National Synchrotron Light Source II

Brookhaven National Laboratory

U.S. DEPARTMENT OF ENERGY

Current Status of Developing Ultrafast Mega-electron-volt Electron Microscope

Xi Yang

on behalf of BNL UEM team

08/10/2022

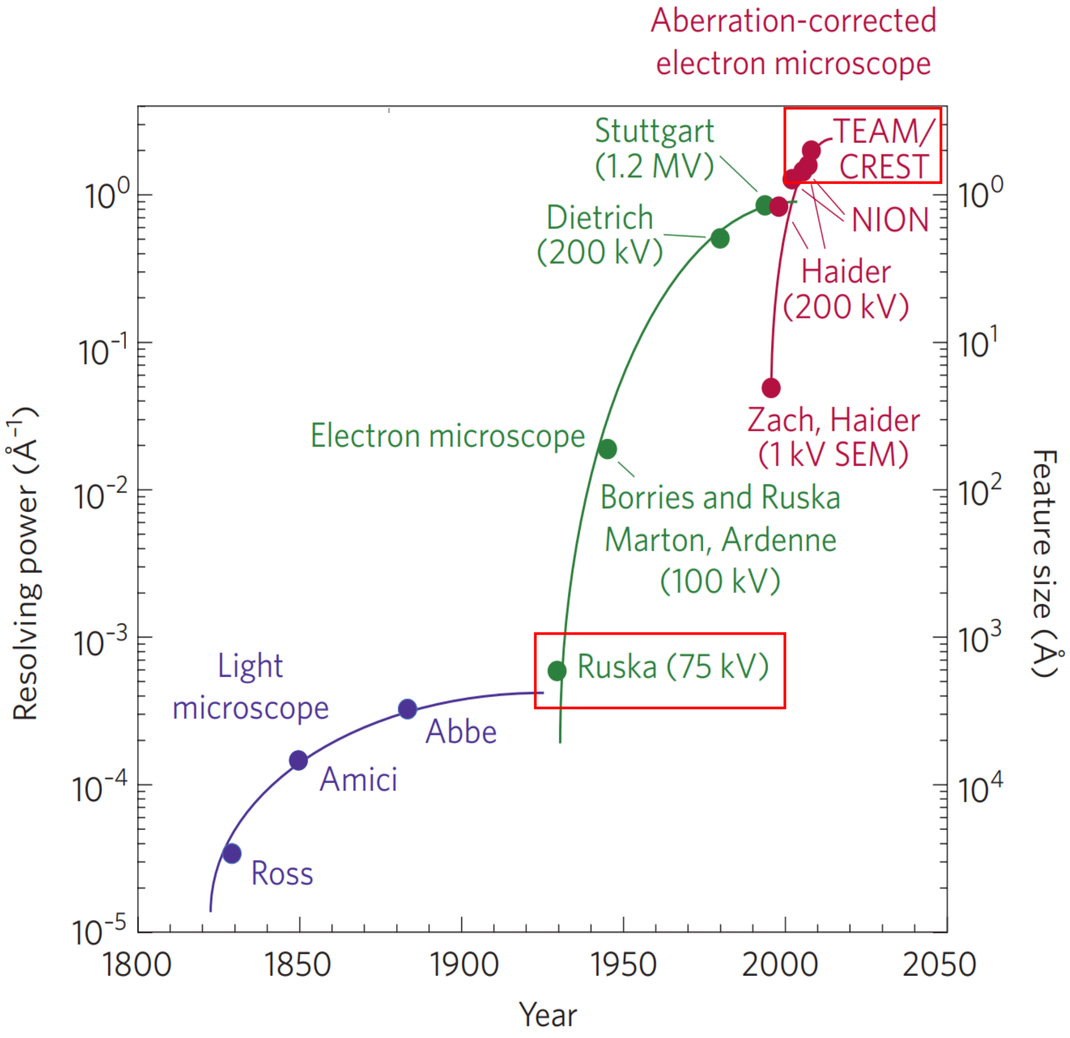
NAPAC 2022

Outline

- **Introduction**
  - **Review of scientific case for conventional TEM**
  - **Promise of MeV-microscopy**
- **History of MeV microscopy**
- **Review of the state-of art in the MeV-UEM field**
- **Physics & engineering challenges & solutions in reaching nm resolution**
  1. **Energy jitter**
  2. **Interaction of MeV-electrons to sample**
  3. **Mechanical stability**
  4. **Detector**
- **Conclusion**

Transmission electron microscopy

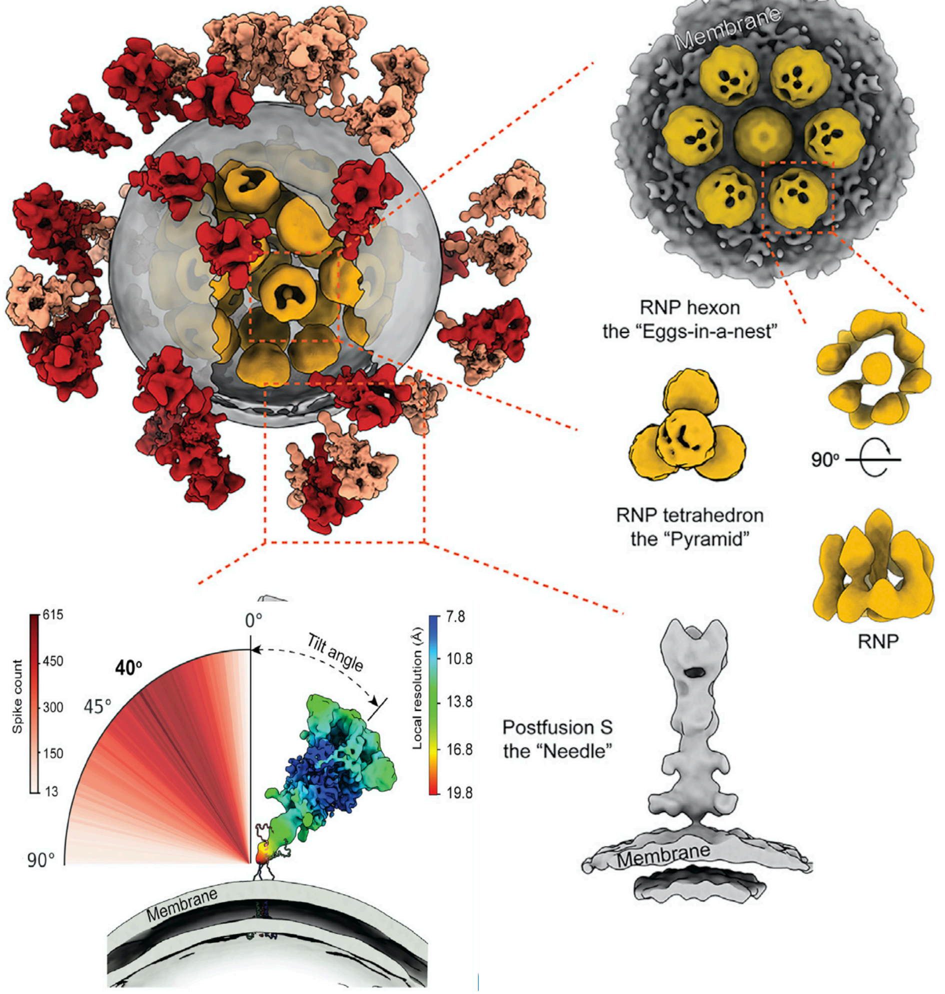
- Imaging tool for material science, chemistry, physics, biology, and industry
- Atomic resolution with aberration correction



D. A. Muller, Nat. Mater. 8, 263 (2009)
Adapted from H. H. Rose (2009)

Cryo-TEM

An intact virus to fight against covid-19

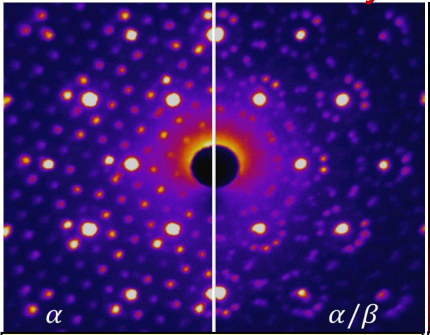


Intact virus applied to EM grids

Yao, H. et al. Cell **183**, 730 (2020)

Ultrafast science enabled by MeV UED

Courtesy of Xiaozhe Shen



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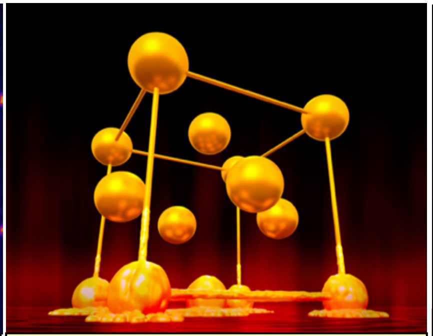

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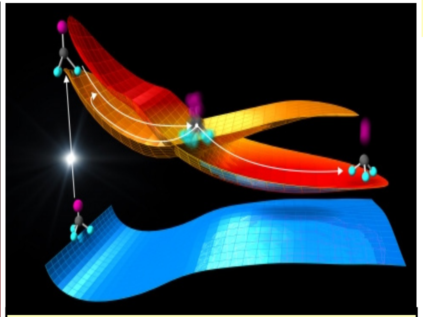

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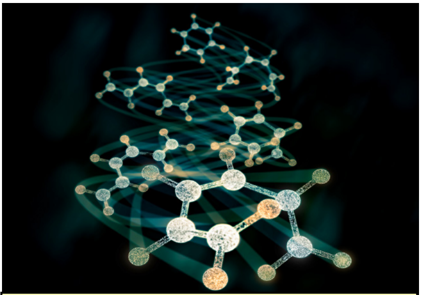
SLAC



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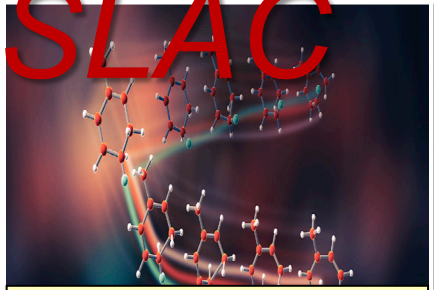

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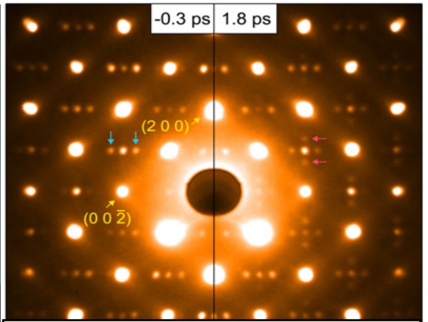

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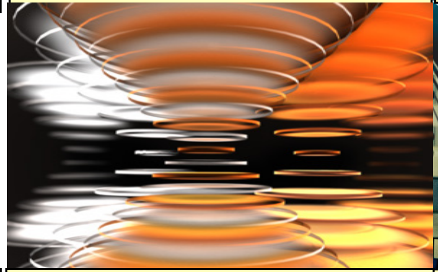

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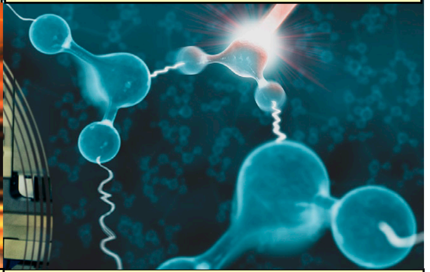

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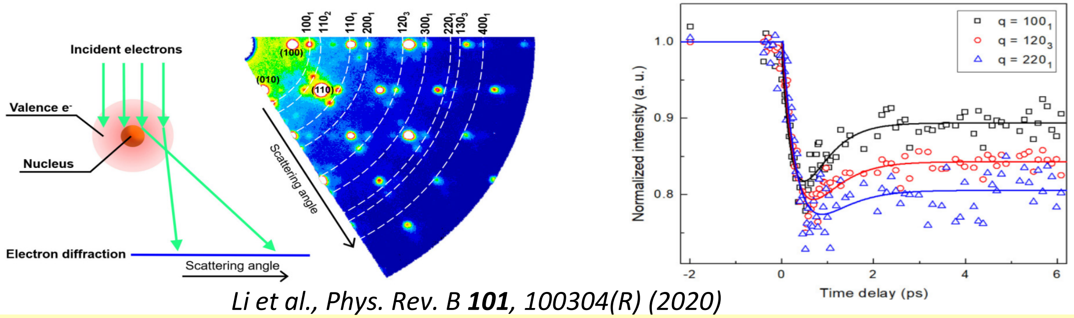

Li et al., Phys. Rev. B ***101****, 100304(R) (2020)*

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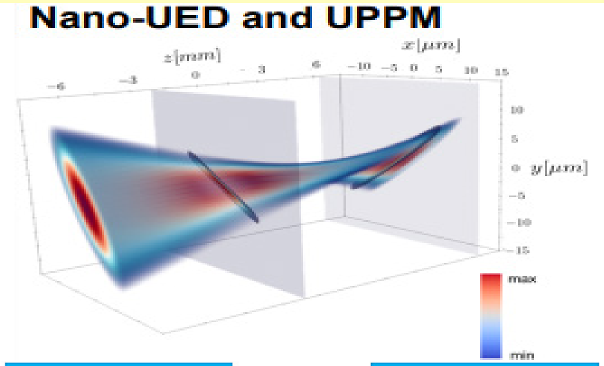
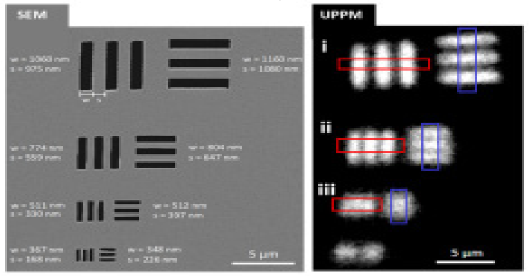
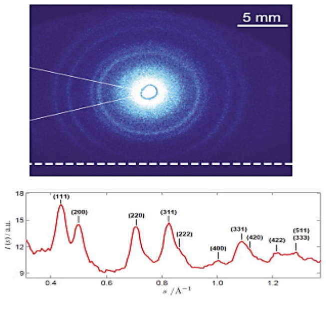


arXiv:1901.03443
arXiv:1903.12610

F. Ji, et al., Commun. Phys. 2, 54 (2019)



THU, 2009



RSI 80, 083303 (2009)

