Counting Electrons and Accounting for Ions in Focused Beam Metrology

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UDRC Themed Meeting on
Quantum Signal Processing

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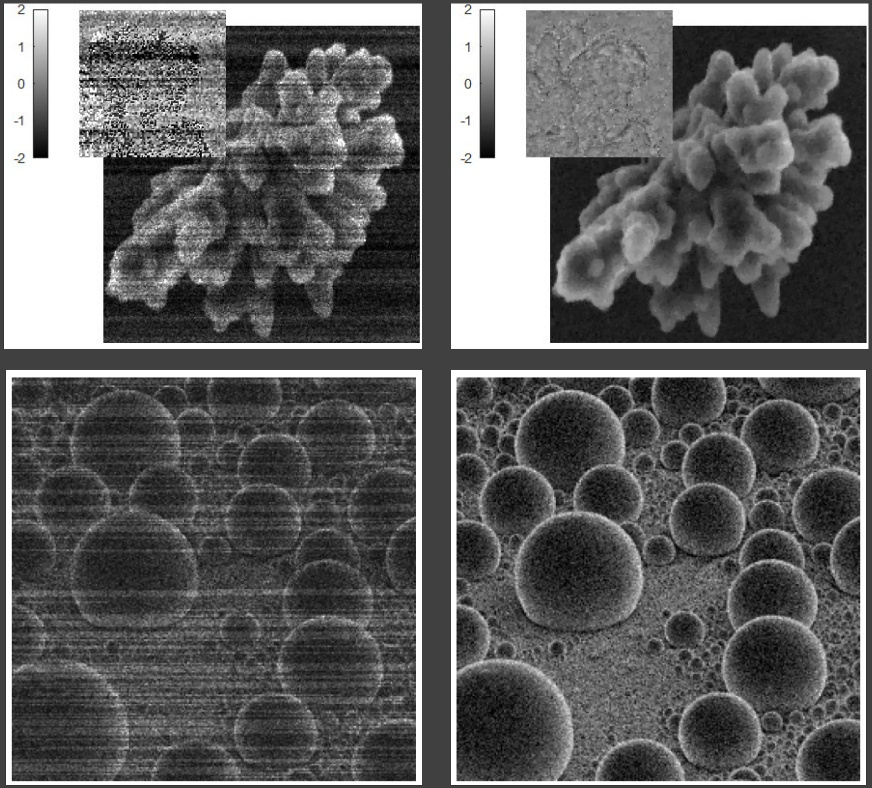
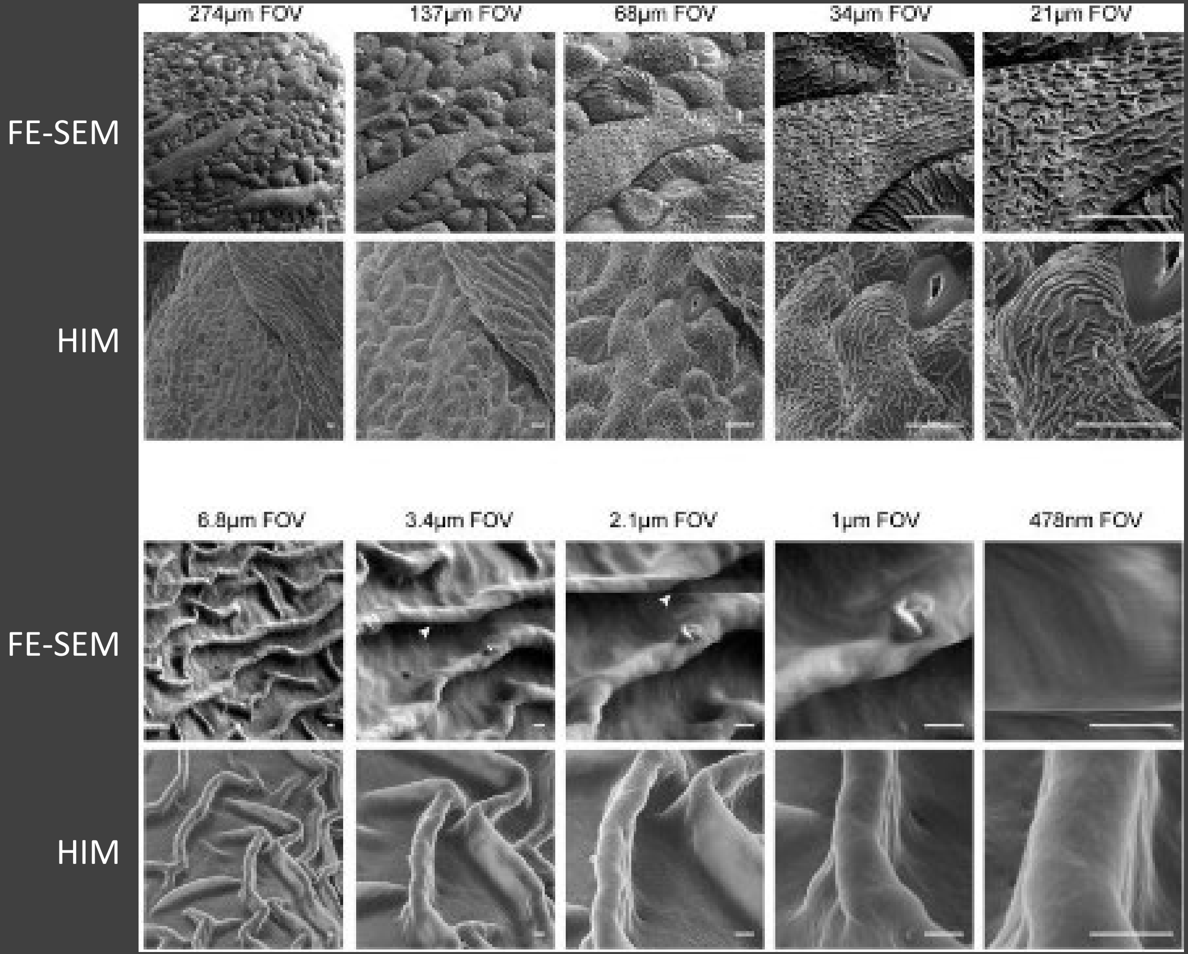


Supported in part by NSF, Draper Lab, and Zeiss

Promise of scanned helium ion beam microscopy

- High resolution, surface sensitivity, and depth of focus
- Image insulators without metal coatings

- short de Broglie wavelength
- low interaction volume
- no charging



Comparison of HIM and FE-SEM imaging in *Arabidopsis thaliana*

Joens *et al.*, Scientific Reports 3(3514), 2013

Limitation of scanned ion beam microscopy

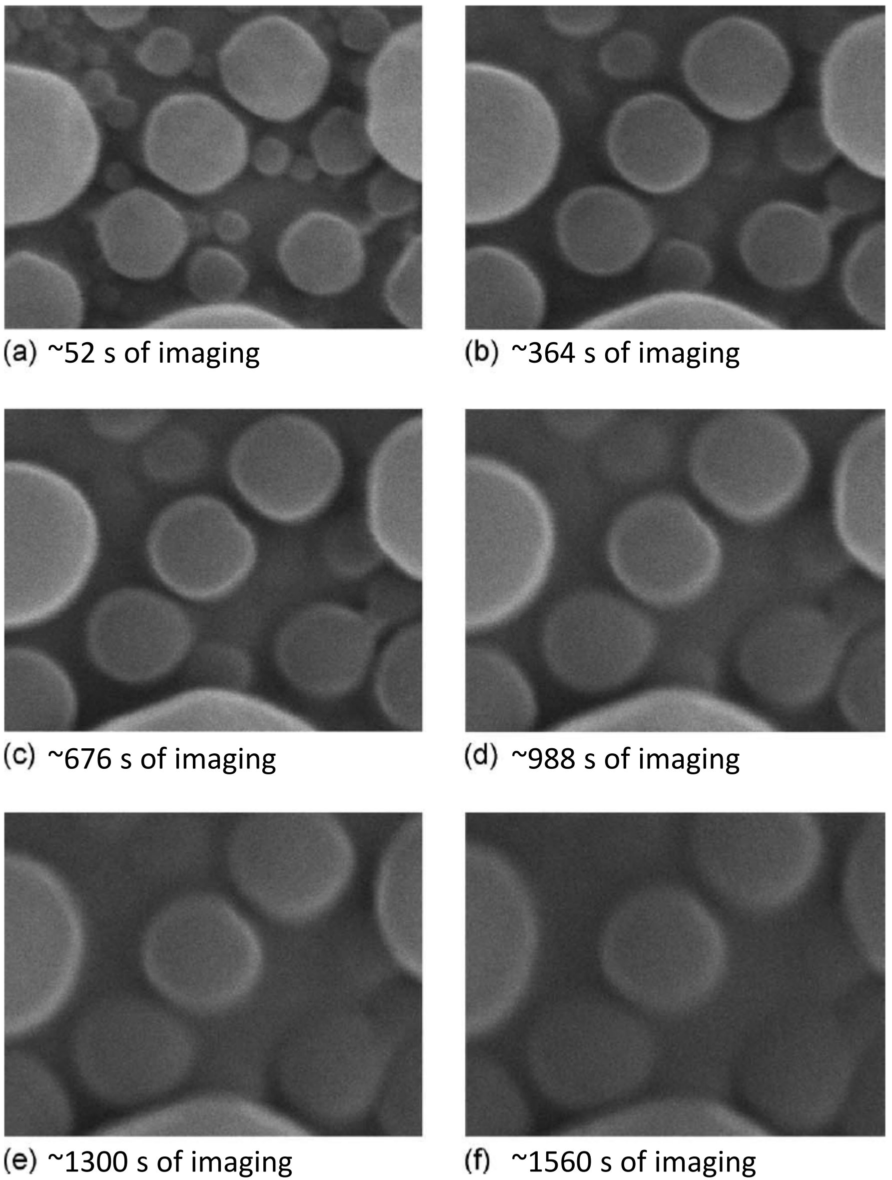
As imaging time increases:

- shot noise reduced by averaging; but
- finer features destroyed by sputtering

Ability to accurately image delicate samples is fundamentally limited

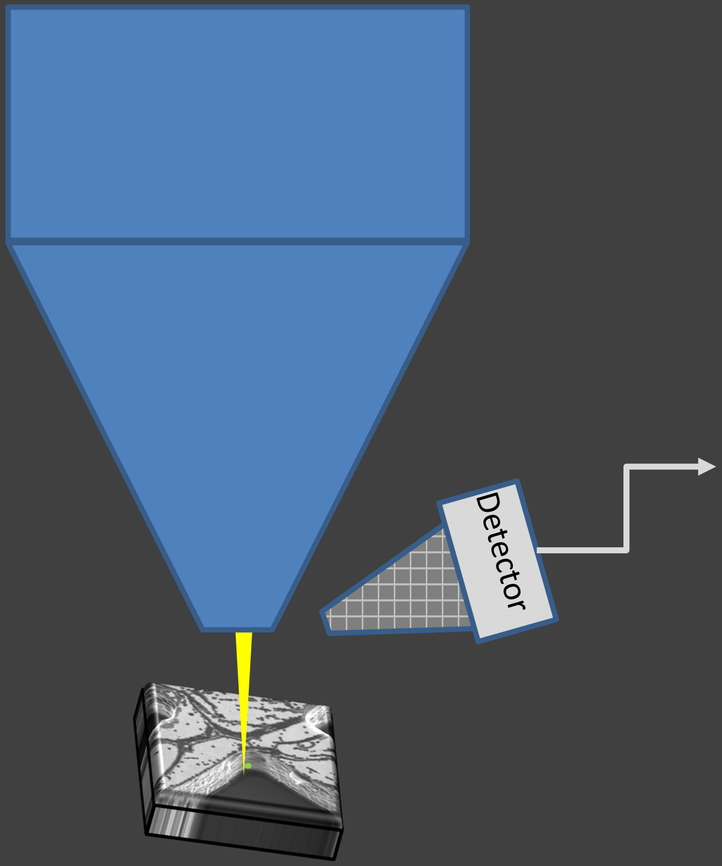
Goal: Make the most of any fixed ion dose, especially a low dose

