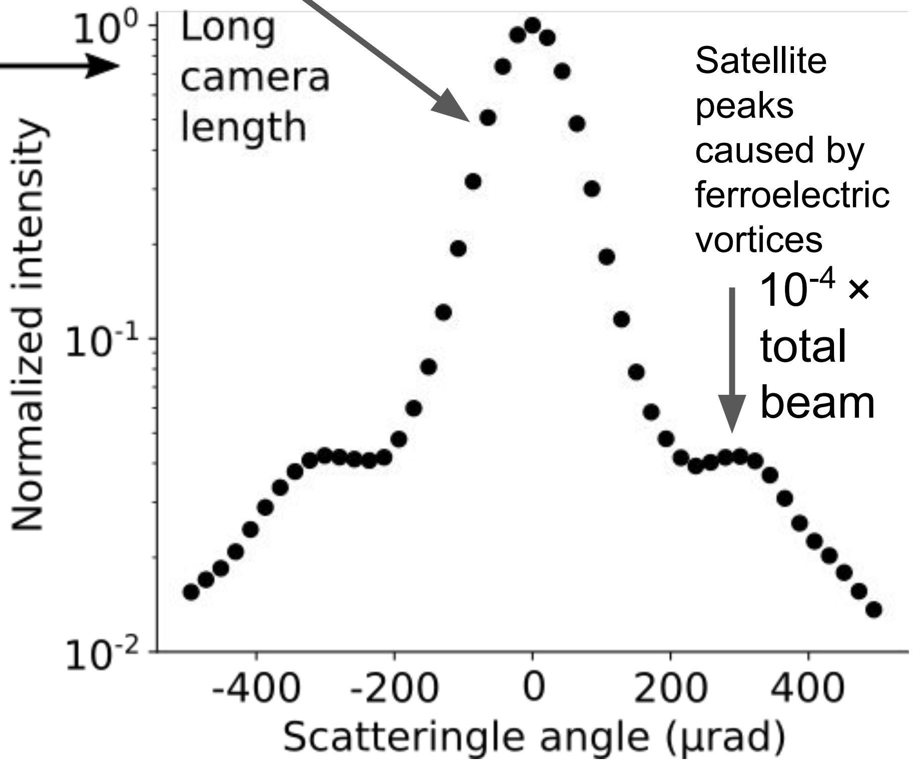
Detector performance

Variable camera length electron optics

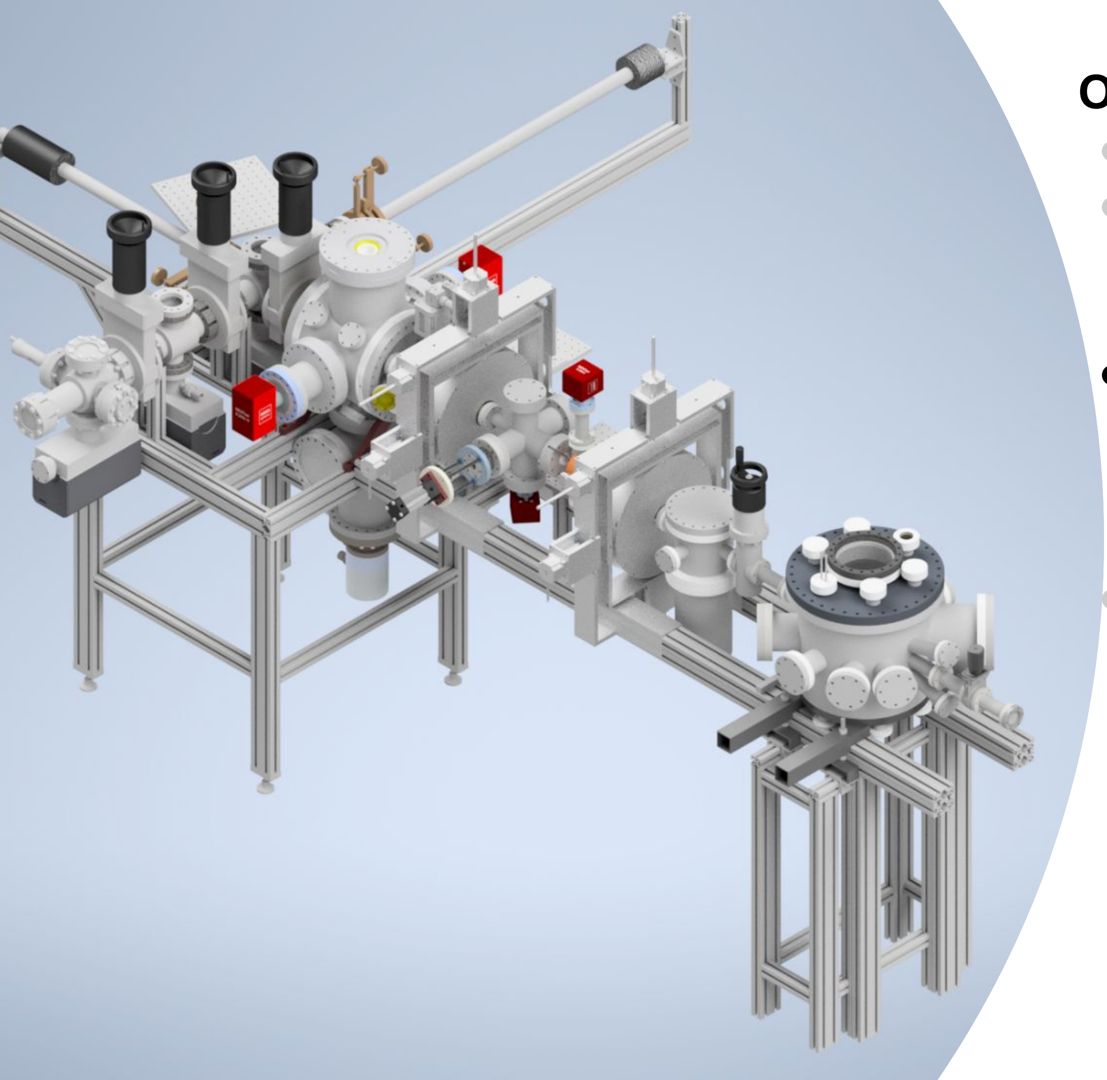
Spot size on sample < 3 μm
Full width at half maximum < 100 μrad
Transverse emittance ~ 100 pm

Sample prepared at Berkeley by Ramesh group





Thanks to Muller group for loaning sample



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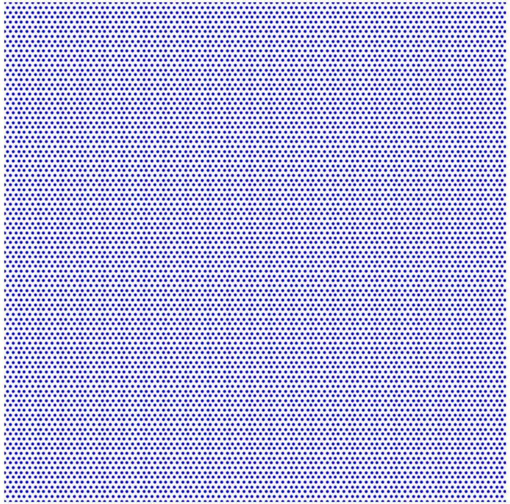
2D Moire material

Probing long-wavelength periodic lattice distortions

Samples prepared at Stanford by Fang Liu and Helen Zeng

$MoSe_2$ monolayer



Real space simulation

2D Moire material

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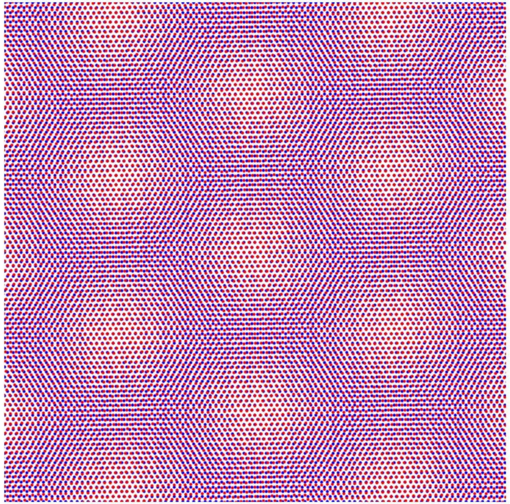

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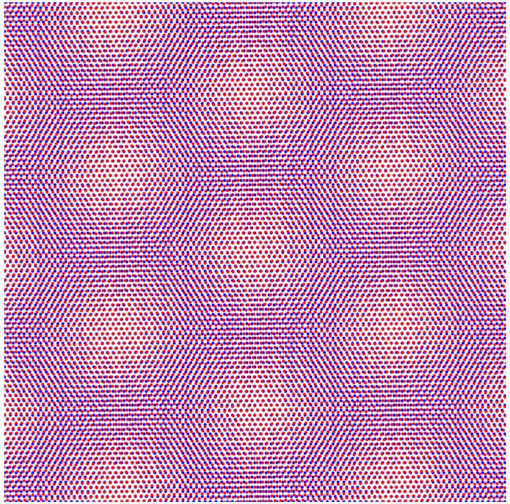

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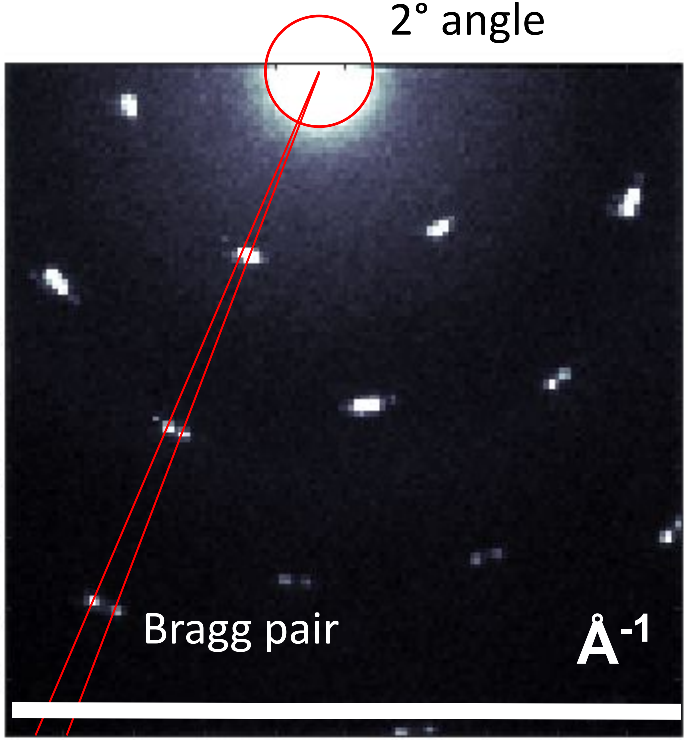

Reciprocal space experiment

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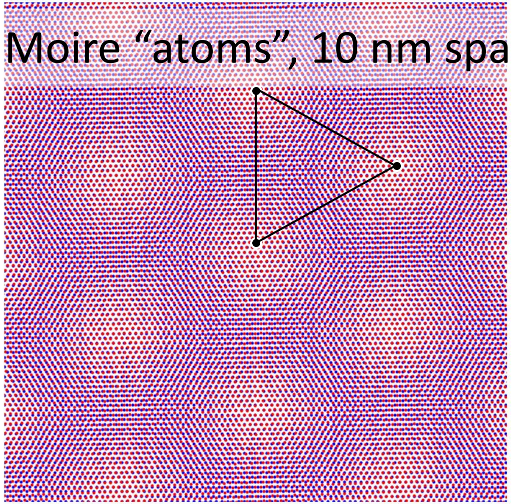

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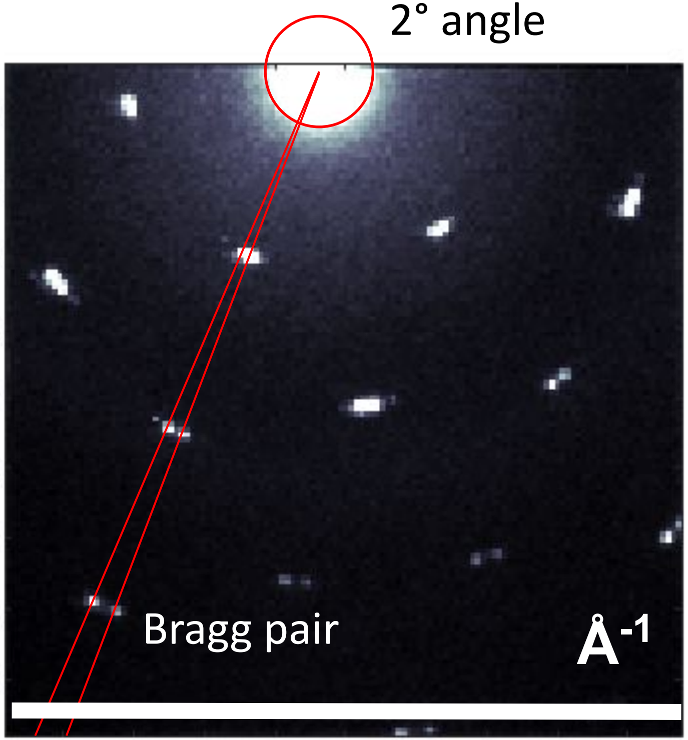