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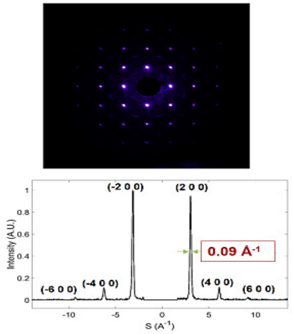

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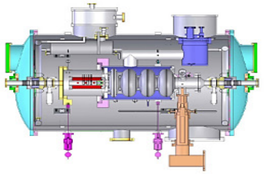

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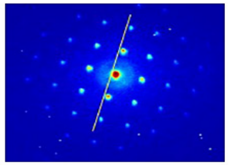
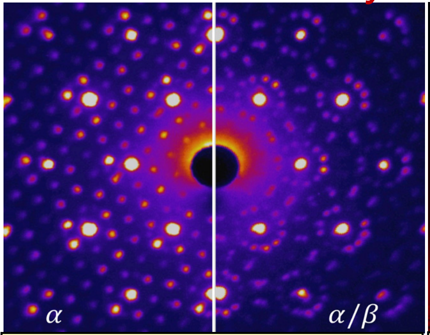


APL 107, 224101 (2015)

Ultrafast science enabled by MeV UED

Toward real space imaging enabled by MeV Microscopy

Courtesy of Xiaozhe Shen



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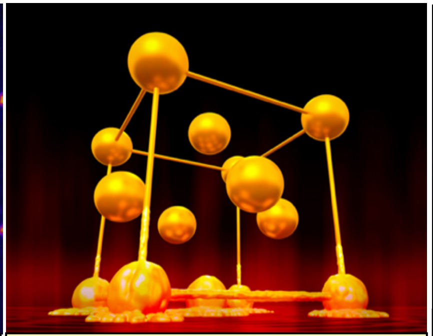

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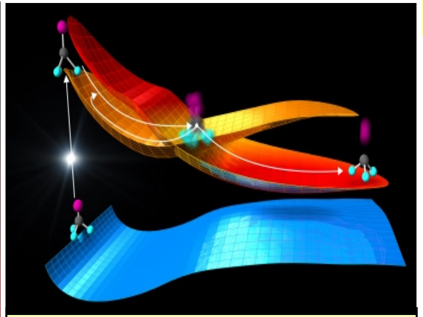

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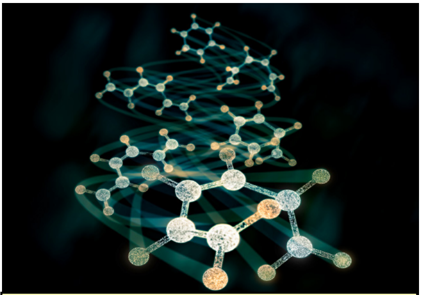

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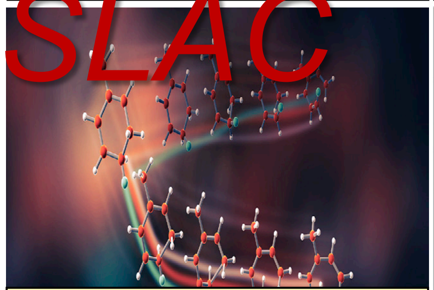

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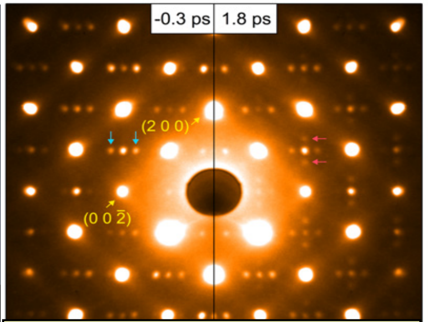

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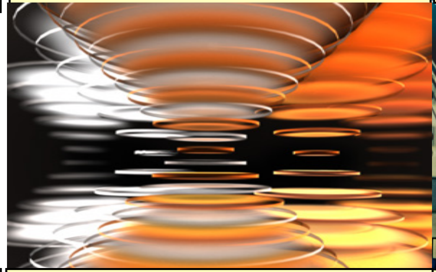

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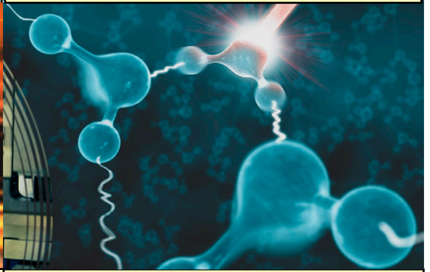

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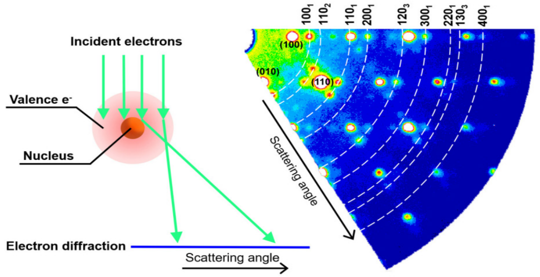

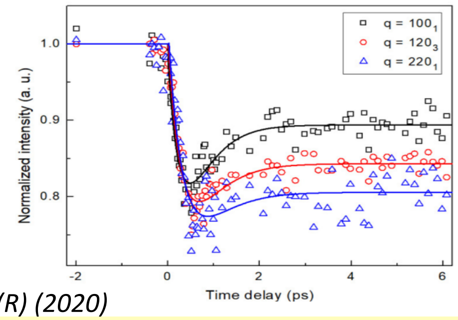

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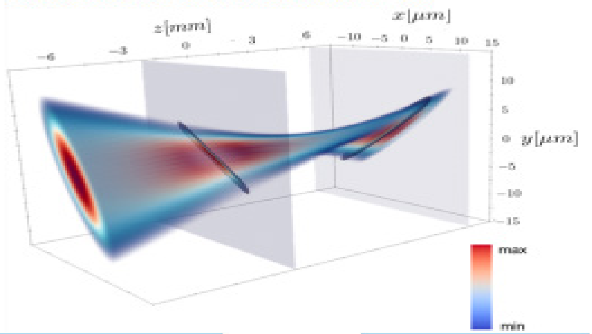

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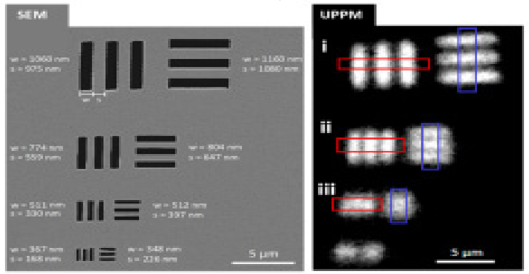

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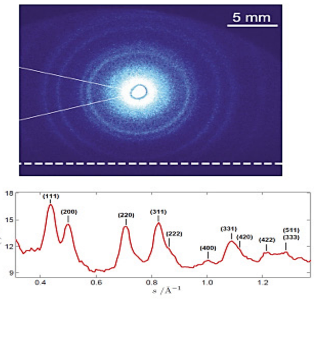

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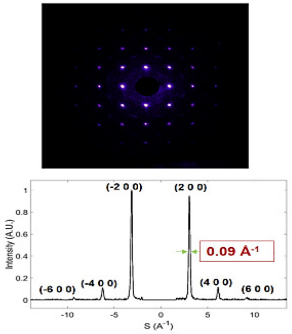

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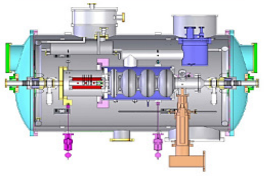

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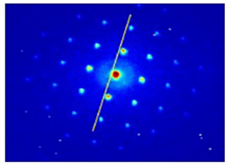

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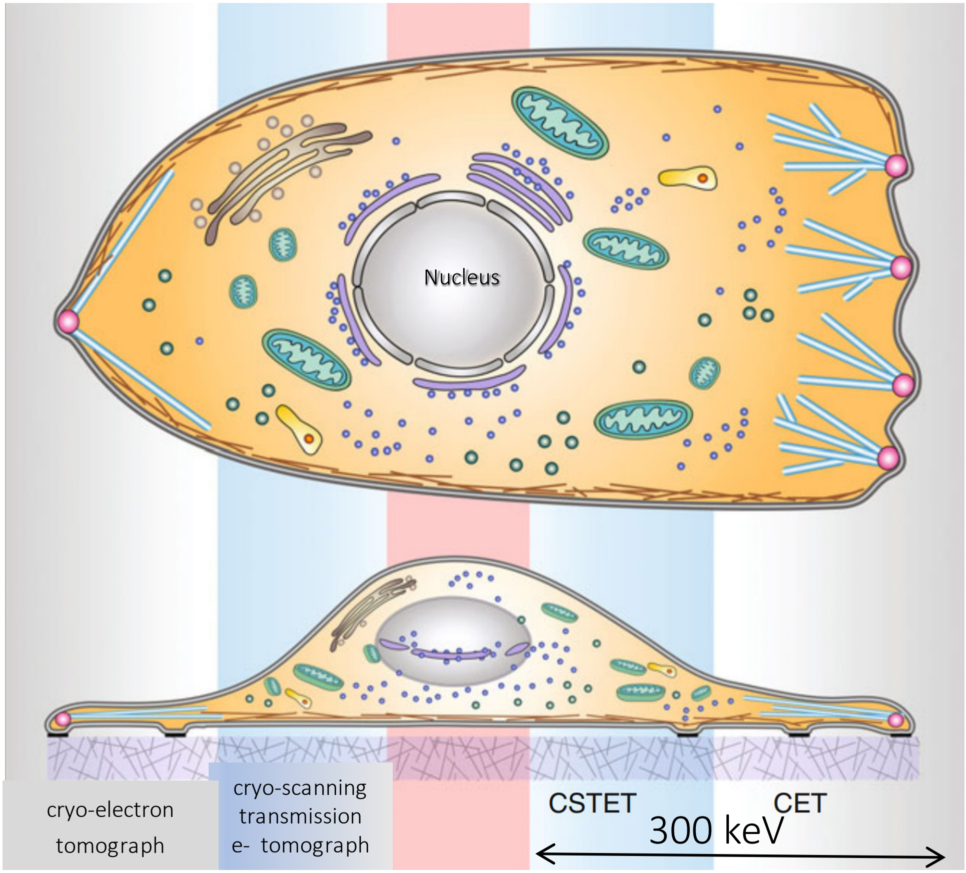
Promise of MeV Microscopy

Two different types of applications

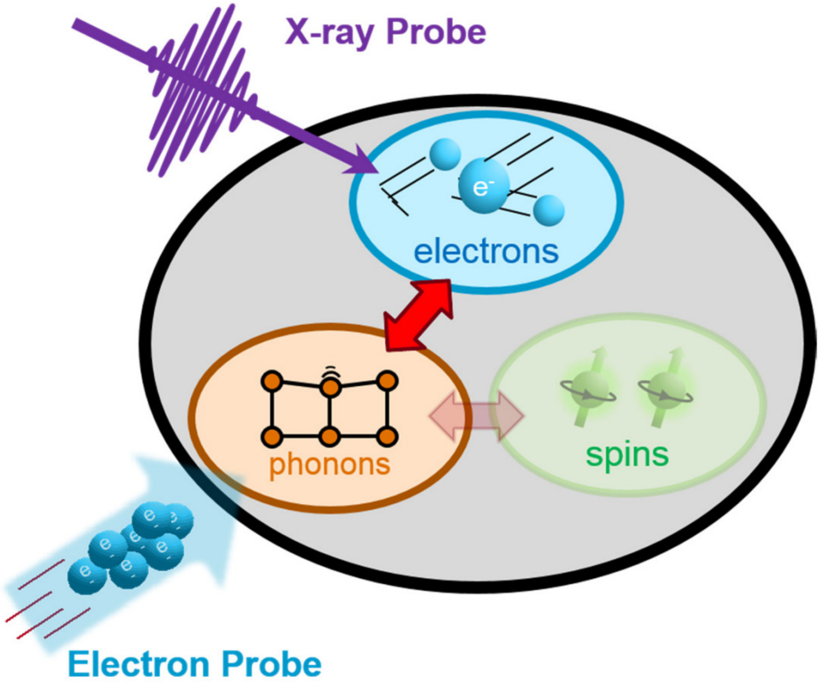
- **Life science application**: 3D-image thick bio-samples
- No need of cryo-FIB (focused ion beam) to slice thick cells
- Limited by 5-10 lamellae/day, "blindly" select target
- Speed up discovery

- Probe into ultrasmall and ultrafast world
- Real-time visualize structural dynamics in real space
- Allow direct probing of charge-spin-lattice interactions



Top and side views of a eukaryotic cell.

