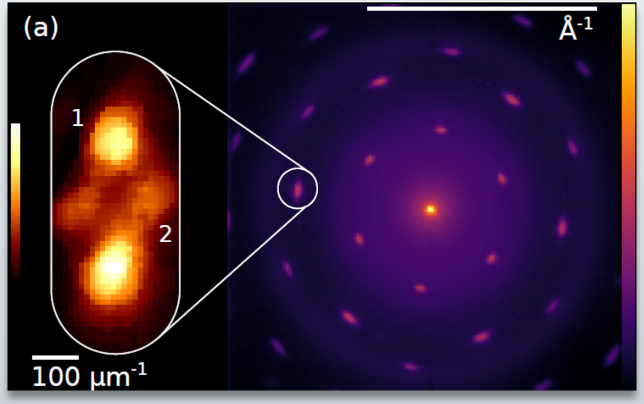
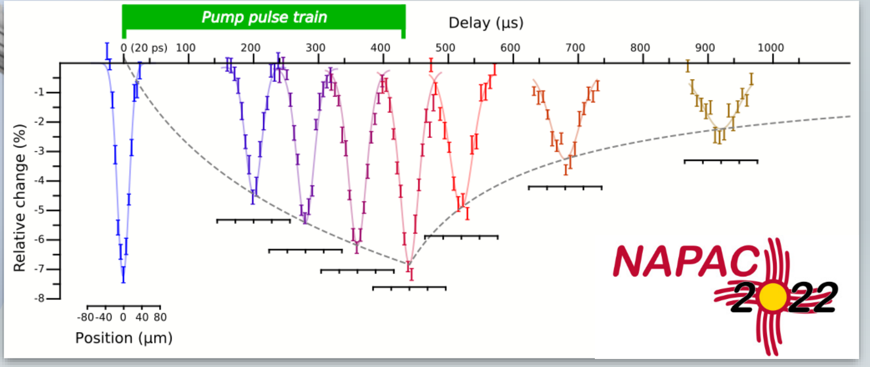
Ultrafast Electron Diffraction with Low Emittance Photocathodes

Jared Maxson, Cornell University





NAPAC 2022

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

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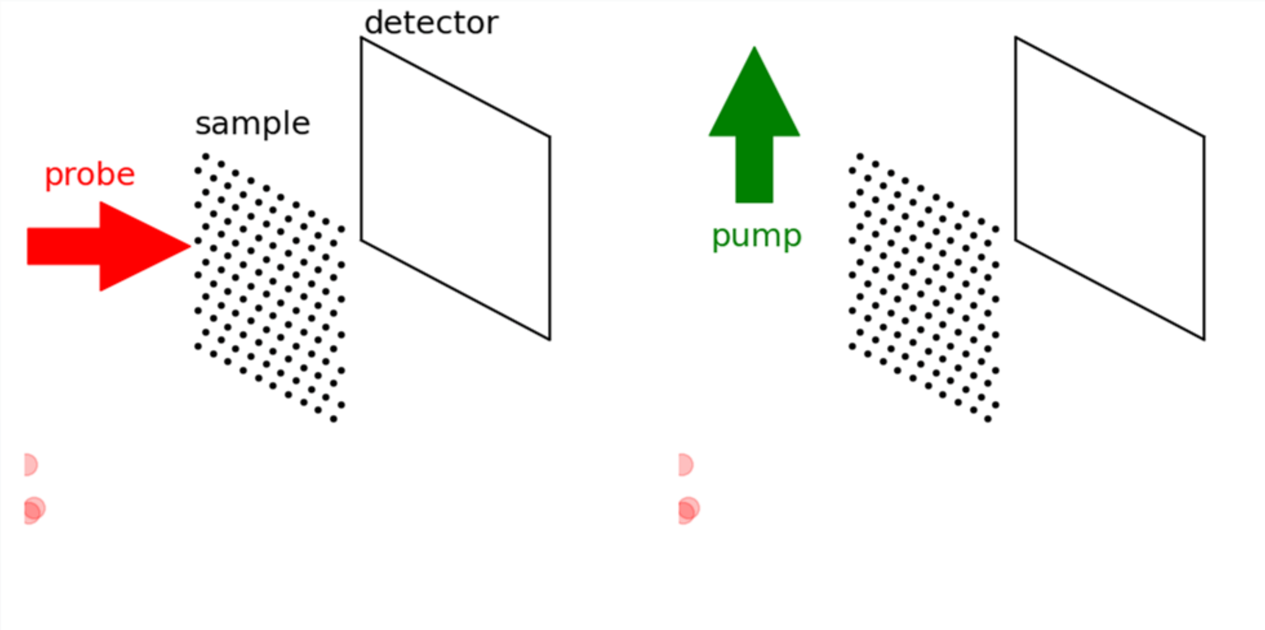
U.S. DEPARTMENT OF ENERGY

Office of Science

Introduction to UED: The need for *brightness*



We want **a small transverse probe size** → some samples are hard to make with large dimensions
We want a **small transverse momentum spread** → need high coherence to see small features in k-space
We want a **short bunch duration** → natural timescale for atomic motion is fs-ps
We want as **many electrons as possible** → large signal to noise for subtle diffraction features

High source brightness is critical for UED!

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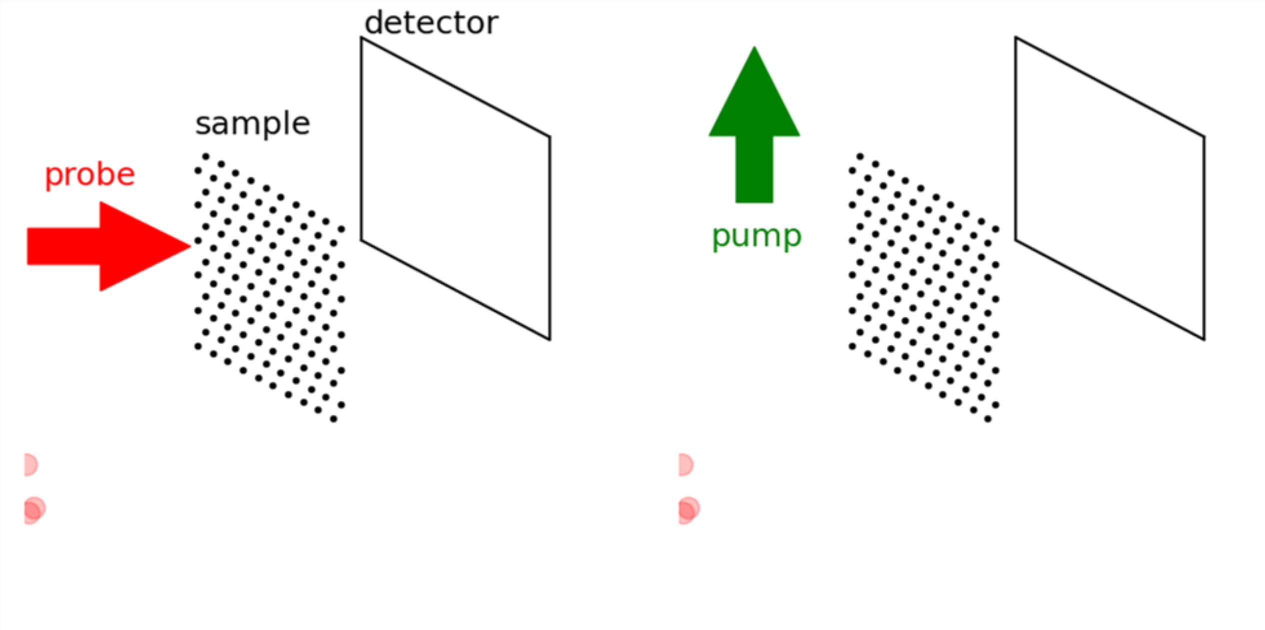

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Much previous UED work

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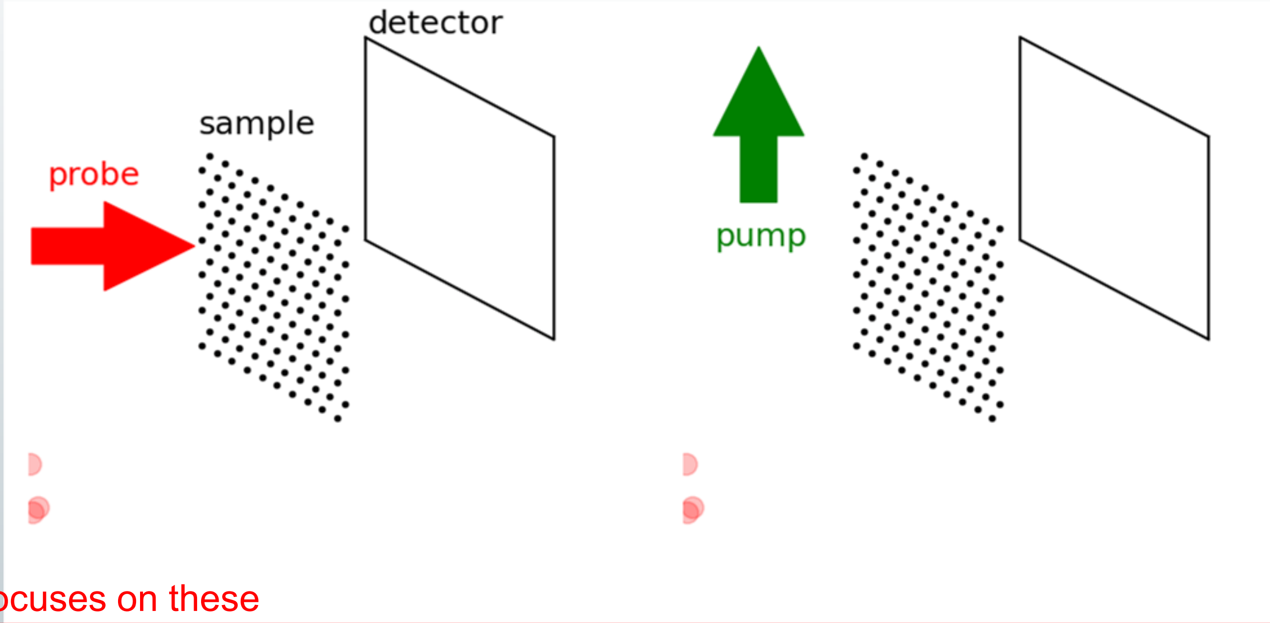

Our device focuses on these

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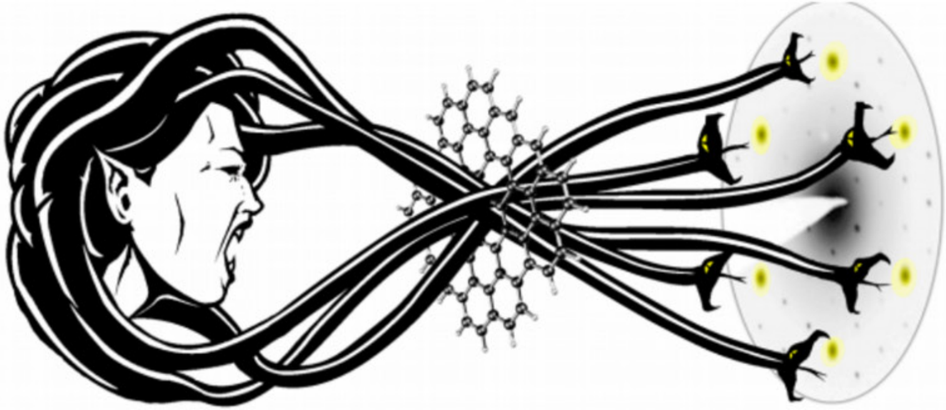
Much previous UED work

High source brightness is critical for UED!

We focus on transverse probe size and coherence, (hence the name)

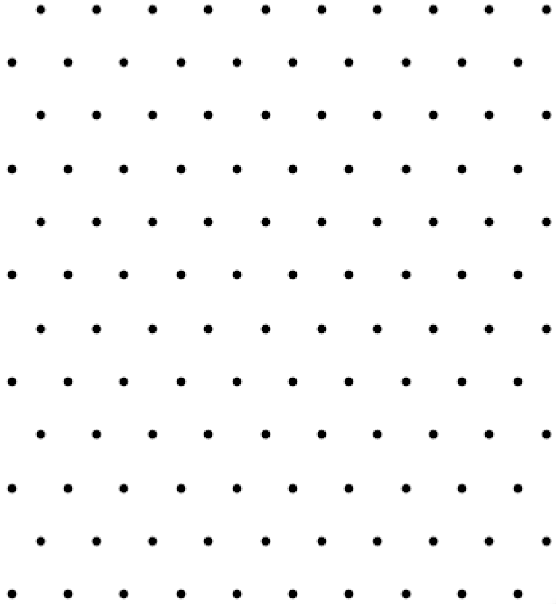
MICRO
ELECTRON
DIFFRACTION FOR
ULTRAFAST
STRUCTURAL
ANALYSIS



Not *just* about Bragg Peaks

Real space

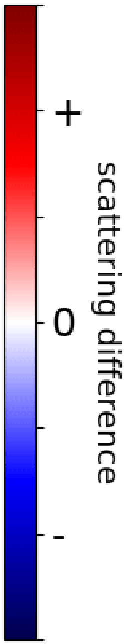
1000 000 000 000 × slow motion



Atomic response occurs on 10^{-13} – 10^{-12} second timescale

(simulation)