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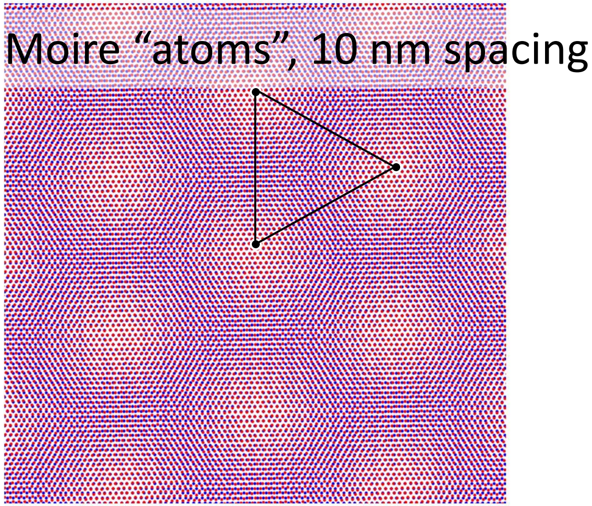

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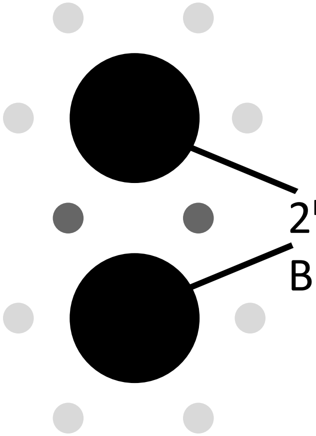
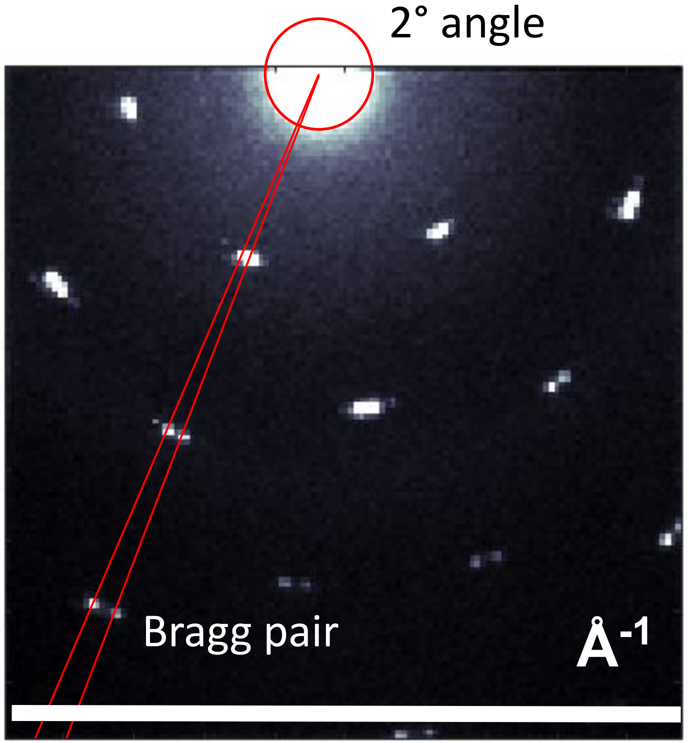


2nd order Bragg pair



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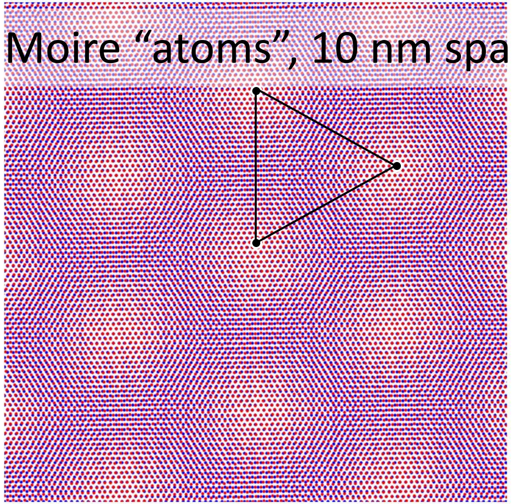

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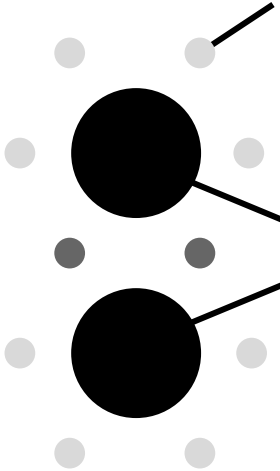

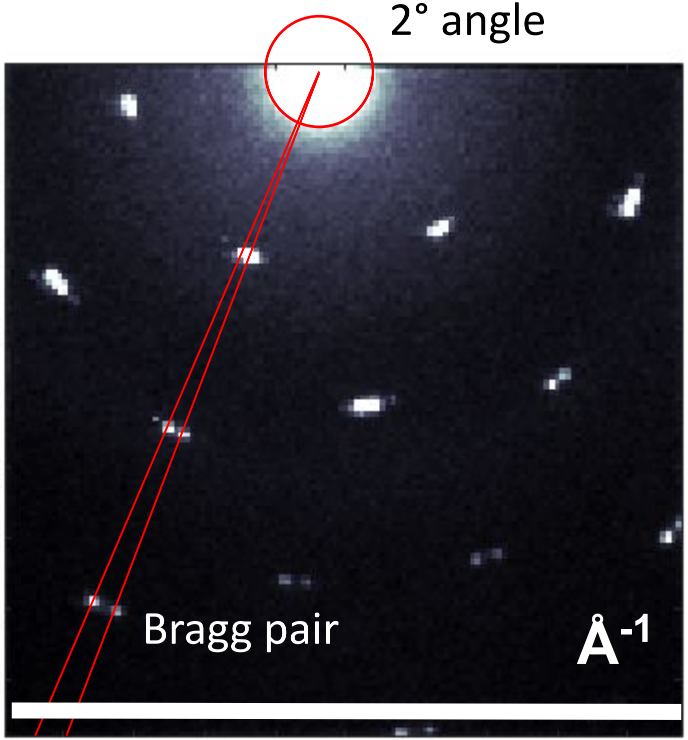

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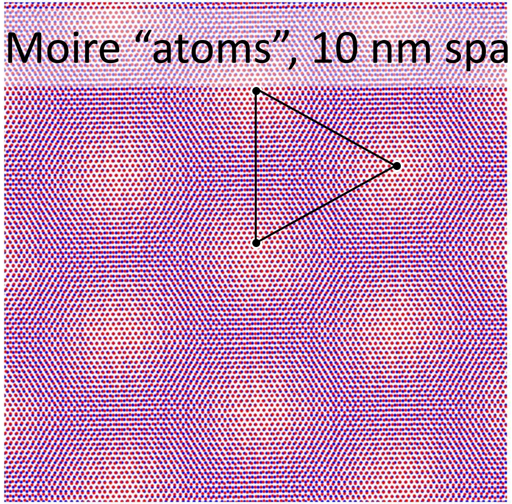

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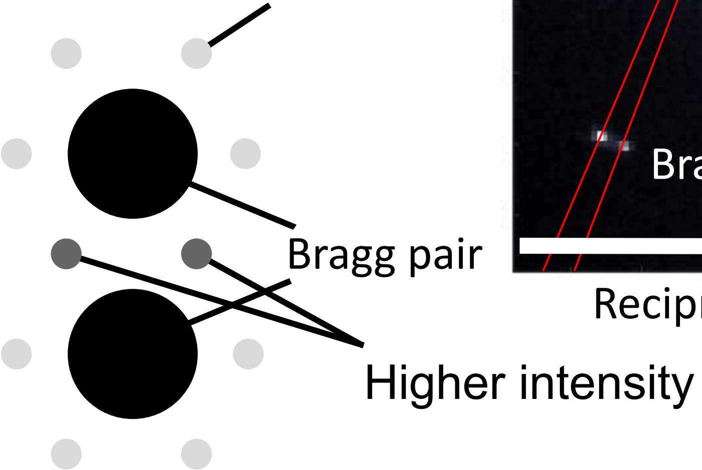

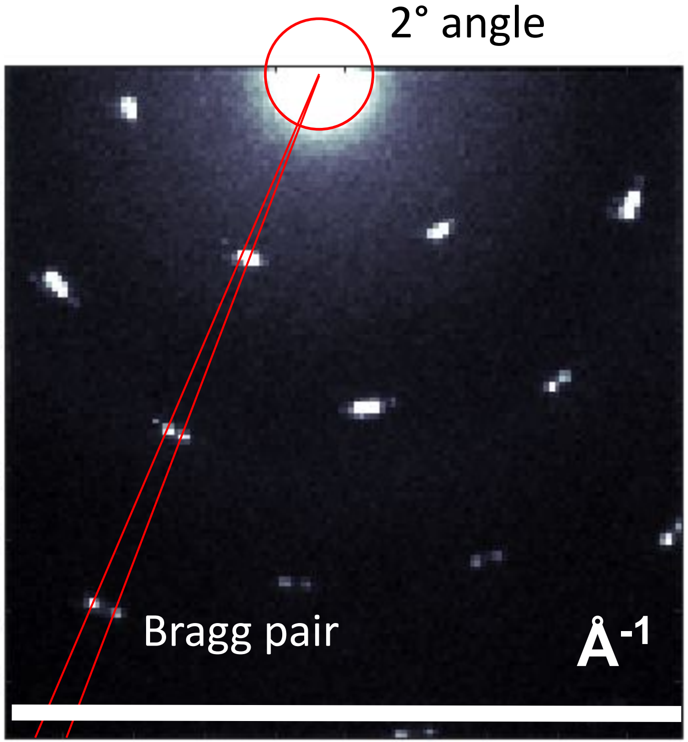

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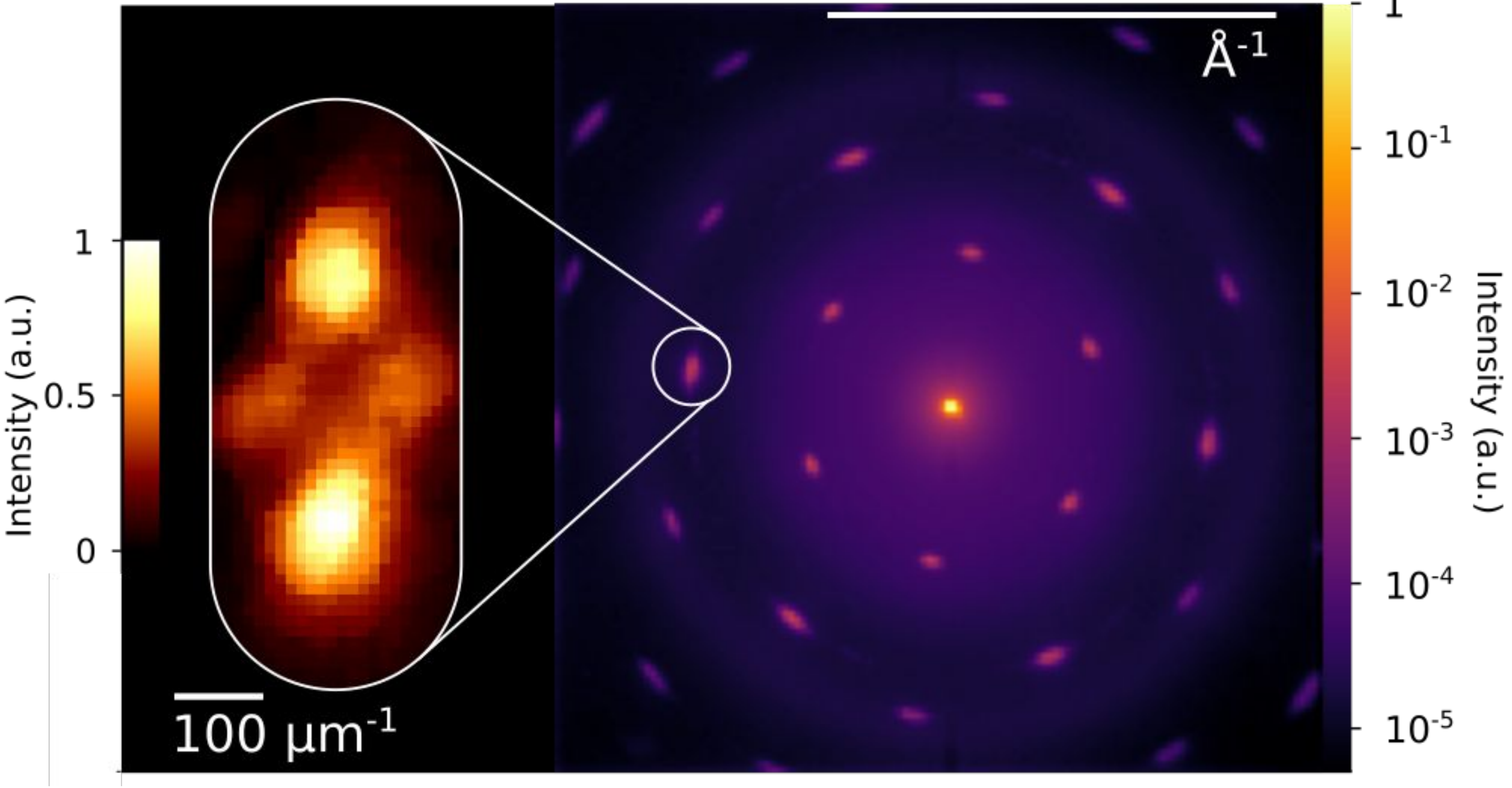