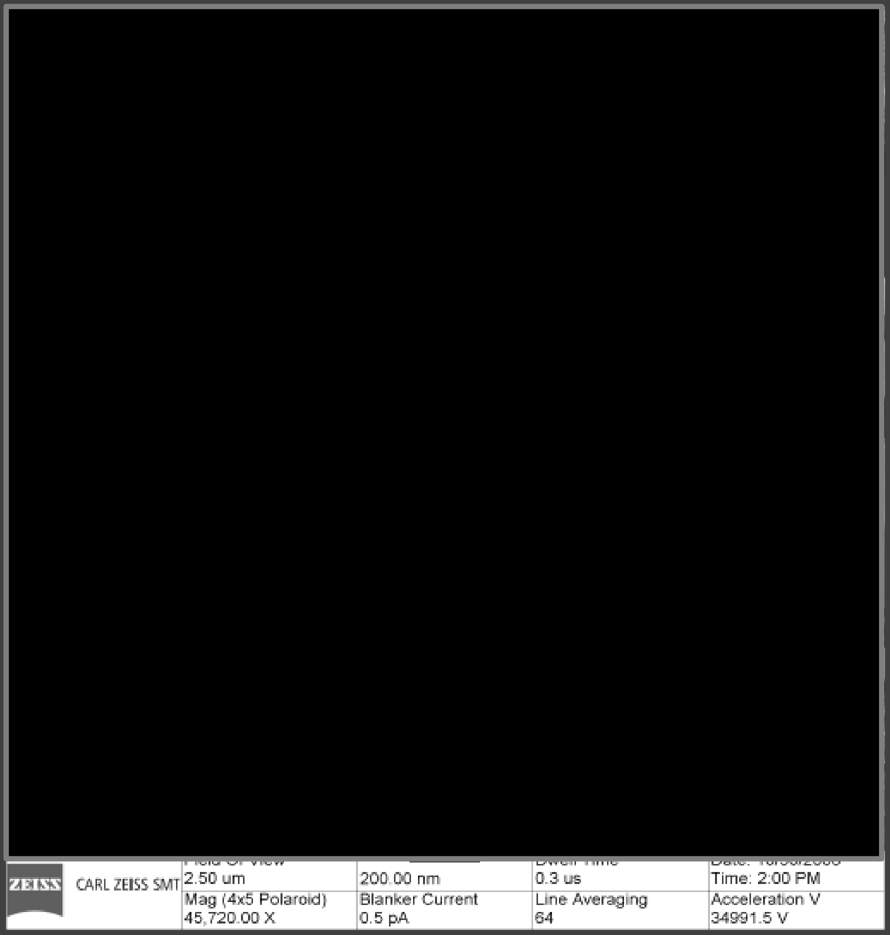
Sn-ball sample
Dwell time: 50 μs
FOV: 500 nm
~0.3 pA He$^+$


(a) ~52 s of imaging (b) ~364 s of imaging
(c) ~676 s of imaging (d) ~988 s of imaging
(e) ~1300 s of imaging (f) ~1560 s of imaging

Castaldo et al., *J. Vac. Sci. Technol. B*, 2009

Helium ion microscope





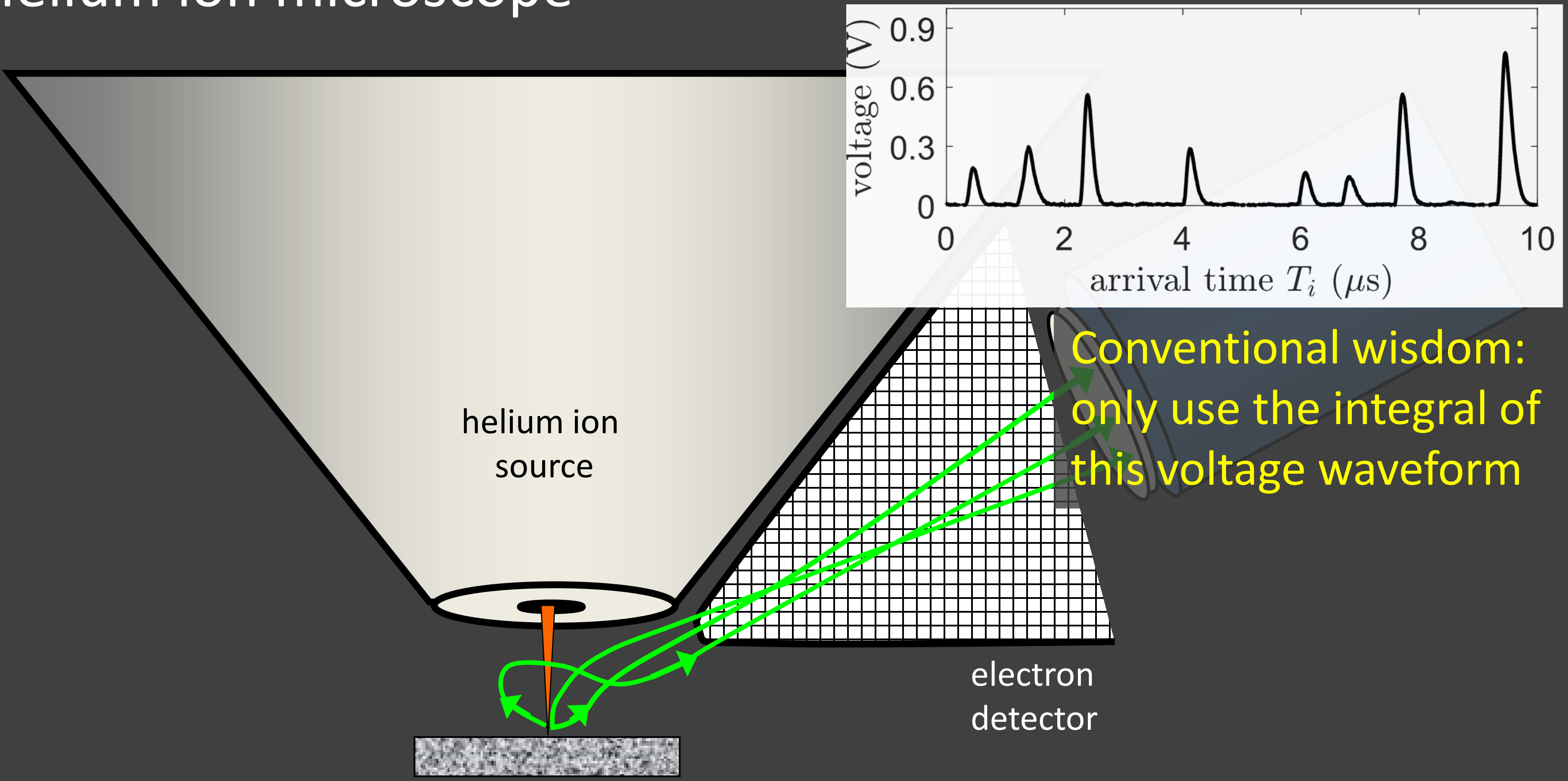
2.50 um
Mag (4x5 Polaroid)
45,720.00 X
200.00 nm
Blanker Current
0.5 pA
0.3 us
Line Averaging
64
Time: 2:00 PM
Acceleration V
34991.5 V

- Beam is raster scanned
- Some dwell time per pixel
- Detected electrons map to grayscale levels
- Like digital camera measuring one pixel at a time

? No! That's defeatist

Notte / Zeiss, 2018

Helium ion microscope



Conventional wisdom: only use the integral of this voltage waveform

Ion-sample interactions cause emission of electrons

- source beam is “primary”
- detected electrons called “secondary electrons”

Interesting things are happening within each dwell time

Key idea:

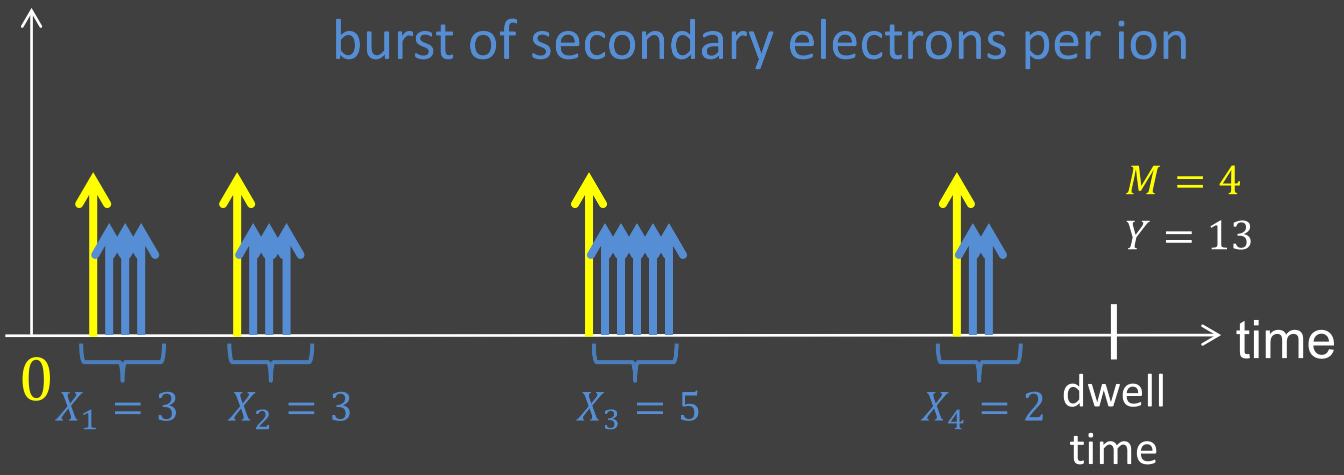
Time-resolved sensing

Helium ion microscopy – abstract model

Helium ion microscopy abstraction

ions incident as Poisson process (hidden)

burst of secondary electrons per ion (total observed)



number of ions $M \sim$ Poisson(λ)

λ: known (“dose”)

due to ion i, number of SEs $X_i \sim$ Poisson(η)

η: parameter of interest

Model: Poisson(η)-distributed marks X_i on rate-λ Poisson process

observation at the pixel: $Y = \sum_{i=1}^{M} X_i$

Goal: compute an estimate of η from Y (for each pixel, separately)

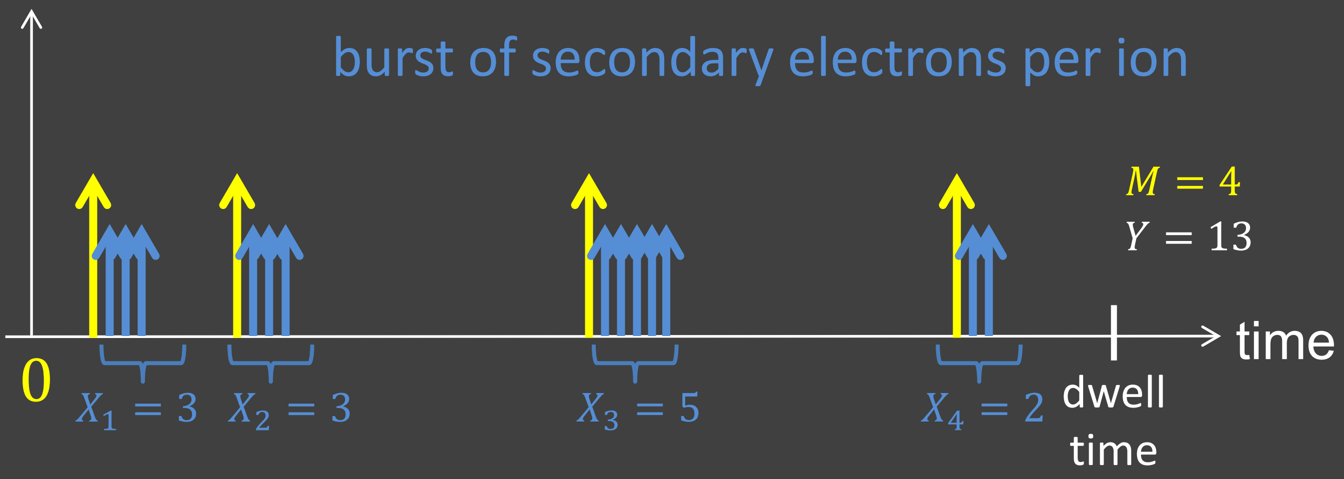
Inspirational thought experiment

An oracle-aided microscope

Helium ion microscopy abstraction – with oracle

ions incident as Poisson process

provided



burst of secondary electrons per ion

$M = 4$

$Y = 13$

0

$X_1 = 3$ $X_2 = 3$ $X_3 = 5$ $X_4 = 2$

time

dwell time

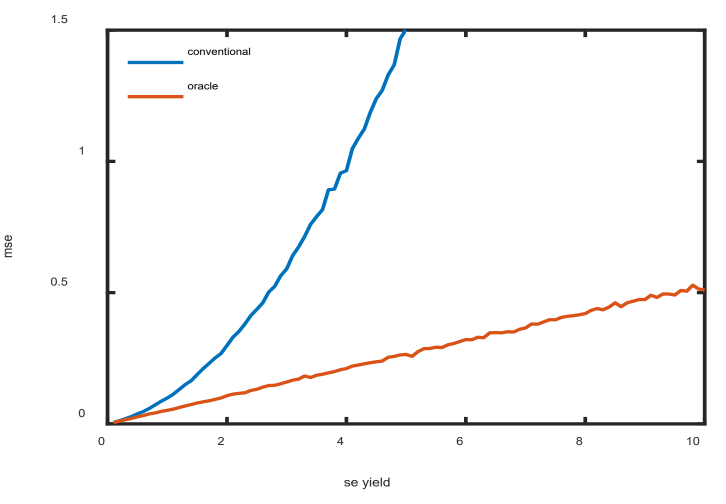
number of ions $M \sim \text{Poisson}(\lambda)$

M: known

due to ion i, number of SEs $X_i \sim \text{Poisson}(\eta)$

η: parameter of interest

observation at the pixel: $Y = \sum_{i=1}^{M} X_i$



Conventional $\hat{\eta}_{conv} = Y/\lambda$

Oracle $\hat{\eta}_{oracle} = Y/M$

Mathematical details of model

Detailed probabilistic generative model

One acquisition at one pixel with dose λ

$$P(Y = y\,;\,\eta) = \frac{e^{-\lambda\eta}}{y!}\sum_{m=0}^{\infty}\frac{(\lambda e^{-\eta})^m m^y}{m!}$$