Reciprocal space



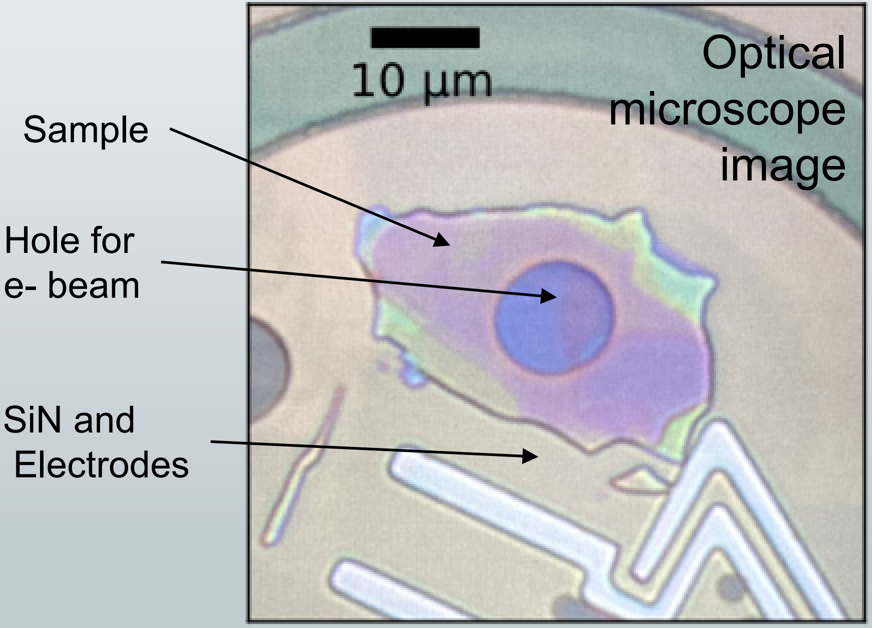
hot - cold

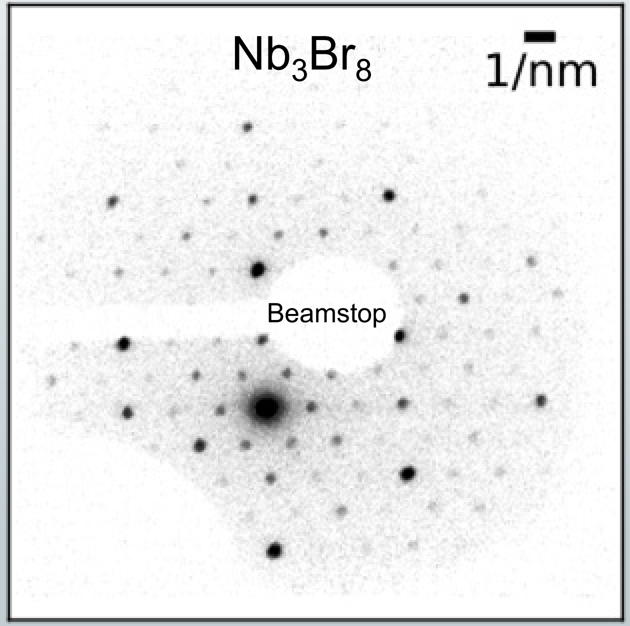
Diffuse scattering encodes *phonon populations*

The need for small probe sizes

Preparing large films of quantum materials for UED can be challenging.
Example: Nb_3Br_8, thin film flakes, exhibits periodic lattice distortion

UED @ MEDUSA





The rich k-space means that one cannot merely focus strongly– need small divergence too.

The need for reciprocal space resolution
Case Study: Moire Materials @ MEDUSA

- Overlapping two monolayers with a small twist can yield remarkable new materials physics: moire materials.

Samples prepared @ Stanford by Fang Liu and Helen Zeng

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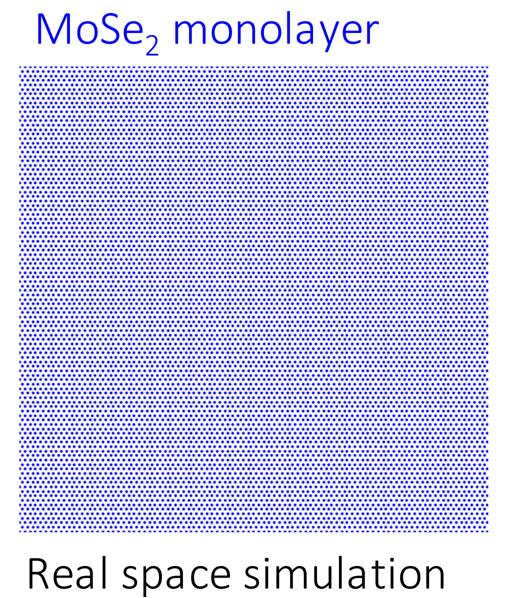

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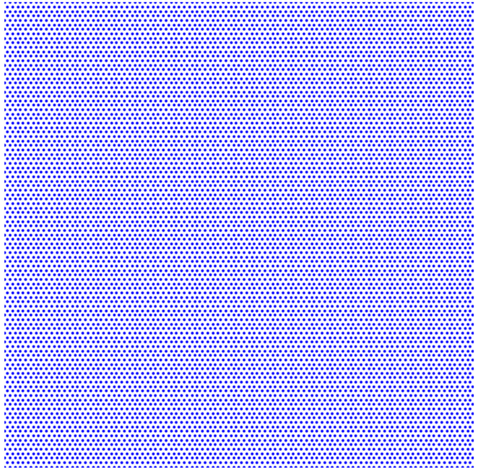
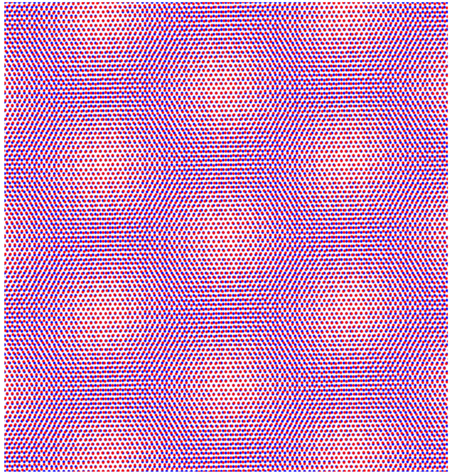
$MoSe_2$ monolayer



Real space simulation

WSe_2 rotated 2° / $MoSe_2$ monolayer



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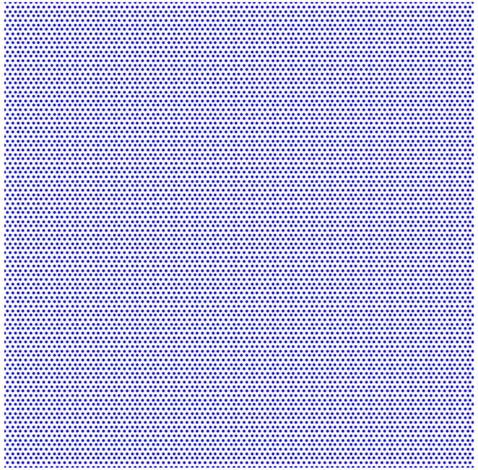

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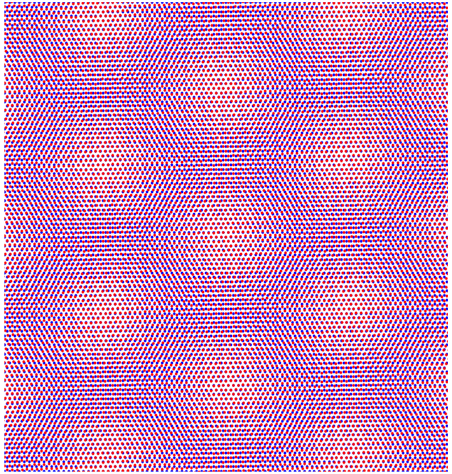
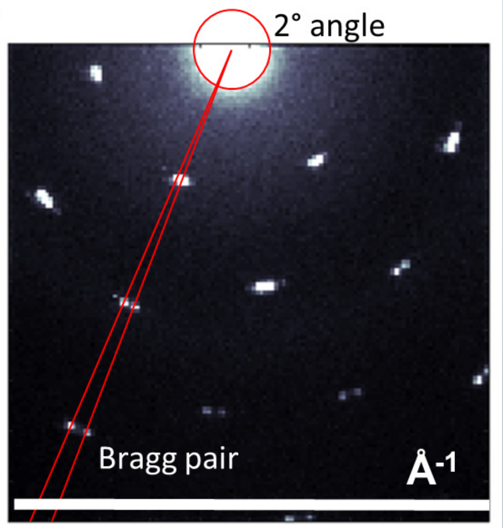


Real space simulation

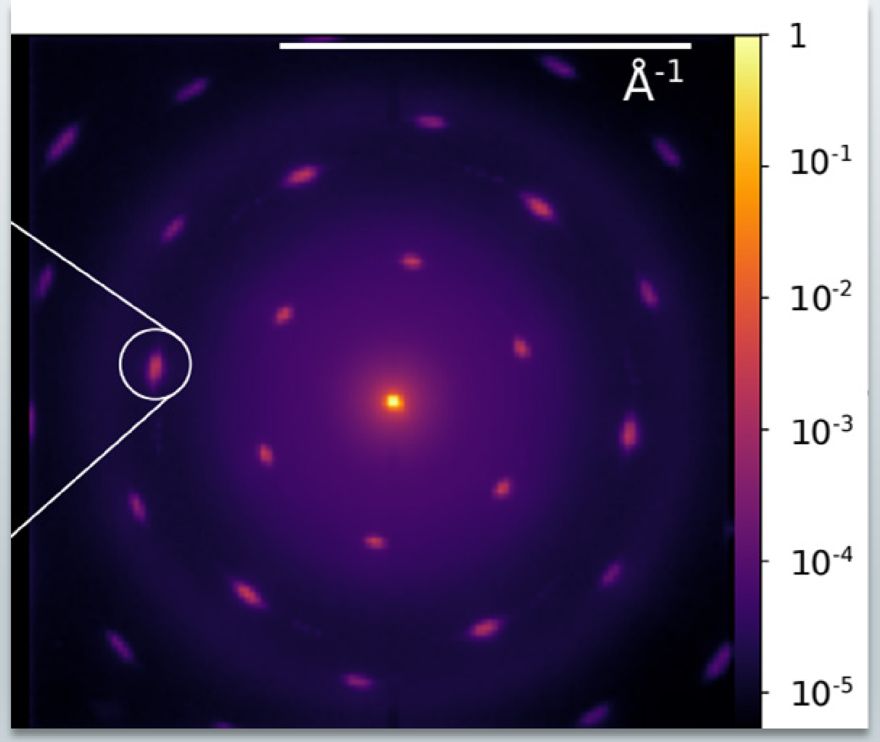


Diffraction experiment

Samples prepared @ Stanford by Fang Liu and Helen Zeng

The need for reciprocal space resolution
Case Study: Moire Materials @ MEDUSA

Interesting physics at multiple scales:



WSe_2 rotated 2° / $MoSe_2$ monolayer

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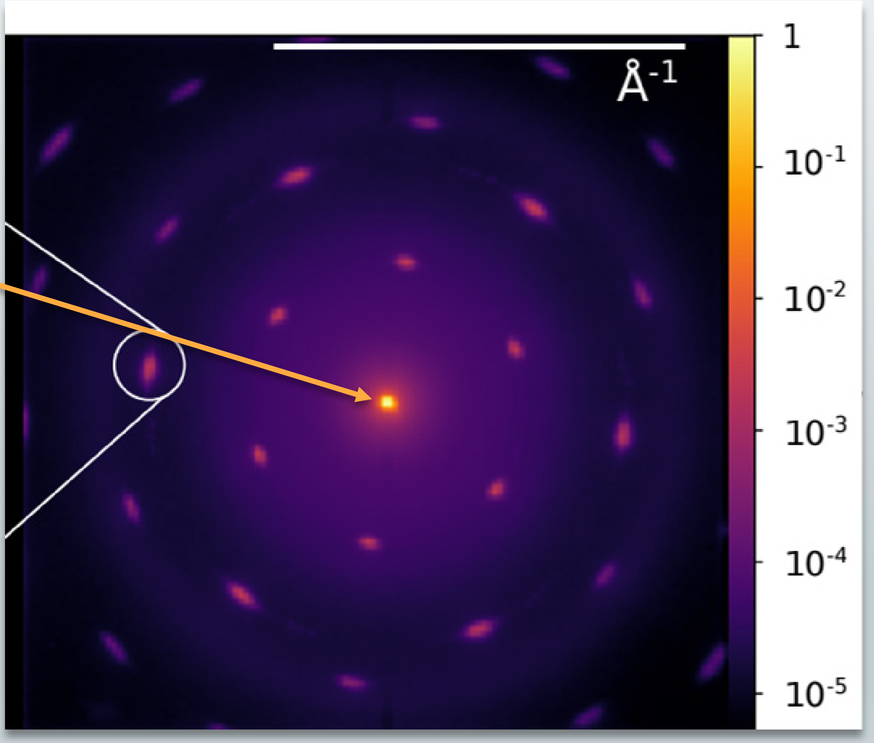
Interesting physics at multiple scales:

Undiffracted beam



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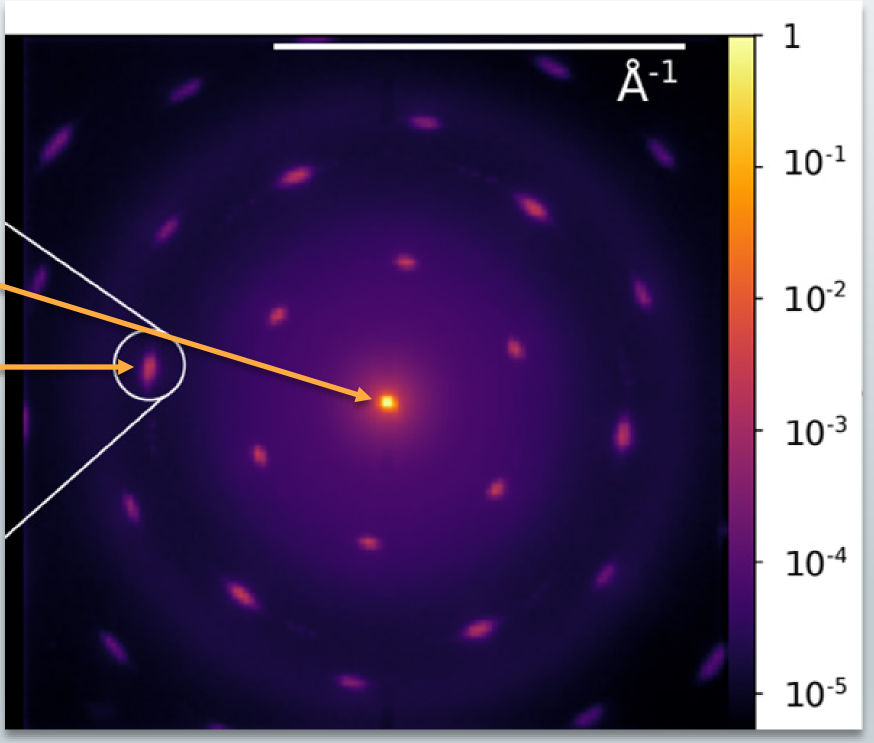

Undiffracted beam

Bragg Peaks: note “smearing” due to moire twist)