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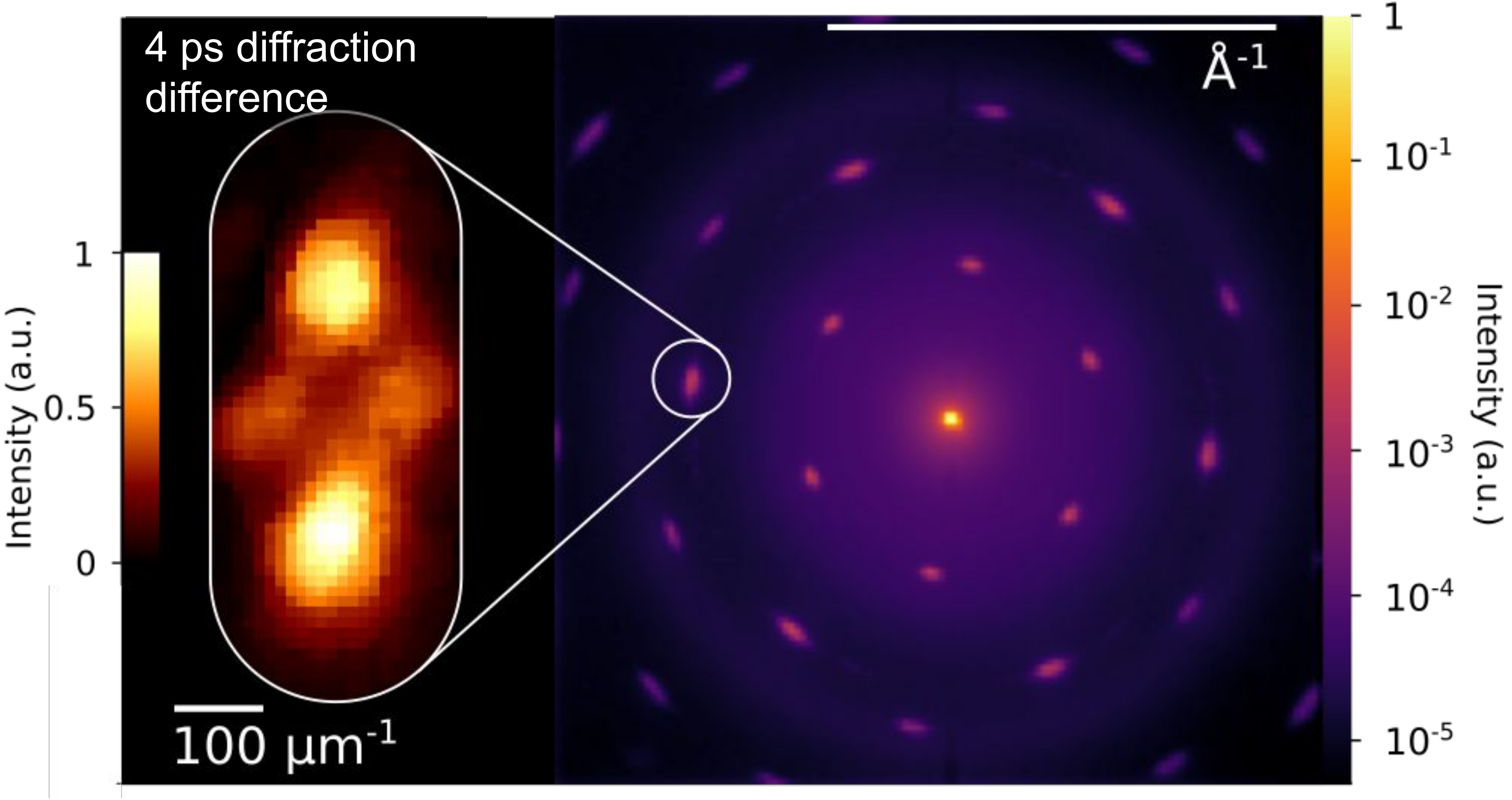
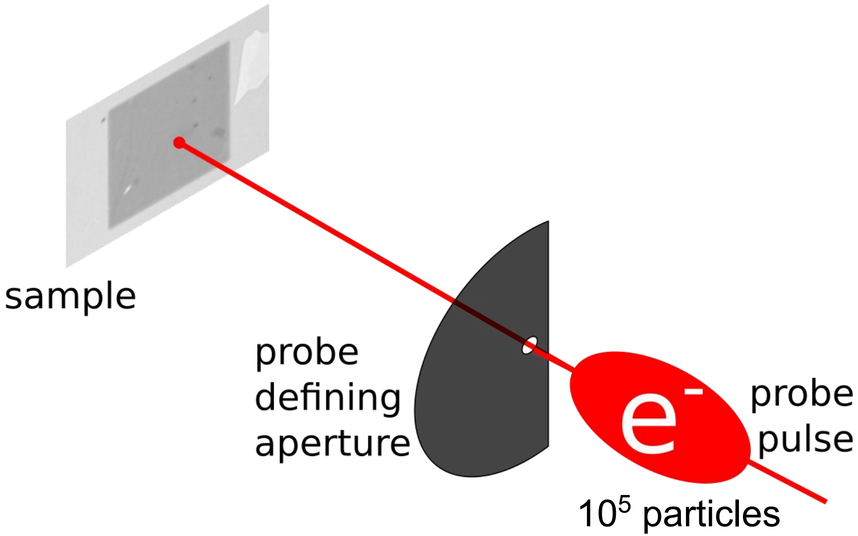


Scanning UED technique



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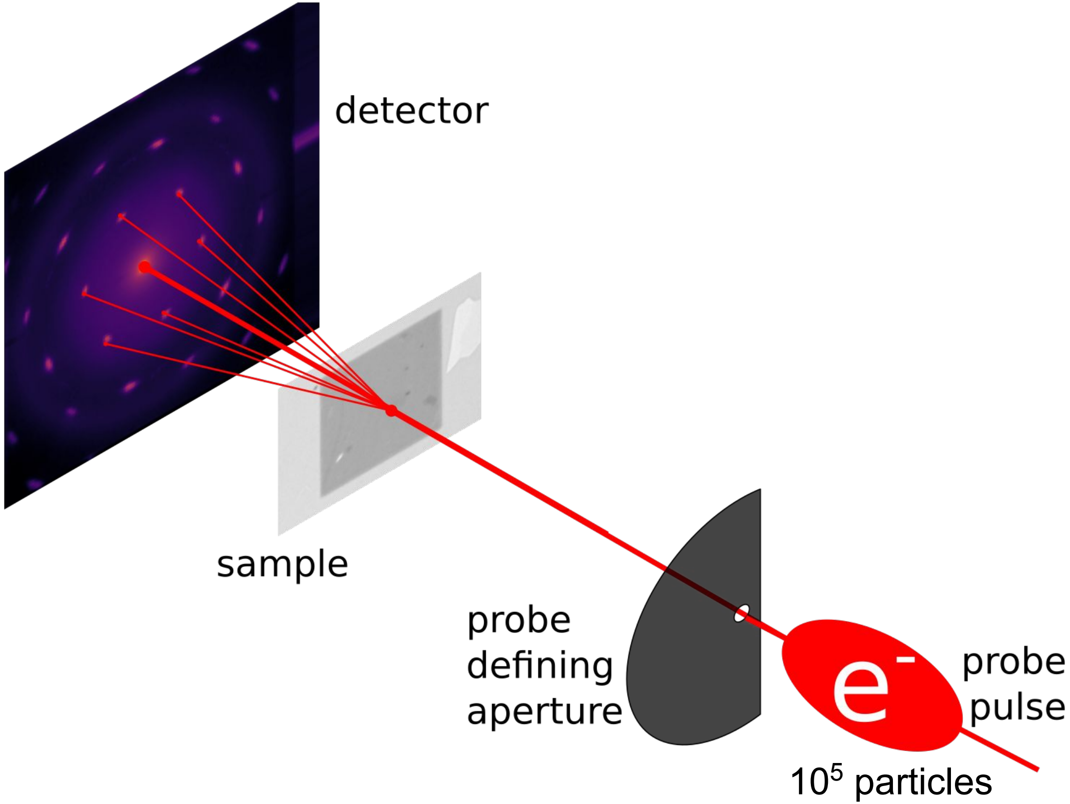

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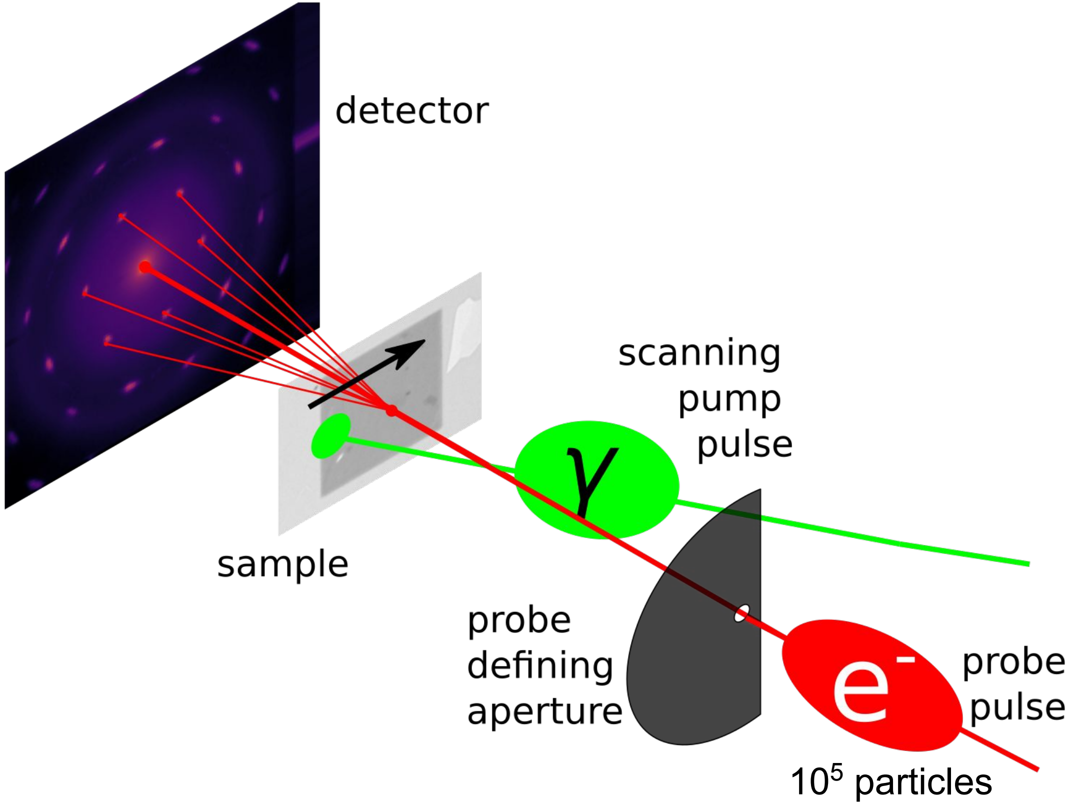

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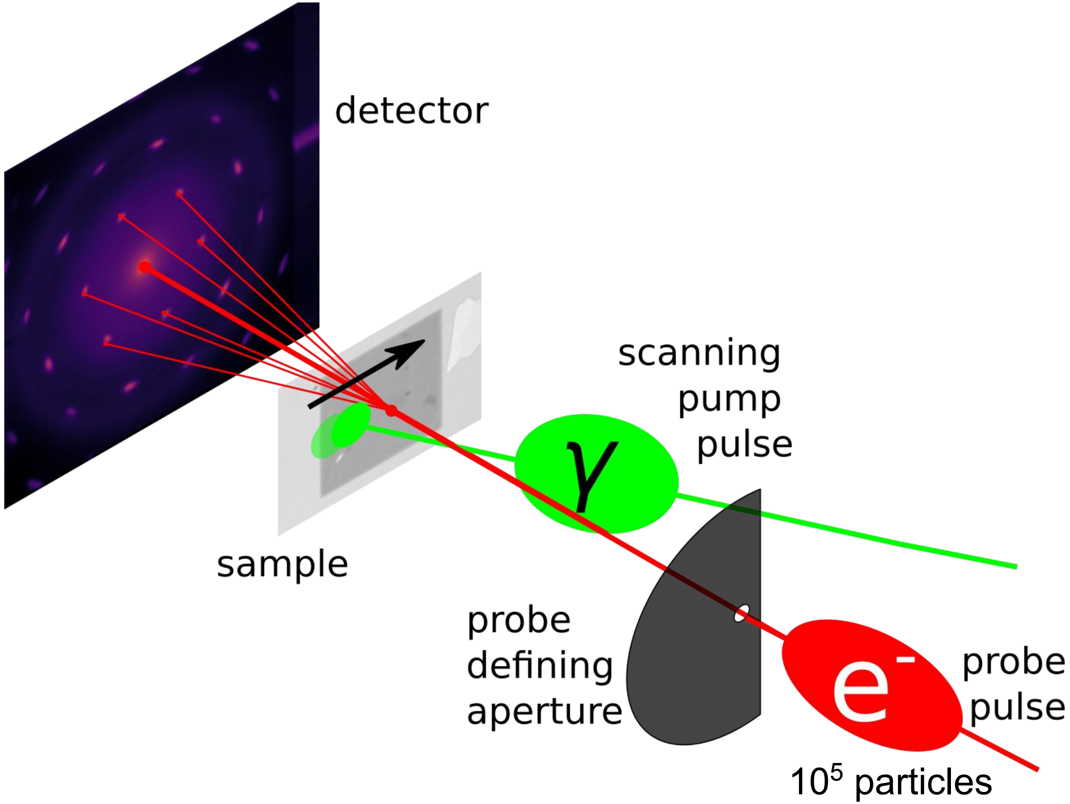