S.G. Wolf, et al., Cellular Imaging, Springer



R.K. Li, et al., IBIC 2019

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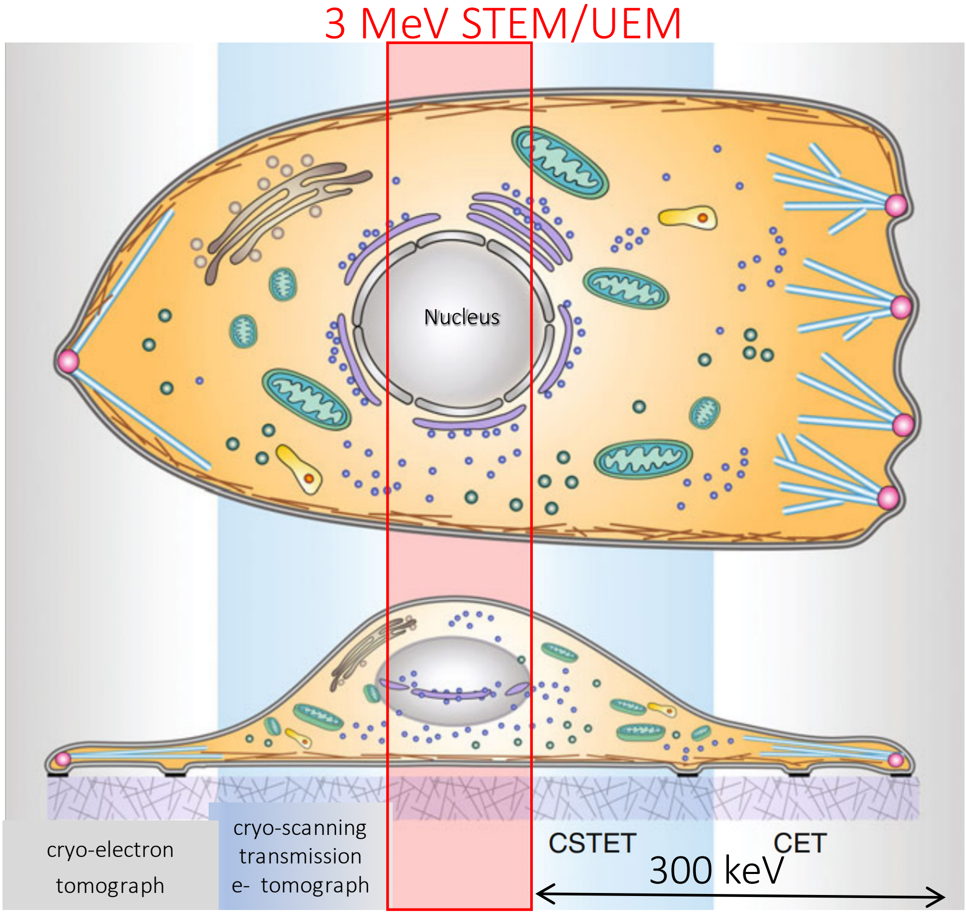

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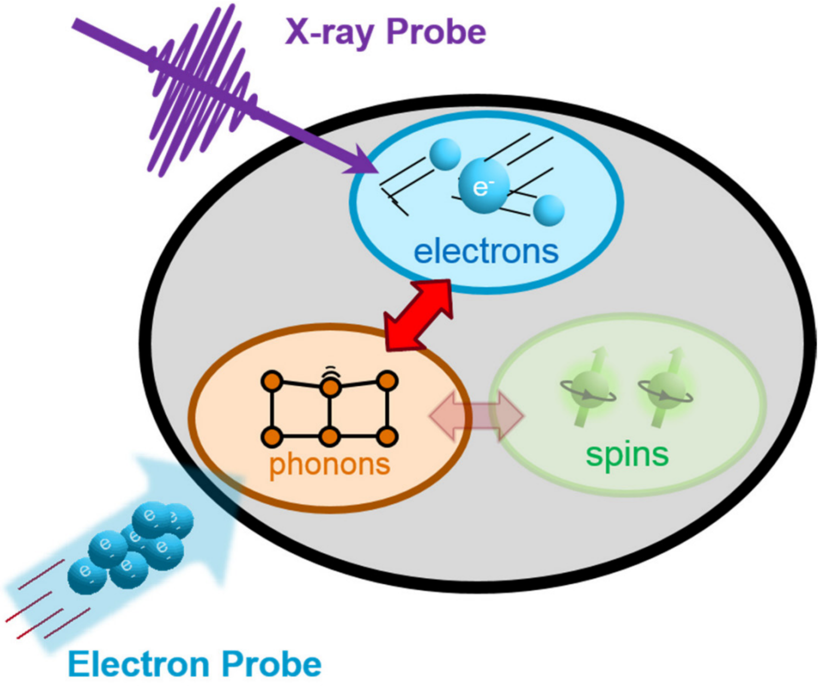
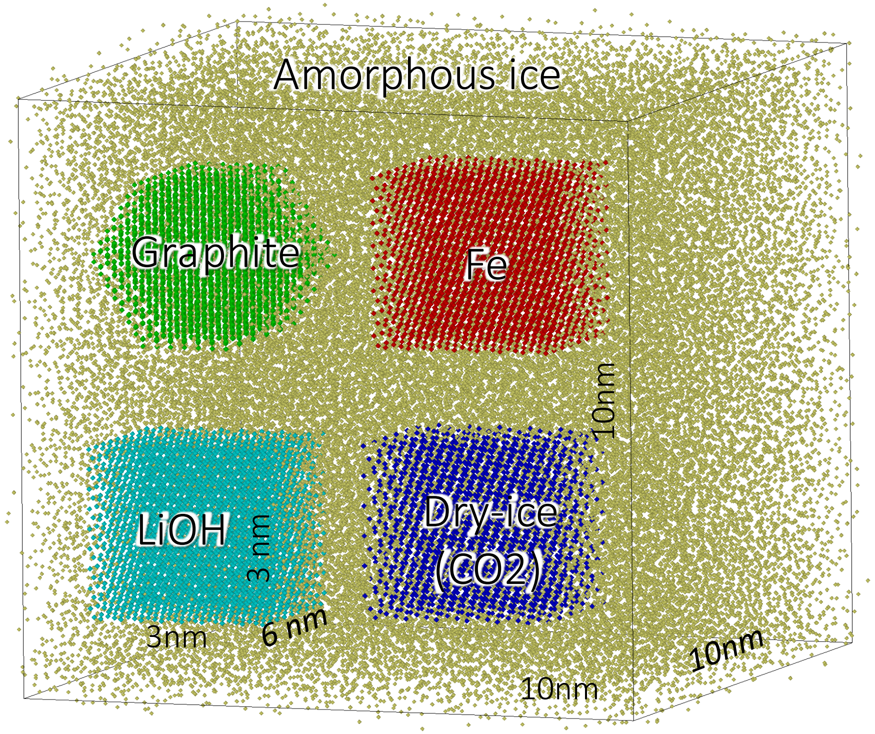


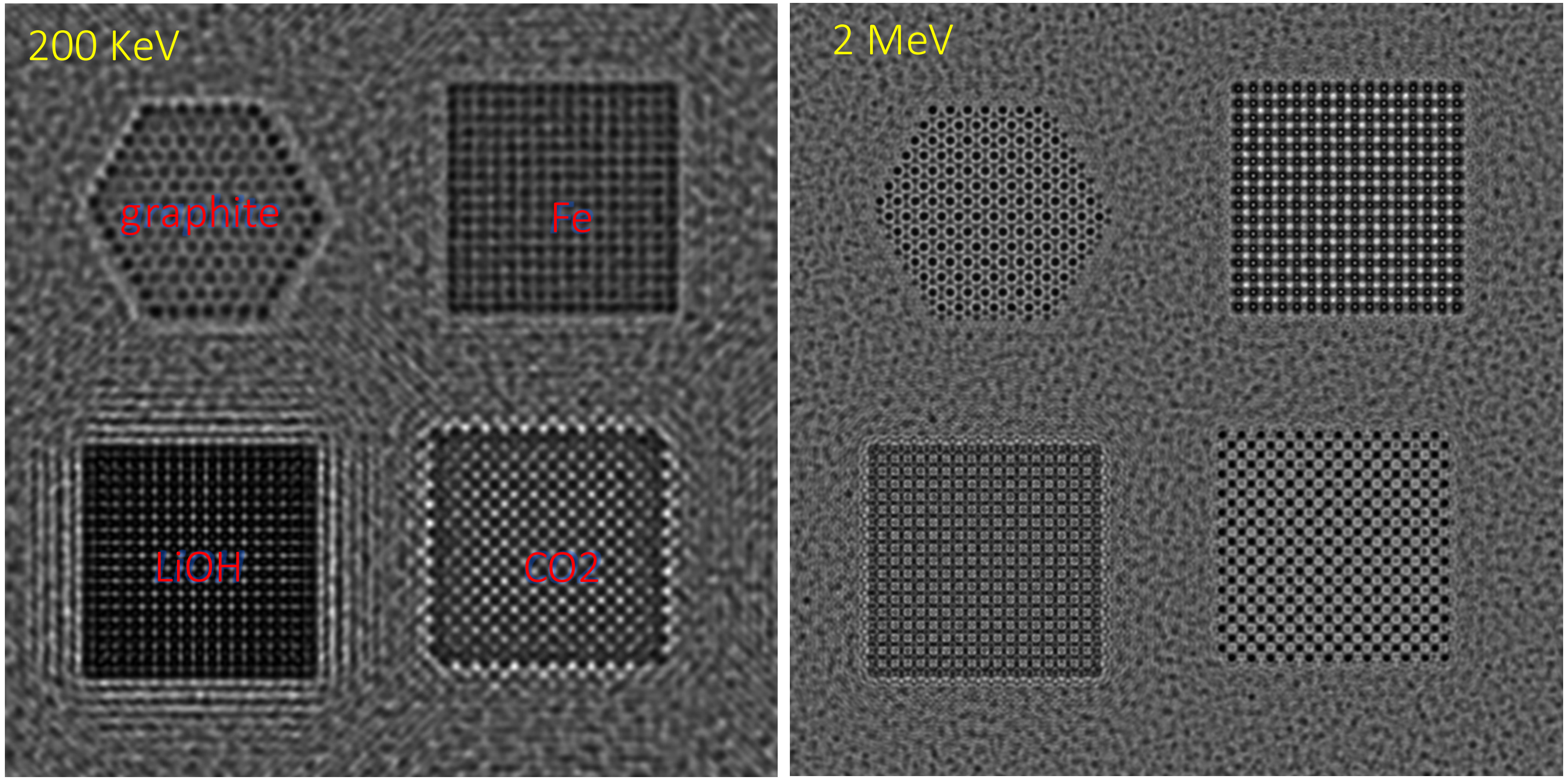
R.K. Li, et al., IBIC 2019

Benefit from increasing electron beam energy to mega-electron volt

Nanoparticle in amorphous ice



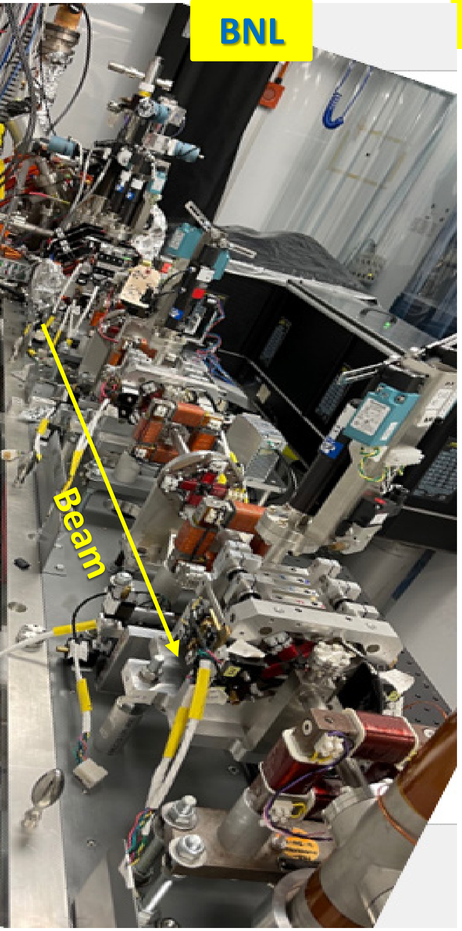
UEM Imaging at different beam energies



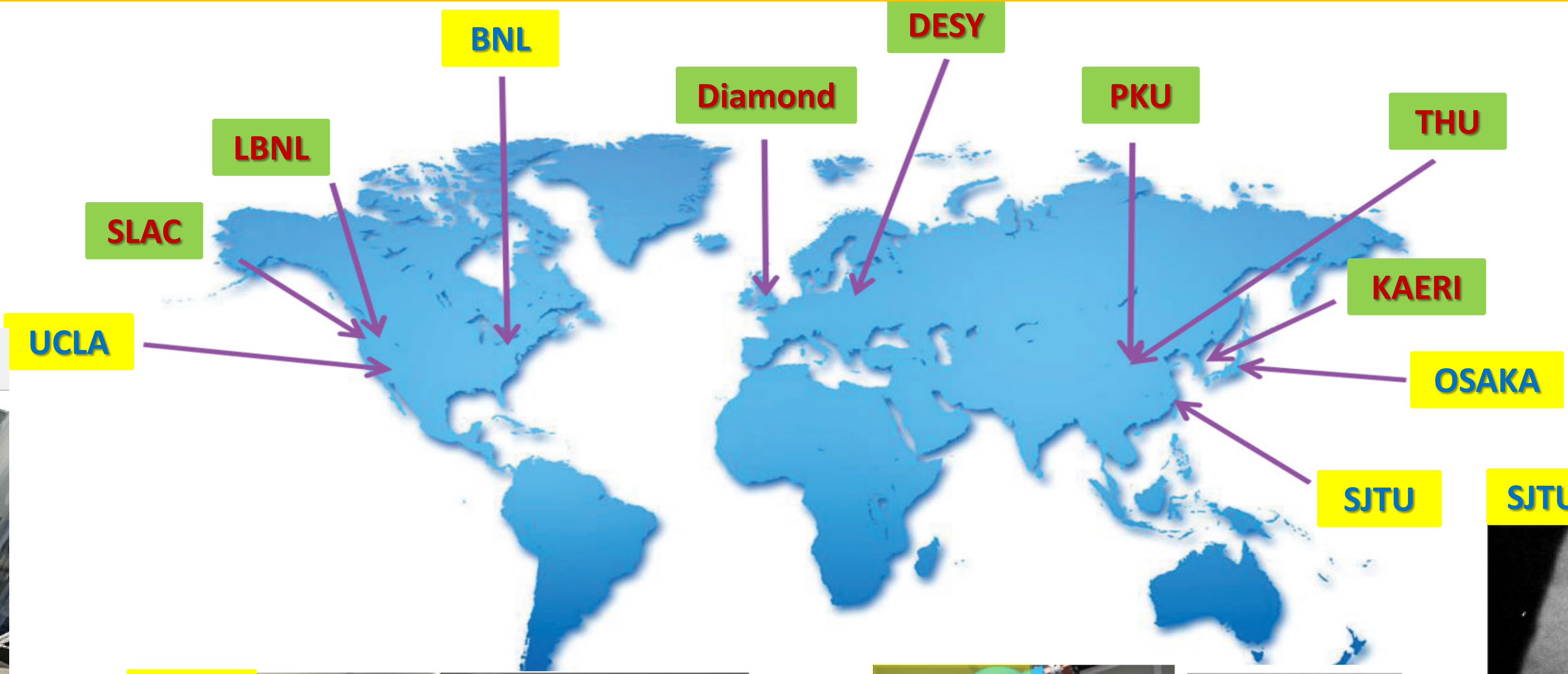
- Graphite, Fe, LiOH and Dry ice (CO_2) with size of ~ 3 nm in-plane and 6 nm thick are embedded in amorphous ice.
- The product of C_s and l is kept constant, we choose Cs=0.5 mm for 200 KeV and Cs=2.5 mm for 2 MeV. D_0=5 nm. Here $\Delta_0 = C_c \cdot {}^{\Delta E}/_{E}$.