WSe_2 rotated 2° / $MoSe_2$ monolayer

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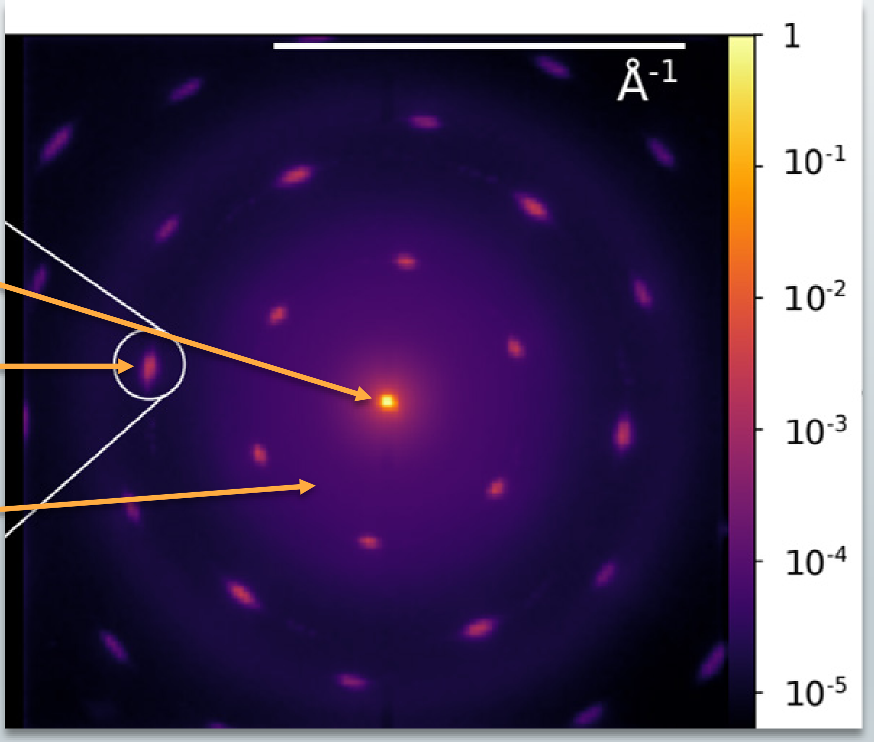

Undiffracted beam

Bragg Peaks: note “smearing” due to moire twist)

Diffuse Scattering

Only two monolayers thick!

WSe_2 rotated 2° / $MoSe_2$ monolayer

The need for reciprocal space resolution

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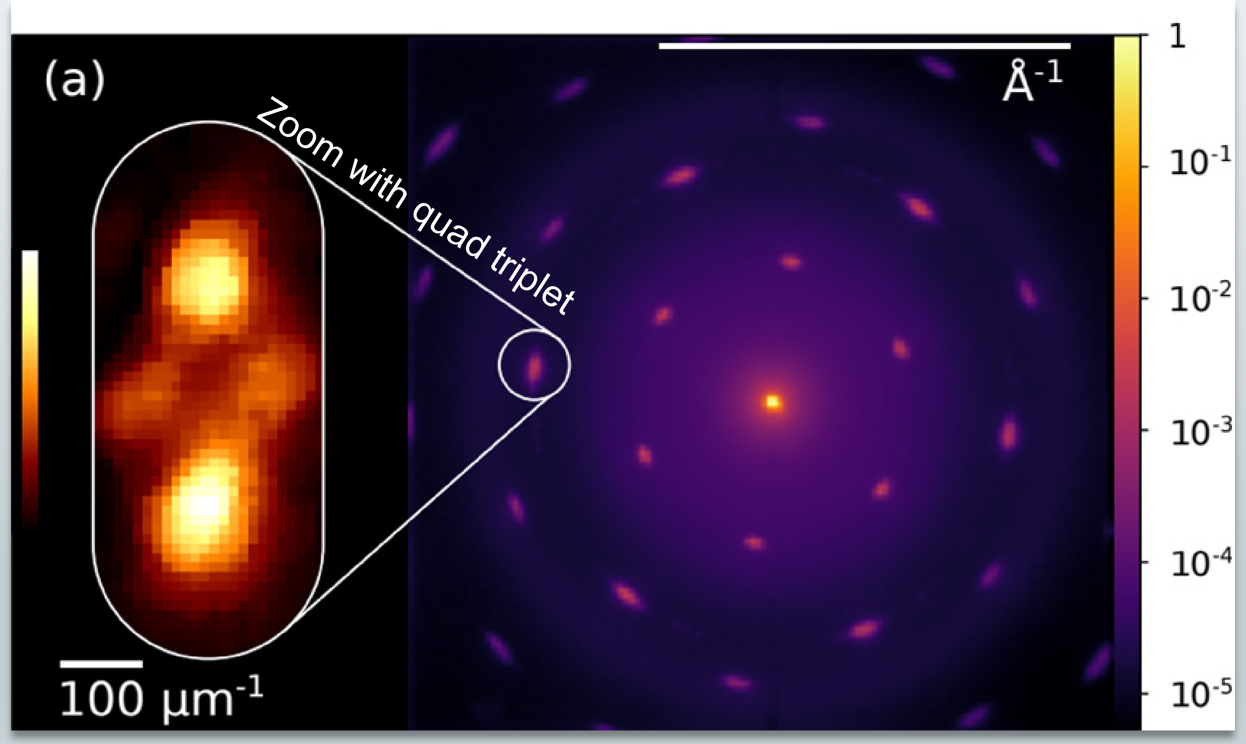


First observation of moire atomic reconstruction with any ultrafast probe!

Requires very high coherence ($\geq 10\ \mathrm{nm}$ coherence length)

MEDUSA Strategy for source brightness: Seek Low MTE

- MTE is rms transverse photoelectron momentum spread expressed in energy units:

$$\epsilon_{n,cath} = \sigma_{laser}\sqrt{\frac{\mathrm{MTE}}{mc^2}}$$

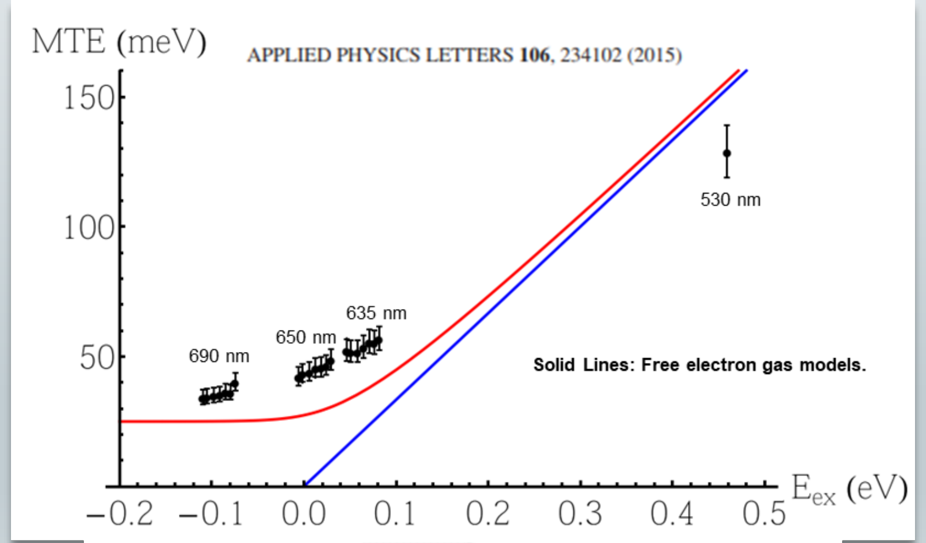
$$\boldsymbol{B_{n,max} \propto \frac{E_{acc}^{n}}{\mathrm{MTE}}}$$

where $1 < n < 2$ depends on bunch aspect ratio.
MTE’s of several hundred meV are common (Cu, Cs-Te).

- But does low MTE actually matter when space charge is present? We will get back to this.

Excellent Low MTE Candidate Materials: Alkali Antimonides

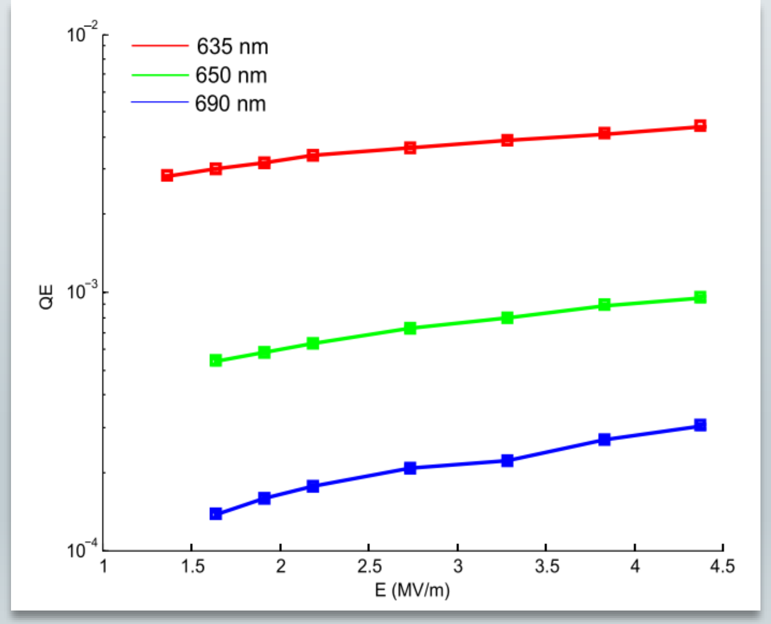
- By reducing the excess energy of photoemission, one can *trade* quantum efficiency for lower MTE.
- Alkali antimonides achieve as low as ~30 meV (shown below: Na-K-Sb, min MTE of 35 meV) with photon energy tuning.
- >10x max. brightness as compared to Cs-Te or polycrystalline Cu in traditional operation.
- Lower QE must be balanced against increased laser energy—ultimate limit is multiphoton photoemission, which spoils low MTE.



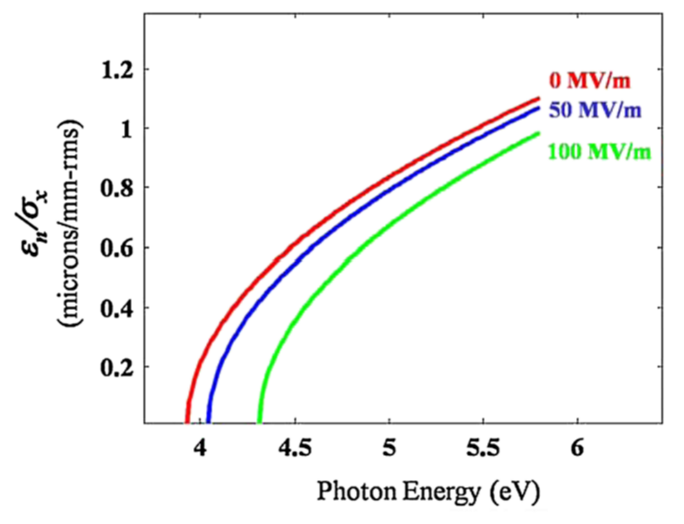
APPLIED PHYSICS LETTERS **106**, 234102 (2015)

Solid Lines: Free electron gas models.

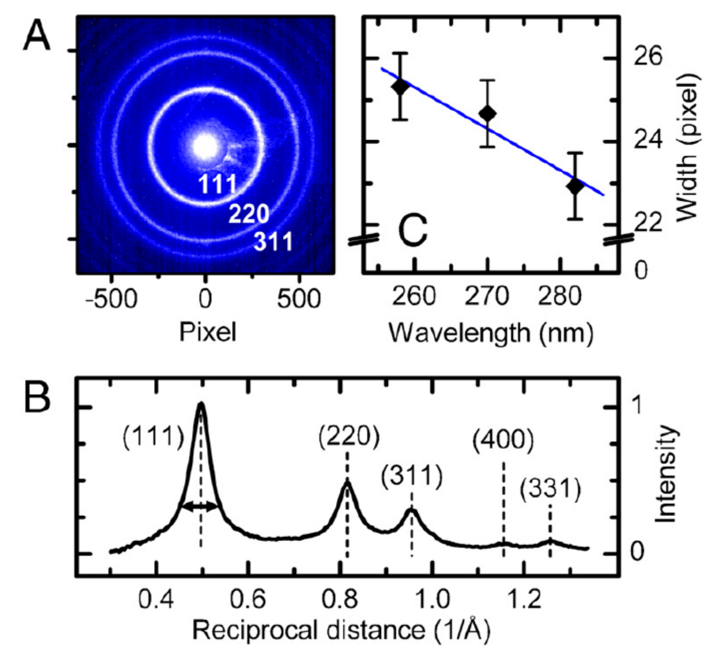
$E_{ex} = h\nu - (\phi - e\sqrt{eE/4\pi\epsilon_0})$. We assume $\phi = 1.95$ eV.