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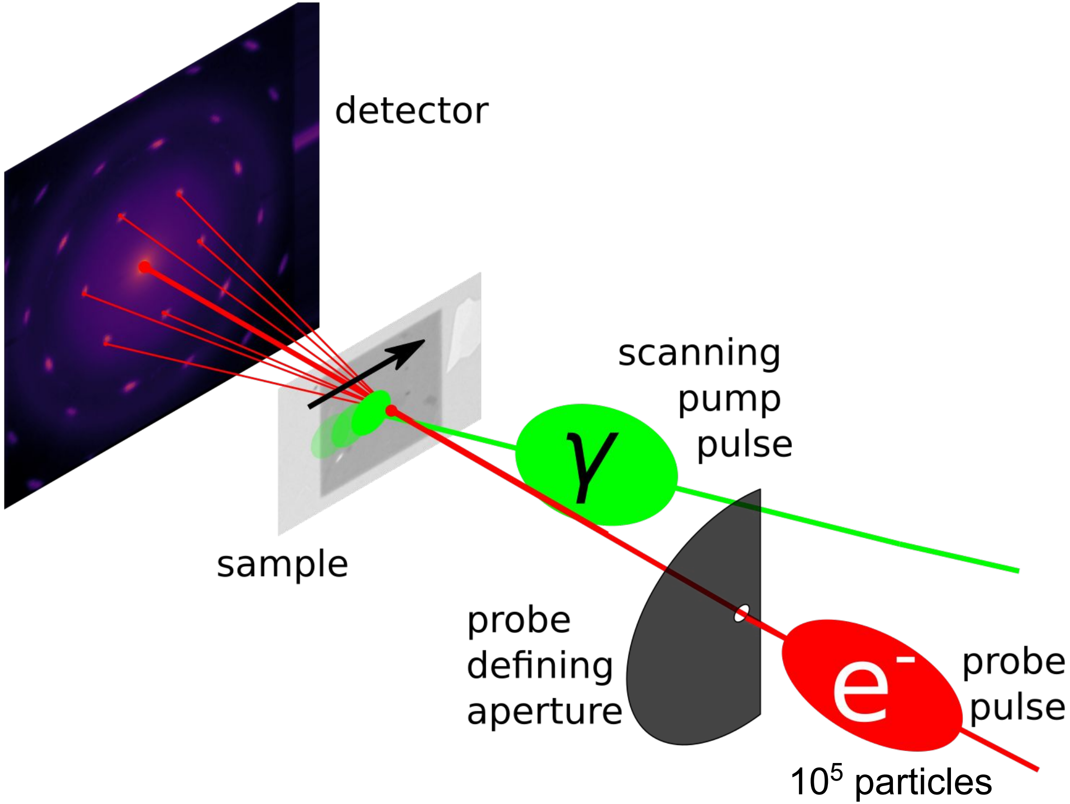

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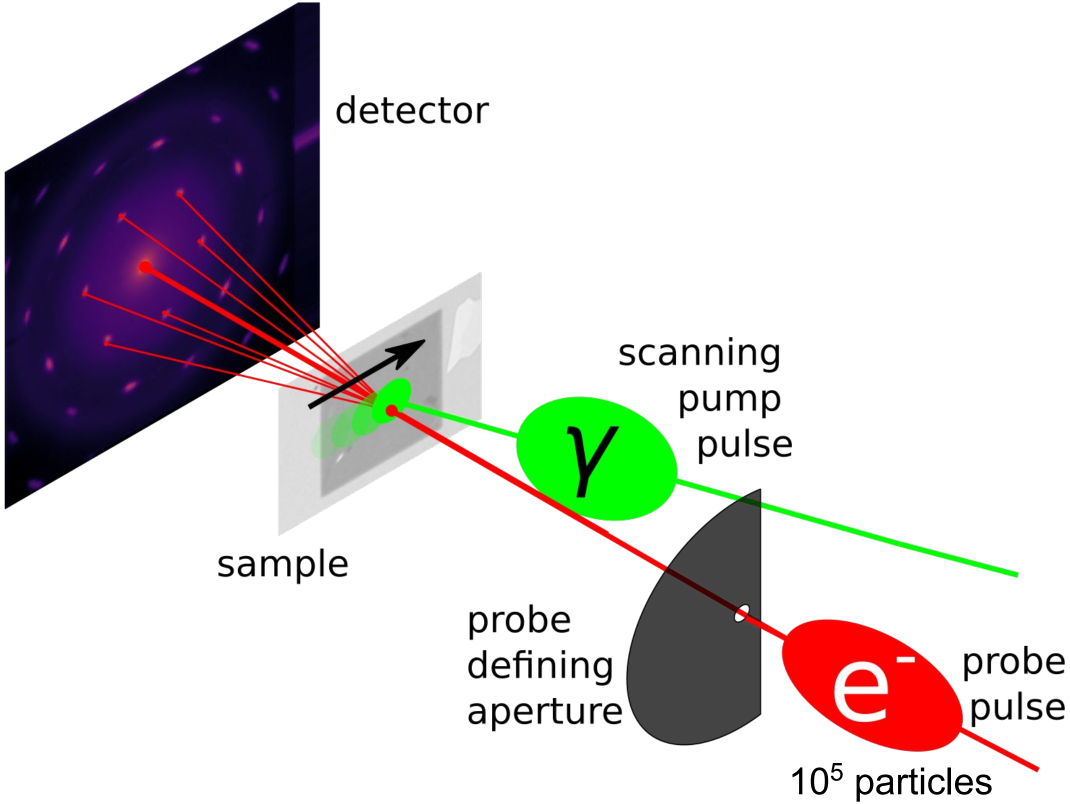

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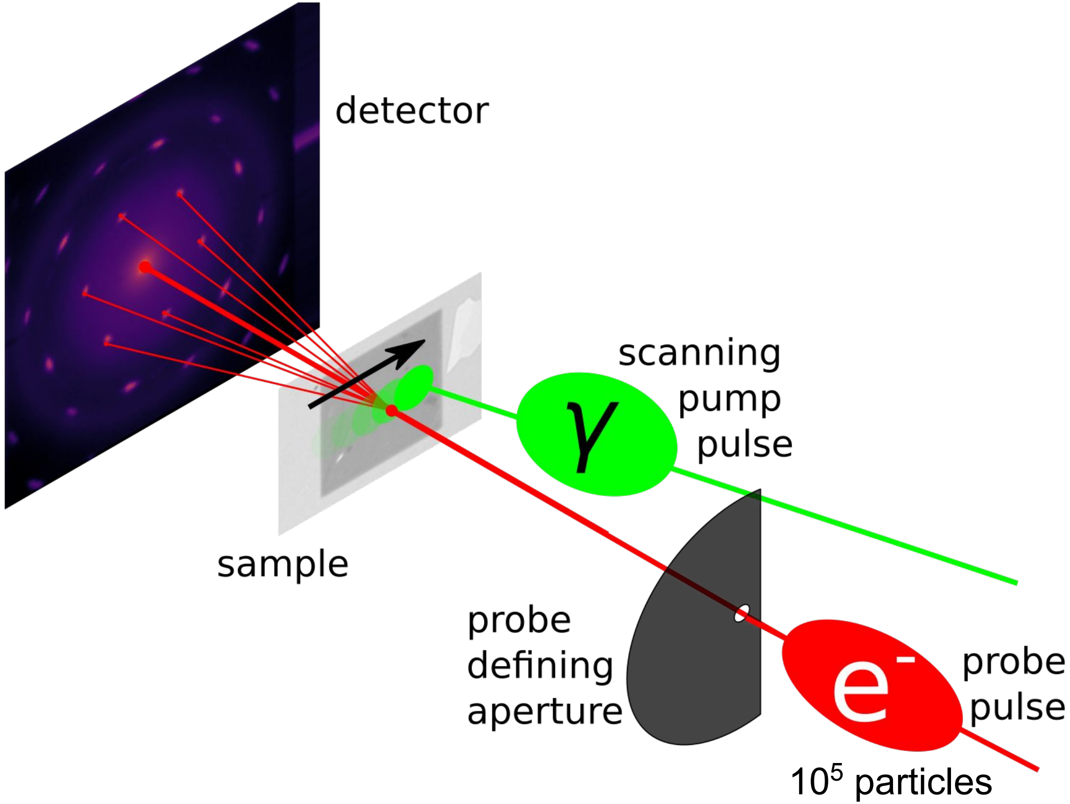

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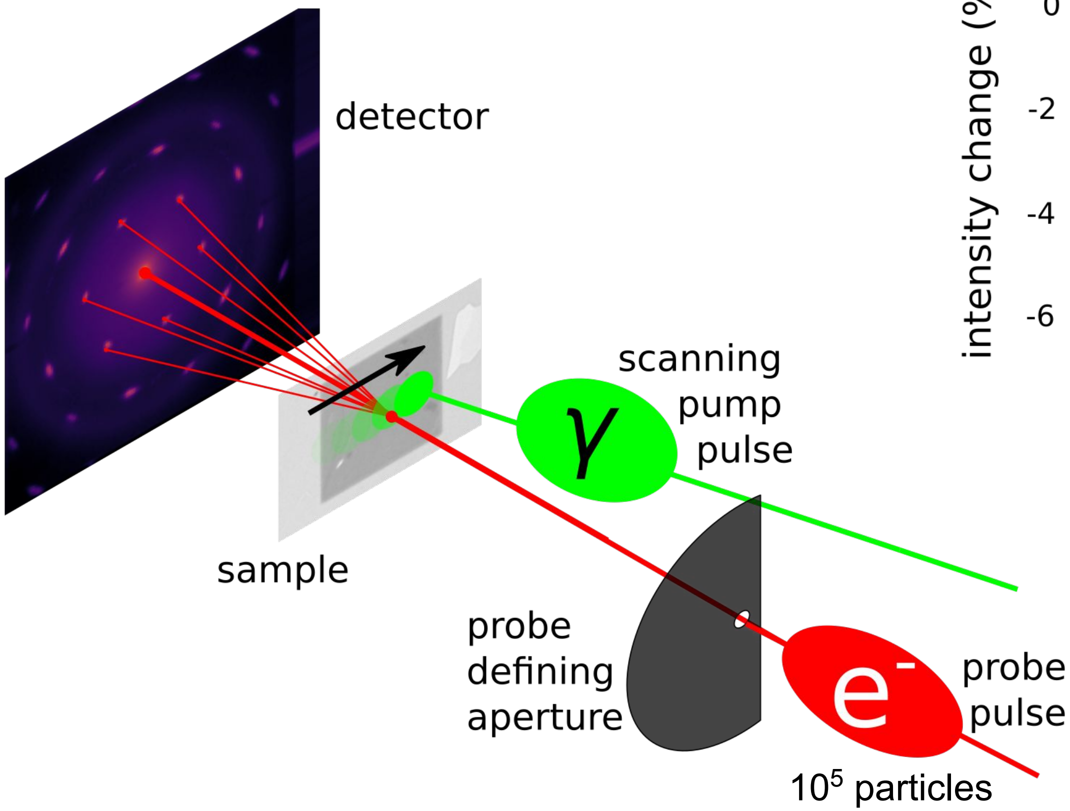
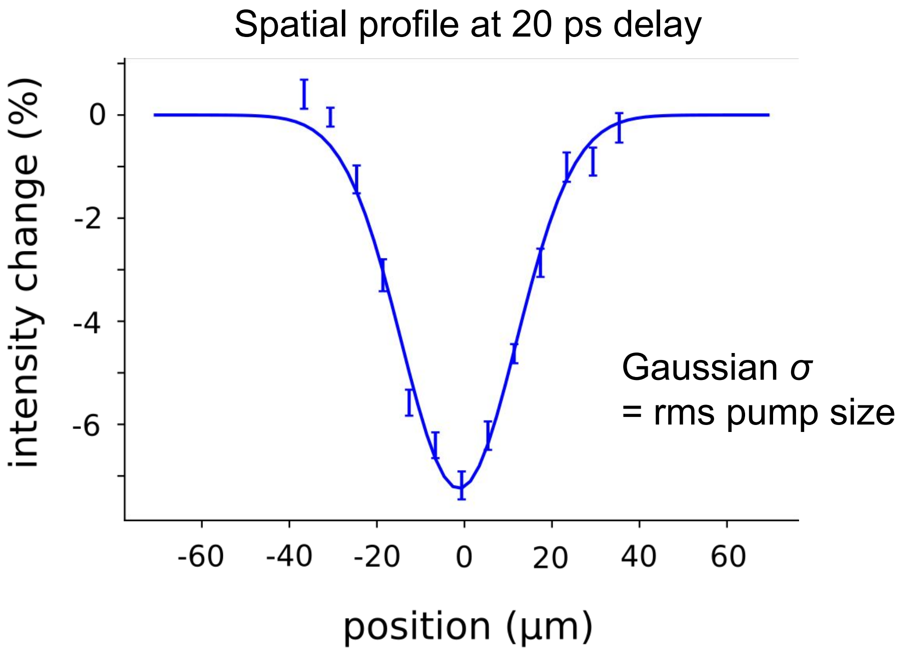
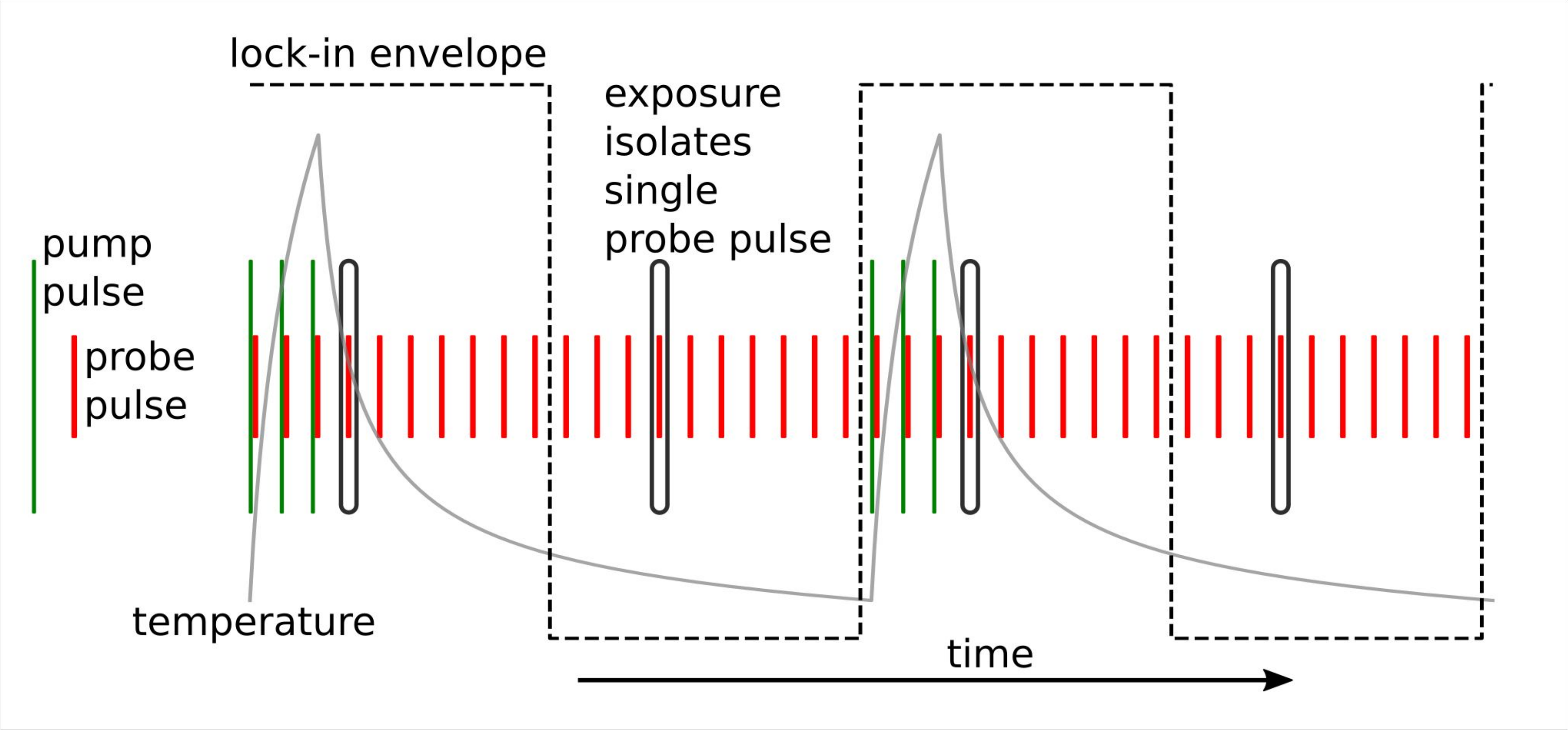