"Neyman Type A"

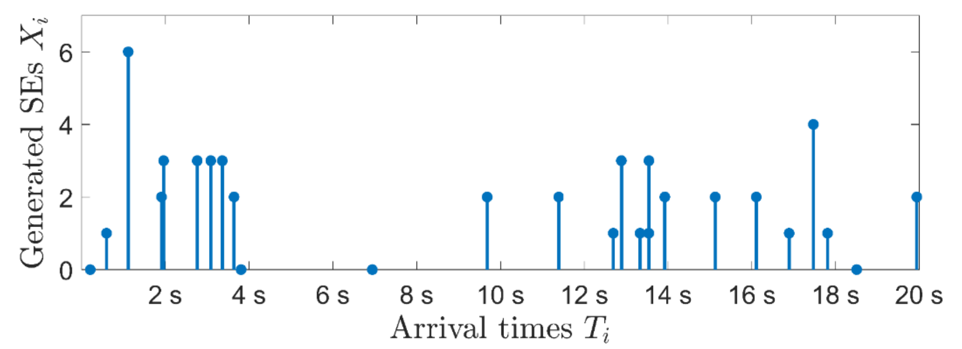
$E[Y] = \lambda\eta$ $\Rightarrow$ baseline estimate $\hat{\eta} = Y/\lambda$ is unbiased

$\Rightarrow$ making an image of Y is reasonable

$\mathrm{var}(Y) = \lambda\eta(\eta+1)$ $\Rightarrow$ estimate $\hat{\eta} = Y/\lambda$ has MSE $\frac{\eta(\eta+1)}{\lambda}$

Hypothetical deterministic beam with λ ions $\Rightarrow \mathrm{var}(Y) = \lambda\eta$

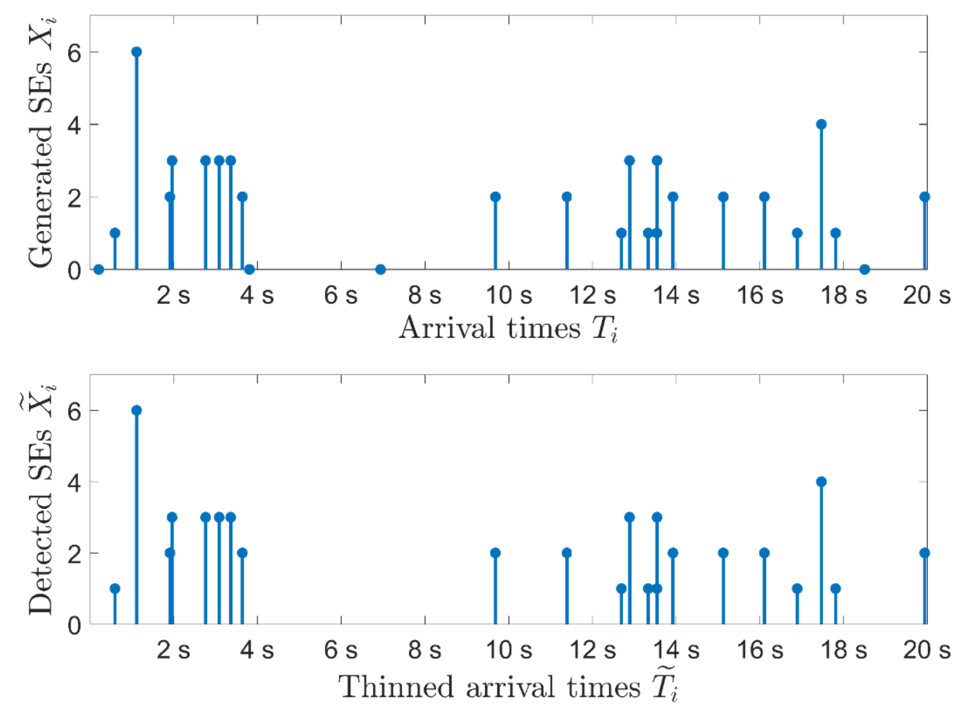
Factor of $(\eta+1)$ is price of a random beam ("source shot noise")



Unobservable underlying process

Poisson(η)-distributed marks X_i on rate-λ Poisson process

Conventional to observe only a single scalar total



Unobservable underlying process

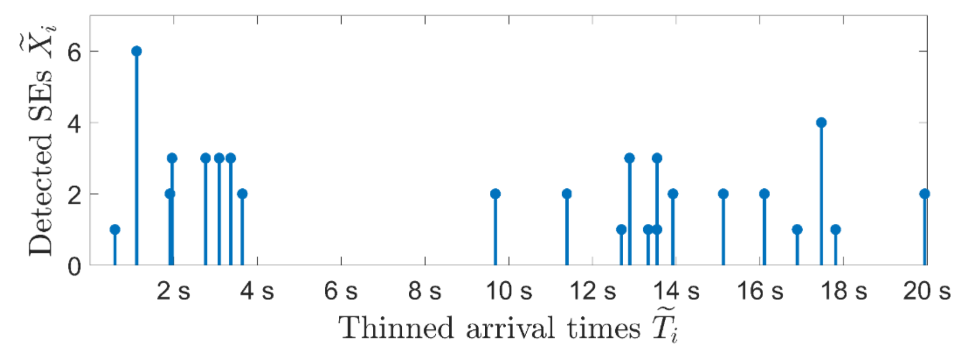
Thinned process (“continuous-time model”)

TruncatedPoisson(η)-distributed marks $\widetilde{X}_i$ on rate-$\lambda(1-\exp(-\eta))$ Poisson process

Continuous-time observation:

$\widetilde{M} \sim$ Poisson$(\lambda(1 - \exp(-\eta)))$

TruncatedPoisson(η)-distributed marks $\widetilde{X}_i$

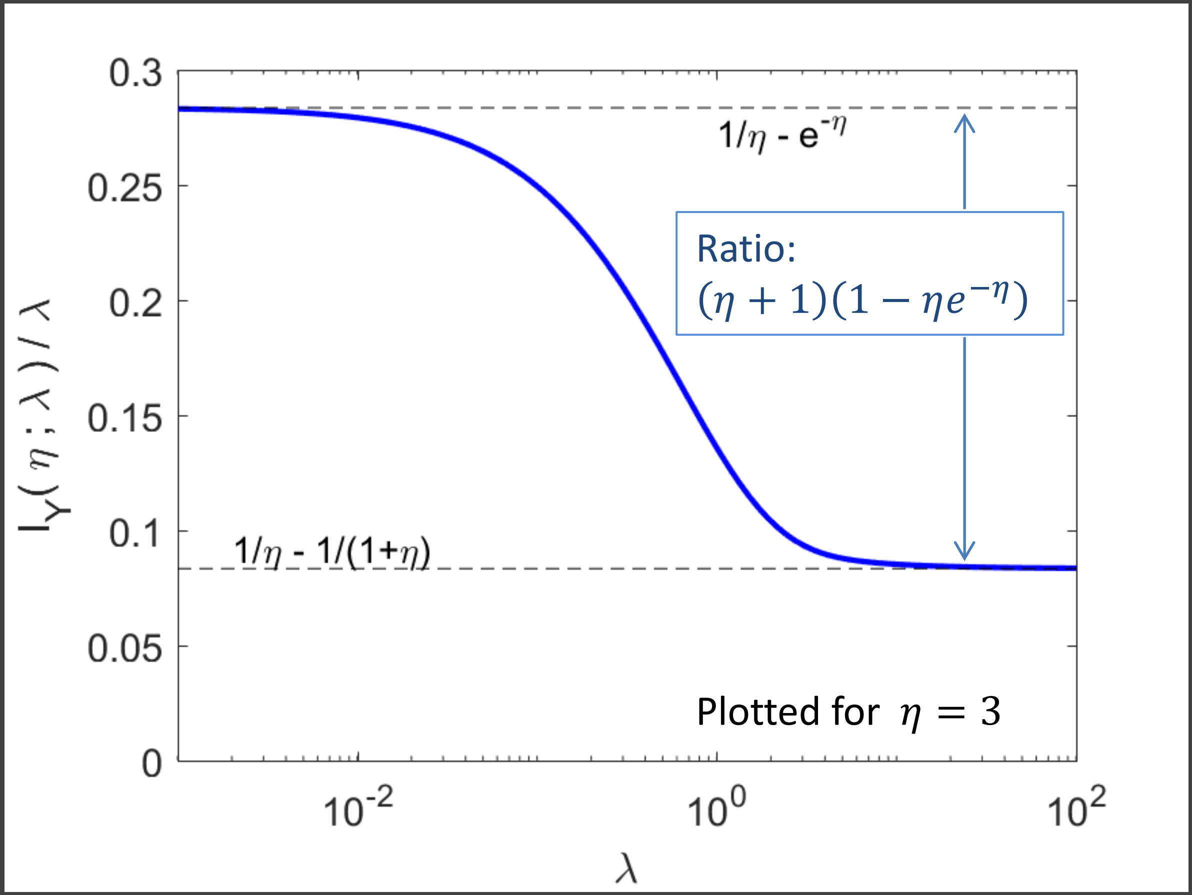


Thinned process
("continuous-time model")

Omitting discrete-time model today (messier)

Analysis through Fisher information

- Fisher information about parameter η in measurement Y
- How does Fisher information per ion behave?



Nontrivial numerical computation, nontrivially derived asymptotes

FI for continuous-time observation is exactly $\lambda\left(\frac{1}{\eta} - e^{-\eta}\right)$

Improvement factor almost the full price of source shot noise

Continuous-time estimators

$\widetilde{M} \sim$ Poisson$(\lambda(1 - \exp(-\eta)))$

TruncatedPoisson(η)-distributed marks $\widetilde{X}_i$

$$Y = \sum_{i=1}^{\widetilde{M}} \widetilde{X}_i = \sum_{i=1}^{M} X_i$$

Quotient mode: Treat $\widetilde{M}$ as the the number of ions:

$$\hat{\eta}_{QM} = Y / \widetilde{M}$$

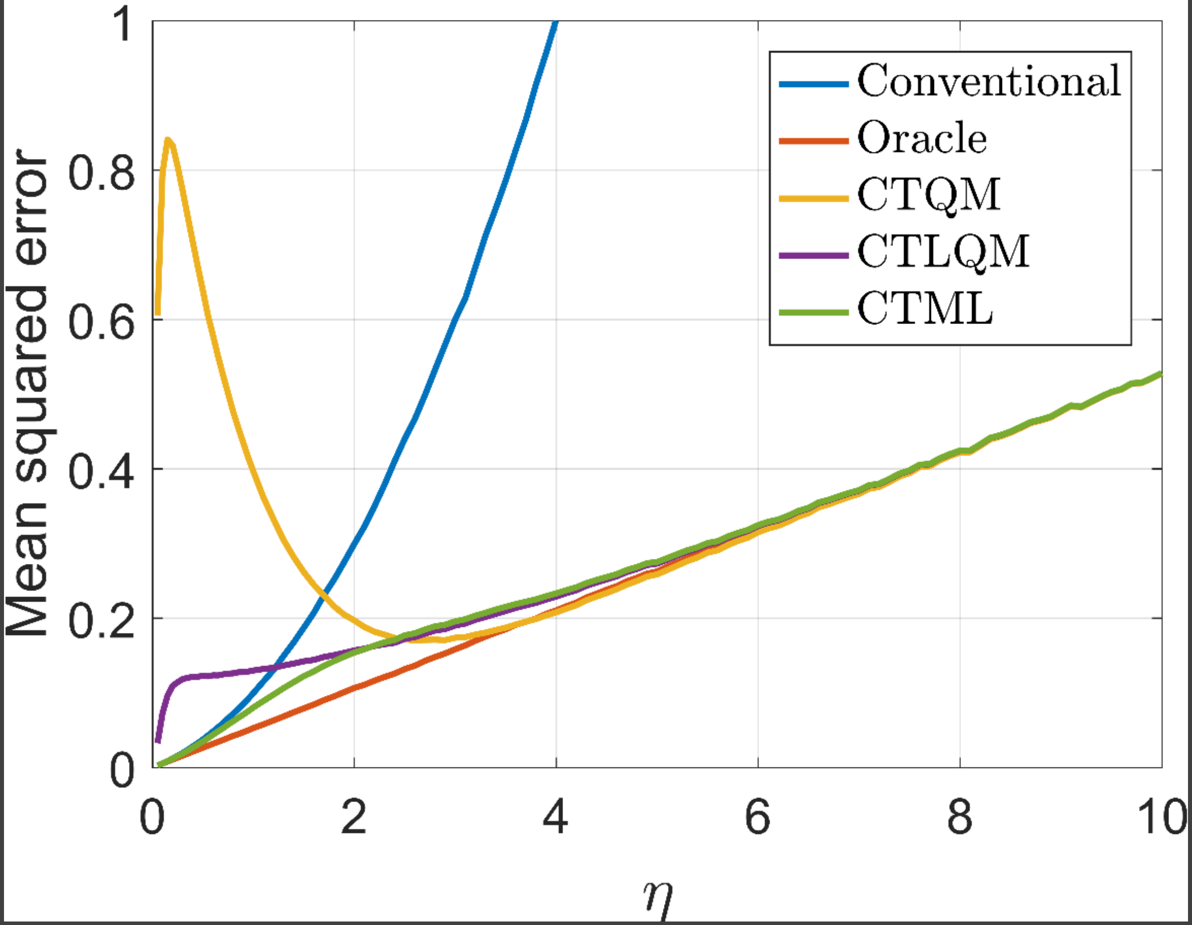
Lambert Quotient mode: Treat $(1 - e^{-\eta})^{-1}\widetilde{M}$ as the number of ions:

$$\hat{\eta}_{LQM} = W\left(-\hat{\eta}_{QM} \exp(-\hat{\eta}_{QM})\right) + \hat{\eta}_{QM}$$