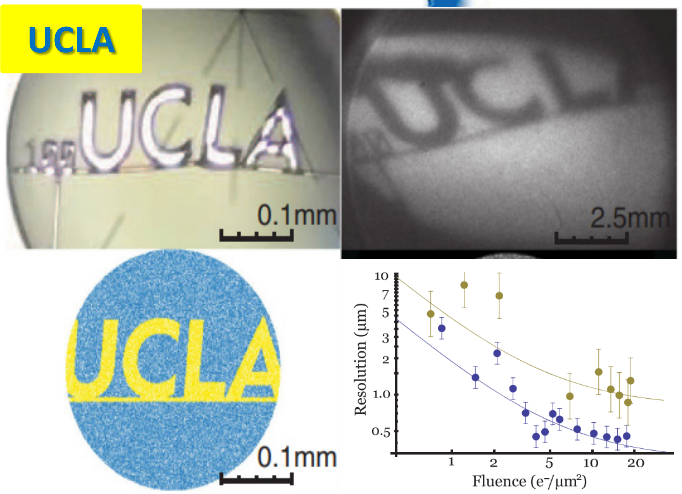
BNL capability: Electrons and sample interaction simulation by Lijun Wu's multislice wave optics code

State of art MeV UEM/UED: world-wide efforts

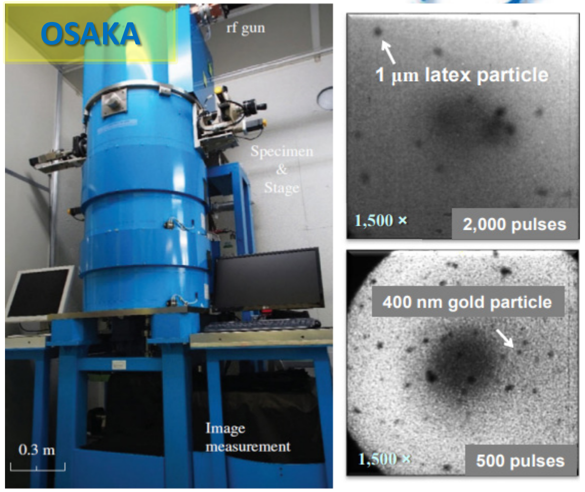


Wait for beam testing



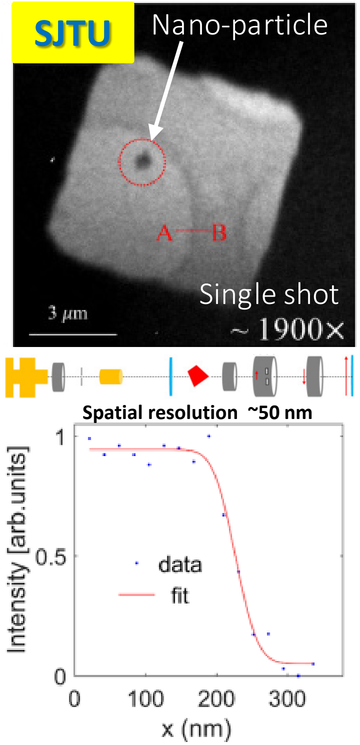


PRL 118, 154802 (2017)



Temporal resolution 180 fs

Microscopy 67, 291 (2018)



$E = 3MeV, \frac{\Delta E}{E} = 5 \cdot 10^{-4}$

Appl. Phys. Lett. 112, 113102 (2018)

Critical parameters and components for MeV UEM

- Beam requirement for single shot UEM

Number of electrons	10^7
Imaging area	$0.01 - 100\ \mu m^2$
Beam divergence	$\leq 1\ mrad$
t - resolution	$10\ fs\ to\ 100\ ps$
Energy spread	$\sim 1 \times 10^{-5}$

- < 10 nm emittance with > 1 pC
- Flattened longitudinal phase space for 1×10^{-5} energy spread

- Four main building blocks of MeV UEM
  - High brightness photoinjector

    Photocathode, high gradient rf gun, drive laser
  - Bright beam transportation

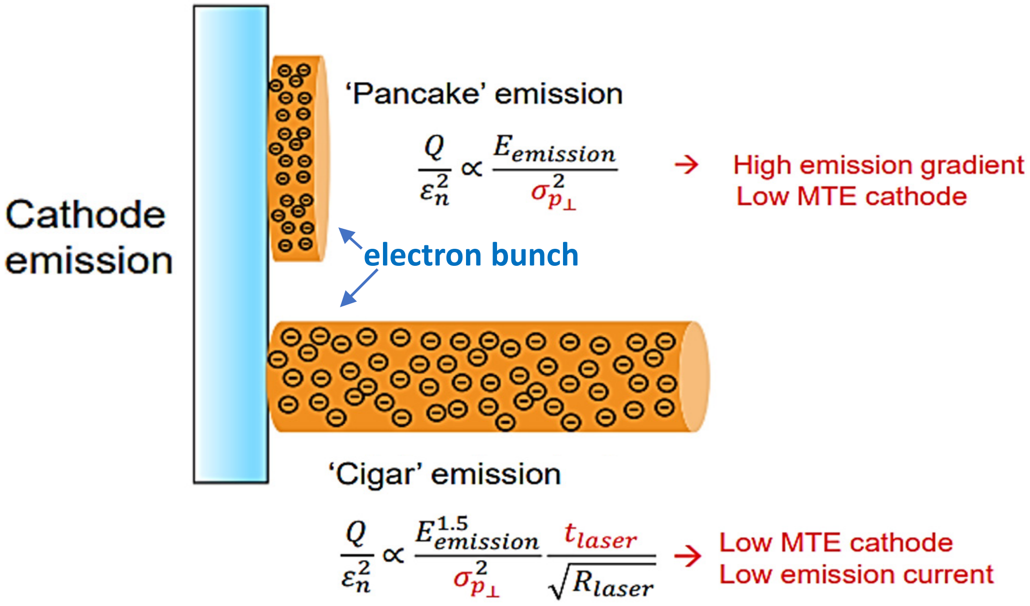
    Condenser, aperture, imaging lens system
  - Electrons and sample interaction

    Time-resolved applications, life science (e.g., bio-samples with up to 10 μm thickness)
  - Detector

    Scintillating, single electron counting (not for > 1MeV)

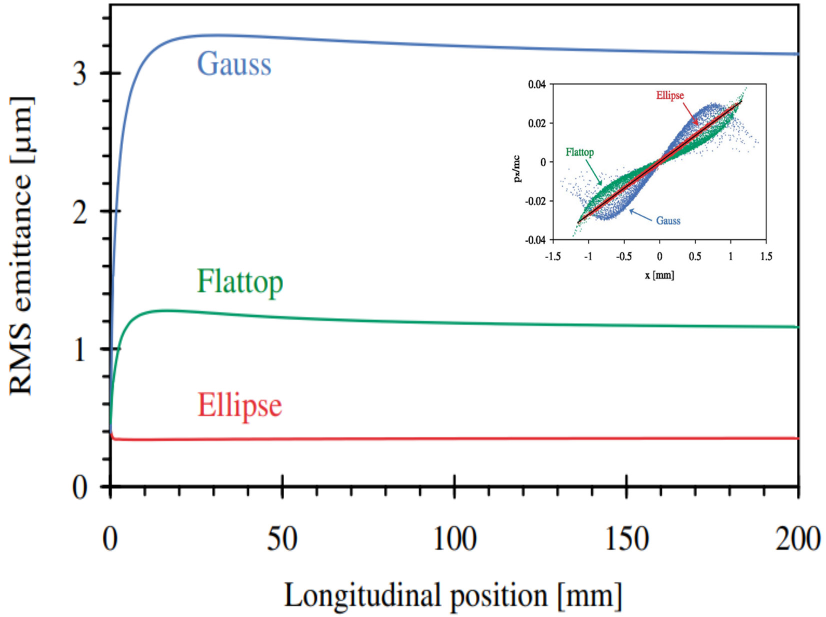
High brightness photoinjector

- Two types of photoinjectors
  - DC gun
  - RF gun (higher gradient)
    - Normal conducting
    - Superconducting



*I. V. Bazarovet al., PRL **102**, 104801 (2009)*
*D. Filippetto et al., PRAB **17**, 024201 (2014)*
Qian and Vogel, IPAC21

- Laser shaping affects tran. & long. dynamics
  - Uniformly filled ellipsoidal
  - Linear self-field



Emittance is preserved during beam transportation with an initial ellipsoidal profile

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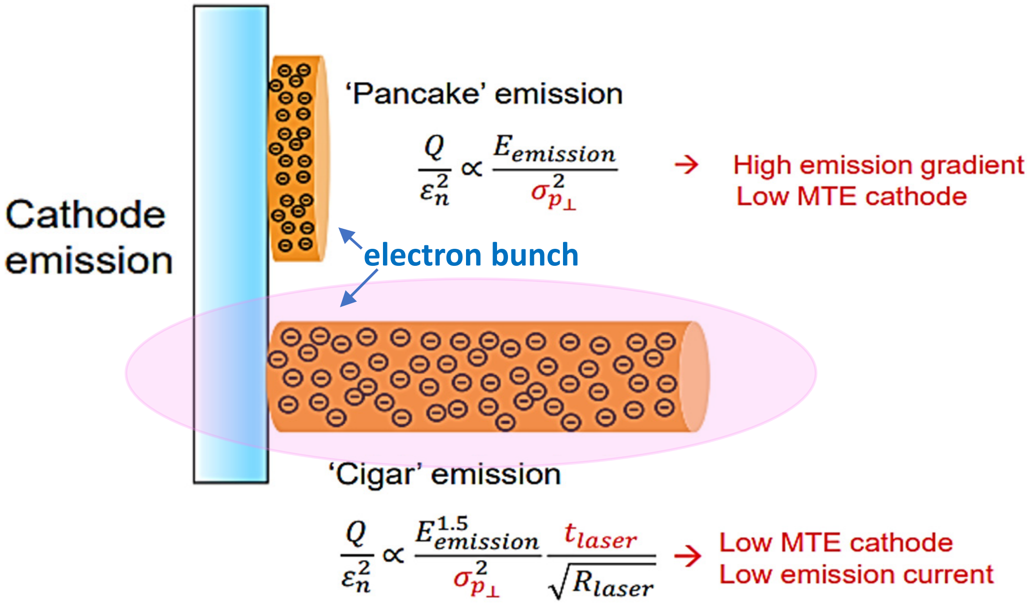

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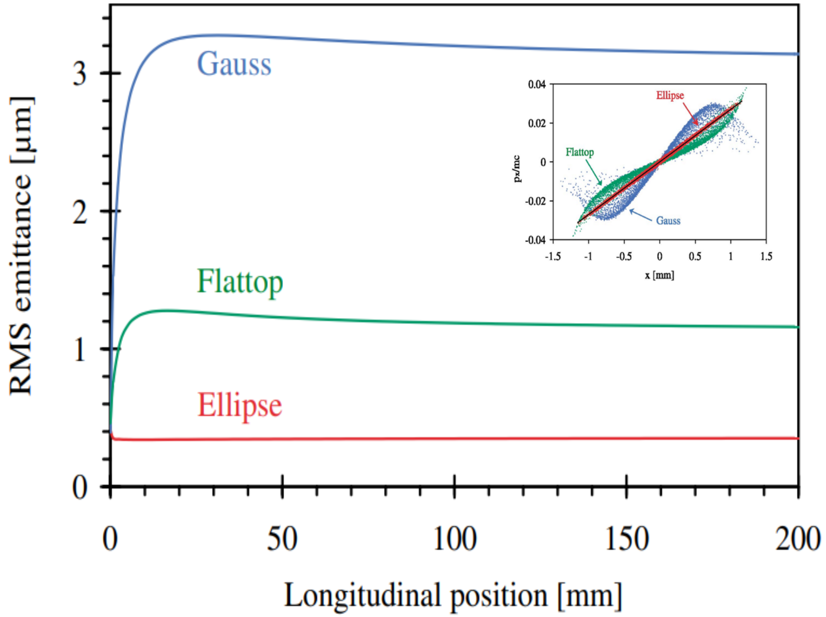

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Challenges of MeV UEM

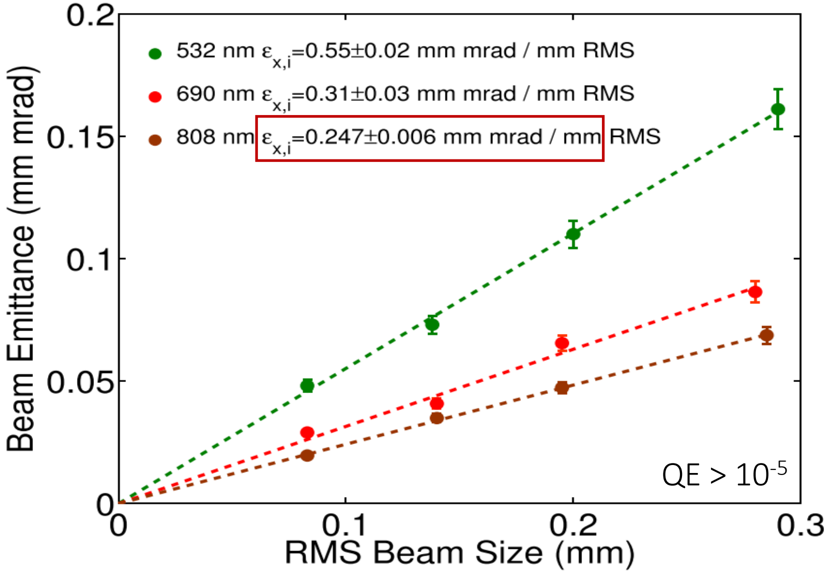
1. Reduce beam emittance to < 10 nm

$\Delta r = C_s \alpha^3$; $\Delta r = C_c \alpha \Delta E/E$; $\varepsilon \propto \sigma_x \sigma_{x'}$

Scherzer' theorem: $C_s < 0$, $C_c > 0$
(Scherzer, Z. Phys. **101**, 593 (1936))

- Reduce thermal emittance (minimize MTE & spot size)
- Reduce space charge induced emittance growth

$$\varepsilon_{nx} = \frac{\sigma_x \sigma_{p_x}}{mc} = \sigma_x \sqrt{\frac{\text{MTE}}{mc^2}}$$