Threshold photoemission for low MTE is not a new idea



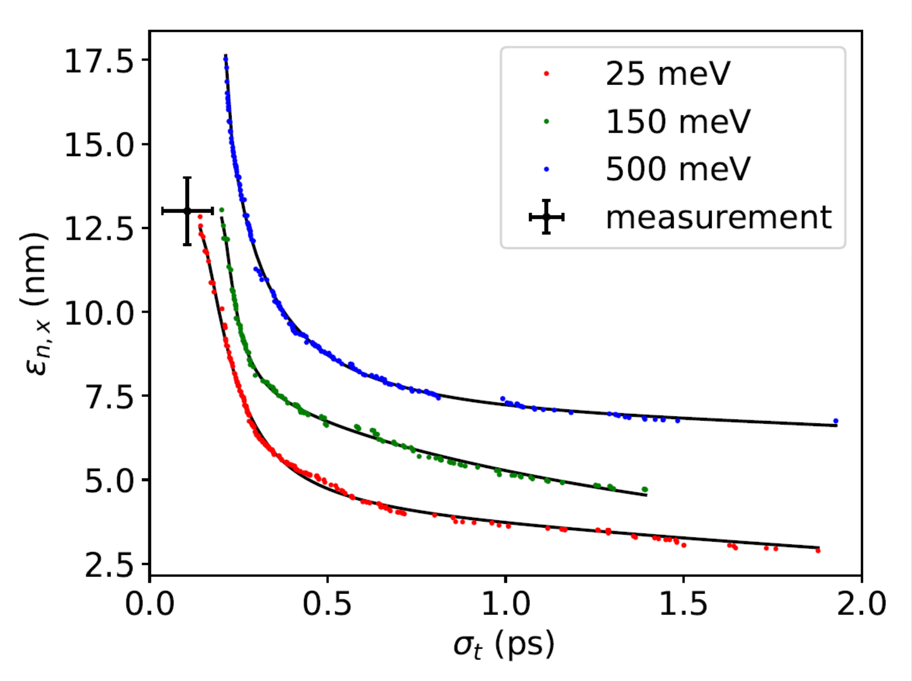
Dowell and Schmerge, PRSTAB 12, 074201 (2009)
See also: J. Feng, APL 107, 134101 (2015)



M. Aidelsburger, PNAS 107 46 (2010).

But even *very low MTE values* remain useful

MOGA optimization: MEDUSA Beamline. DC Gun (140 kV) + RF buncher, 100,000 e/bunch
(space charge very much not negligible)



Lower MTE provides access to:

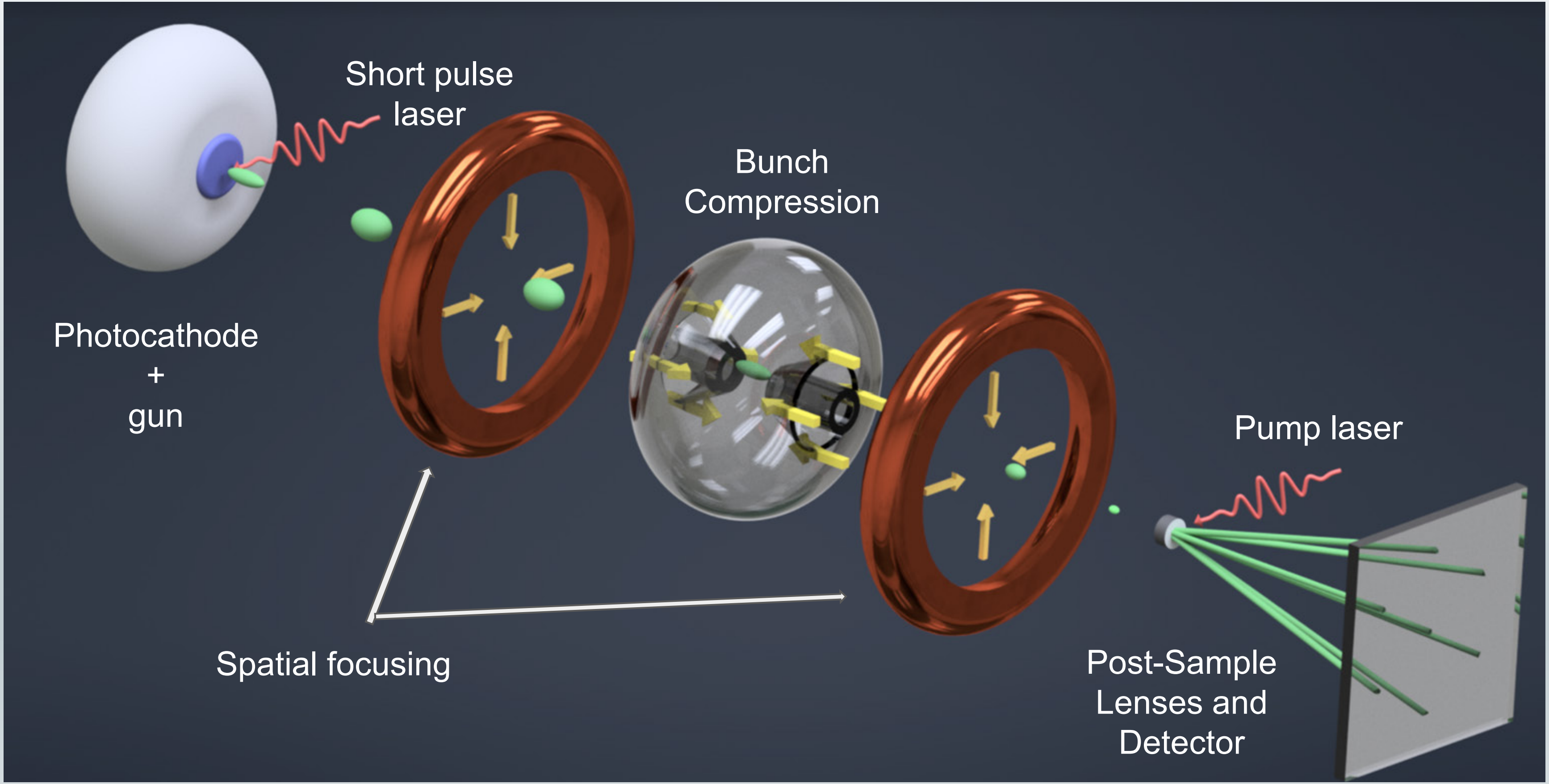
Smaller bunch length for a given emittance, or vice versa

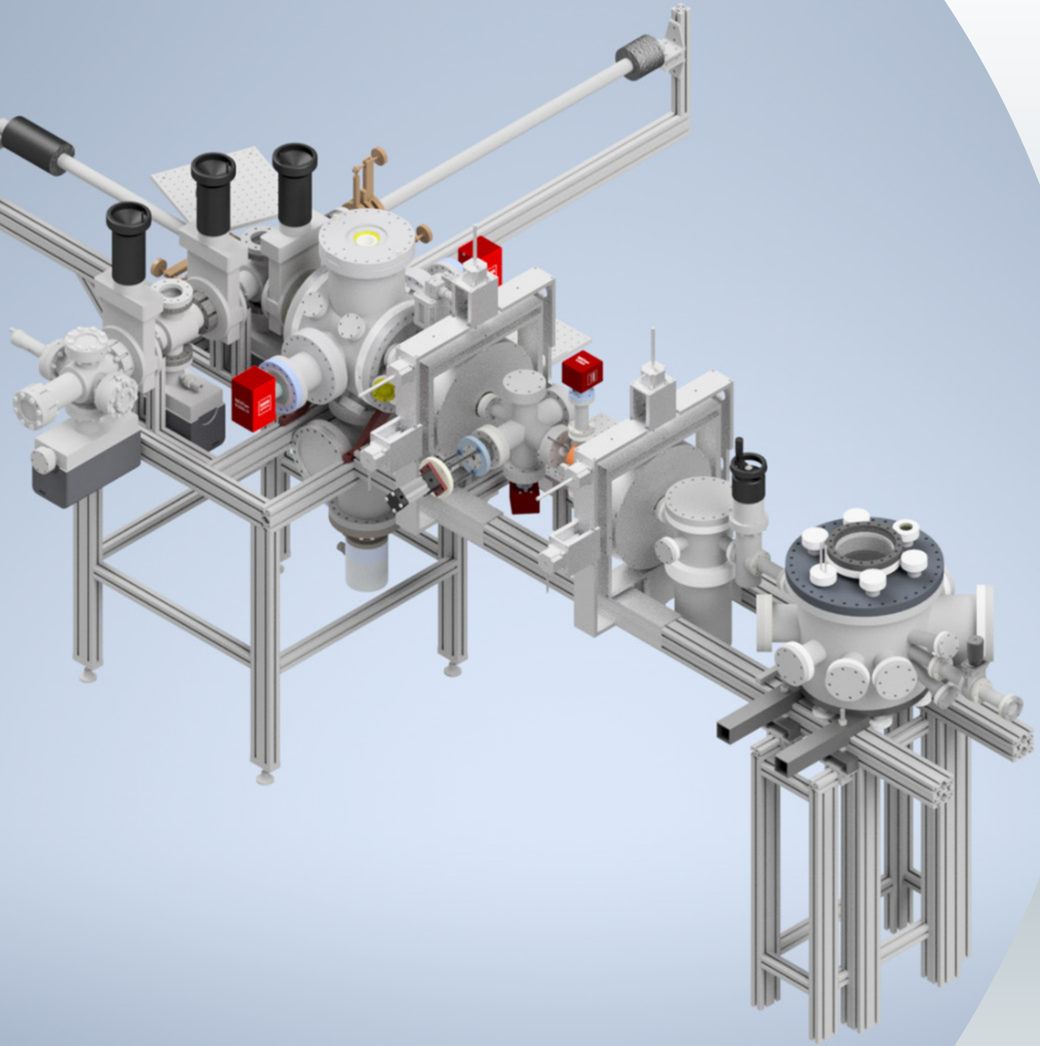
An important effect even below 100 meV MTEs

We regularly measure emittances of 12-14 nm depending on transverse optics, and bunch length between 100-200 fs rms.

Infer MTE significantly less <100 meV

Cartoon of MEDUSA: The critical components





Our device: piece by piece.

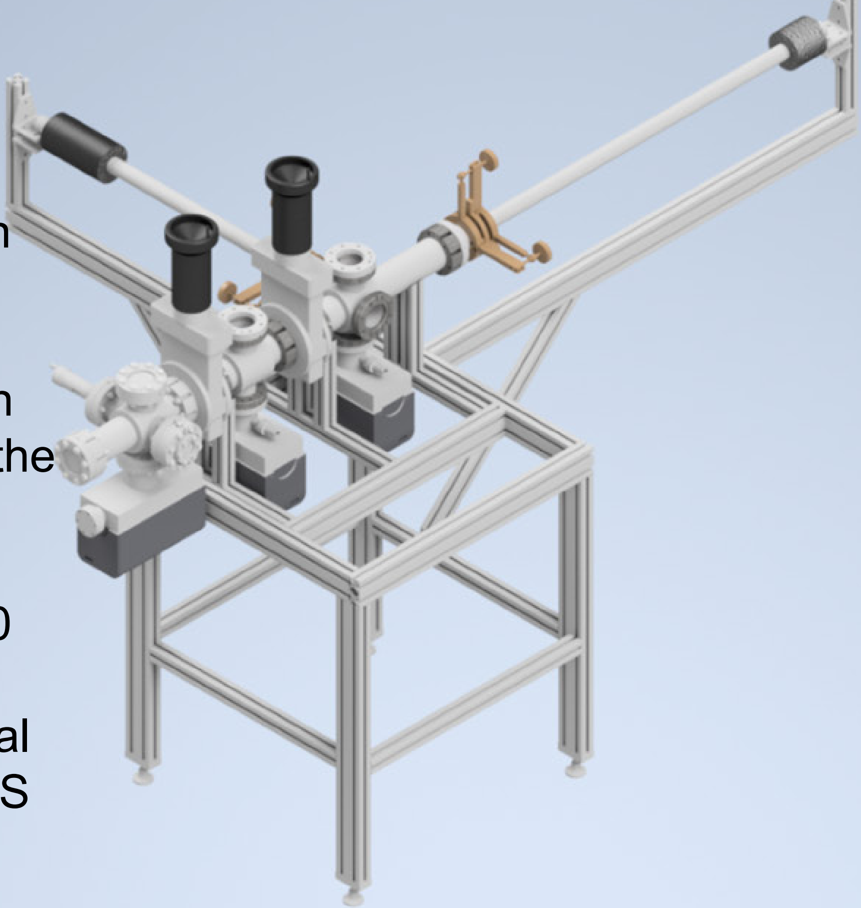
Beamline

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

650 nm photo-emission wavelengths matches the cathode bandgap

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

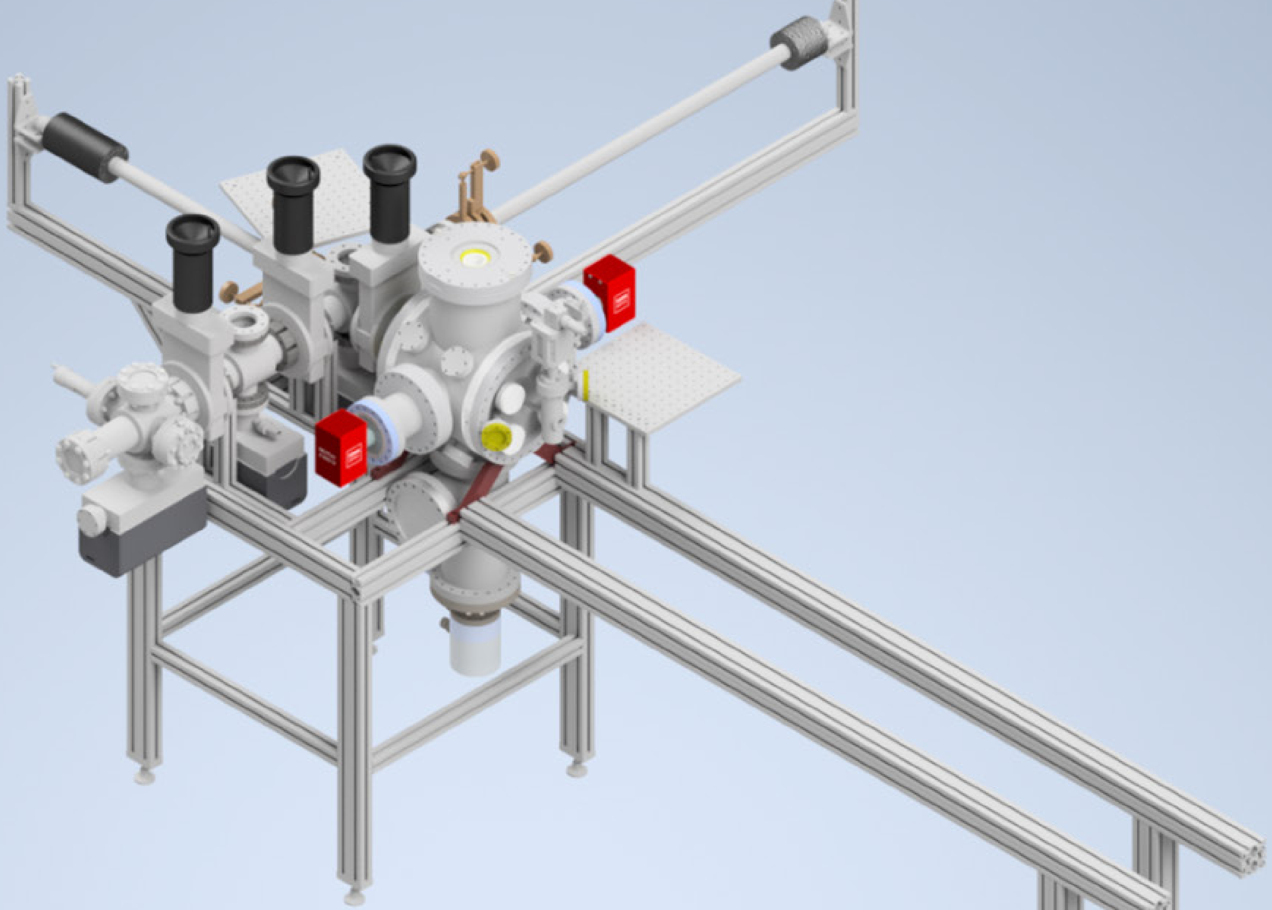
Source is extremely vacuum sensitive (XHV)