Maximum likelihood: Root of

$$\hat{\eta}_{ML} = \frac{Y}{\widetilde{M} + \lambda \exp(-\hat{\eta}_{ML})}$$

Continuous-time estimator performances



Conventional $\hat{\eta}_{conv} = Y/\lambda$

Oracle $\hat{\eta}_{oracle} = Y/M$

Quotient mode $\hat{\eta}_{QM} = Y/\widetilde{M}$

Lambert Quotient mode

Maximum likelihood

$\lambda = 20$

Roughly: electrons $\eta \in [0.2, 1]$
helium ions $\eta \in [1, 8]$
heavier ions larger

Peng, Murray-Bruce & Goyal, *IEEE Trans. Computational Imaging*, 2021

Recap

- Particle beam microscopy as a quantitative imaging modality is the estimation of secondary electron yield η
- Randomness of incident beam (“source shot noise”) is a nuisance
- Using time-resolved data mitigates source shot noise
- MSE lower roughly by factor of $\eta + 1$ (uniform across λ)
- Could be used to lower dose roughly by factor of $\eta + 1$

Peng, Murray-Bruce, Berggren & Goyal, *Ultramicroscopy*, 2020

Peng, Murray-Bruce & Goyal, *IEEE Trans. Computational Imaging*, 2021

Remainder of the talk – more time-resolved sensing

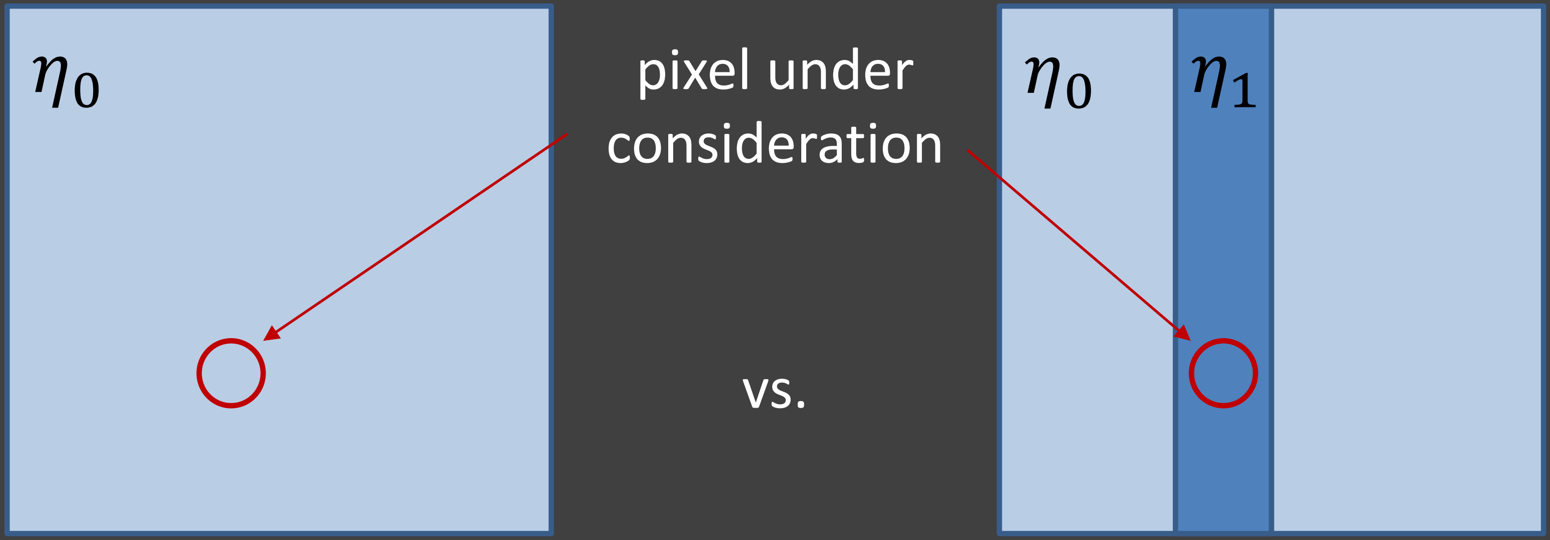
- Improved feature detection (abstracted as hypothesis testing)
- Improved resolution
- Online estimation of beam current
- Combining with regularization

Watkins, Seidel, Peng, Agrawal, Yu & Goyal, *Microscopy & Microanalysis*, 2021
Seidel, Watkins, Peng, Agrawal, Yu & Goyal, *Microscopy & Microanalysis*, 2022
Seidel, Watkins, Peng, Agrawal, Yu & Goyal, *IEEE Trans. Computational Imaging*, 2022
Agrawal, Peng & Goyal, *IEEE J. Sel. Areas Inform. Theory*, 2023
Peng, Kitichotkul, Seidel, Yu & Goyal, *IEEE Trans. Computational Imaging*, 2023
Agarwal, Kasei, Schultz, Feldman & Goyal, *Microscopy & Microanalysis*, 2023

Feature detection

(heavily abstracted)

A detection problem



Substrate with SE yield $\eta = \eta_0$

Does the pixel have SE yield $\eta = \eta_0$ or $\eta = \eta_1$?

Study through probability of missed detection P_{MD} with probability of false alarm held constant

Error exponents – Kullback-Leibler divergence

$$\lim_{n\to\infty} -\frac{1}{n}\log P_{\mathrm{MD}} = D_{KL}\underbrace{(p_0||p_1)}_{\text{relevant distributions}}$$

Conventional observation:

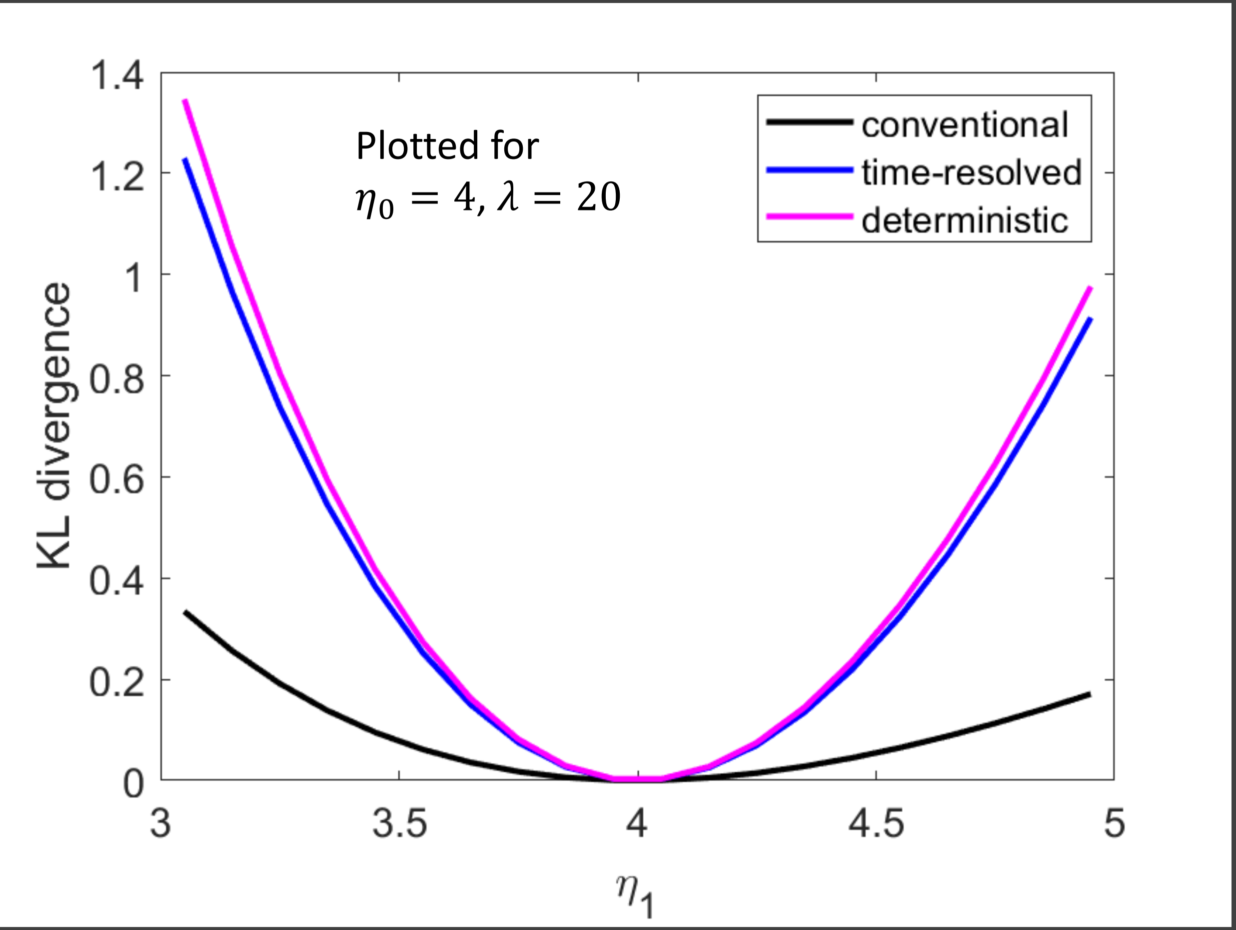
$$P(Y = y\,;\,\eta) = \frac{e^{-\lambda}\eta}{y!}\sum_{m=0}^{\infty}\frac{(\lambda e^{-\eta})^m m^y}{m!}$$

Continuous-time observation:

$\widetilde{M}$ ~ Poisson($\lambda(1-\exp(-\eta))$)

TruncatedPoisson(η)-distributed marks $\widetilde{X}_i$

Analysis through Kullback-Leibler divergence



$\eta_0 = \eta_1$: decision is hopeless

Probability of error decreases with increasing $|\eta_0 - \eta_1|$

Improvement with time-resolved observations

Almost matches deterministic beam

Gap can be arbitrarily large

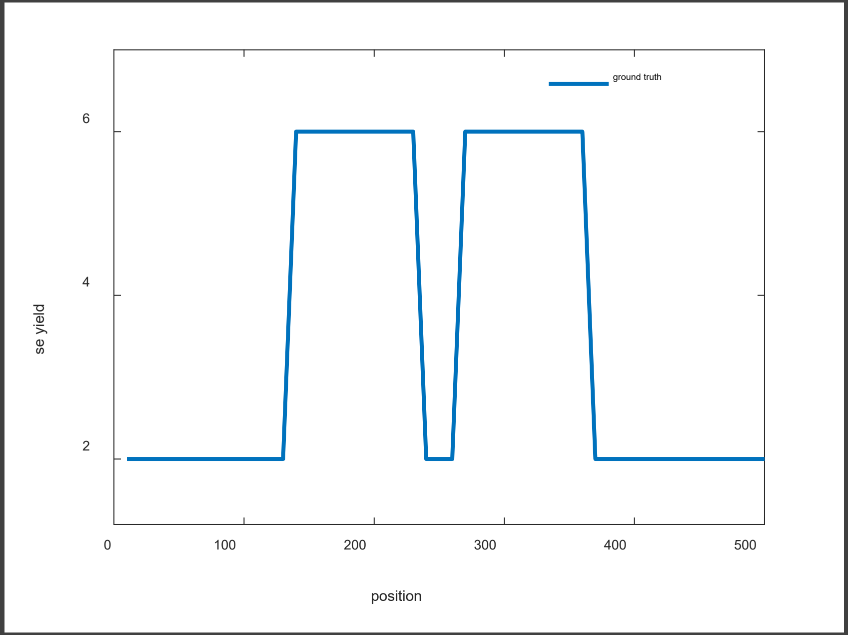
Agrawal, Peng & Goyal, *IEEE J. Sel. Areas Inform. Theory*, 2023