0.25 µm/mm rms emittance is close to the limit set by e- finite *T=300 k*
0.12 µm/mm rms emittance when *T=90 k*

Map to accelerator terminology

- Imaging requires: $R_{12} = 0$
- Chromatic aberration: $C_c = T_{126}$
- Spherical aberration: $C_s = U_{1222}$

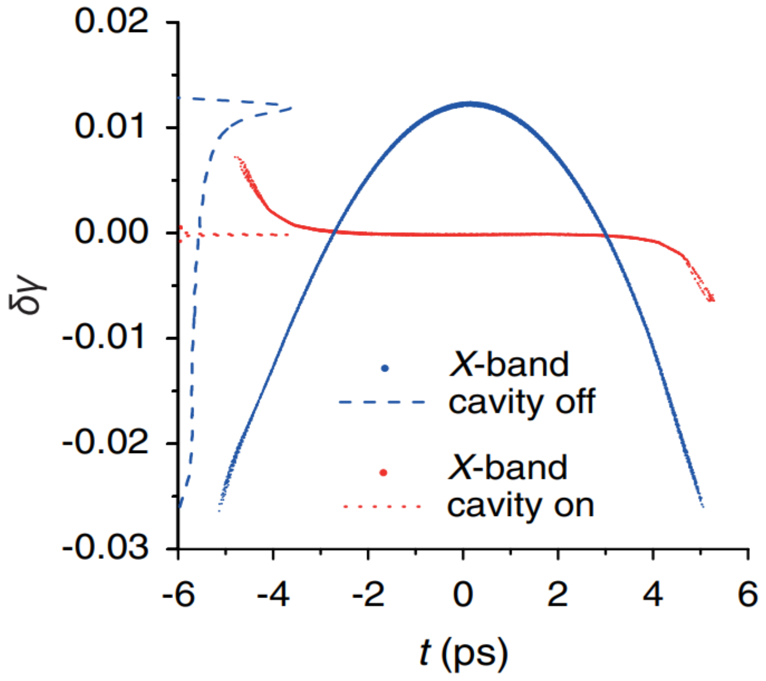
Assume rotational symmetry
Objective lens defines the aberrations

*L. Cultrera et al., Phys. Rev. ST – Acc. Beams **18** (2015) 113401*
*H. Lee et al., Rev. Sci. Instrum. **86** (2015) 073309*
Xiang et al., Nucl. Instrum. Methods A, 759, 74 (2014)

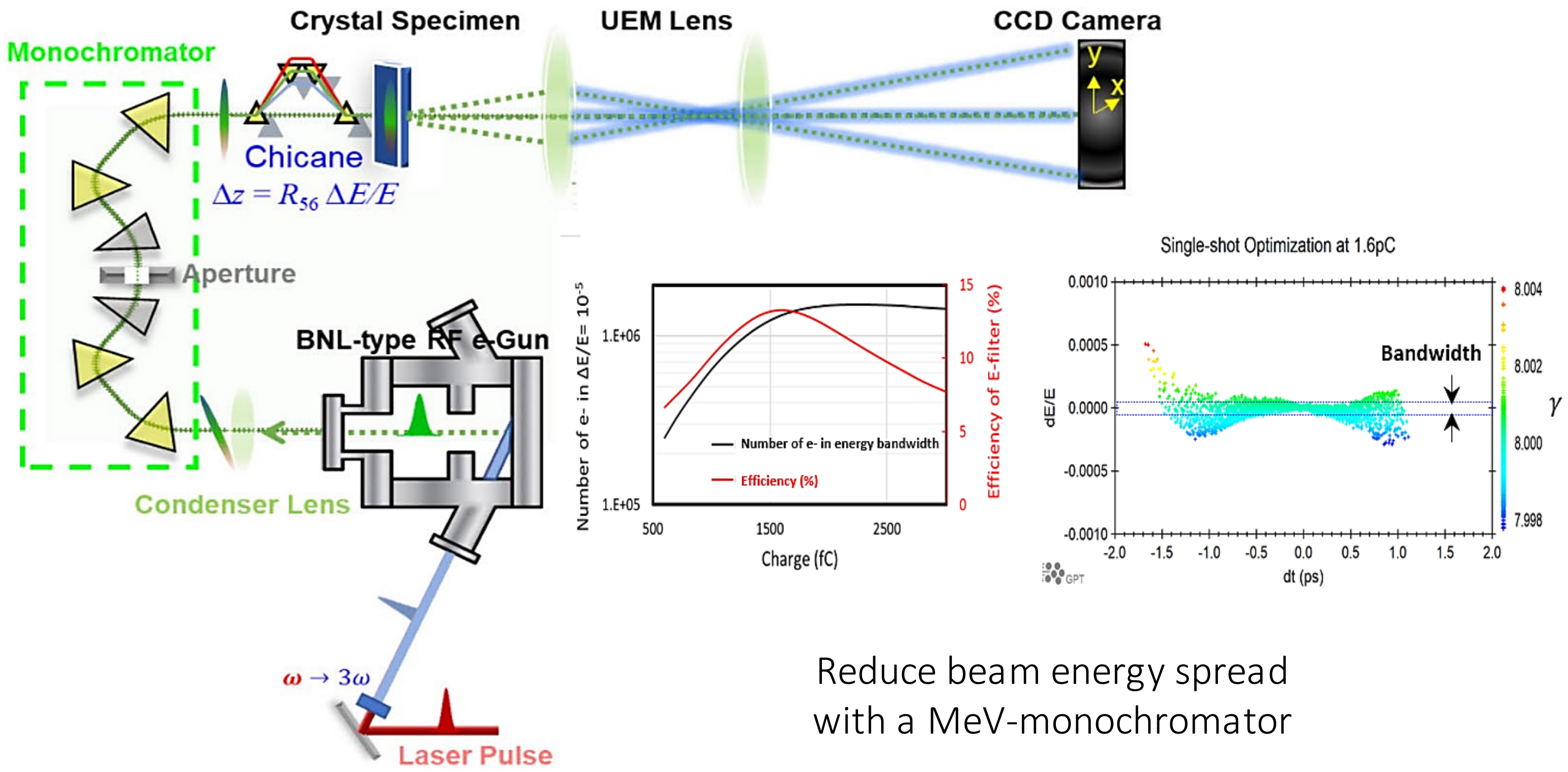
Challenges of MeV UEM

2. Reduce beam energy spread to < 10^{-4}

- Remove quadratic energy chirp with a harmonic cavity (single shot)
- Remove off-energy electrons with a monochromator (single- and multi- shot)
- Excellent RF and Low-level RF system



Reduce beam energy spread with a harmonic cavity



Reduce beam energy spread with a MeV-monochromator

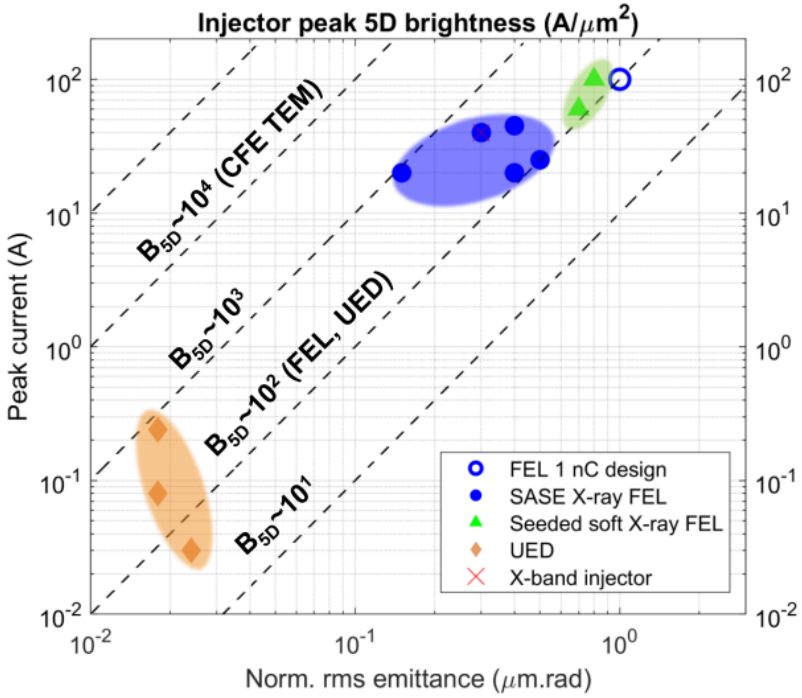
Li and Musumeci, Phys. Rev. Applied, 2, 024003 (2014)

Yang, et al., Sci. Rep. 10, 16171 (2020)

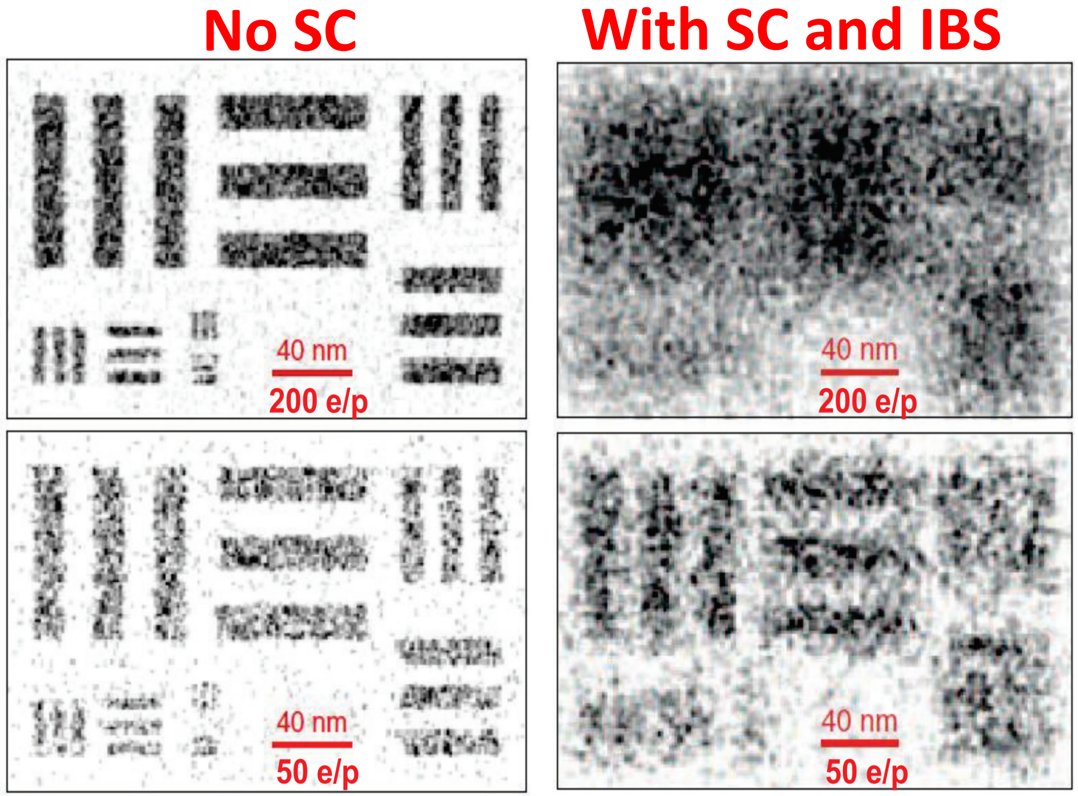
3. High beam density and space charge effect

- Rose theorem: >100 electrons/pixel to make a useful image
- Single shot UEM: B_{6D} $\underline{10^2\text{-}10^3}$ $A/(keV*\mu m^2*rad^2)$
- Ultimate beam density (e-/nm^3) limited by space charge effect
- Spatial resolution also limited by stochastic space charge effect

	Charge	Duration	ε_n (nm)	dp/p
Diffraction	0.01-1 pC	0.1-1 ps	<100	$<10^{-2}$
Single shot Imaging	>1 pC	10~100 ps	<10	$\underline{\leq 10^{-4}}$



Qian and Vogel, IPAC21

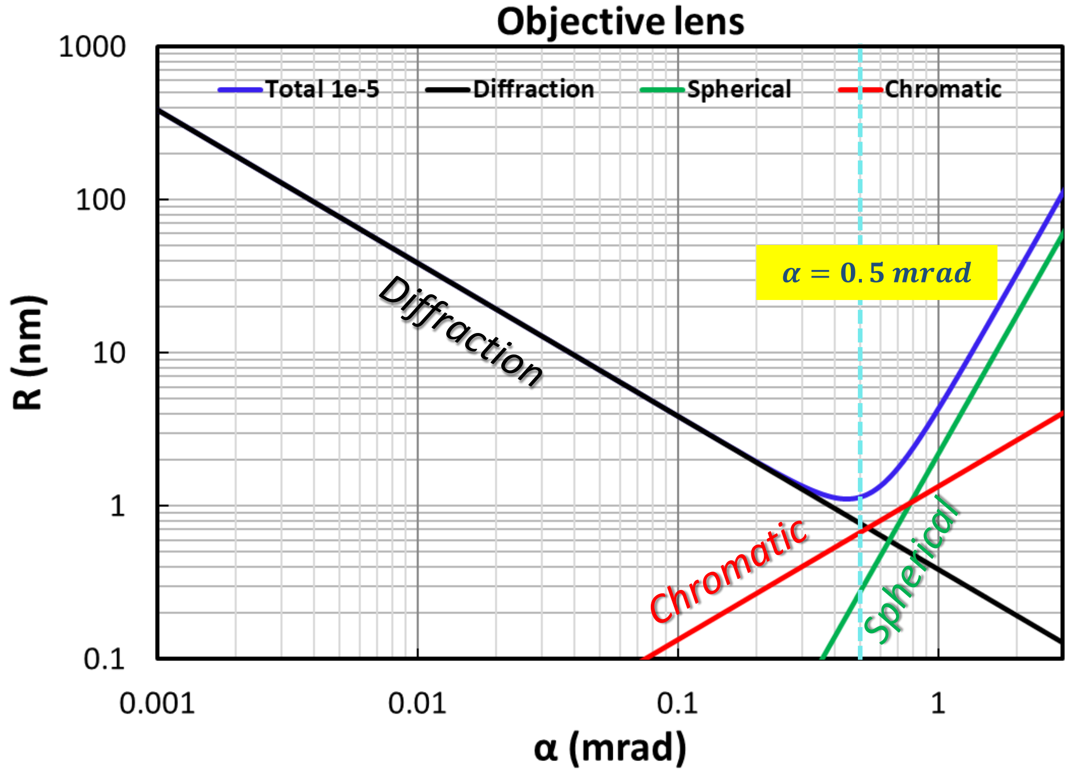


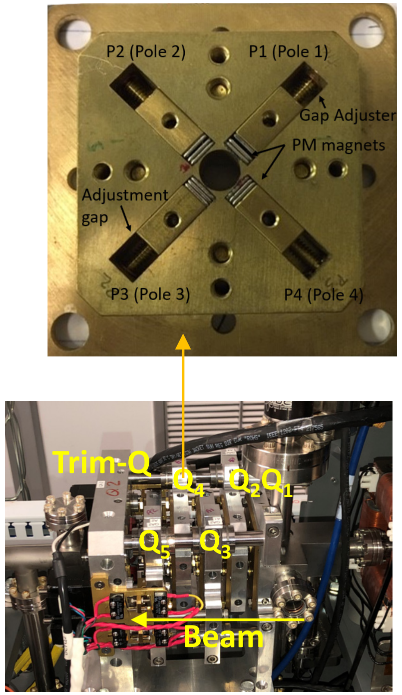
Li and Musumeci, Phys. Rev. Applied, 2, 024003 (2014)