Beamline

150 kV DC gun

Base pressure:
8×10^{-12} Torr



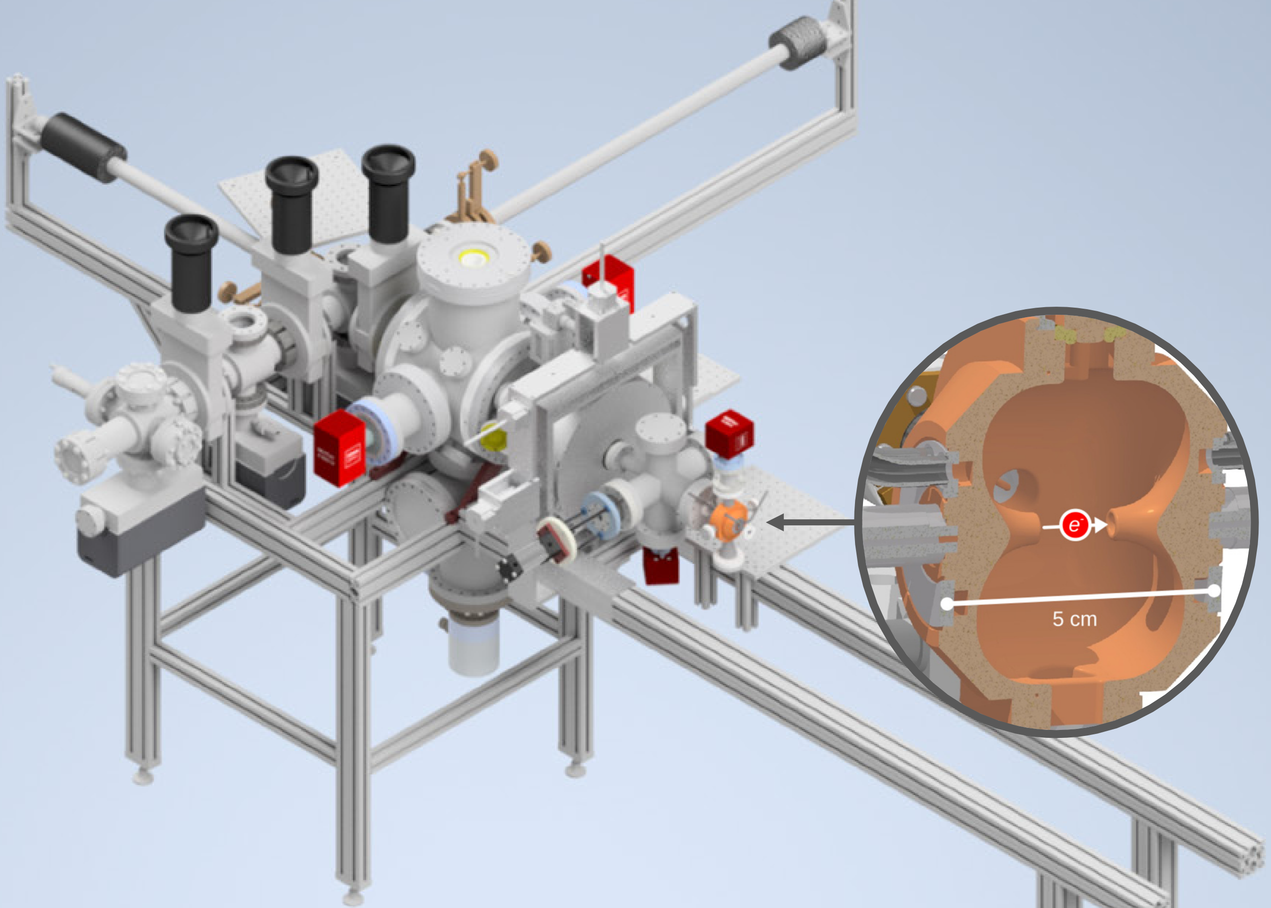
Beamline

3 GHz bunching cavity

Long pulse length at cathode: ~10 ps

Bunching after acceleration mitigates space-charge

~100 fs rms bunch-length at sample



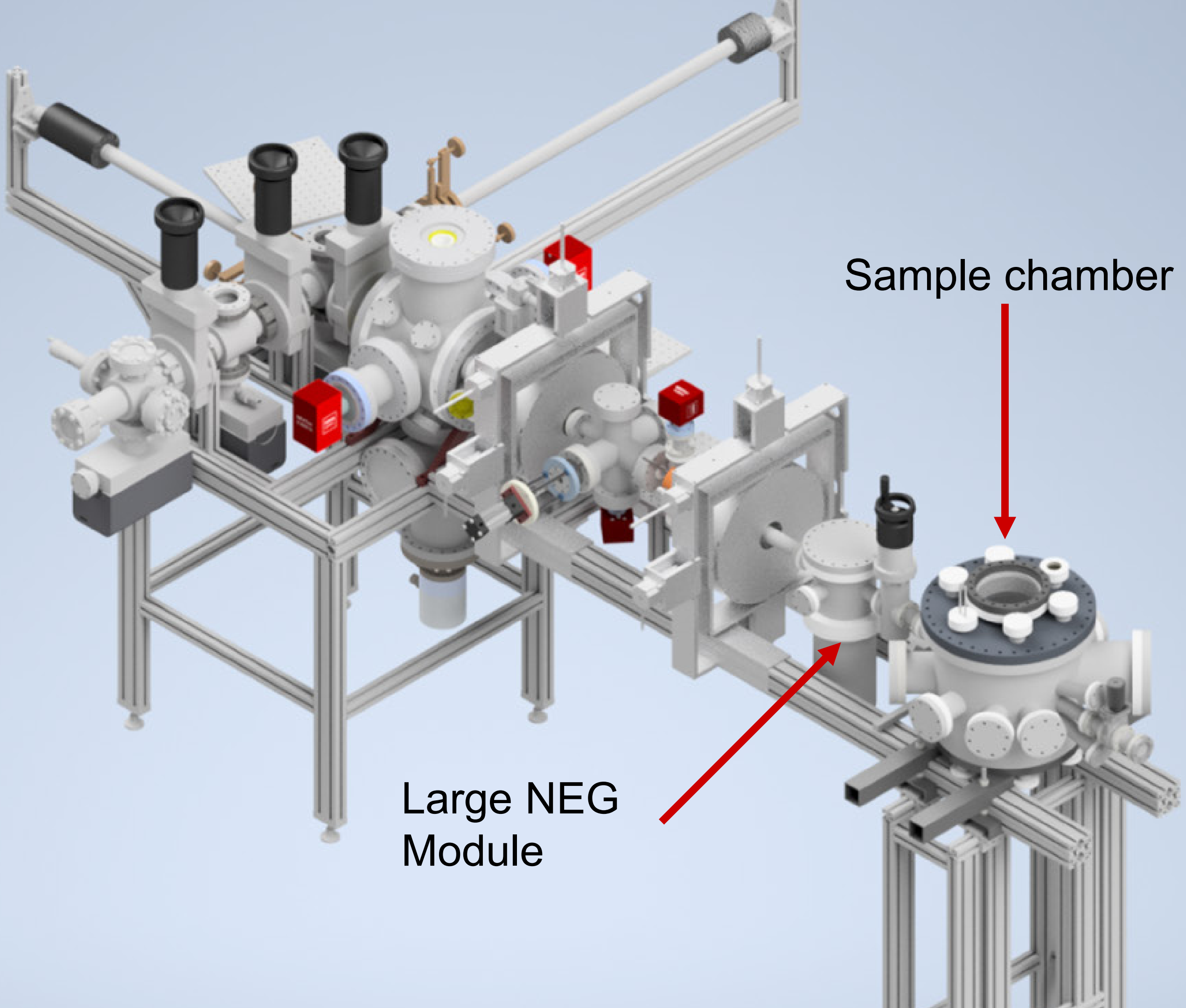
Beamline

Sample chamber

Pressure 10^4 × higher than gun

515 nm pump pulses, second-harmonic of Tangerine

(future upgrade: Visble and NIR OPA)

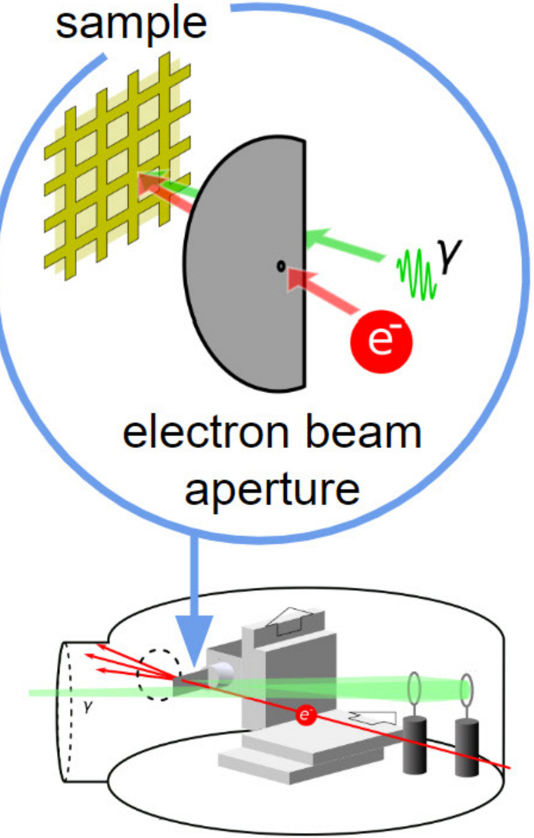


Sample chamber

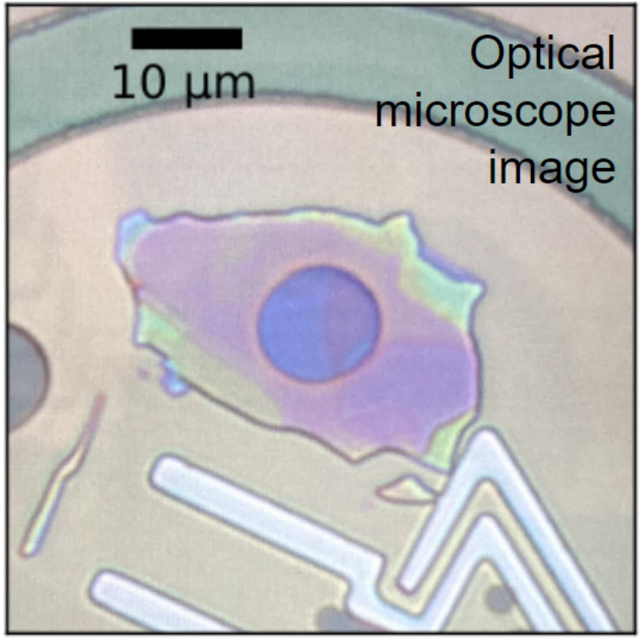
Large NEG Module

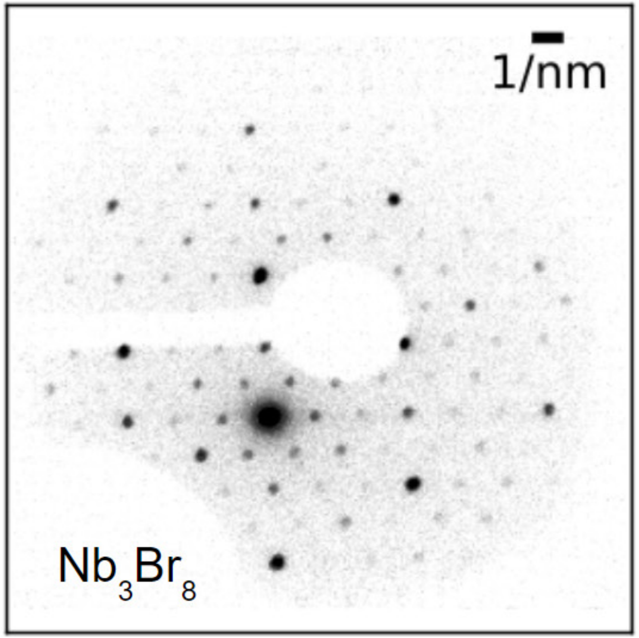
Spatial Resolution

Microdiffraction probe



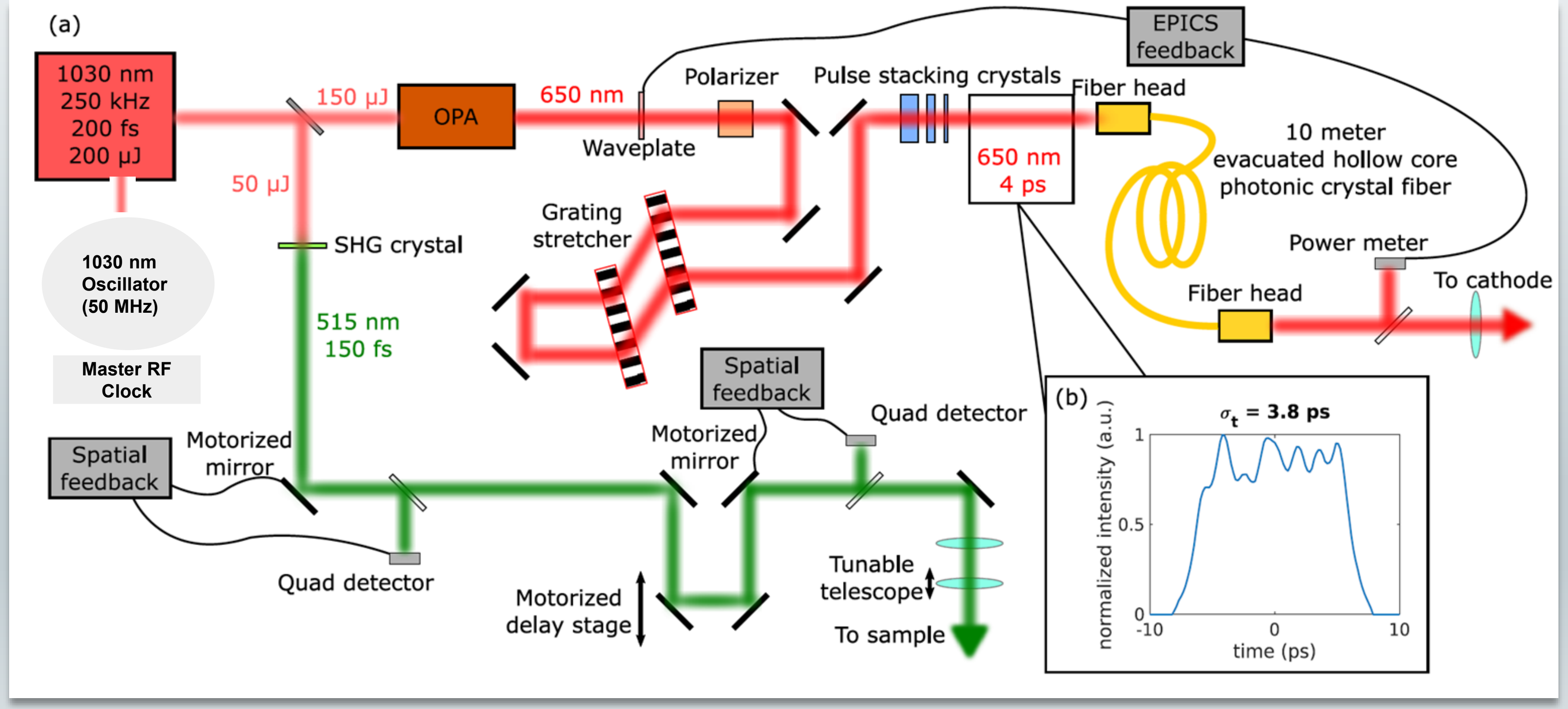
	With aperture	No aperture
Spot size on sample (r.m.s.):	3 µm	40 µm
Electrons on sample per pulse:	500	10^5