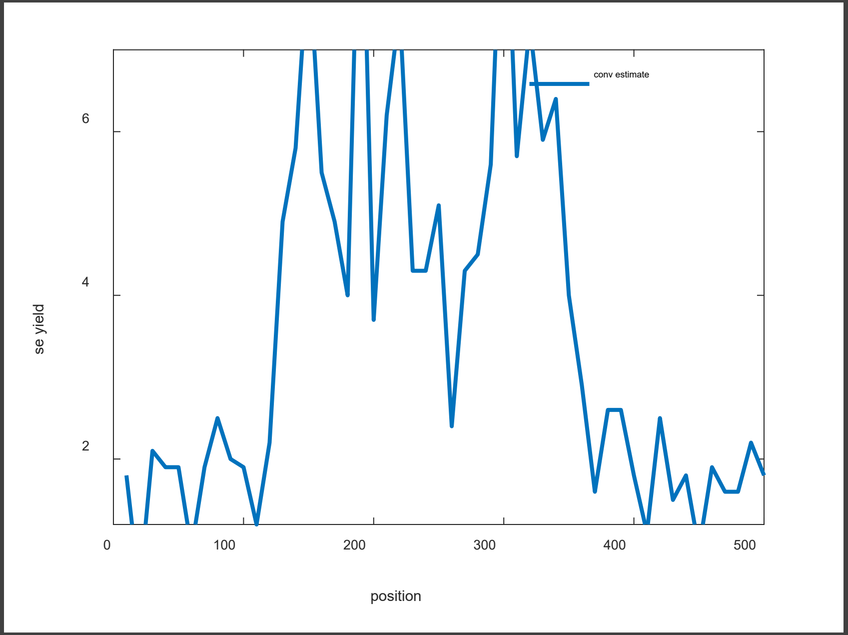
Resolution improvement

Resolution as distinguishing a feature

Size of smallest feature reliably determined to be present

One scan line cross-section

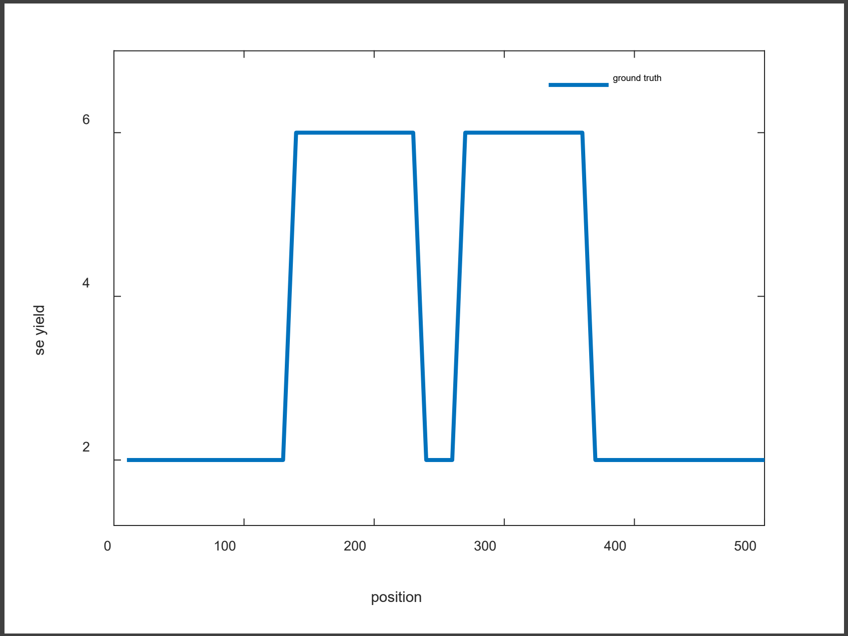


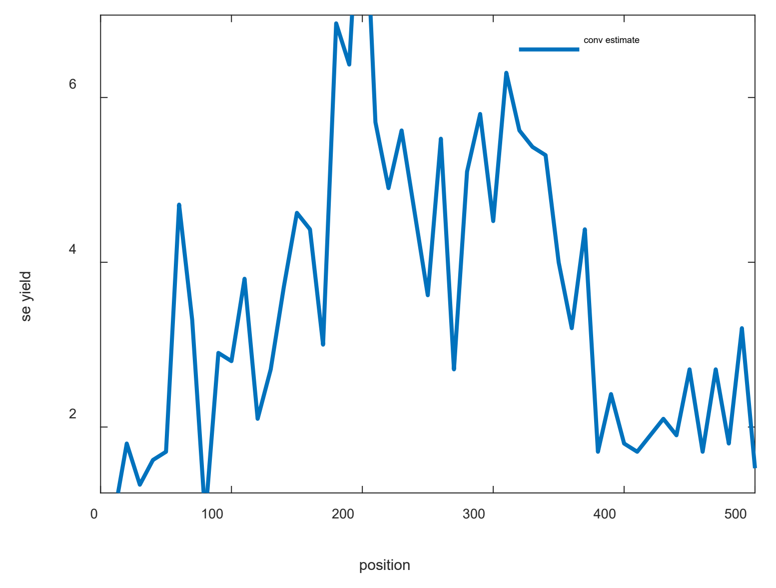


Acquisition not easily interpretable

Resolution as distinguishing a feature

Size of smallest feature reliably determined to be present

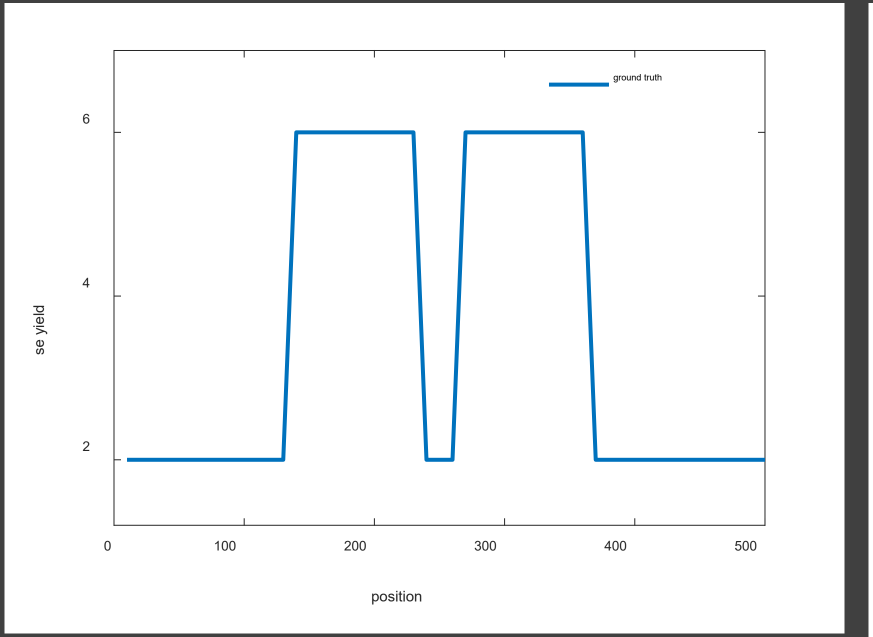


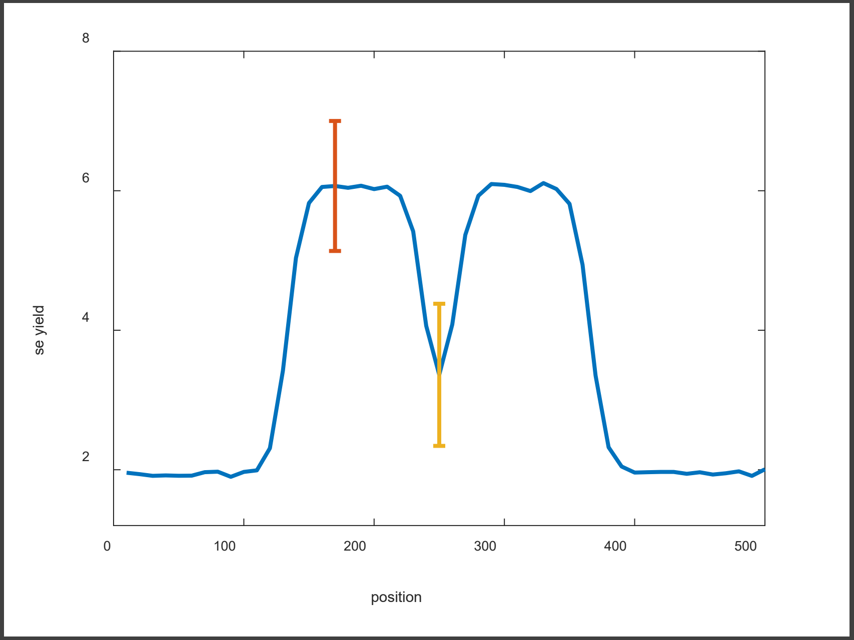


Acquisition not easily interpretable

Statistics

- Simulations with Gaussian beam spatial cross-section
- Compute mean and standard deviation per pixel
- Non-overlapping error bars: confident feature is present
- Resolution: smallest feature giving non-overlapping error bars





Statistics

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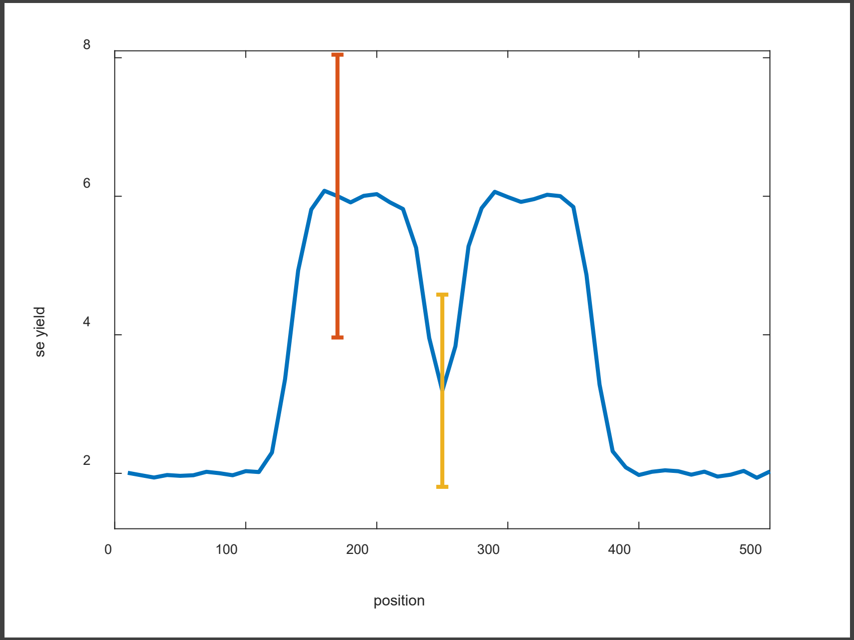
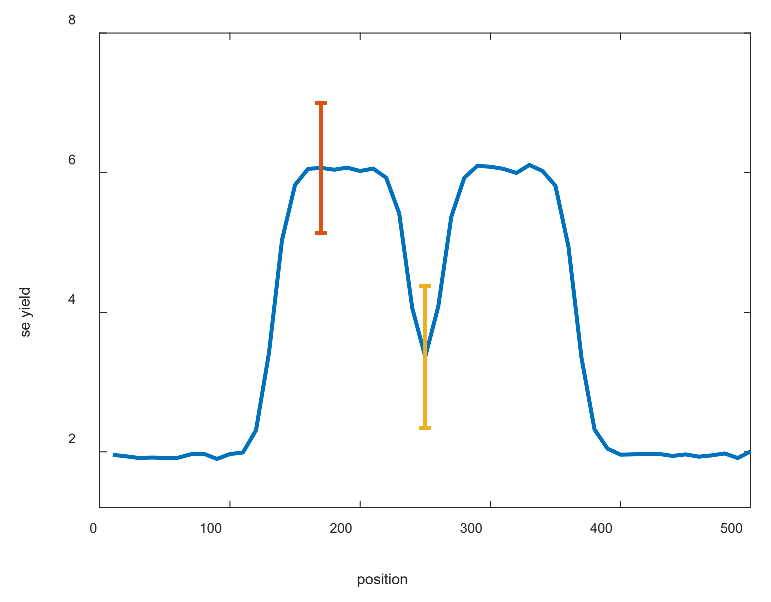