Multi-time-scale Electron Diffraction *with 1 kHz detector framerate*



Pump pulse train is gated with acousto-optic modulator (AOM)

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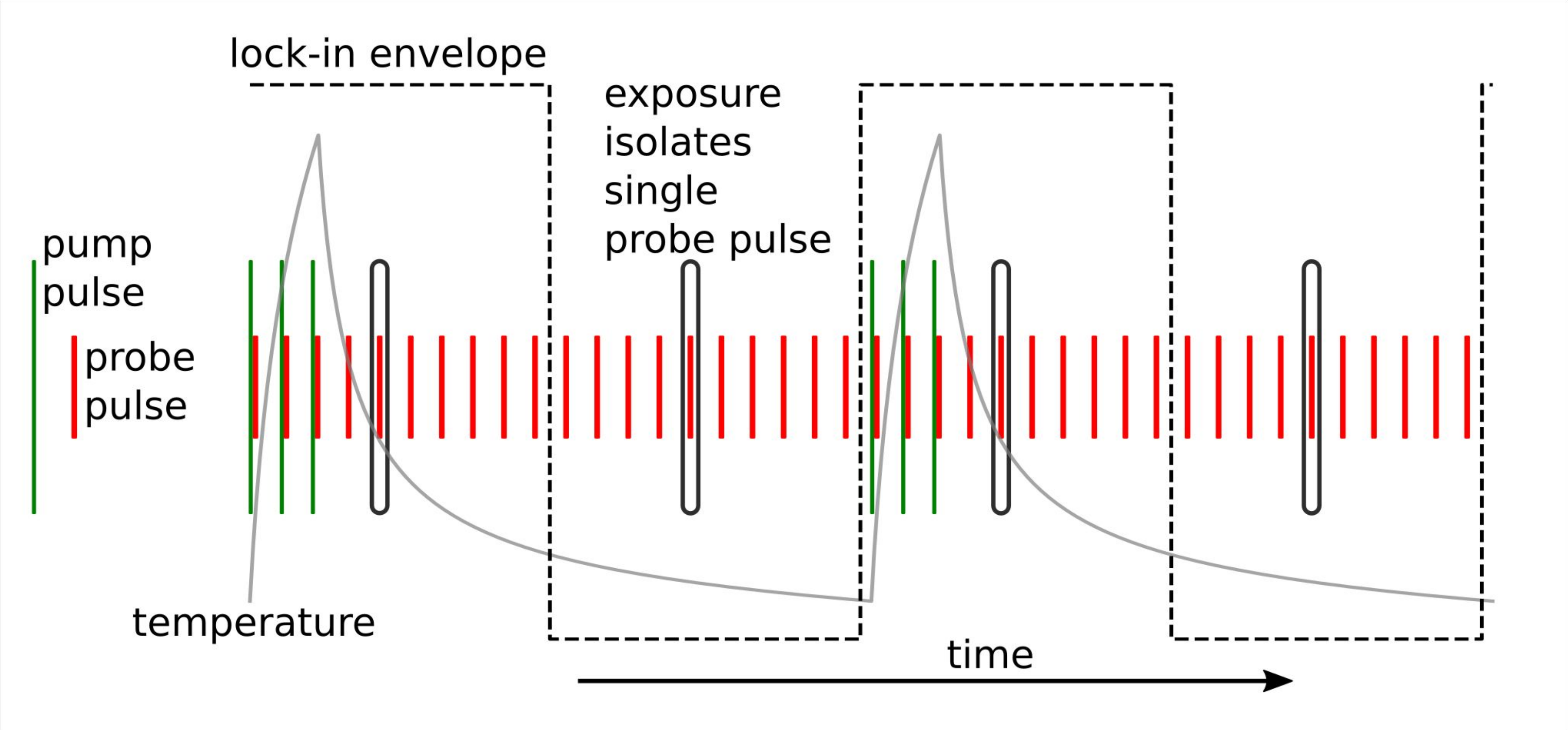

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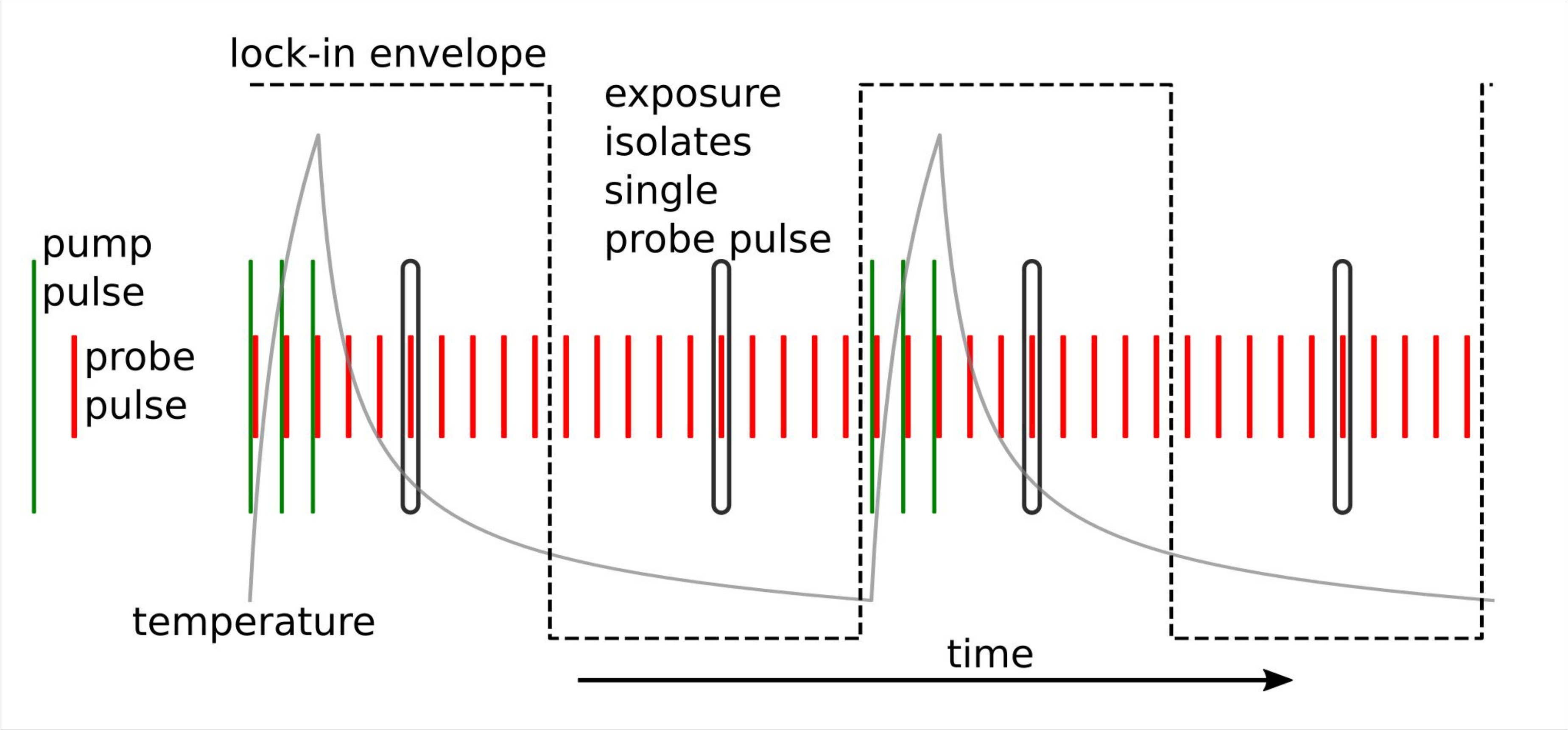

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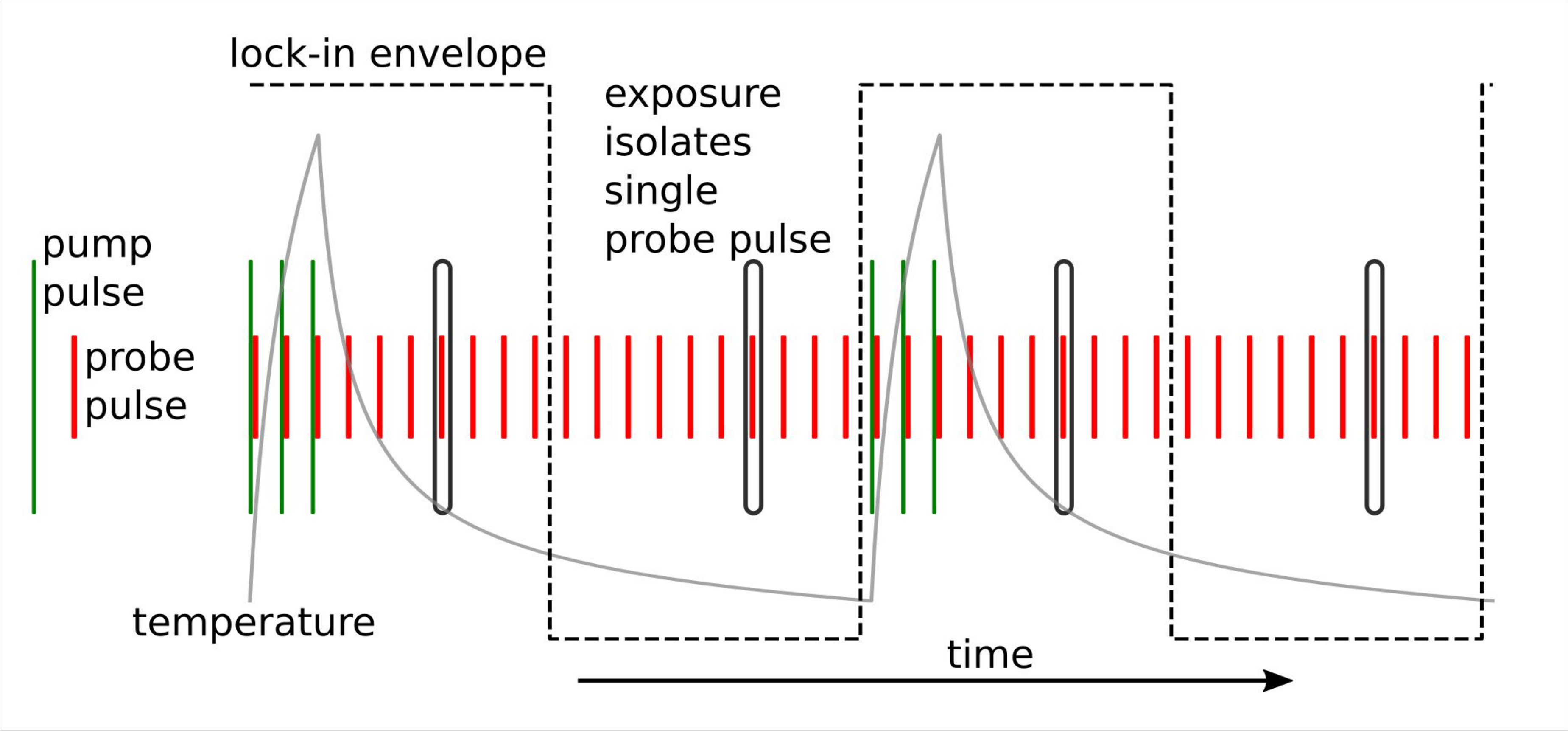