Thanks to Lena Kourkoutis and Elisabeth Bianco for providing the Nb_3Br_8 sample

The UED Laser system: “Much ado about the photocathode drive laser”



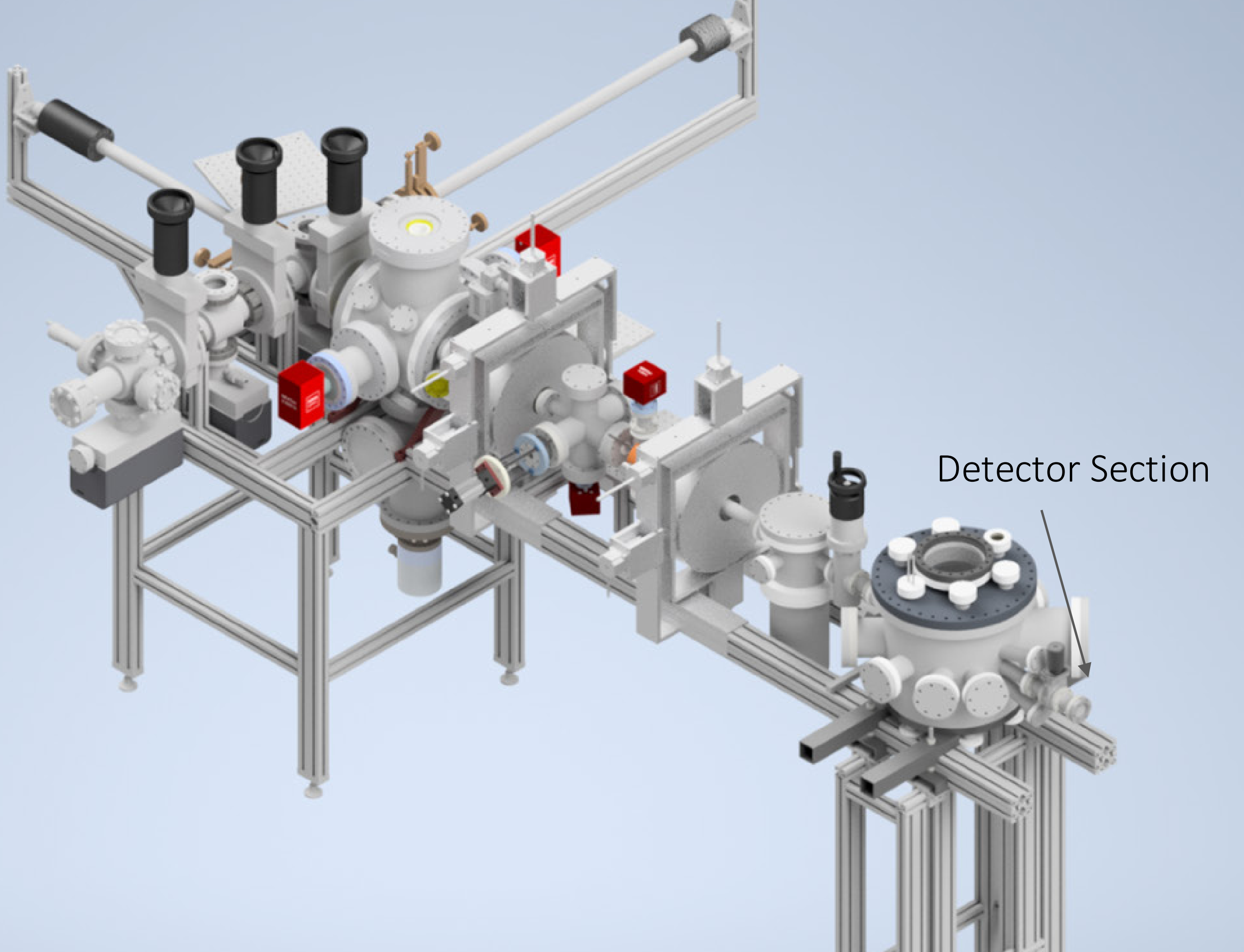
Beamline

Sample chamber

Pressure 10^4 × higher than gun (UHV)

Direct Electron Detector (EMPAD) deployed in collab. with Gruner/Thom/Muller

Quad triplet post-sample not shown

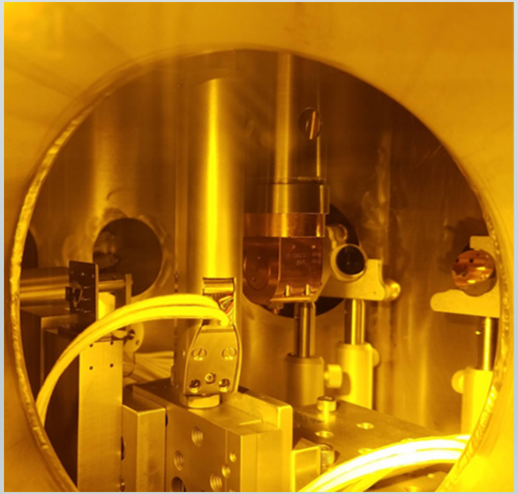


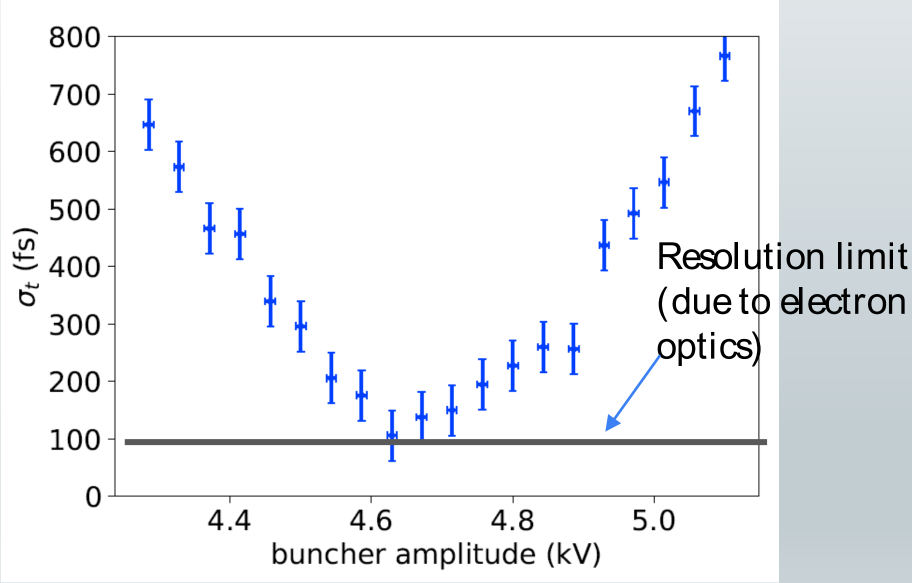
Bunch length measurements: compact RF deflector cavity

3 GHz *insertable deflection cavity manufactured by Dr. X. Works, Eindhoven*.

Drops in just upstream of sample location

Looking through laser entrance window
Note 1'' light optics for scale





Resolution limit (due to electron optics)

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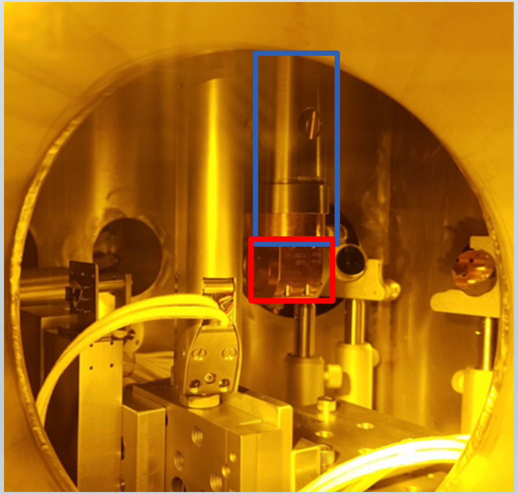
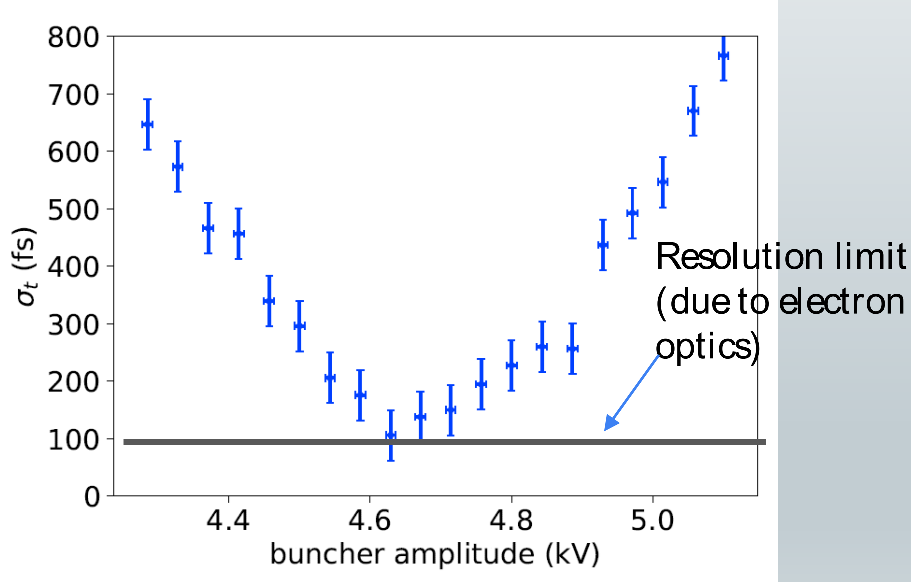
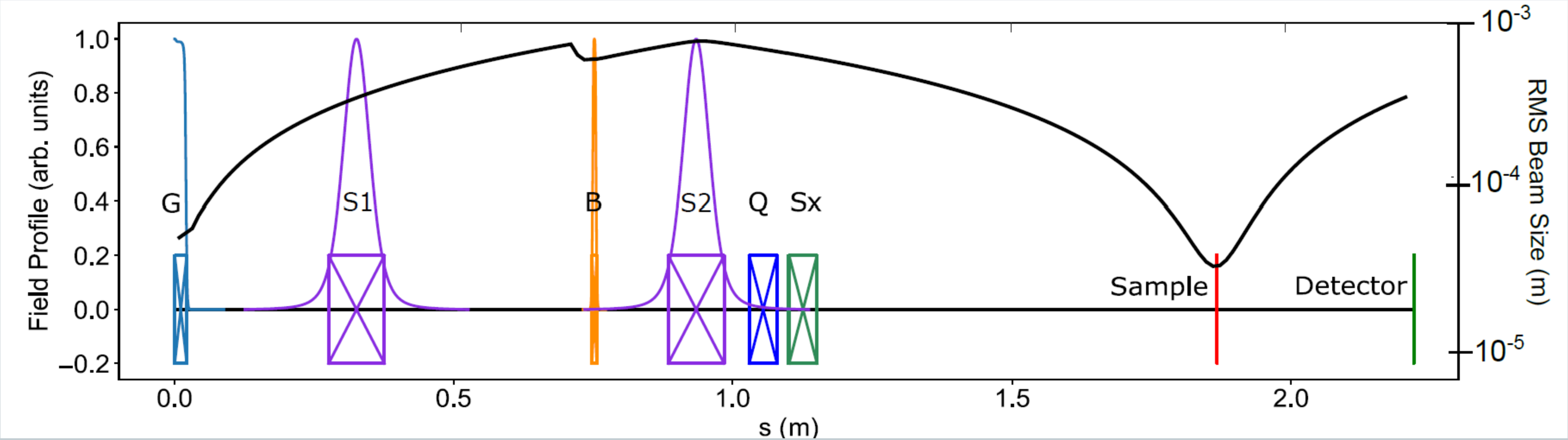


Mounting Pole
Cavity



Aberrations: an important obstacle

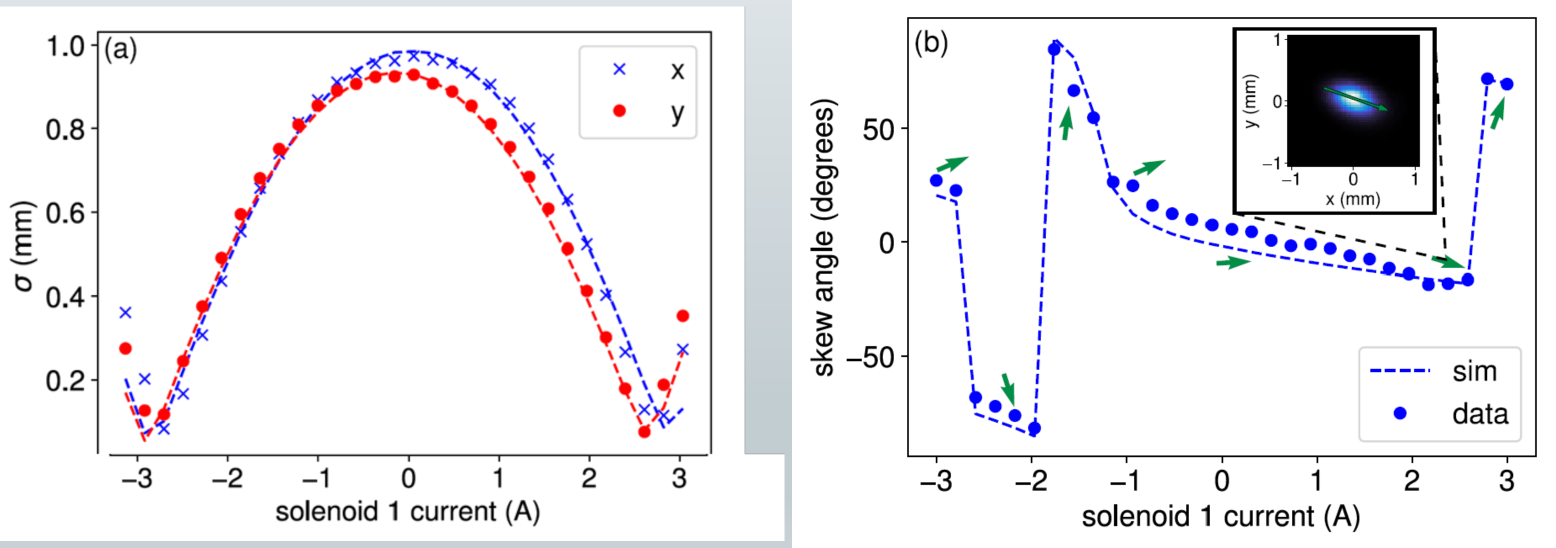
- Our microdiffraction optics rely on large changes in beam size:



- This leaves us *very vulnerable to field aberrations.*
- We have dedicated correction magnets for quadrupole, skew quadrupole, and sextupole moments.