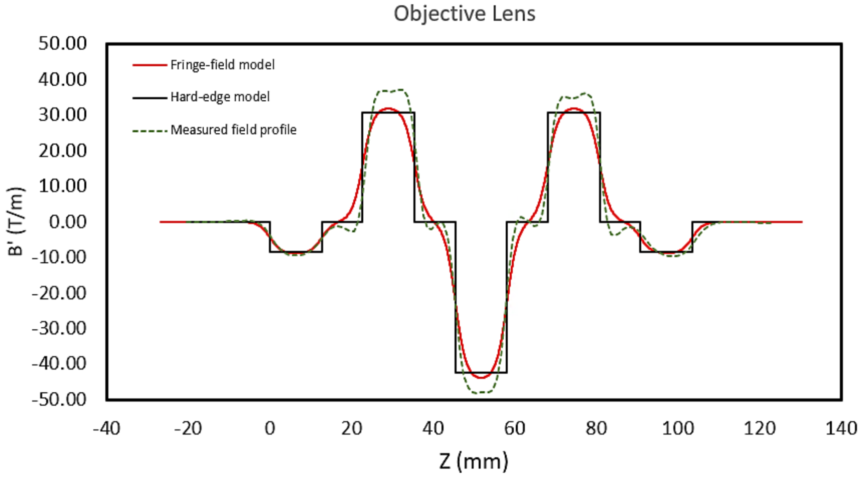
Challenges of MeV UEM

4. Build low-aberration strong lenses for MeV UEM at BNL

- Novel approach: compact low-cost imaging lens based on PMQ quintuplets.
- Minimum aberrations: strong lens with the highest achievable gradient and shortest focal length.
- Tunability of the magnification: +/- 5% by adjusting drift spaces between magnets and trim quadrupole.







Focal length *f* and main aberrations C_s and C_c

	f (cm)	$C_{s,x}$ (cm)	$C_{s,y}$ (cm)	$C_{s,xy}$ (cm)	$C_{c,x}$ (cm)	$C_{c,y}$ (cm)
Round lens	1.36	3.36	3.36	3.36	2.22	2.22
Quintuplet_BNL (measurement)	5.70	121	76.5	197	10.6	7.4
Quintuplet_BNL (analytical model)	6.20	215	106	336	13.4	9.7

Wan, W., Chen, F. & Zhu, Y., Ultramicroscopy. 194 143-153 (2018).

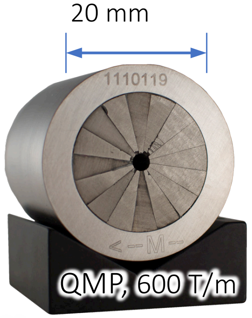
Building a compact MeV UEM with small business

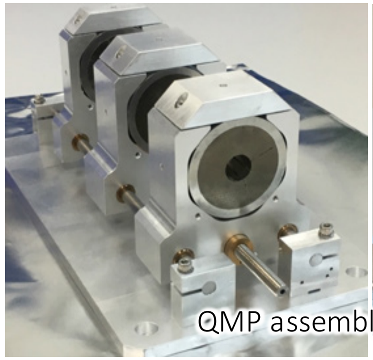
We are collaborating with small business to develop and construct a state-of-the-art MeV UEM.
The collaborations are supported DOE SBIRs and the final products will be delivered to BNL for testing and optimization.

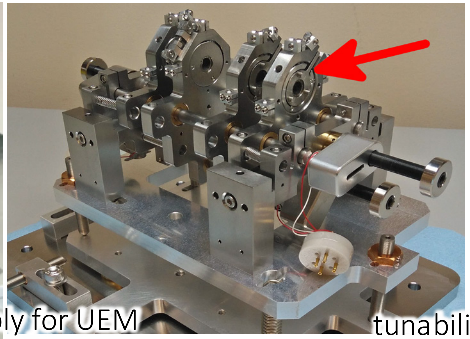
<u>Quadrupole based focusing and imaging lens systems with Radiabeam</u>

Quadrupoles are small, but have much stronger focusing capability for high energy electrons than round lens as their focusing power is inversely proportional to the momentum rather than momentum squared

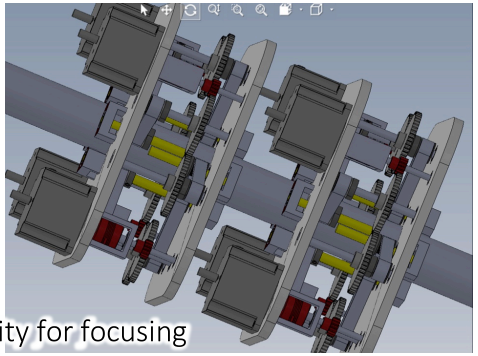
radiabeam TECHNOLOGIES





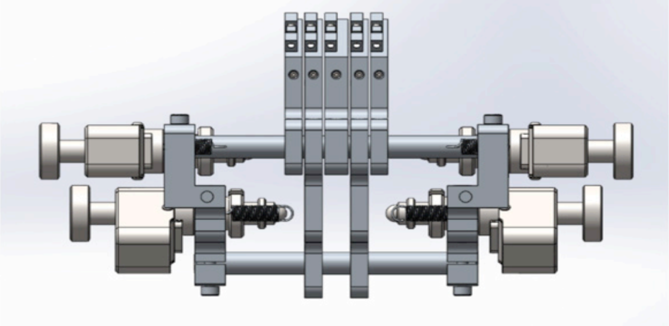


QMP assembly for UEM



tunability for focusing

Quintuplet design for MeV imaging



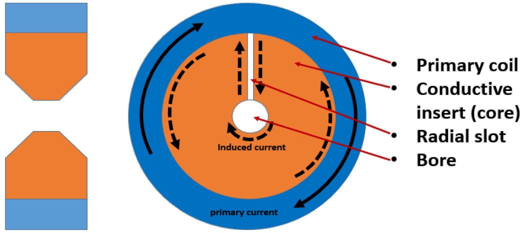
DOE SBIR Phase II Grant #DE-SC0018562

- Focusing power of quadrupoles is asymmetric, thus difficult for imaging
- The required high field gradients, short length, small bore, and high stability make the tunable lens design extremely challenging
- Using modern genetic algorithms Radiabeam has optimized PMQ based objective lens with nm resolution
- Continued efforts on combining adjustable PMQ positions and field gradient

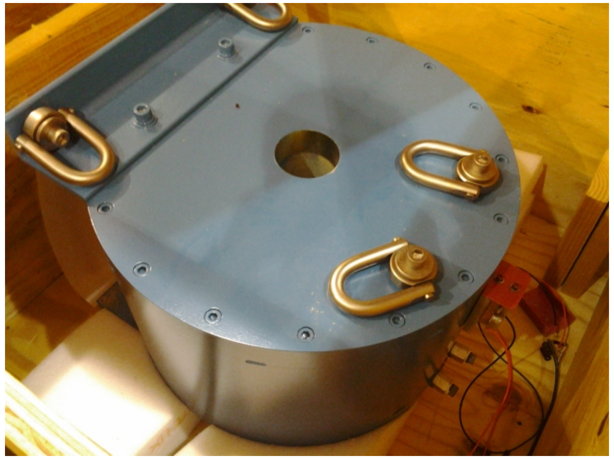
Ultracompact Objective Lens for MeV UEM/UED*

Motivation:

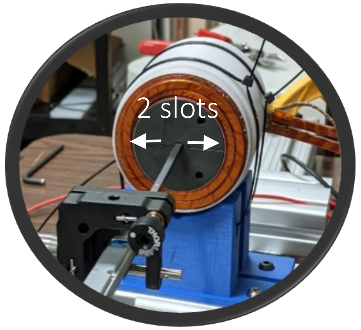
- Reduce the weight and size of a bulky Tesla DC objective lens by 100 times using a flux concentrator based pulsed solenoid.



1Tesla DC solenoid with weight of 1Ton and 1m of overall diameter



2Tesla Flux Concentrator with weight of 1.5kg and 8cm of overall diameter

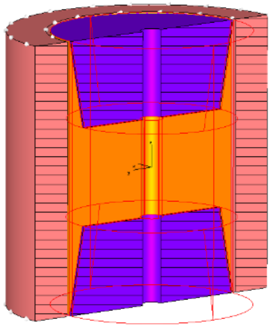


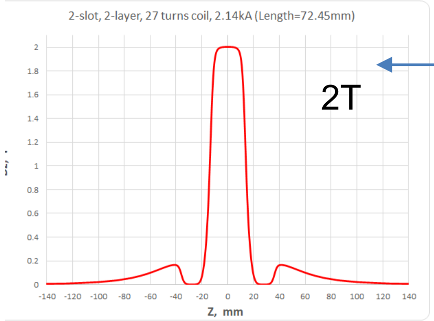
- Euclid *2T* pulsed solenoid can achieve 2.3 Å resolution with $\frac{\Delta E}{E} = 10^{-5}$
- Limited repetition rate to a few Hz

Euclid's Design:

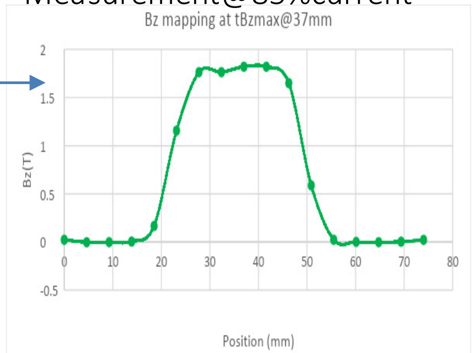
- Using 2 slots to eliminate the dipole fields.
- Using ferrite to reduce the fringe fields
- Using high stability power supply and pulser to reach **10ppm** field stability.

Simulation

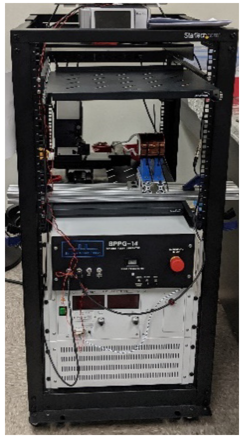




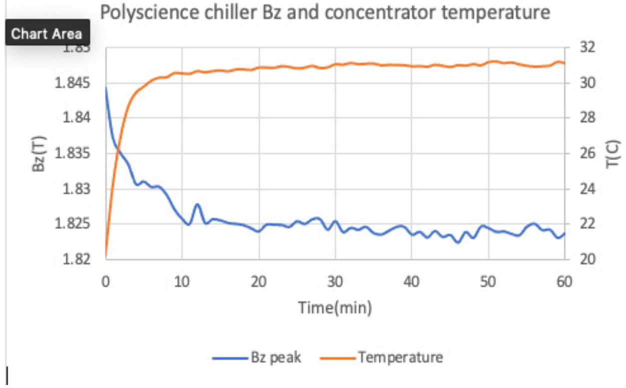
Measurement@85%current



Orange – copper concentrator
Purple – ferrite insert
Red – water cooled coil



Stability test over 1-hour

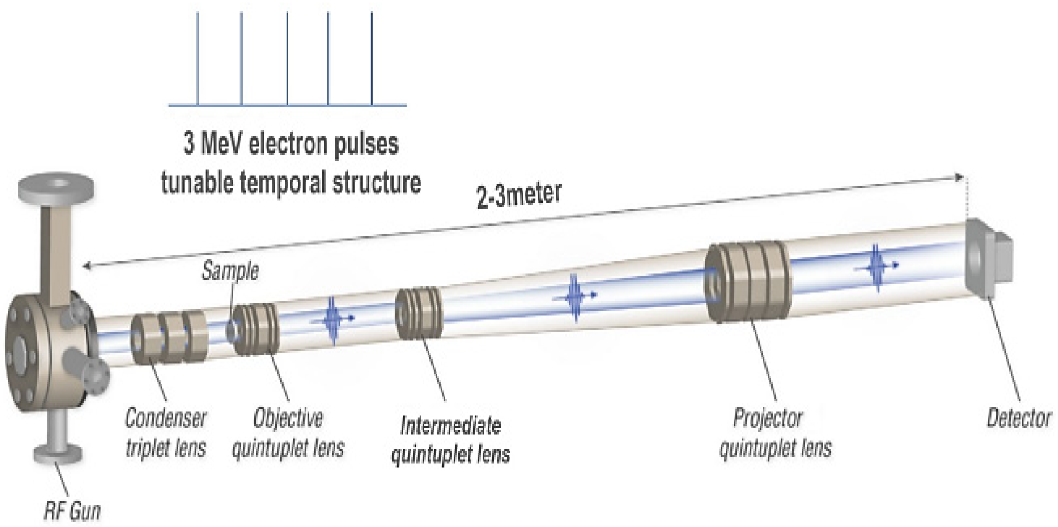


Challenges of MeV UEM

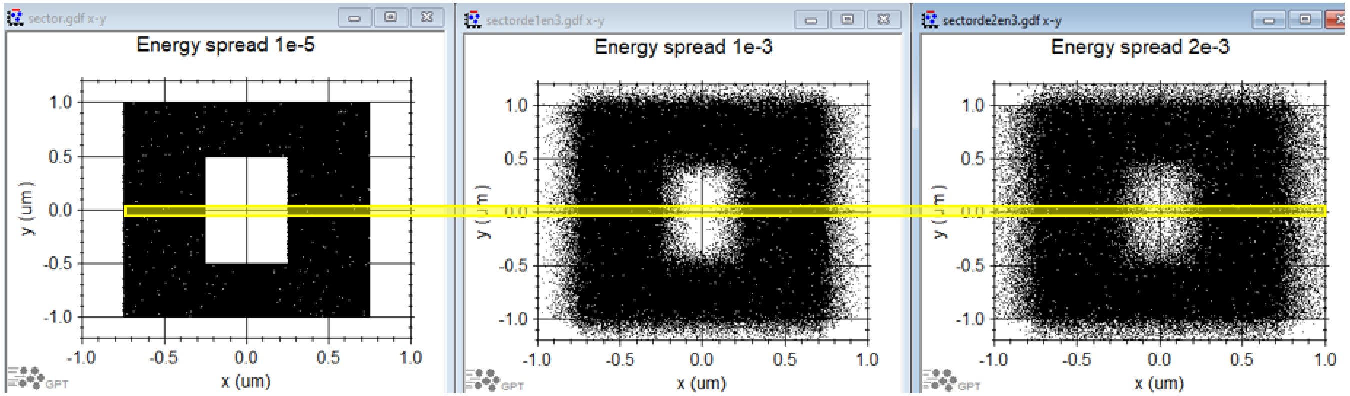
5. Design ultrafast microscope for single and multi-shot imaging with MeV electrons