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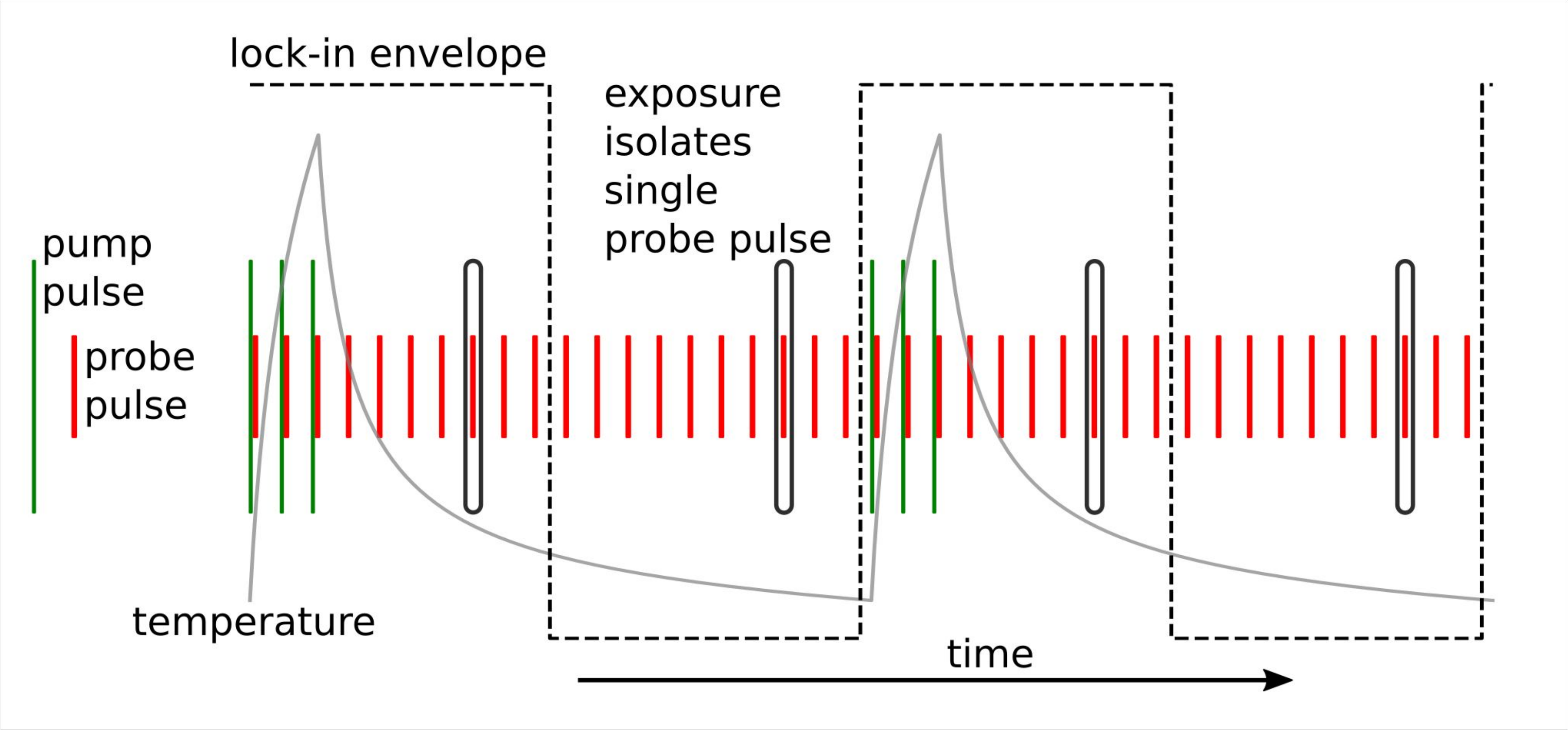

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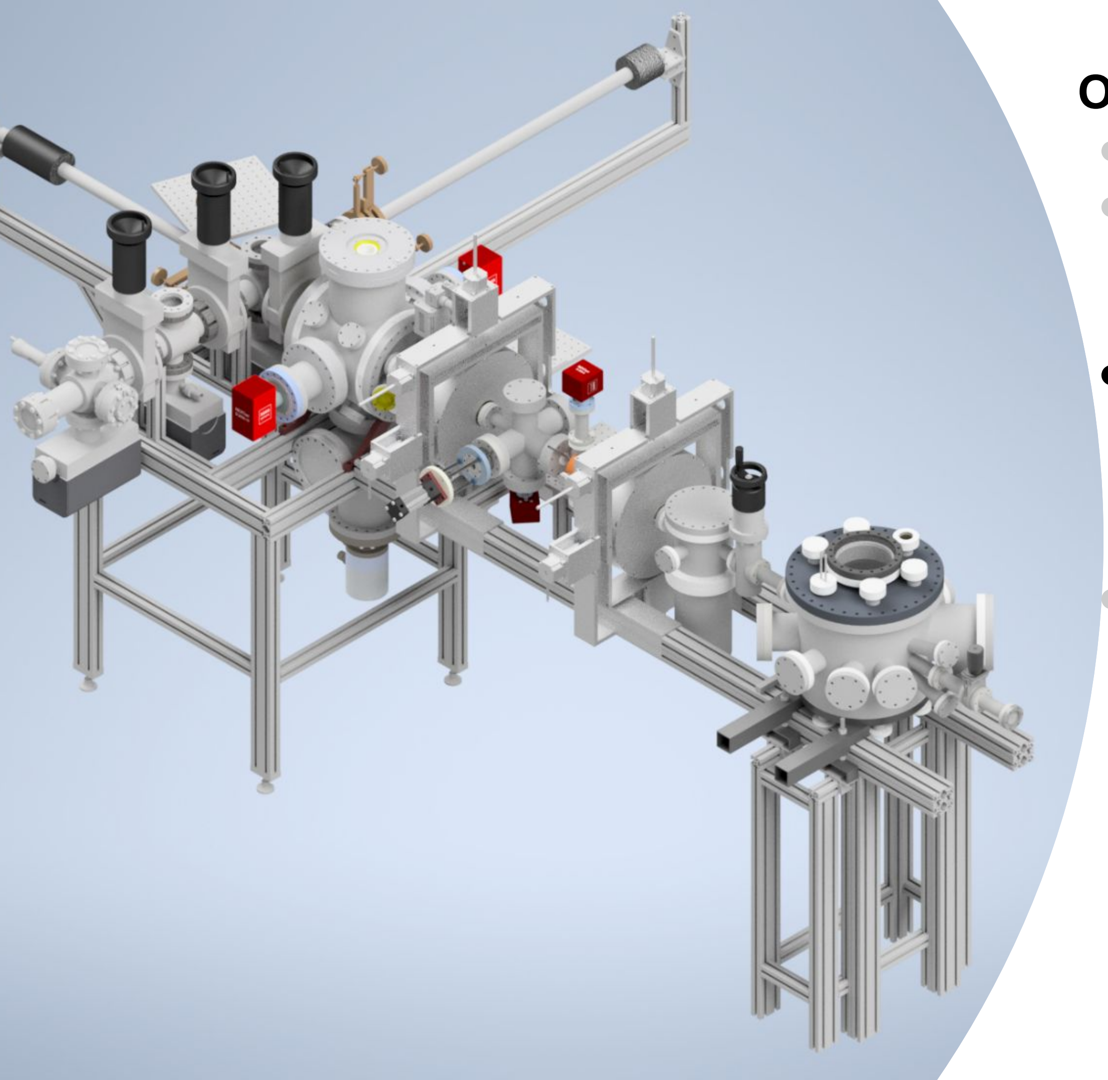

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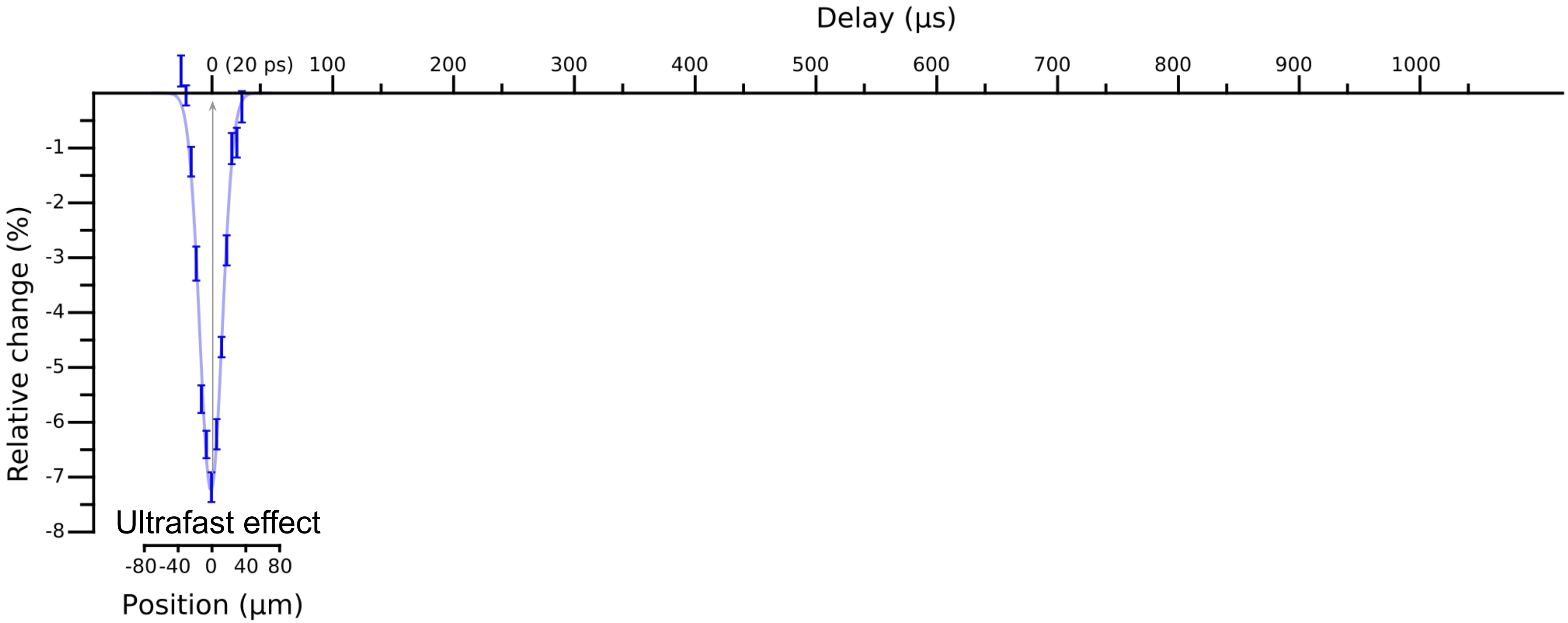

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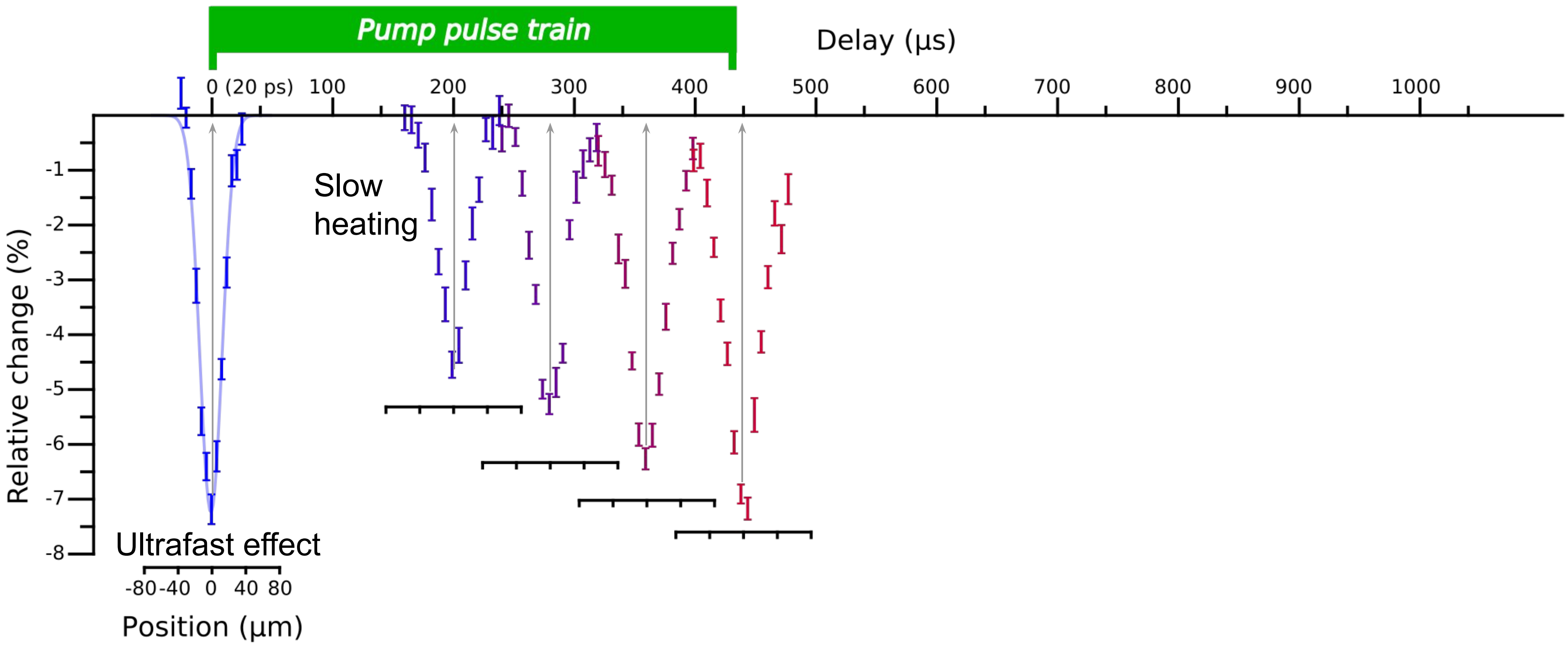