Aberrations: Normal and Skew Quadrupole

- Erroneous quads are found in our solenoids *and* due to the coupler kick of the bunching cavity where beam size is large.



Lines: simulation, dots: measurement

Aberrations: Sextupole

- We use quadrupole correctors just downstream of our second solenoid→ as previously demonstrated [L. Zheng, PRAB **22**, 072805 (2019)] very effective in removing transverse coupling.
- Sextupole moment primarily arises from buncher coupler
- Once we do that, our beam looked like this on our diffraction detector:

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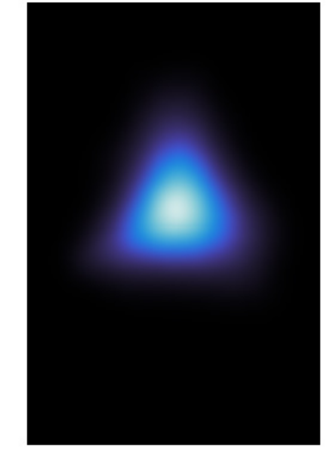
No Correction



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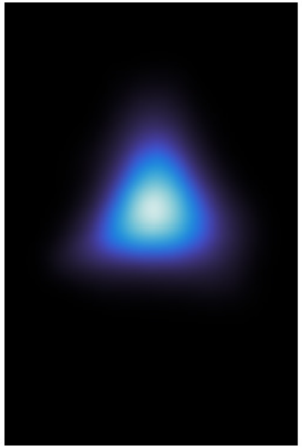

Overcorrection

Aberrations: Sextupole

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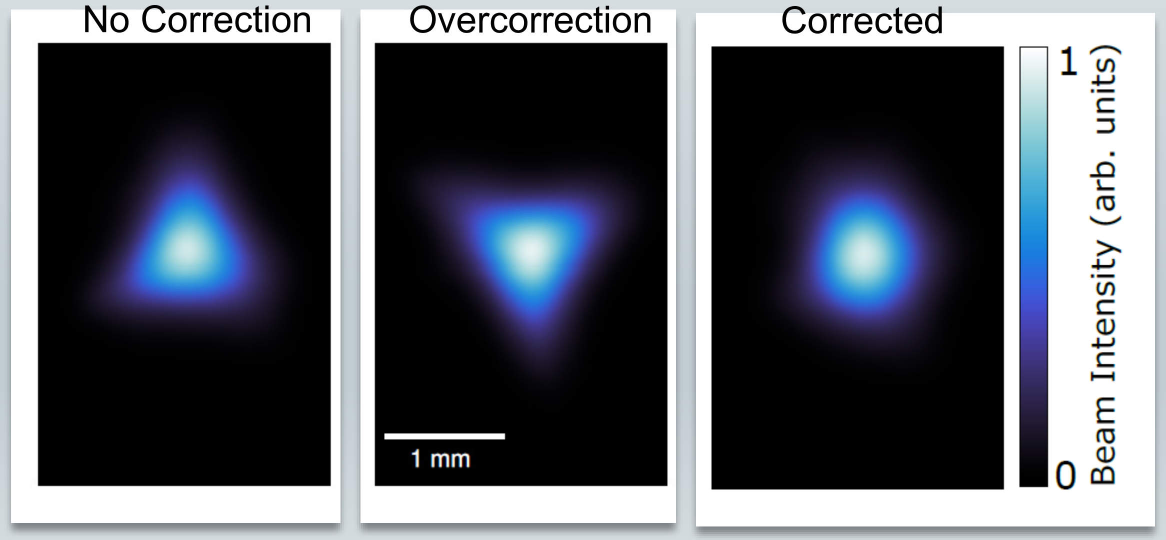
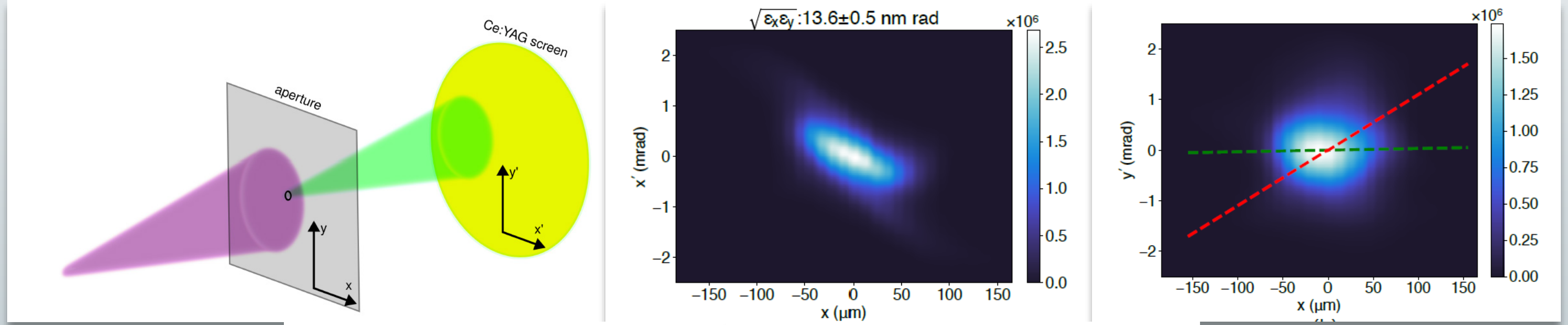
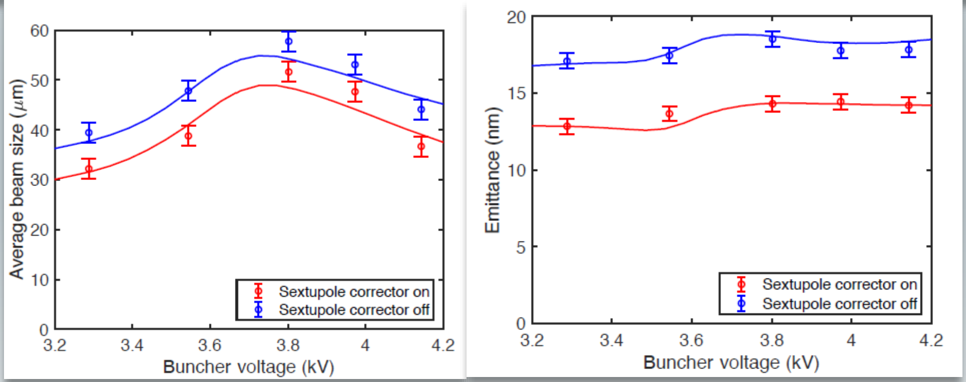


4D Phase Space Measurements

- We use 4D transverse phase space mapping to ensure cancellation of skew quadrupoles and sextupole moment

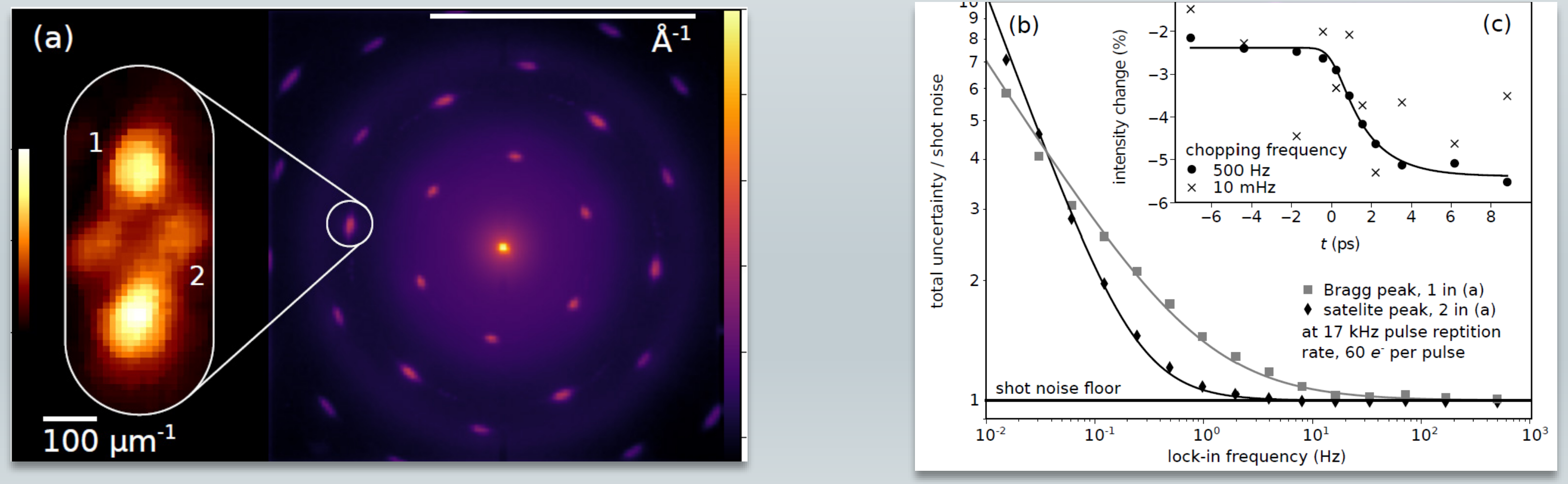




Lines: simulation, dots: measurement

Critical Step Forward: Direct Electron Detection

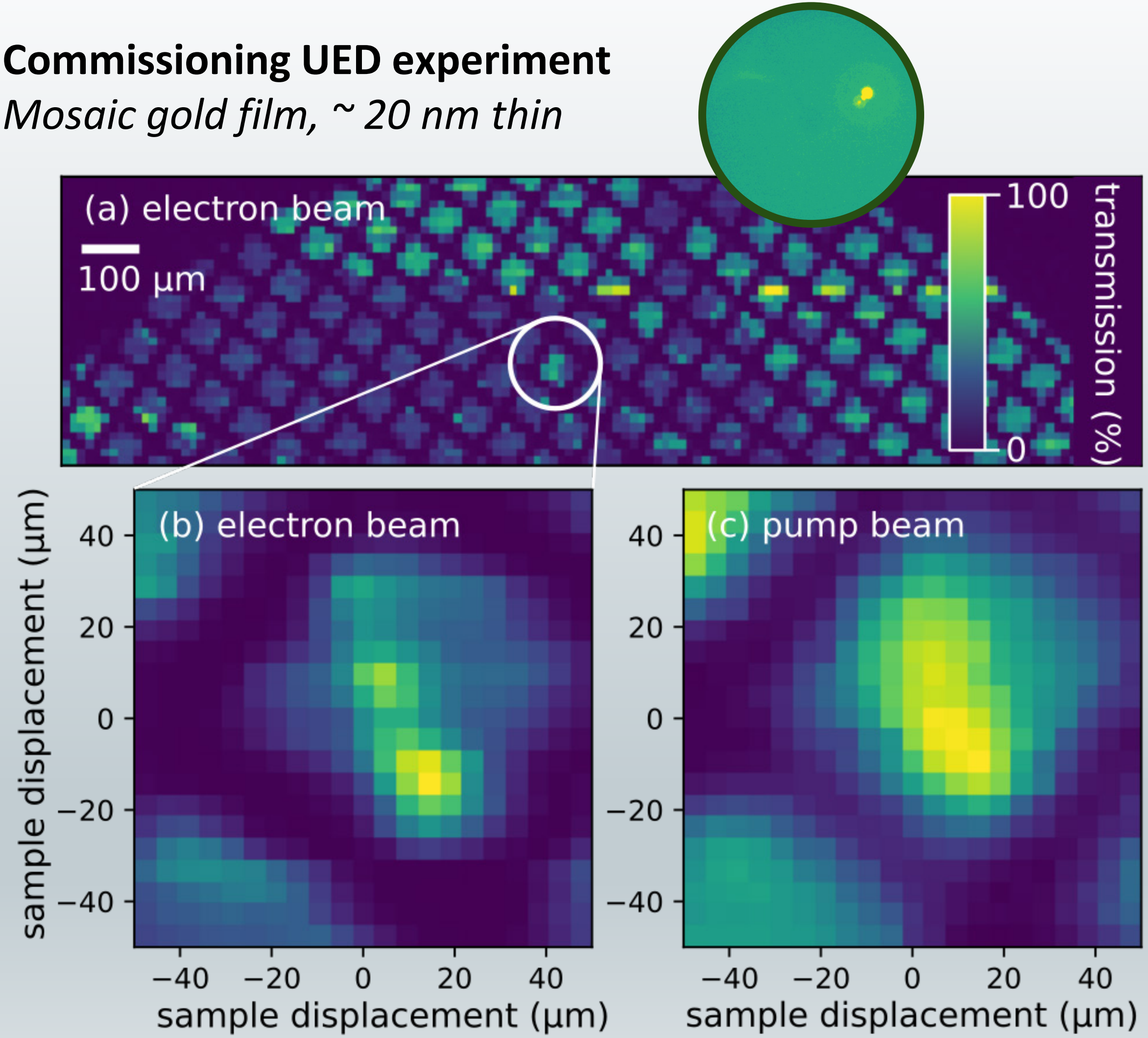
- Brightness in UED is only as good as your detector.
- A direct electron detector called the EMPAD, has been a huge step forward for MEDUSA**.** Collab with Gruner, Thom-Levy, and Muller at Cornell.
- Single Particle sensitivity (SNR ~100 per electron), and very high dynamic range (10^6)
- Images up to **1000 frames/second→ outrun noise!**



To Conclude: Some examples of what you can do with a UED microprobe

Commissioning UED experiment

Mosaic gold film, ~ 20 nm thin



Why gold?