not confident



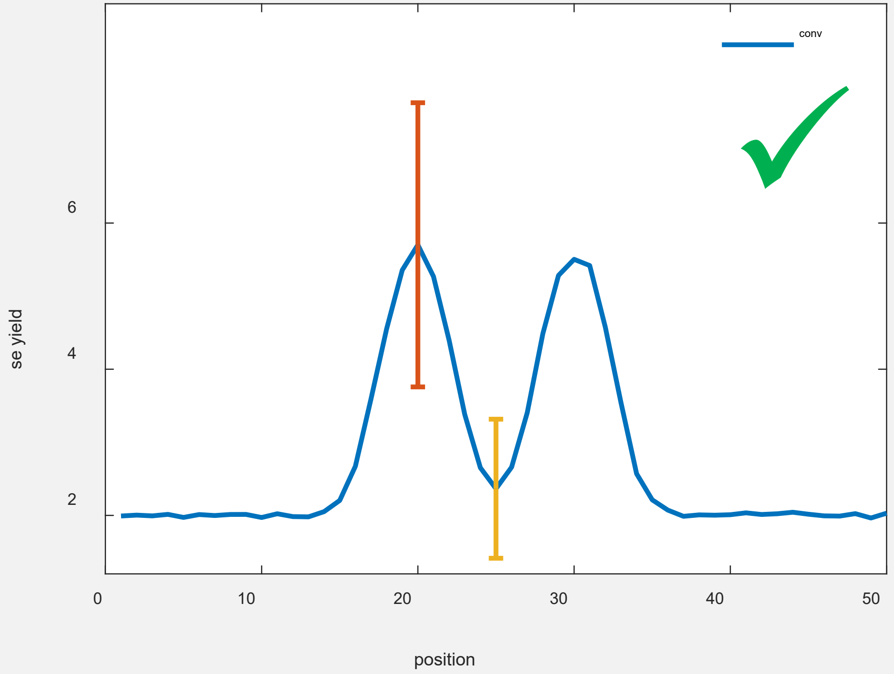
confident

Resolution improvements

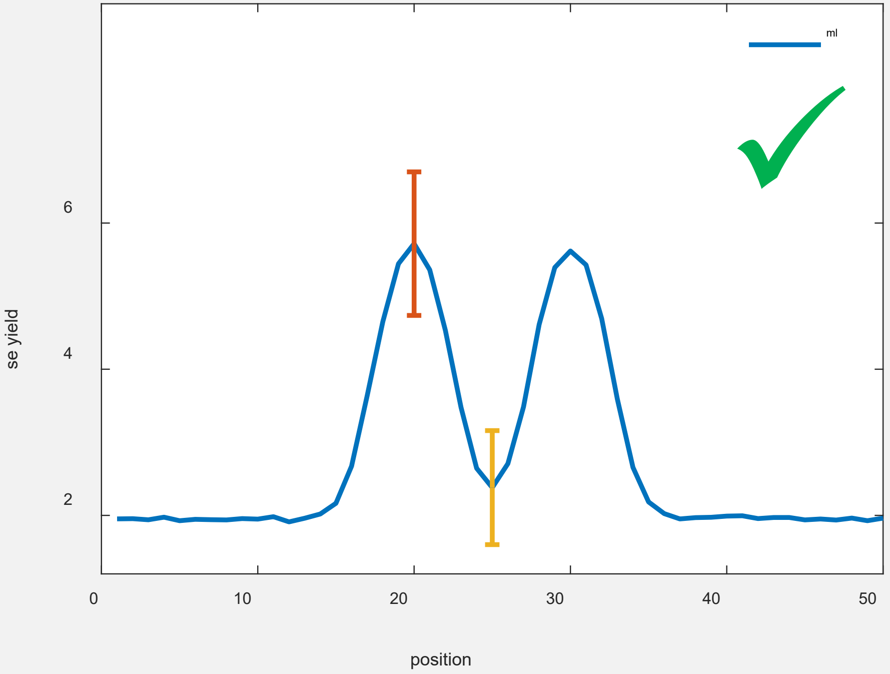
Scan steps: 10

Gaussian beam σ: 15

Size of feature: 50



conventional

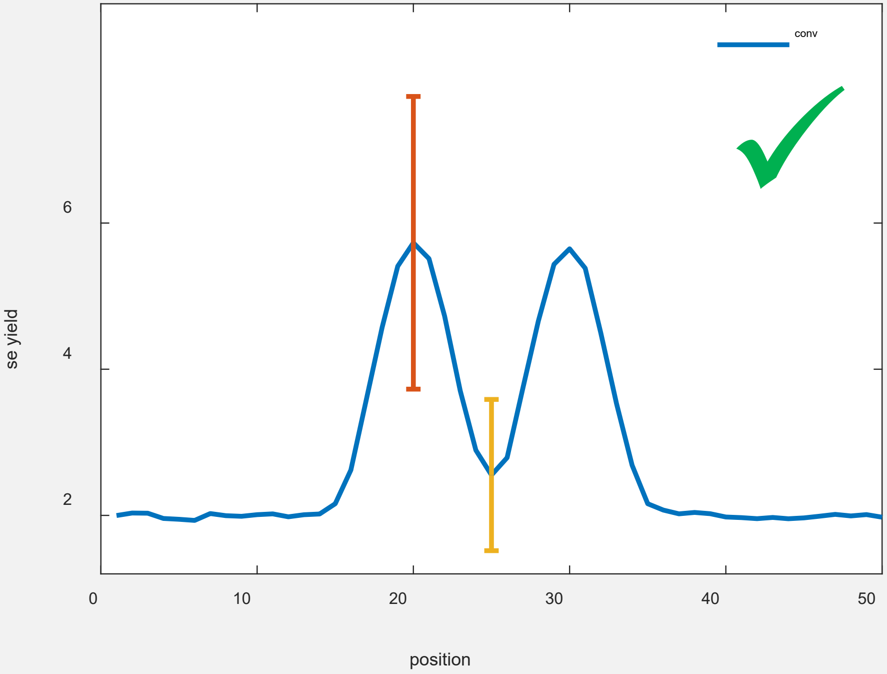


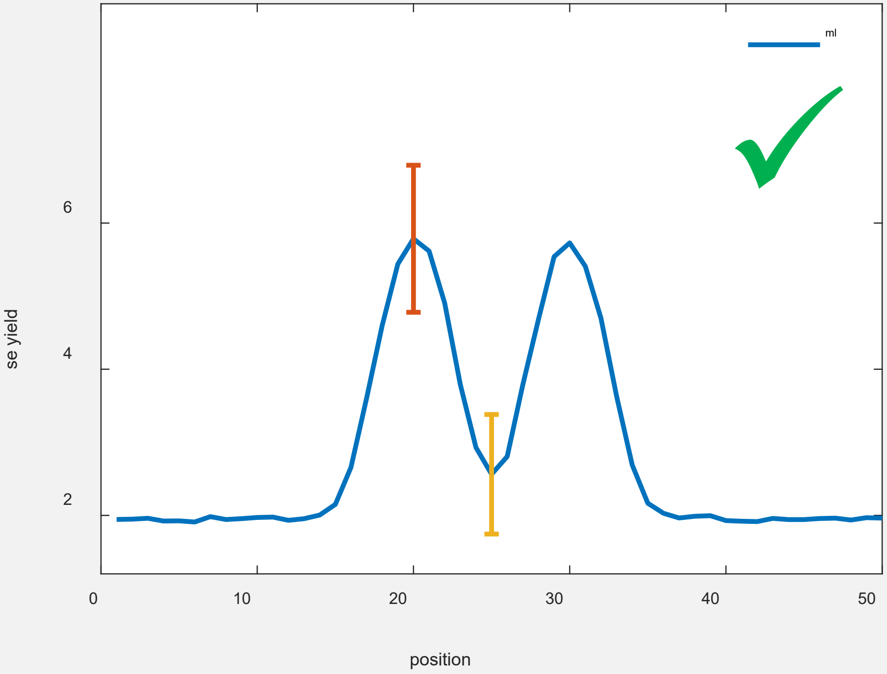
improved

Resolution improvements

Scan steps: 10
Gaussian beam σ: 15

Size of feature: 45





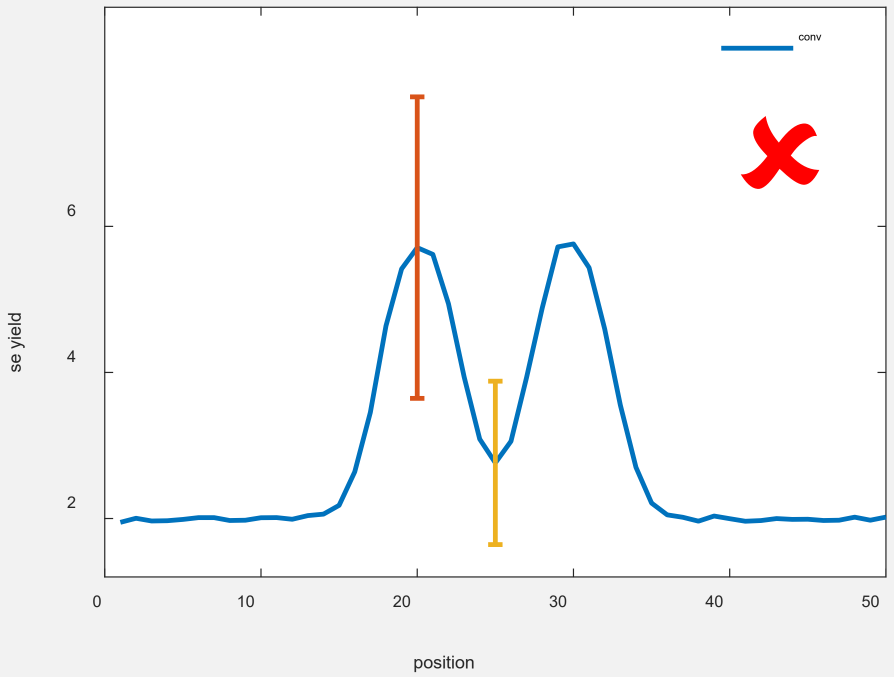
conventional

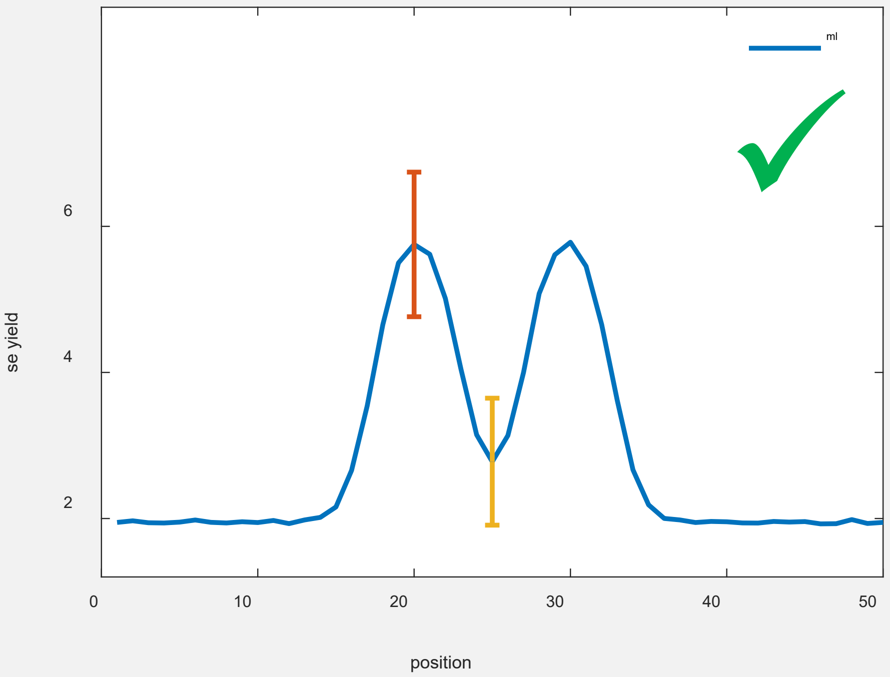
improved

Resolution improvements

Scan steps: 10
Gaussian beam σ: 15

Size of feature: 40





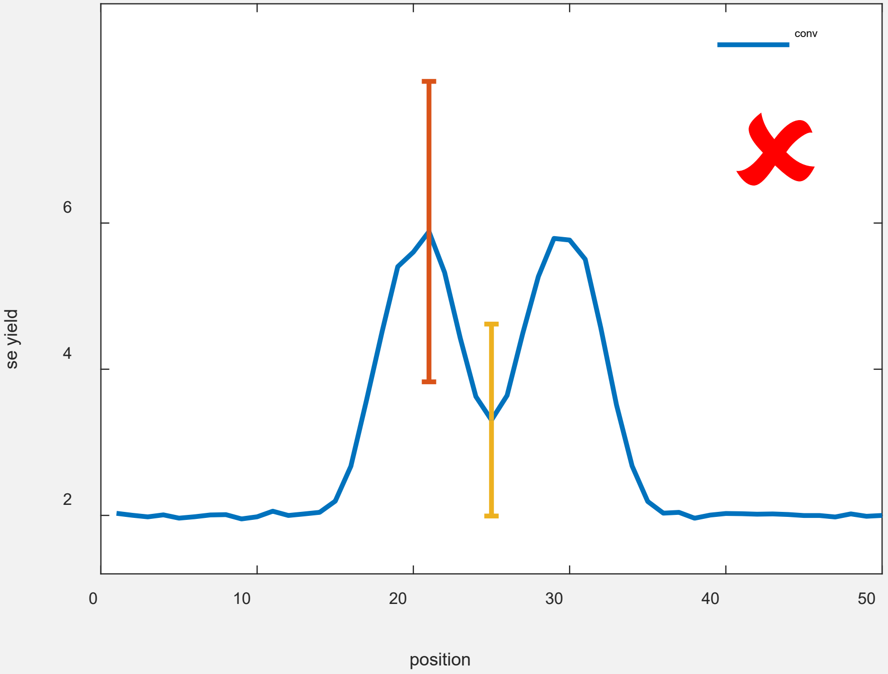
conventional

improved

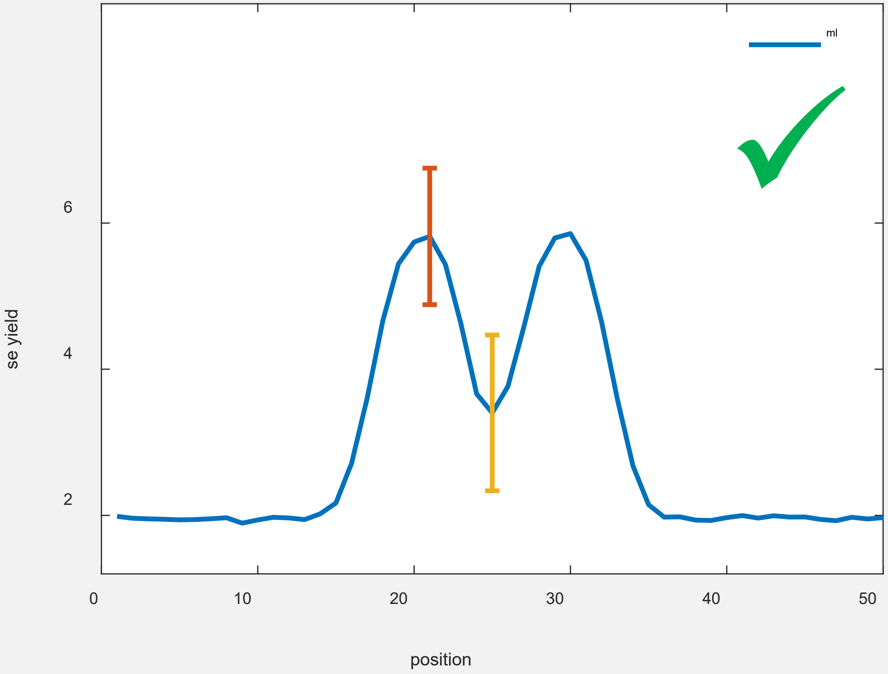
Resolution improvements

Scan steps: 10
Gaussian beam σ: 15

Size of feature: 30



conventional

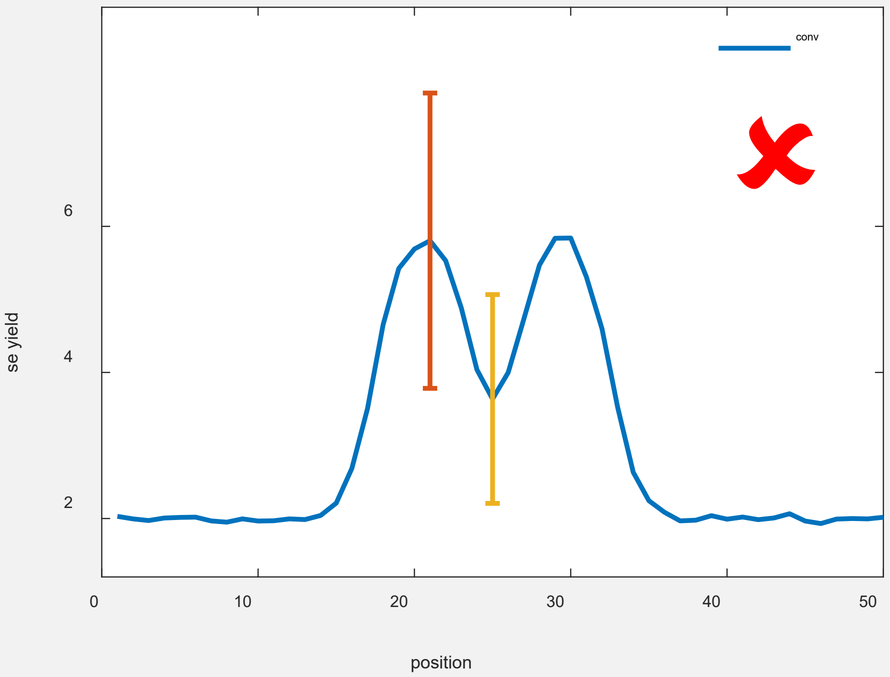