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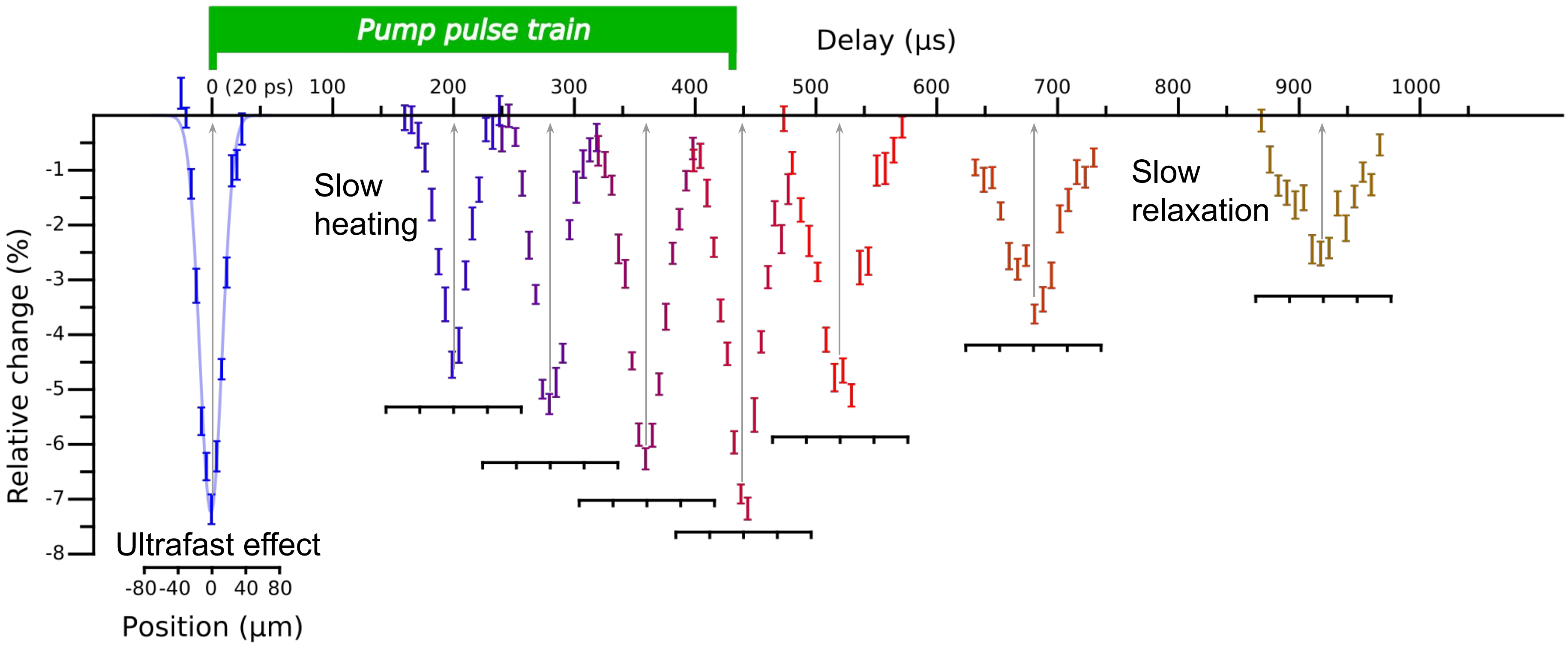
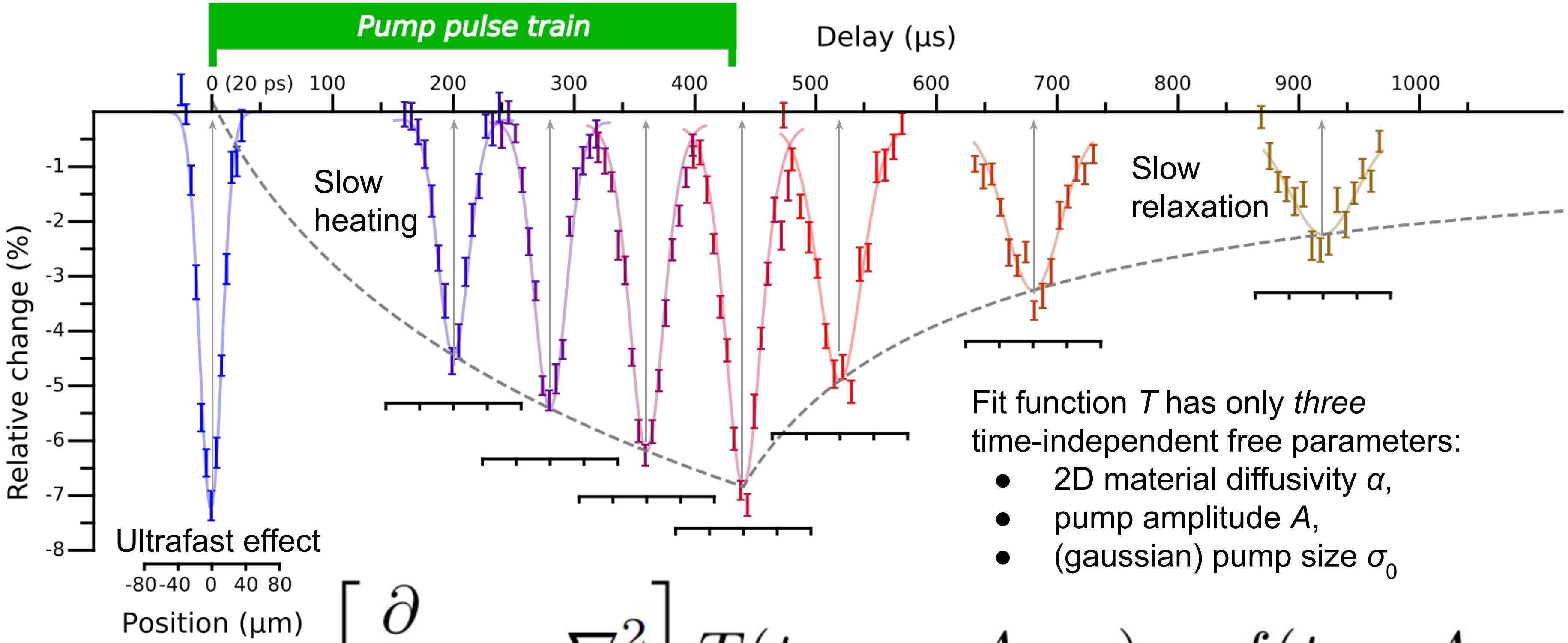


Scanning Multi-time-scale Electron Diffraction *on $WSe_2/MoSe_2$ bilayer*



Fit function T has only *three* time-independent free parameters:

- 2D material diffusivity α,
- pump amplitude A,
- (gaussian) pump size σ_0

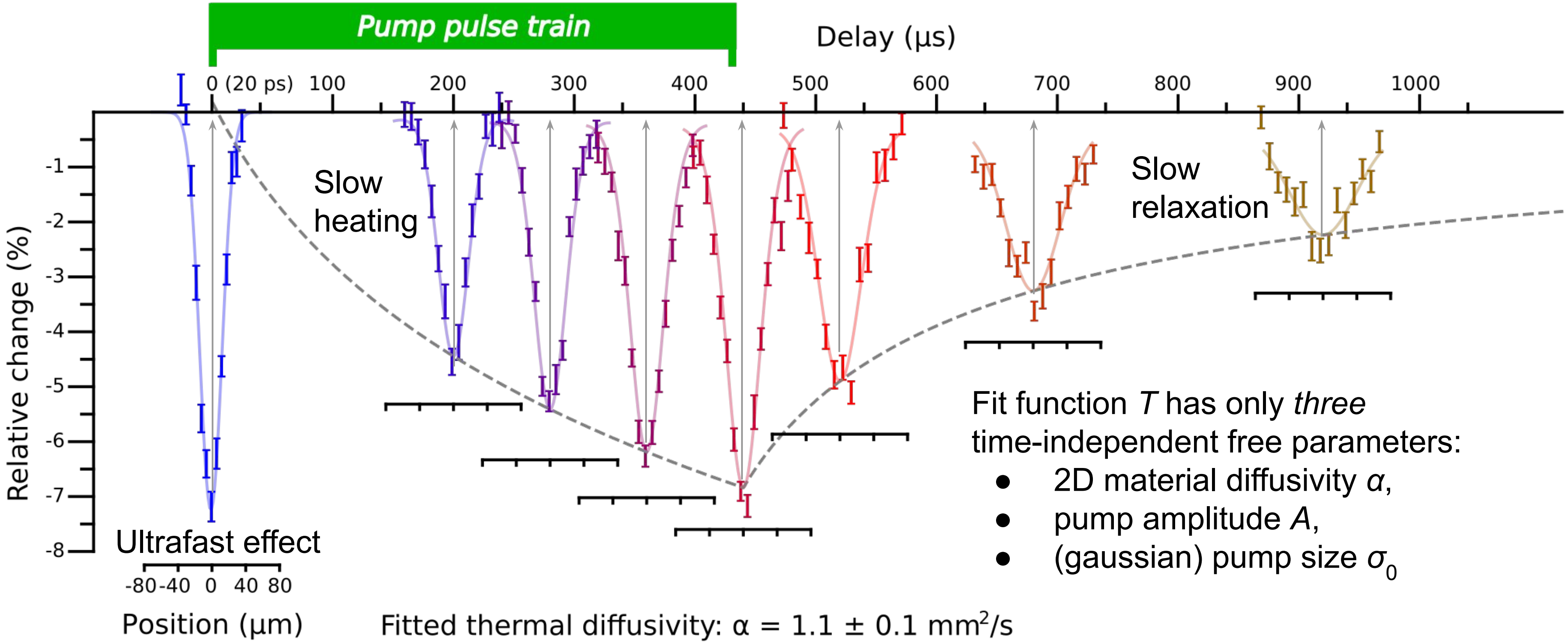
$$\left[\frac{\partial}{\partial t} - \alpha\nabla^2\right] T(t, \mathbf{x}; \alpha, A, \sigma_0) = f(t, \mathbf{x}; A, \sigma_0)$$

Sample response

Pump beam

2D inhomogeneous heat equation

Scanning Multi-time-scale Electron Diffraction *on $WSe_2/MoSe_2$ bilayer*



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Fitted thermal diffusivity: $\alpha = 1.1 \pm 0.1$ mm^2/s

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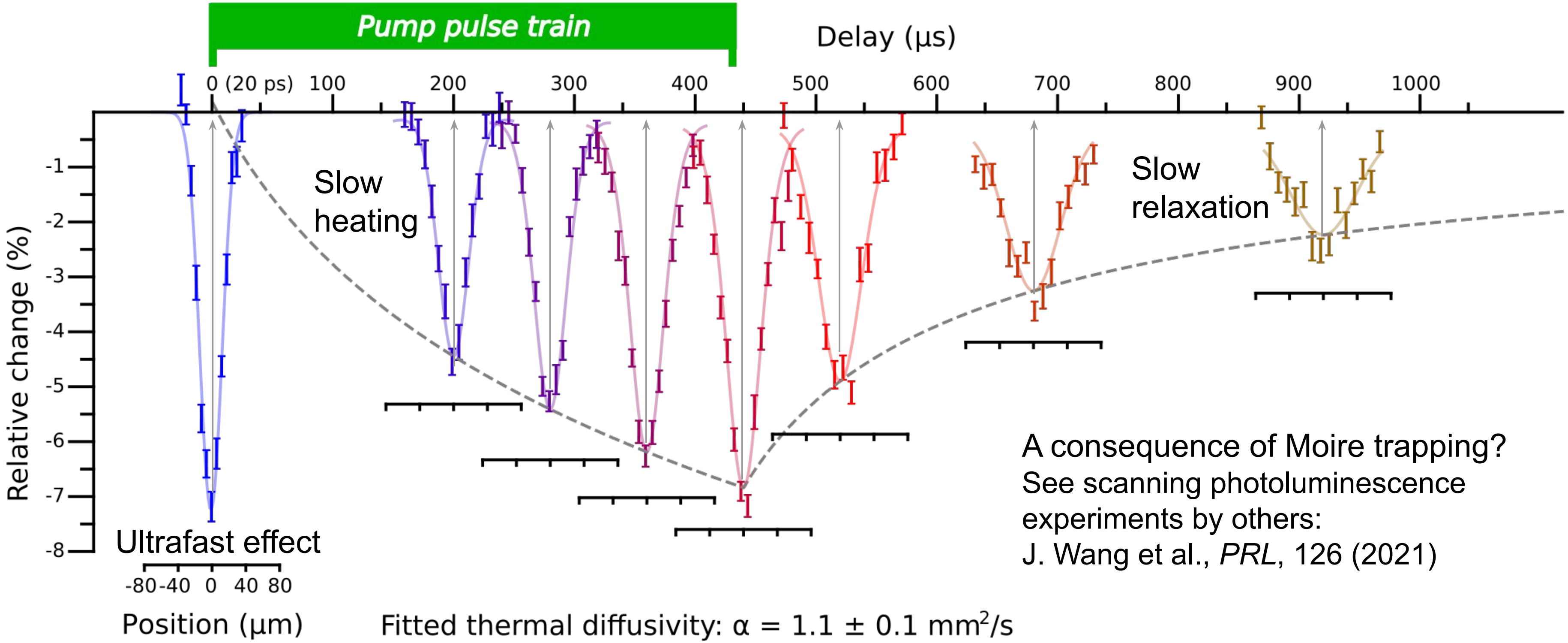