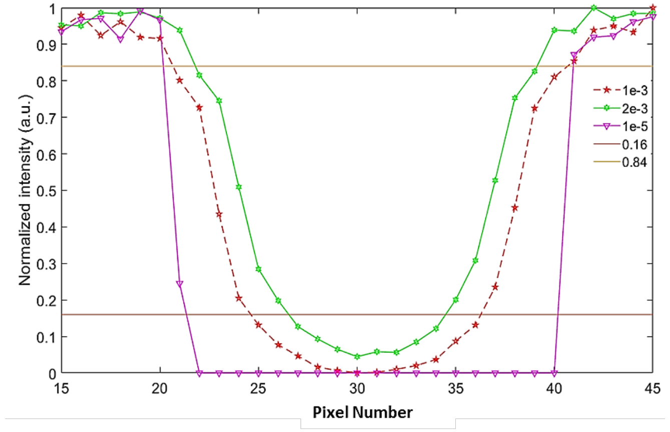
- Modular design: future upgrade to higher magnification, towards atomic resolution.
- Real image at each stage: easy for commissioning.
- Simple design: make the objective and projector lenses identical.
- **The design resolution of** the lens system: **1.2 nm** with the low energy spread 10^{-5} and optimal aperture angle α 0.5 mrad.

Schematic of the UEM beamline



Start to end simulation





With 25 nm/pixel

- R = 130 nm ($\frac{\Delta E}{E} = 2 \cdot 10^{-3}$).
- R = 90 nm ($\frac{\Delta E}{E} = 1 \cdot 10^{-3}$).
- R = 25 nm ($\frac{\Delta E}{E} = 1 \cdot 10^{-5}$), limited by pixel size.

BNL efforts: construction of tunable quadrupole-based UEM

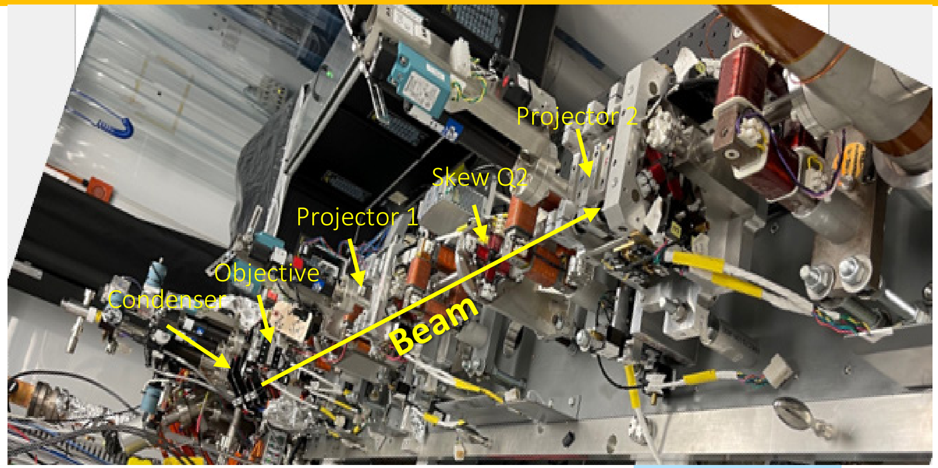
- A MeV UEM: simultaneous high spatial and high temporal resolution

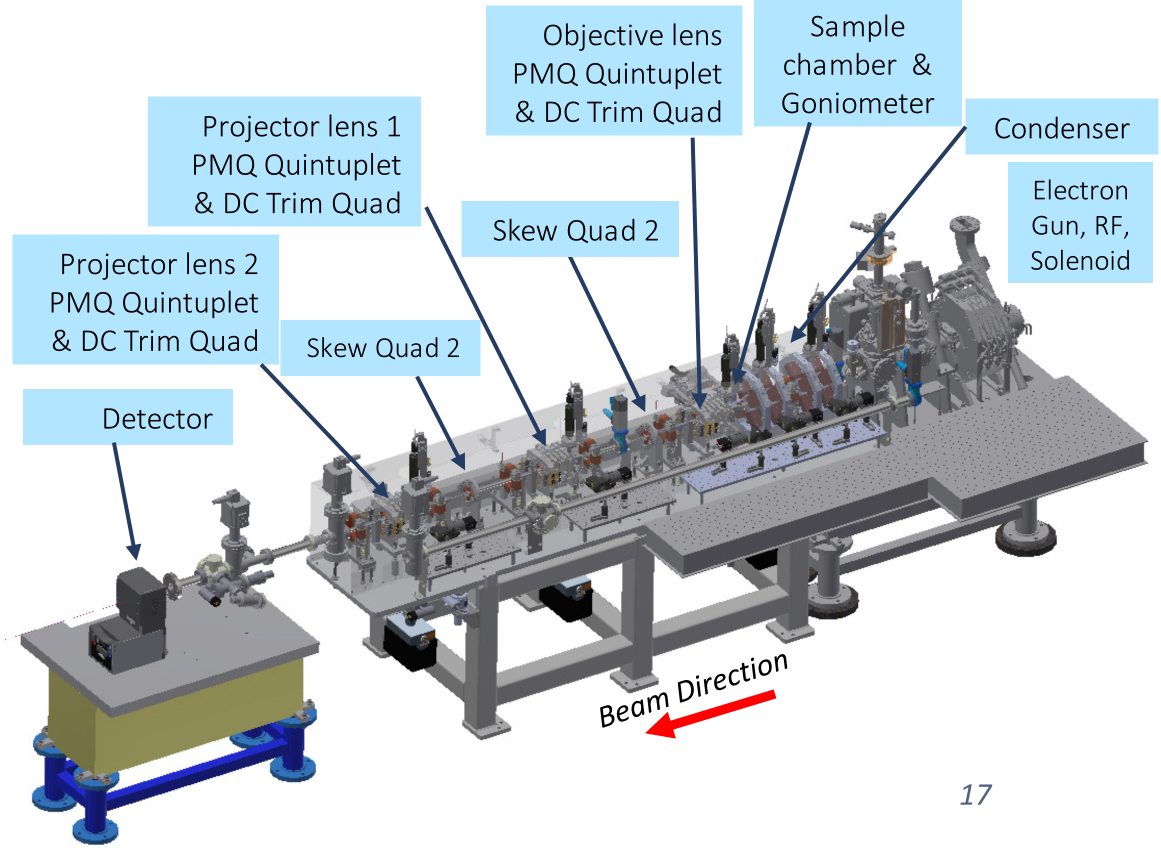
  for probing ultra- fast and small world

- Constructed the world first compact 3 MeV quadrupole-multiplets UEM system based on
  - Our conceptual design (*Wan, Chen & Zhu, 194 Ultramicroscopy, 143-153, 2018*)
  - Success of UED LDRD
- The UEM consists of tunable condenser-, objective- and projector-lenses and various steering magnets.
- A modular design allows the future upgrade from 2000X to 100,000X

  Using the high spatial resolution compact lens system constructed by Radiabeam and/or Euclid supported by DOE SBIRs.

- Design resolution of 1.2 nm lens can be achieved with $^{\Delta E}/_{E} = 10^{-5}$.
- Preliminarily designed a monochromator with energy acceptance of 10^{-5}.





Beam Stability Requirement for nanometer UEM

Energy and spatial-pointing stability requirement

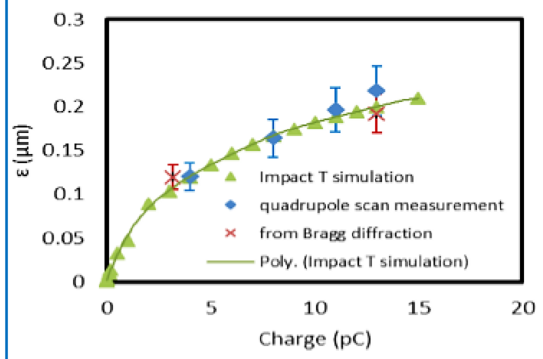
- **The RF system is the dominant source of shot-to-shot pulse jitter**
- **We use two methods for the energy jitter measurement:**
  - Bragg Diffraction Method
  - Direct RF jitter measurement
- **Nanometer resolution needs RF stability in the level of 10^{-5}**
- **Future improvement of RF system toward nm resolution**
  - HVPS (ScandiNova SS-K300@30ppm)
  - RF harmonic cavity
- **Implement monochromator with $1 \cdot 10^{-5}$ energy acceptance**
- **Stable source + narrow-band optics can achieve nm resolution in either single-shot or multi-shot mode.**
- **Shot-to-shot spatial pointing jitter is ~10 μrad**

Yang, et al., Scientific Report. 10, 16171 (2020)

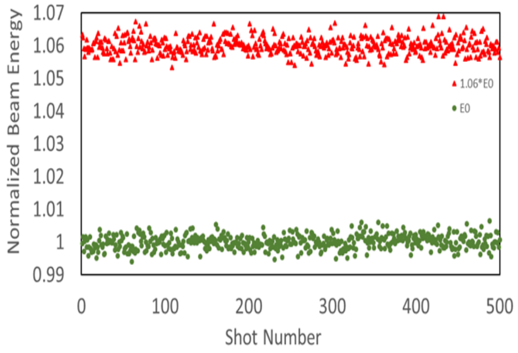
Development of real time, nondestructive diagnostic methods for MeV electron pulses

BDM allows us to optimize beam quality including the beam emittance, energy spread, shot-to-shot pulse fluctuation and jitter in real time.

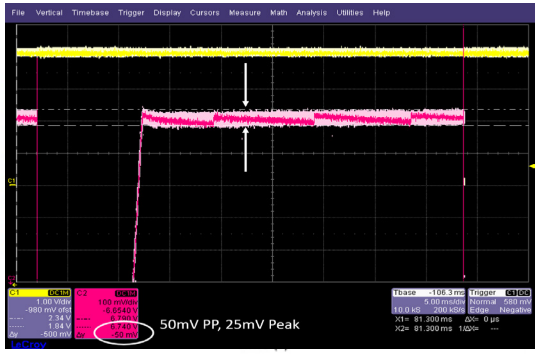
Beam emittance



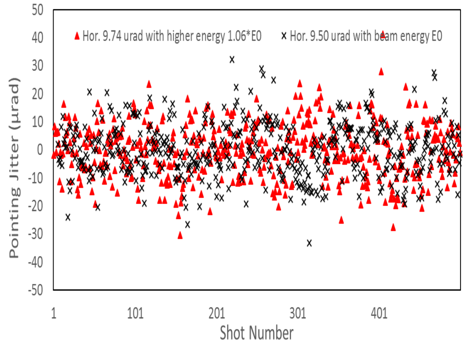
Beam energy jitter



RF jitter measurement



Beam pointing jitter



Yang, et al., Scientific Reports 9, 5115 (2019).
Yang, et al., Scientific Report. 9, 17223 (2019)

Mechanical Tolerances for nanometer UEM

- **Magnet tolerances:**
  - Manufacture and measurement tolerances
  - Alignment tolerances
- **The lens system with 5% tunability requires**
  - Manufacture tolerance of the gradient error $\Delta B'/_{B'} = \pm 2\%$
  - Measurement tolerance (Table)
- **Alignment tolerance is determined by sample plane defocus:** $\Delta D = \alpha \cdot {}^{R_{12}}/_{R_{11}} \leq \textit{Resolution}.$
- **UEM system dimension scales with magnet gradient (for round lens)**
  - $1/_f = \frac{eGL}{p}$. Short focal length needs high gradient (HG)
  - UEM LDRD builds experience with low gradient (LG) (up to *44 T/m*) f (= 6.2 cm)
  - Total beamline L = 4.2 m
- **Toward nm UEM with a compact beamline design.**
  - Two SBIRs for high gradient short focal length objective lenses.
  - Stability prefers compact system

magnification

angle-to-position spread

$$M^1 = \begin{pmatrix} R_{11} & R_{12} \\ R_{21} & R_{22} \end{pmatrix}$$

Alignment	Q1	Q2	Q3	Q4	Q5
Roll (mrad)	13	2.8	1.1	1.5	3.5
Pitch-x (mrad)	10.5	5	2.8	4	6.6
Yaw-y (mrad)	10.5	4.3	4.3	3.5	6.7
Δx (μm)	11	6	6	9	40
Δy (μm)	13	4	5	11	40
Δz (μm)	56	5	5.6	6.6	36
Manufactory ΔB'/B' (%)	±2	±2	±2	±2	±2
Measured ΔB'/B' (%)	±1.4	±0.5	±0.2	±0.5	±1.5