improved

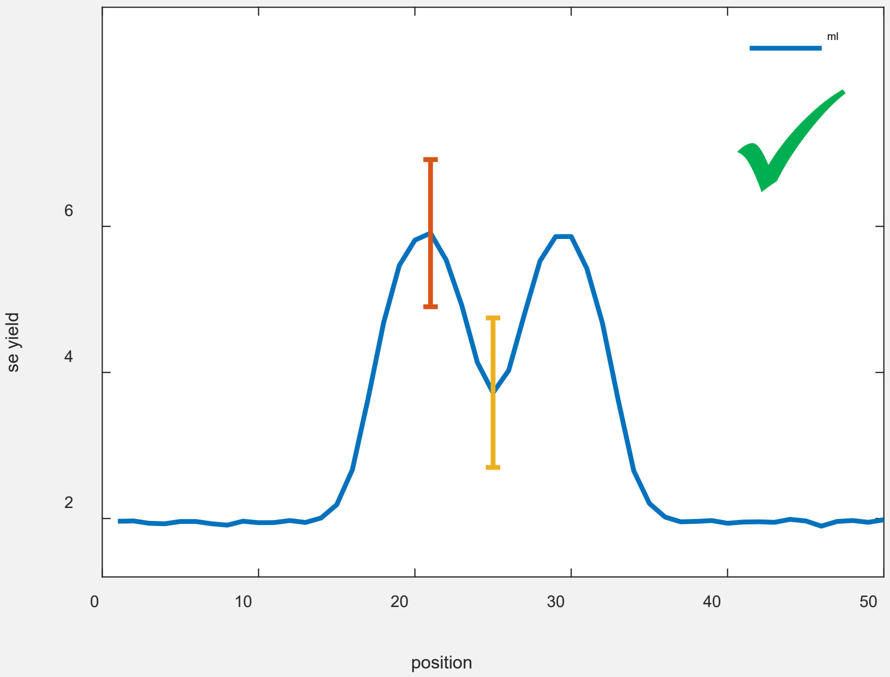
Resolution improvements

Scan steps: 10
Gaussian beam σ: 15

Size of feature: 25



conventional

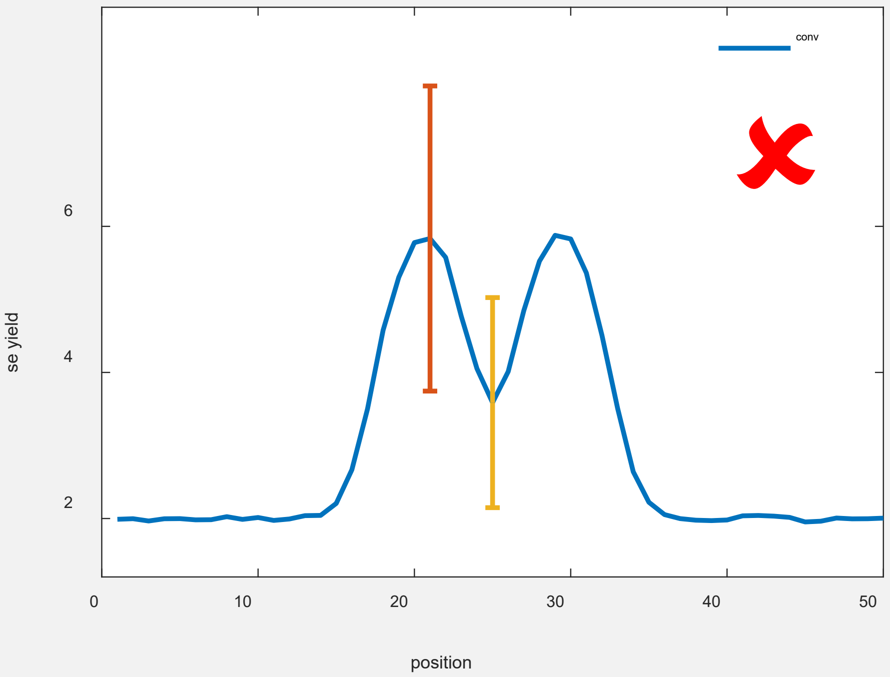


improved

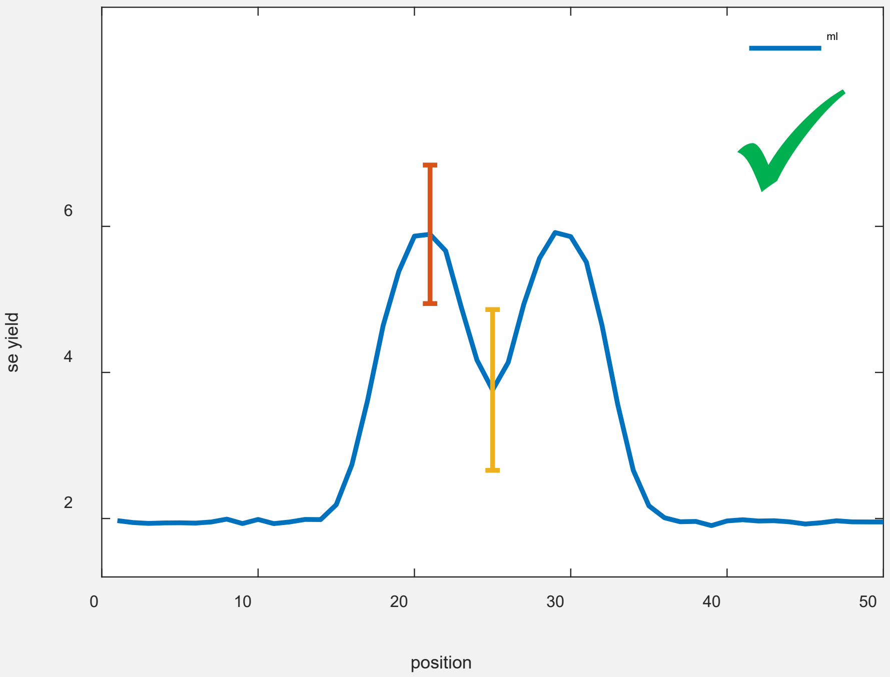
Resolution improvements

Scan steps: 10
Gaussian beam σ: 15

Size of feature: 22



conventional



improved

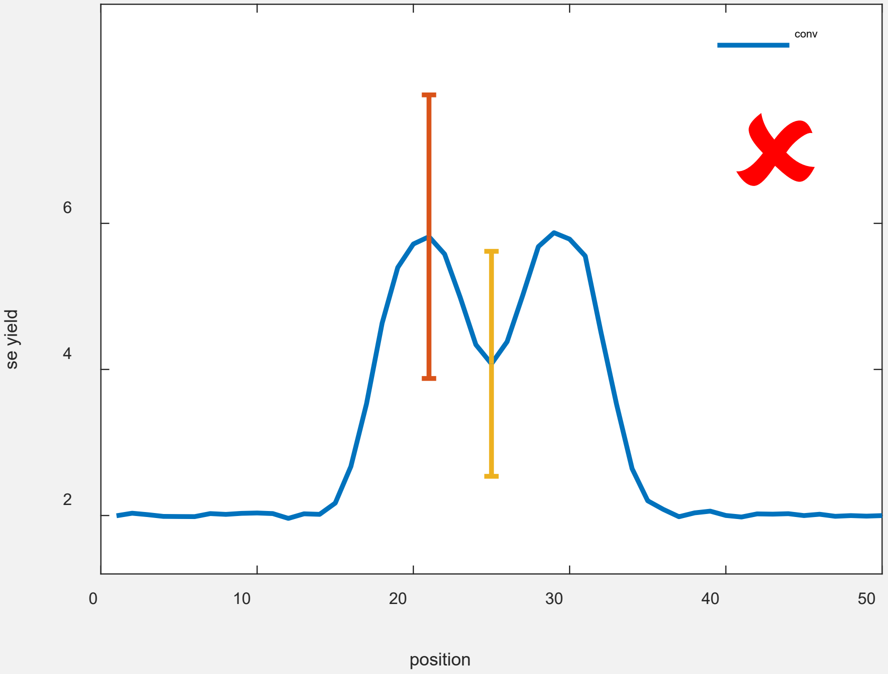
Resolution improvements

Scan steps: 10

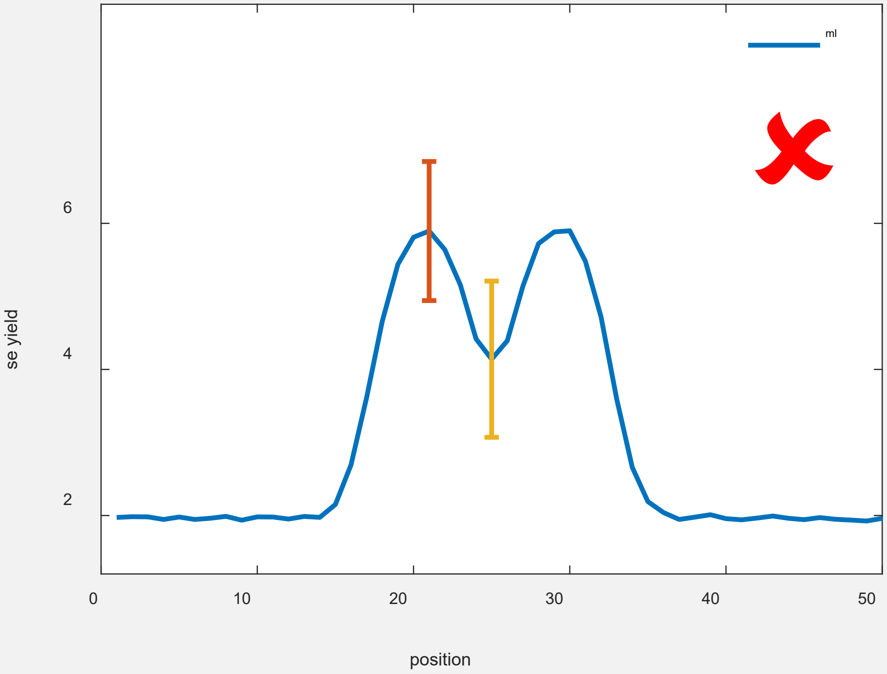
Gaussian beam σ: 15

Size of feature: 20

Resolution has been improved from ~45 to ~22



conventional



improved

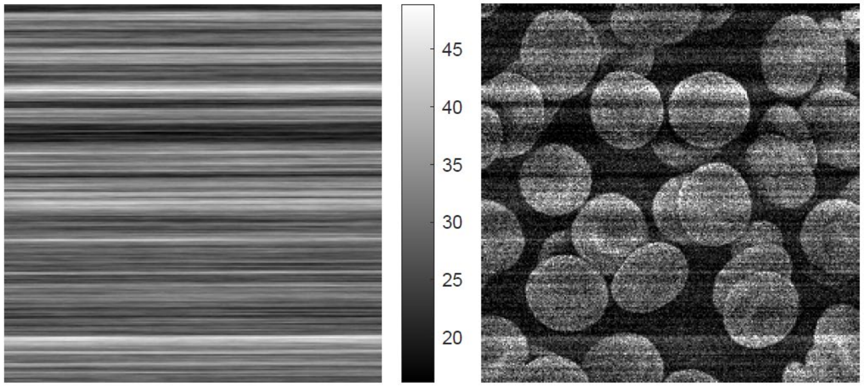
Mitigating beam current fluctuation

Beam current fluctuation

- Beam current lacks stability
  - Worse with contamination
  - Source tip ages
  - Alleviated by re-forming (baking) the trimer