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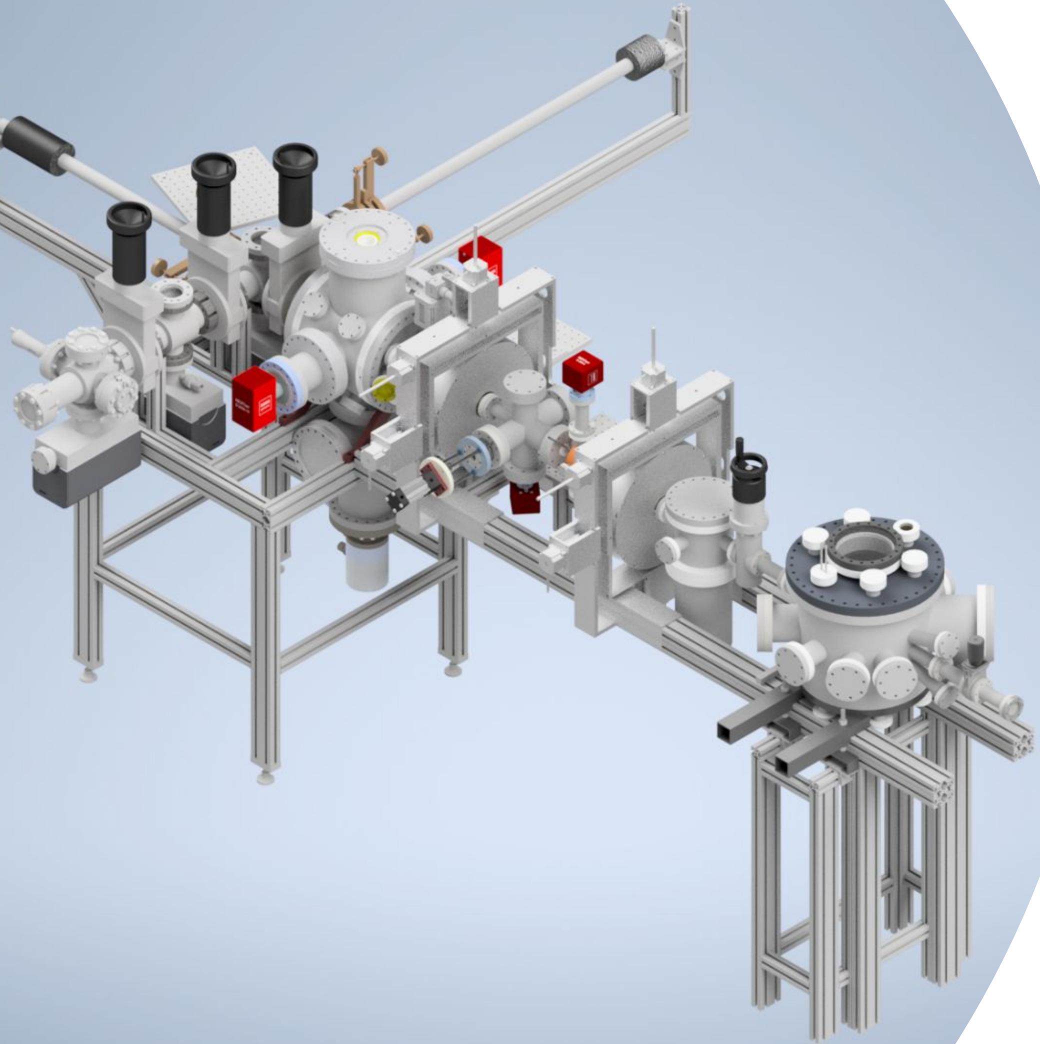
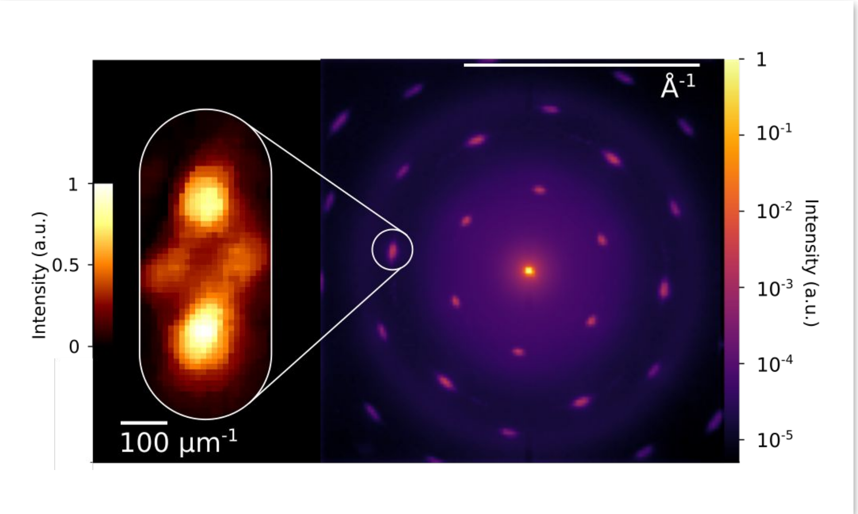
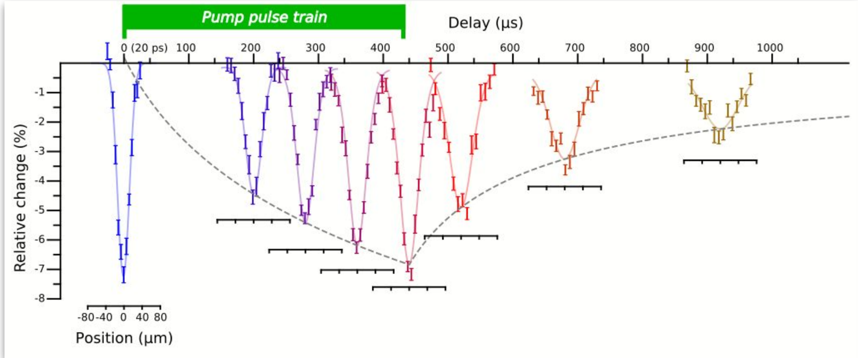
A consequence of Moire trapping?
See scanning photoluminescence experiments by others:
J. Wang et al., *PRL*, 126 (2021)



Outline:

- Overview of the apparatus
    - Beamline
    - Direct electron detector
- First science results
    - 2D Moire material
    - Scanning UED technique
    - Thermal diffusion data
- Summary and Outlook

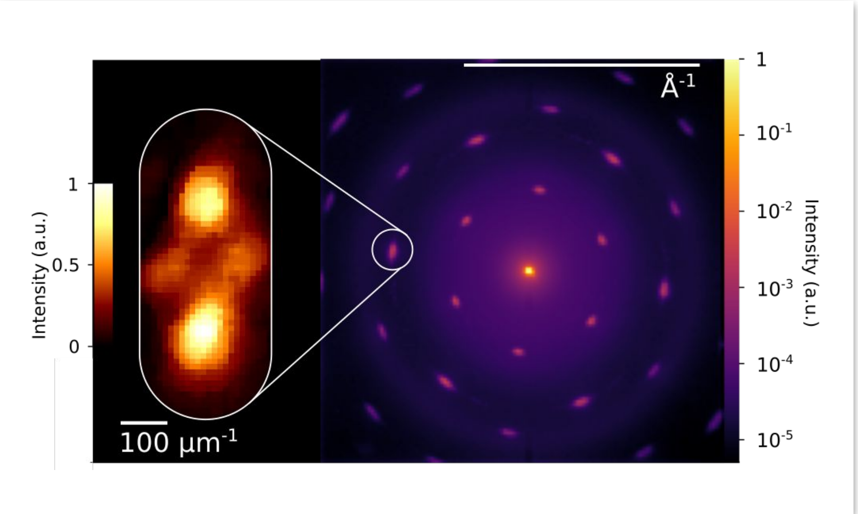


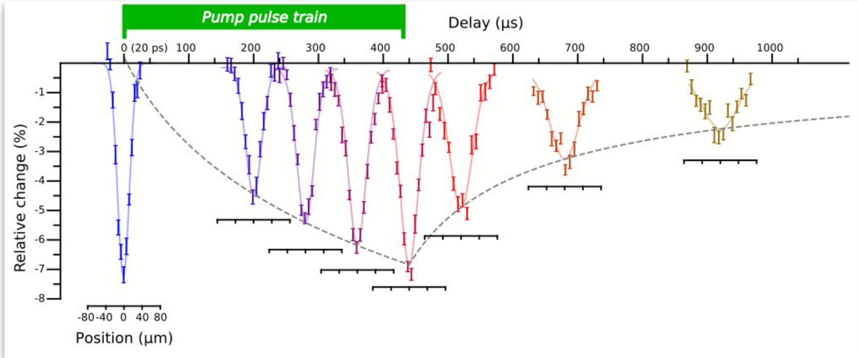


Summary

Ultrafast Electron Diffraction @Cornell

- Design incorporates a high brightness, pulsed electron source
- Direct electron detection enables:
    - fine momentum resolution
    - high sensitivity to diffuse and superlattice signals
- 1 kHz framerate enables:
    - Lock in detection
    - Pulse picking for delay scans
- Science results on Moire material
    - Superlattice structure
    - Thermal transport

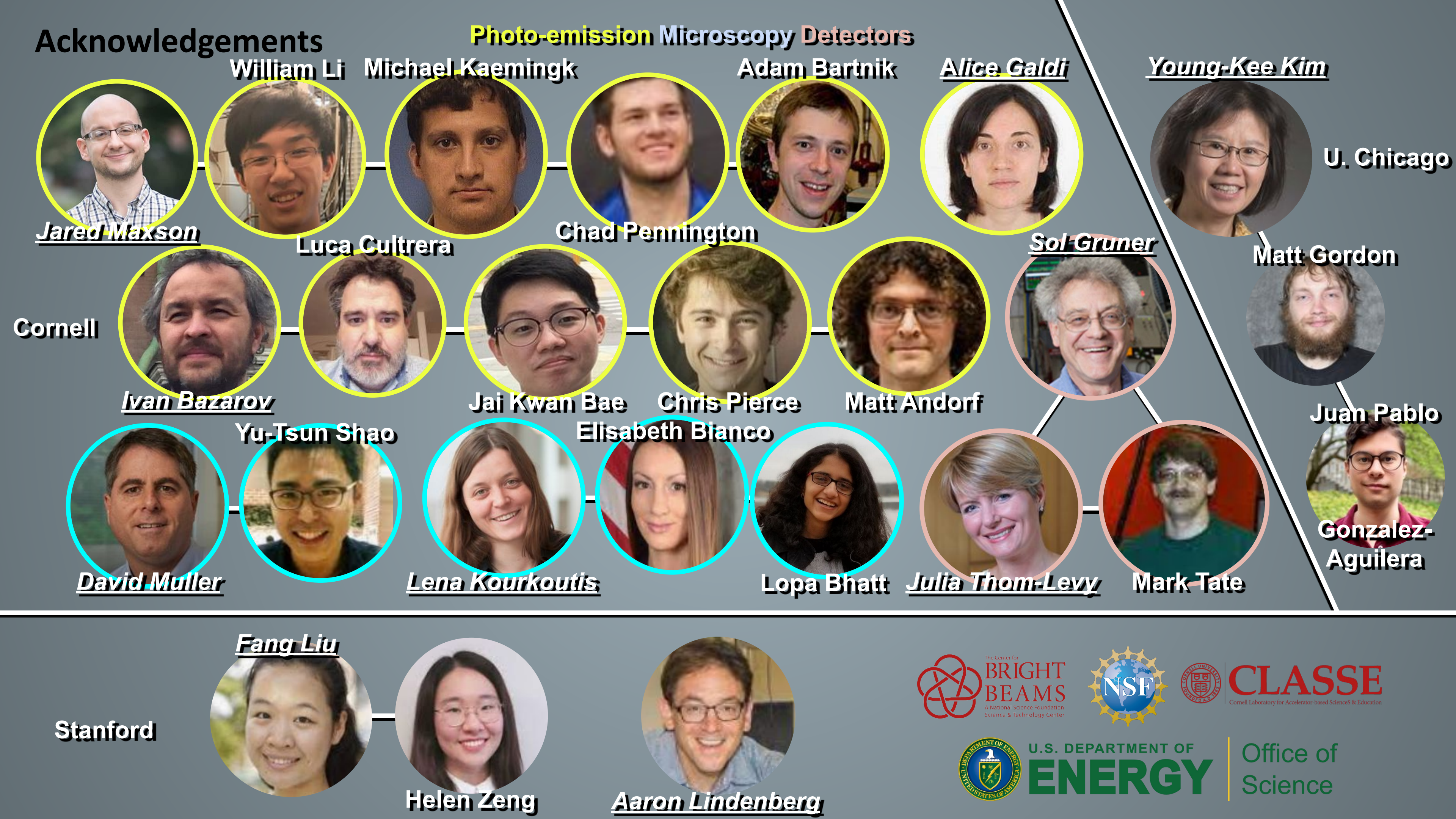




Outlook

Ultrafast Electron Diffraction @Cornell

- Currently collecting time-resolved Moire data
- Follow-up measurements to study dependence of sample response on twist angle
- New pump wavelengths
  - UV
  - Terahertz
- Other samples with nm scale superlattice structure
  - Polar skyrmions
  - Your favourite sample!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