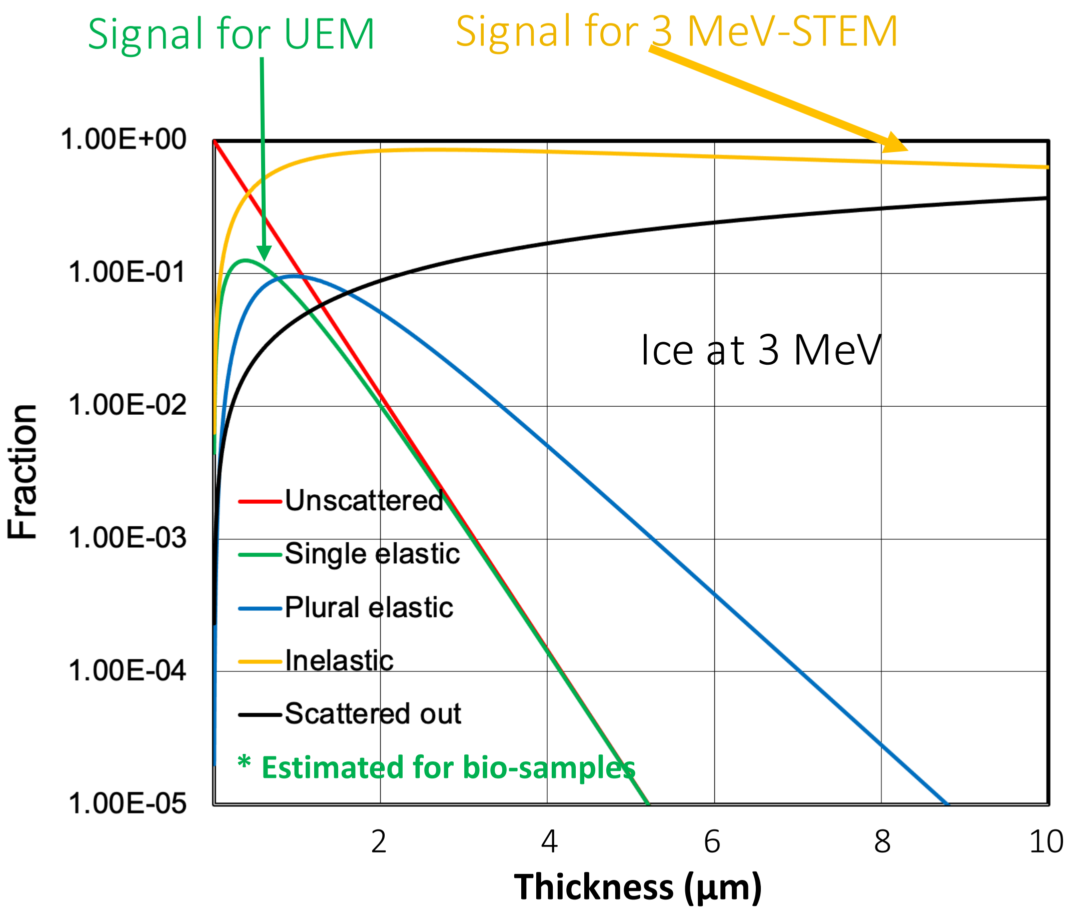
** Q1, Q2, Q3, Q4 and Q5 are quadrupole #1, 2, 3, 4 and 5 in a UEM lens assembly

All tolerances are estimated using COSY INFINITY.

MEV STEM to visualize intact cells

Understand cell biology and microbiology in cellular context
(imaging of intact plant and bacteria cells)

- Beam size and converging angle on sample determines resolution.
- To achieve 2-10 nm with up to 10 µm sample thickness:
  1. Electron dose: 200 e^-/nm^2 / µs
  2. Probe on sample: 1-5 nm
  3. Beam convergence angle: 1 mrad
  4. Beam flux: 200 - 5,000 e^-/ µs
  5. Scanning accuracy: 1 nm



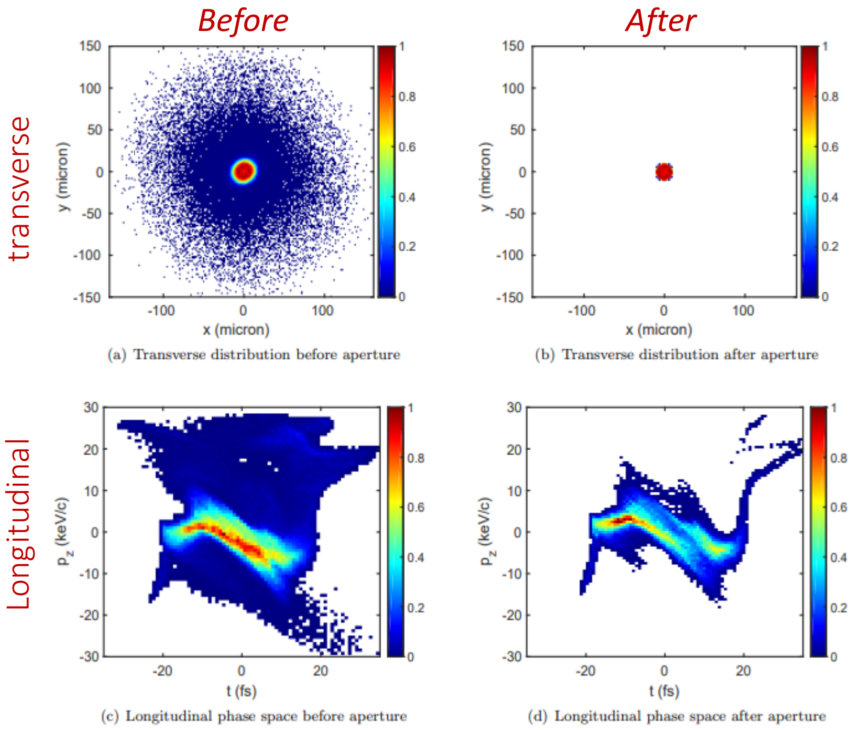
Challenges of MeV STEM

1. Reduce beam emittance to ~ 2 pm with current of 30 – 750 pA
2. Photocathodes & beam dynamics to improve brightness by >1000

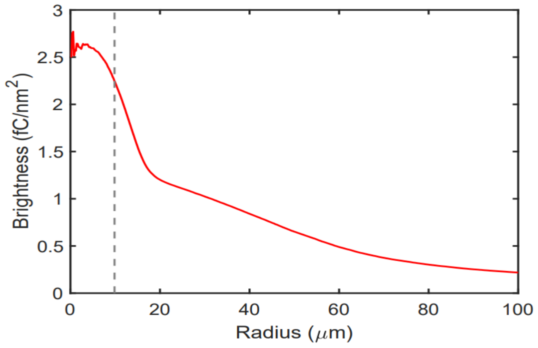
Probe size: $\sigma_r = 2\ nm$; divergence: $\sigma_{r'} \leq 1\ mrad$; emittance: $\varepsilon \leq 2\ pm$

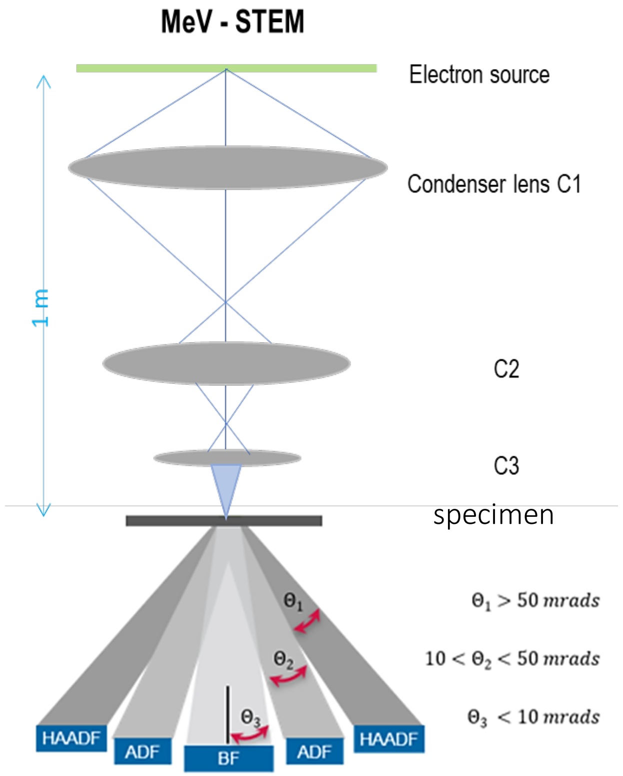
- Reduce laser spot size and MTE from photocathode → ε ≈ 2 pm·rad
- Increase QE ≥10^{-5}
- Beam dynamics come hand in hand with improved emittance
    - Apply aperture to increase both transverse and longitudinal brightness

- Preliminarily MeV-STEM design (reversal of UEM to cathode to sample)
- Assume 20 μm spot at cathode
- Dose rate 200 to 5k e-/μs



(a) Transverse distribution before aperture
(b) Transverse distribution after aperture
(c) Longitudinal phase space before aperture
(d) Longitudinal phase space after aperture



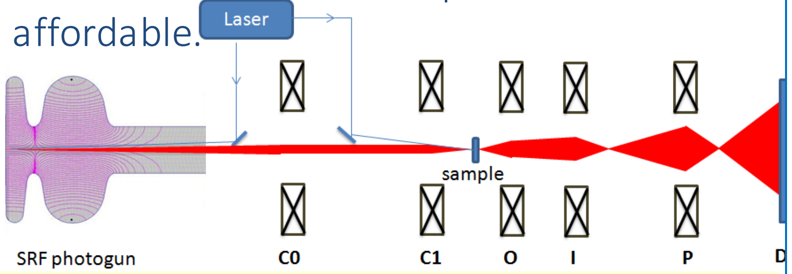


Bartnik, et al., arXiv:2108.06414v2

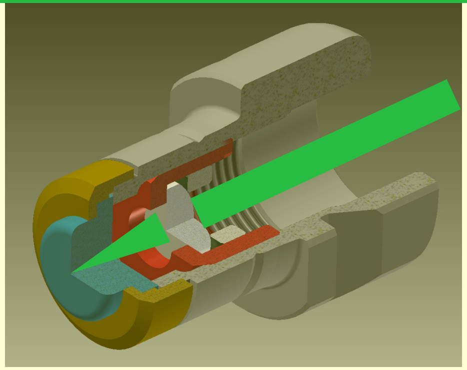
A promising source for UEM/STEM and UED: SRF photogun

Introduction:

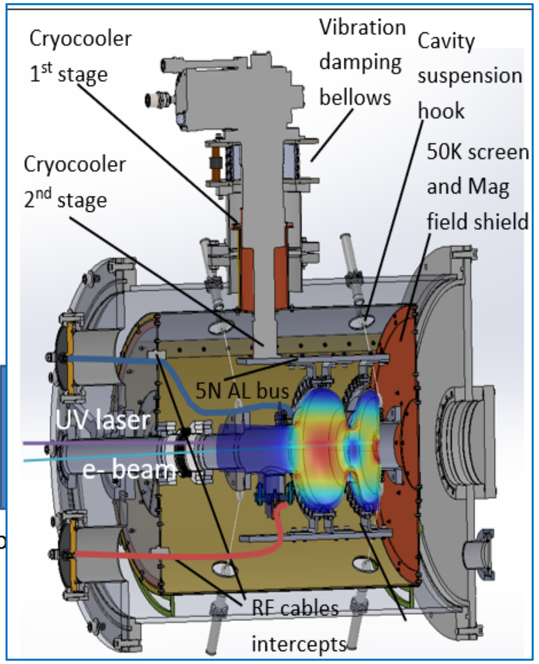
- Aimed for the first CW MeV UEM/UED application with RF frequency **1.3 GHz**.
- Using **conduction cooled Nb_3Sn** SRF photogun. Only 2W of RF power and one cryocooler are required, which makes the beamline compact and affordable.

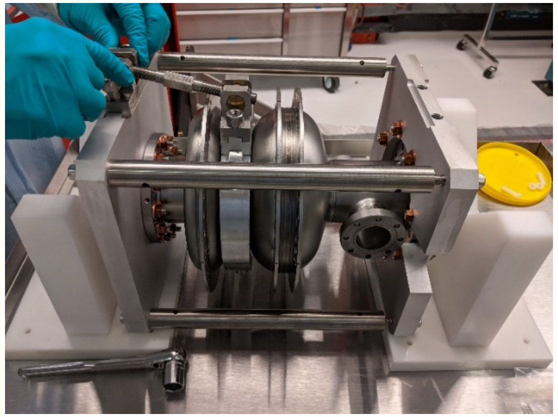


Low-ε (alkali antimonides) photocathodes in high field RF guns were operated with 100s of hours at CERN.
A UCLA variant of INFN/DESY cathode plug allows μm spot size in transmission mode with a short *f* lens.



H. Lee et al., Rev. Sci. Instrum. ***86*** *(2015) 073309*





Current Status:

- The cryomodule was commissioned: cooled down to 2.5K; magnetic field at room temperature is below 5mG.
- SRF gun (pure Nb) was tested at 2K resulted in $Q_0=10^{10}$ and reached E_z=47MV/m on axis field.
- 1st Nb_3Sn test was conducted but low $Q_0=4{\cdot}10^9$ at 4K (the film was damaged during the gun assembly)
- 2nd Nb_3Sn coating has been applied; test is coming.

Parameter	***Value***	
Application	*UED*	*UEM*
Beam energy	*1.7MeV*	*1.7MeV*
Charge	*5fC*	*0.5pC*
Energy spread (relative)	***1.3e-5***	***6.4e-5***

Details refer to a talk by Roman Kostin in this conference.

***DoE SBIR Phase II Grant #DE-SC0018621; In tight collaboration with BNL and Fermilab.**

Summary

- Accelerator based MeV UEM/STEM hold great potential for solving the challenges in probing matter at ultrafast temporal and ultrasmall spatial scales and studying life science
- Expected better performance than low-energy UED/UEM for thick samples
- Several MeV UED user facilities are being built and deliver high impact results
- MeV UEM efforts are ramping up
- MeV STEM is just in the beginning
- A great complement to XFELs
- Inter-discipline collaboration generates creative idea: real-time nondestructive Bragg diffraction based diagnostic methods

BNL UED/UEM Team

Xi Yang, Victor Smaluk, Timur Shaftan, Liguo Wang, Sean McSweeney, Lihua Yu, Ganesh Tiwari, Yoshiteru Hidaka, Guimei Wang, Lewis Doom, Jim Rose, David Bergman, Michael Fulkerson, George Ganetis, Bernard Kosciuk, Danny Padrazo, Belkacem Bacha, Charles Spataro, King Wilson, Mikhail Fedurin, Marcus Babzien, Robert Malone, Karl Kusche, Christian Cullen, Junjie Li, Lijun Wu, Yimei Zhu, Luca Cultrera, Qun Liu, Weishi Wan

Thank you!