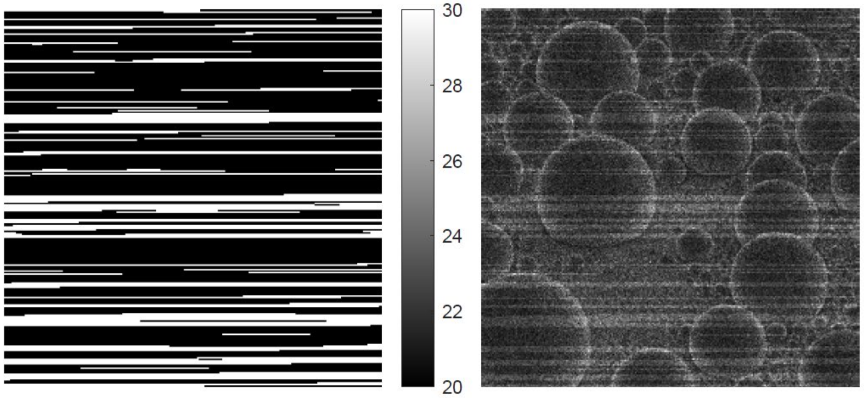
- Roughly:
  - Electron and helium ion beams have continuous fluctuation
  - Neon ion beams “flicker” between discrete values

- Existing techniques are post facto removal of horizontal stripe content without physical modeling



(a) Beam current incident on raster scanned sample in HIM

(b) Micrograph resulting from beam current in (a)



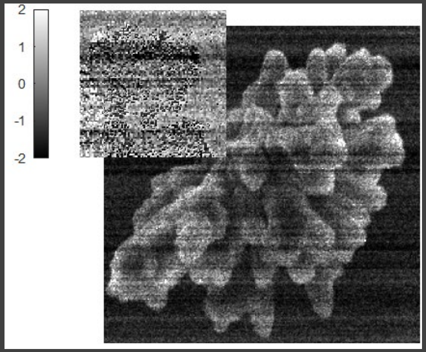
(c) Beam current incident on sample in Neon beam system

(d) Micrograph resulting from beam current in (c)

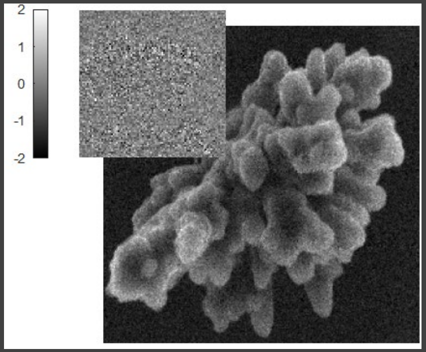
Synthetic examples of stripe artifacts in models for helium and neon beam microscopes.

Physics-based model + principled estimation

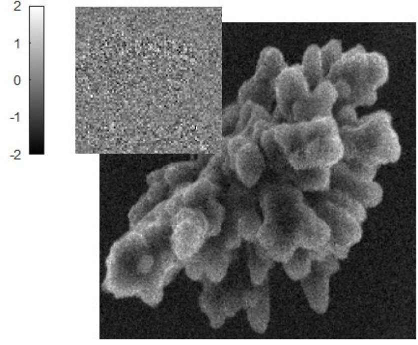
Continuous (helium ion beam-inspired)



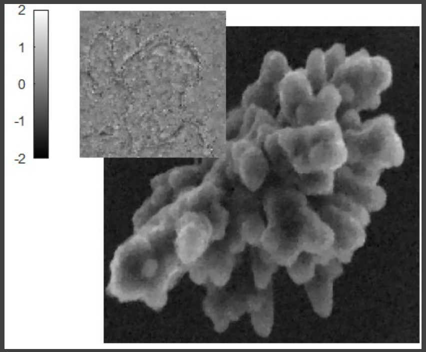
Conventional estimate
(RMSE: 1.1129)



TR ML “oracle”
(RMSE: 0.4874)

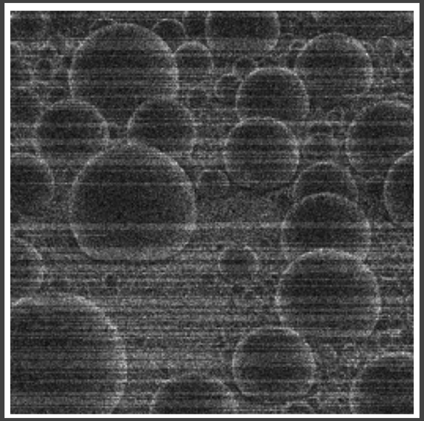


Joint estimation
(RMSE: 0.4979)

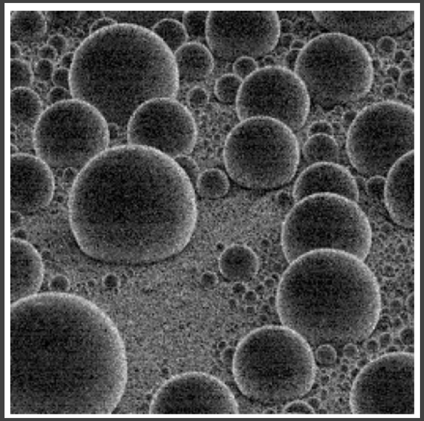


Joint estimation w/TV
(RMSE: 0.2296)

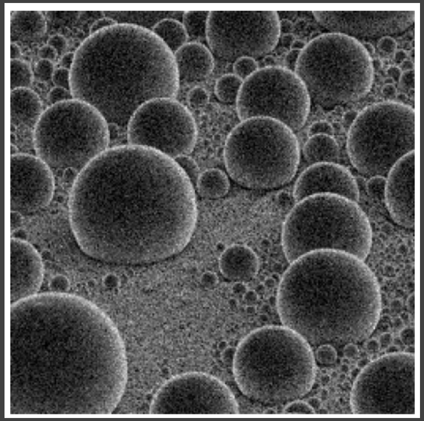
Discrete (neon ion beam-inspired)



Conventional estimate
(RMSE: 1.0689)



TR ML ”oracle”
(RMSE: 0.5147)



Hidden Markov chain estimation
(RMSE: 0.5149)

Seidel, Watkins, Peng, Agrawal, Yu & Goyal, *IEEE Trans. Computational Imaging*, 2022

Spatial regularization

Plug-and-play methods for particle beam microscopy

PnP methods combine a denoiser with maximizing a likelihood

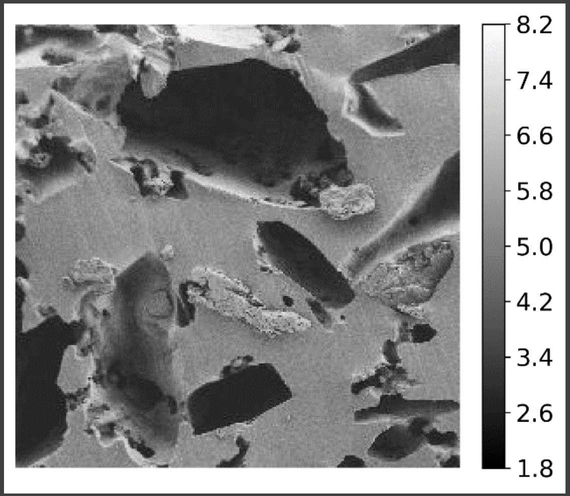
Need gradient or proximal operator of (troublesome) log likelihood

- Derived a simple approximation through Touchard polynomials

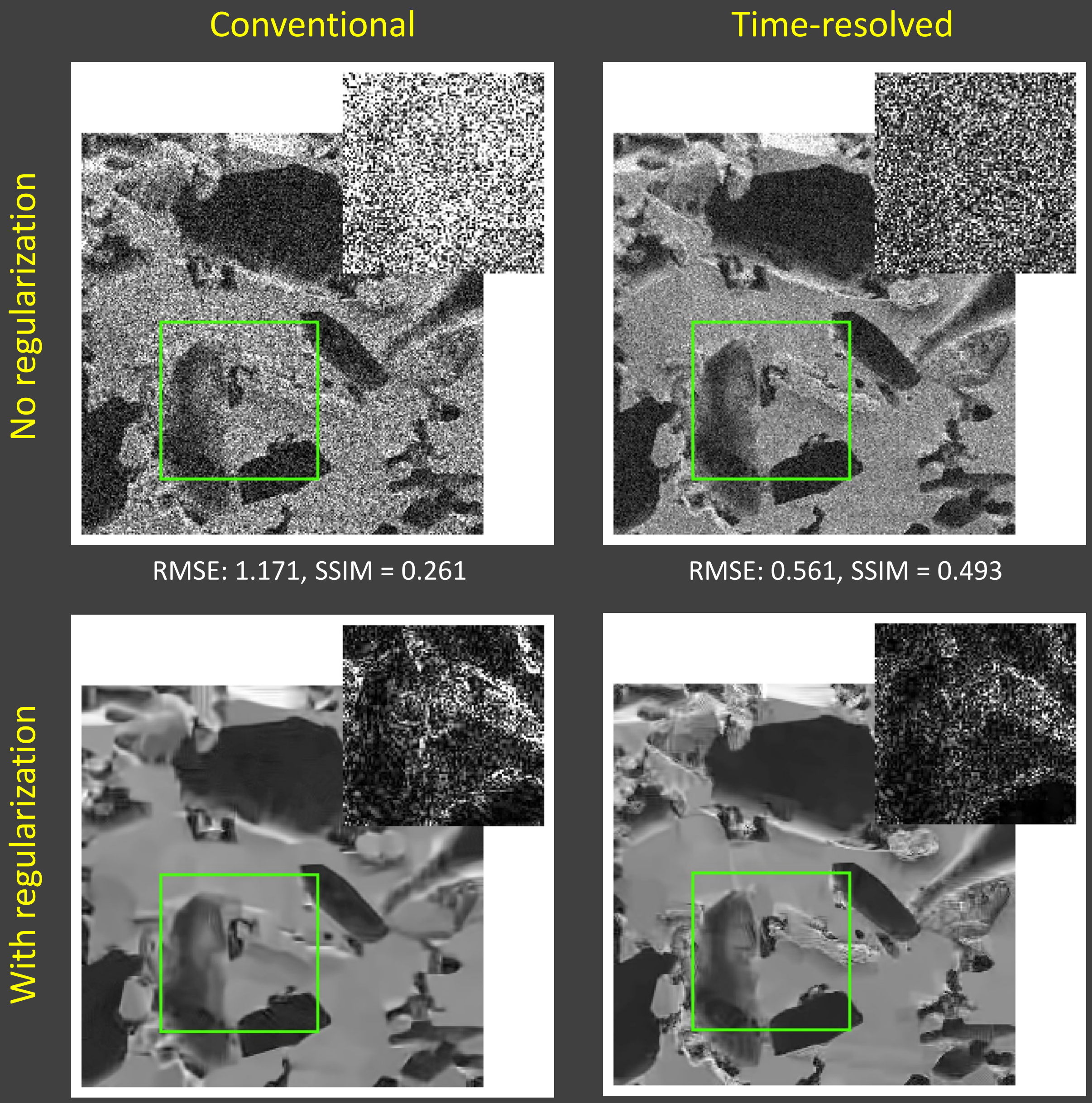
Peng, Kitichotkul, Seidel, Yu & Goyal, *IEEE Trans. Computational Imaging*, 2023

PnP FISTA with BM3D denoiser

Ground truth



$\lambda = 20$, insets are absolute error



RMSE: 1.171, SSIM = 0.261

RMSE: 0.561, SSIM = 0.493

RMSE: 0.363, SSIM = 0.675

RMSE: 0.265, SSIM = 0.776

Peng, Kitichotkul, Seidel, Yu & Goyal, *IEEE Trans. Computational Imaging*, 2023

Take-home messages

Time resolution finer than the pixel dwell time changes particle beam microscopy substantially:

- MSE lower roughly by factor of $\eta + 1$ (uniform across λ)
- Improves feature detection and resolution
- Mitigates beam current variation

Peng, Murray-Bruce, Berggren & Goyal, *Ultramicroscopy*, 2020
Peng, Murray-Bruce & Goyal, *IEEE Trans. Computational Imaging*, 2021
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