Responds strongly to temperature changes that are small compared to the melting point, 100 K vs 1300 K

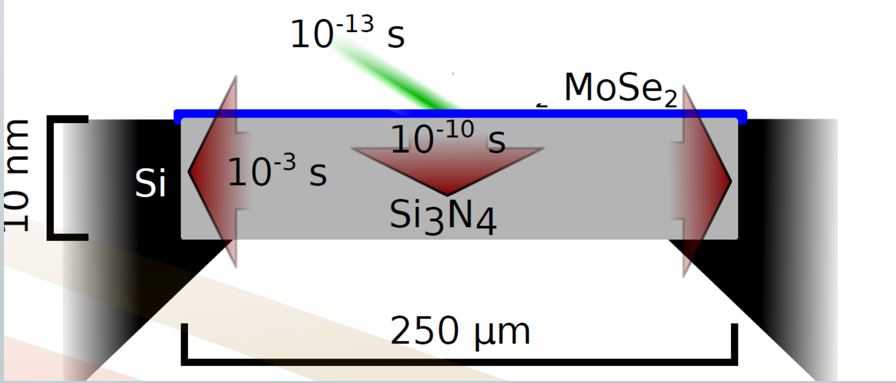
We can pick out individual grains of a mosaic material

We can make our pump beam very small! (10 micron rms)

→ Reduces average power needed for pumping!

Mapping the full life cycle of optical excitation

- With a high rep-rate laser *and detector* need both, you can use pulse-picking to extend UED delays out to microsecond-millisecond-second timescales.
- This allowed us to watch the full “life cycle” of optical excitation in thin films.
- Example 1: Monolayer $MoSe_2$ atop SiN



Step 1: light absorption and e-ph scattering, few ps

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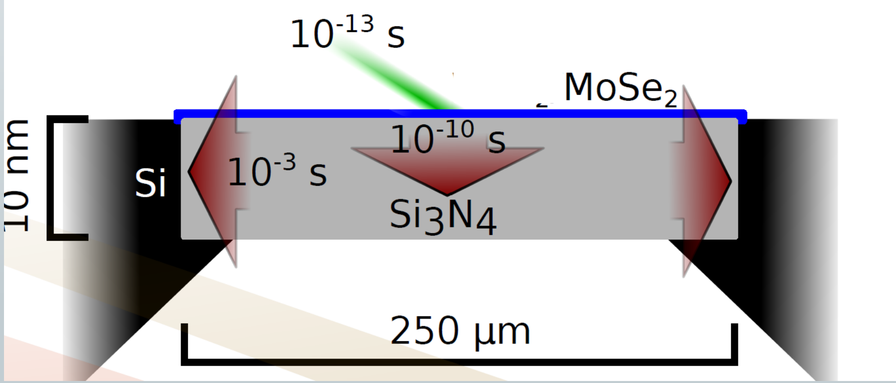

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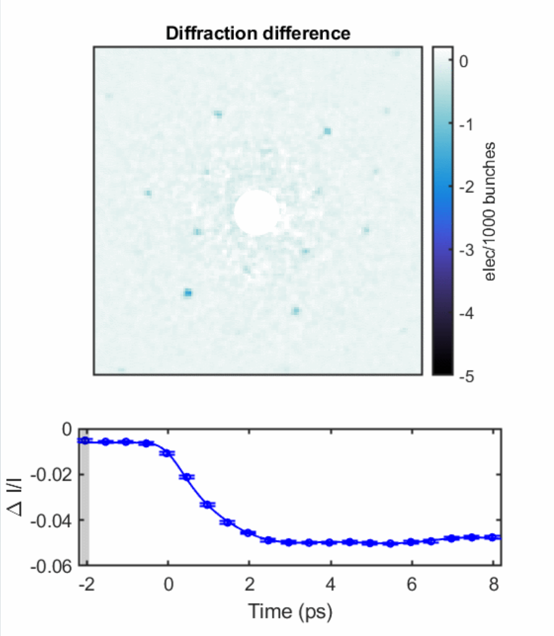

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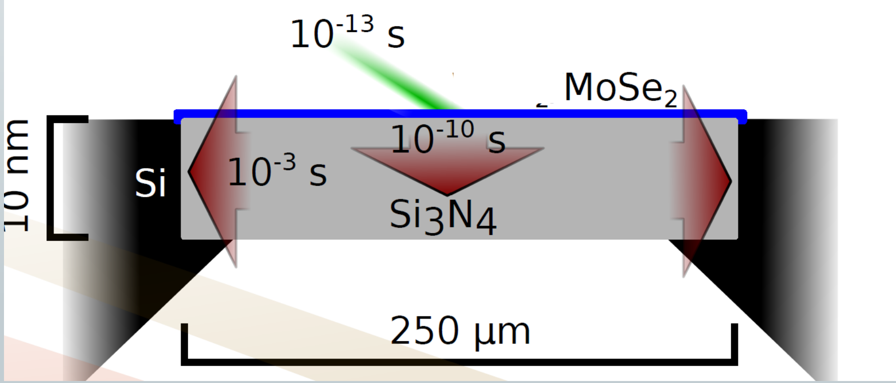
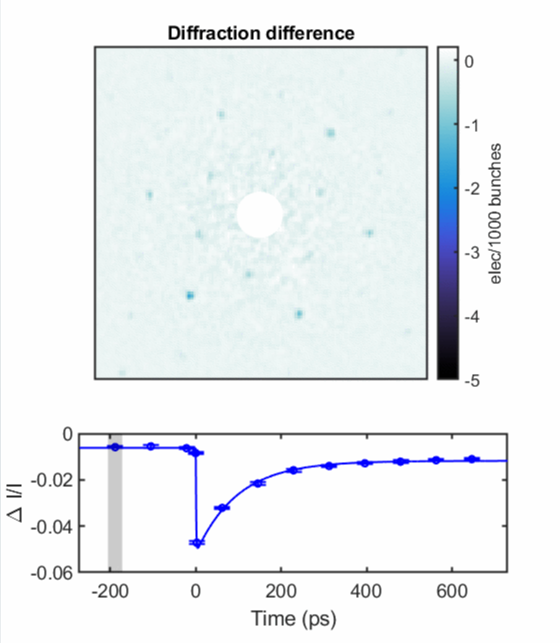
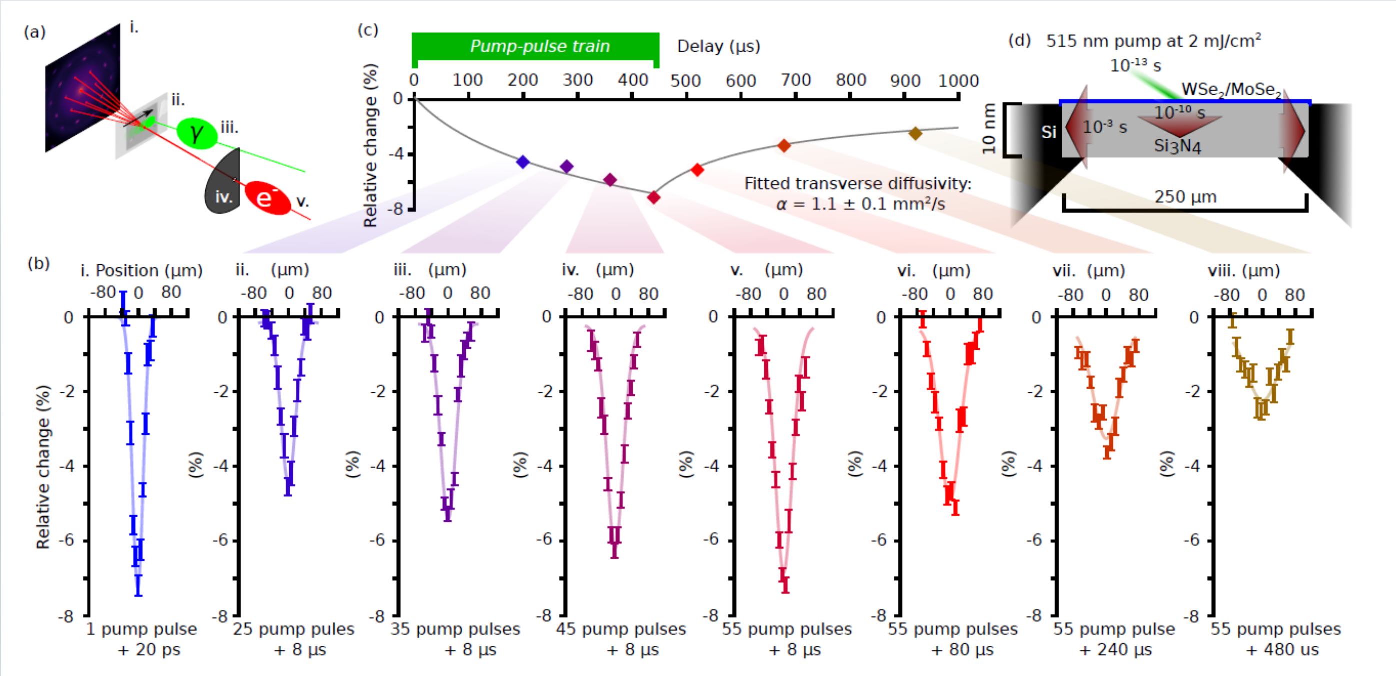


Step 2: *Longitudinal* heat transfer to SiN (100s of ps)



Mapping the full life cycle of optical excitation

- Example 2: Monolayer ***WSe_2/$MoSe_2$ moire bilayers*** atop SiN
- We use pulse picking, small spot sizes, and high rep rate to track heat transfer out to millisecond timescales, and 10s of microns in space.



Conclusions

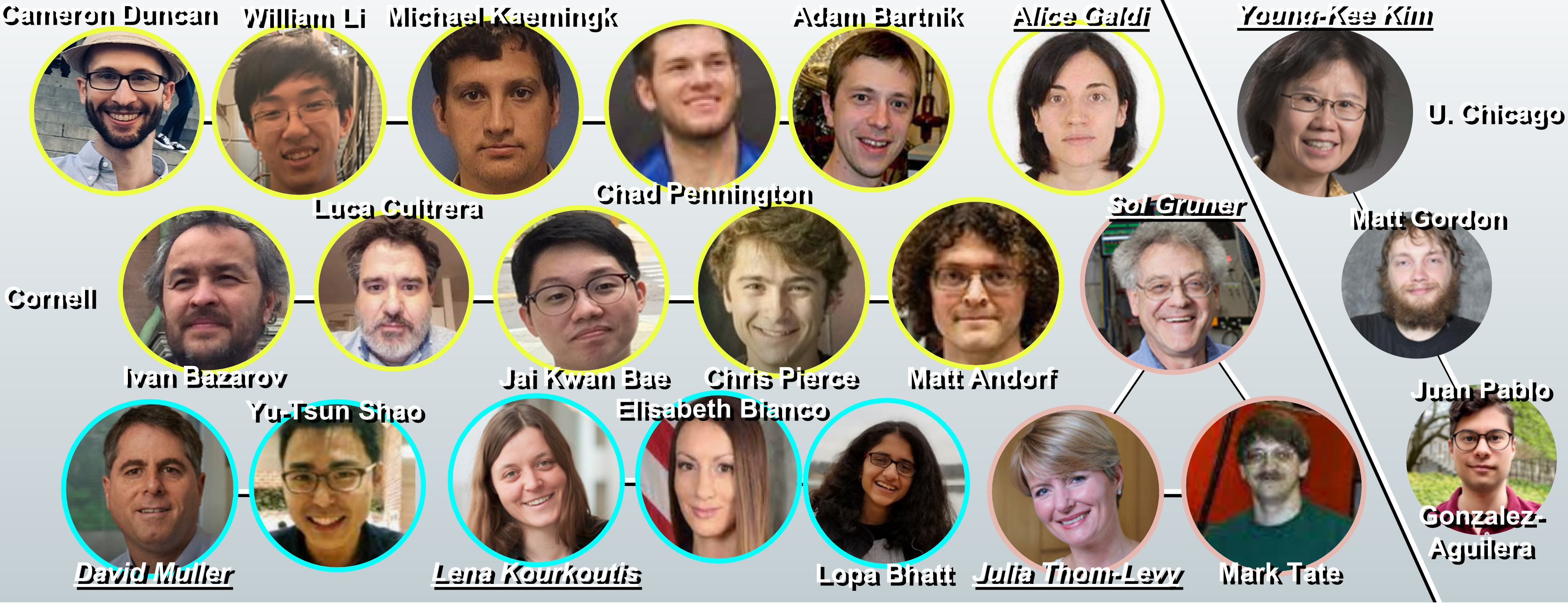
- Operating alkali antimonides at threshold gave us dramatic improvement in beam quality in UED.
- Diagnosis and correction of aberrations out to sextupole order was critical.
- Transversely small, coherent ultrafast electron probes are very useful, particularly for UED on quantum materials.
- Our electron source, coupled with a state-of-the-art direct electron detector, enabled a novel study of moire materials.

Papers this talk draws from:

- W. H. Li et al., Structural Dynamics **9**, 024302 (2022)
- M. Gordon et al., PRAB, Accepted (2022) [https://arxiv.org/abs/2207.13634]
- C.J. R. Duncan et al., in review [https://arxiv.org/abs/2207.13634]

Acknowledgements

Photo-emission Microscopy Detectors



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Thank you!