

## Supporting Information

### Code and Data

Datasets used in this study and video recordings of live imaging session using riCOM can be found online at the open data platform Zenodo:

authors: Chu-Ping Yu and Thomas Friedrich  
title: Real Time Integration Center of Mass (riCOM) Reconstruction for 4D-STEM  
doi: 10.5281/zenodo.5572123  
url: <https://doi.org/10.5281/zenodo.5572123>

The Source Code and precompiled binaries for Windows 10 and Ubuntu OS can be found on GitHub:

authors: Thomas Friedrich and Chu-Ping Yu  
title: riCOM\_cpp  
license: GNU GPL3  
url: [https://github.com/ThFriedrich/riCOM\\_cpp](https://github.com/ThFriedrich/riCOM_cpp)

### Derivations

To prove equation 2 holds, we demonstrate moving the gradient out of the cross correlation with this example. Here we assume three functions, a, b, and c, with the relationship

$$a(x, y) = b(x, y) \star c(x, y).$$

By taking Fourier transform on both side we have

$$A(\hat{x}, \hat{y}) = \bar{B}(\hat{x}, \hat{y}) \times C(\hat{x}, \hat{y}),$$

with which the equation can be rewritten with function a and the inverse Fourier transformed B and C

$$\begin{aligned} a(x, y) &= F^{-1}\{\bar{B}(\hat{x}, \hat{y}) \times C(\hat{x}, \hat{y})\} \\ &= \int \int \bar{B}(\hat{x}, \hat{y}) C(\hat{x}, \hat{y}) e^{ix\hat{x}} e^{iy\hat{y}} d\hat{x}d\hat{y}. \end{aligned}$$

Taking partial derivative of x from both side,

$$\begin{aligned} \frac{\partial a(x, y)}{\partial x} &= \frac{\partial}{\partial x} F^{-1}\{\bar{B}(\hat{x}, \hat{y}) \times C(\hat{x}, \hat{y})\} \\ &= \int \int \bar{B}(\hat{x}, \hat{y}) C(\hat{x}, \hat{y}) \frac{\partial e^{ix\hat{x}}}{\partial x} e^{iy\hat{y}} d\hat{x}d\hat{y} \\ &= i\hat{x} \int \int \bar{B}(\hat{x}, \hat{y}) C(\hat{x}, \hat{y}) e^{ix\hat{x}} e^{iy\hat{y}} d\hat{x}d\hat{y}. \end{aligned}$$

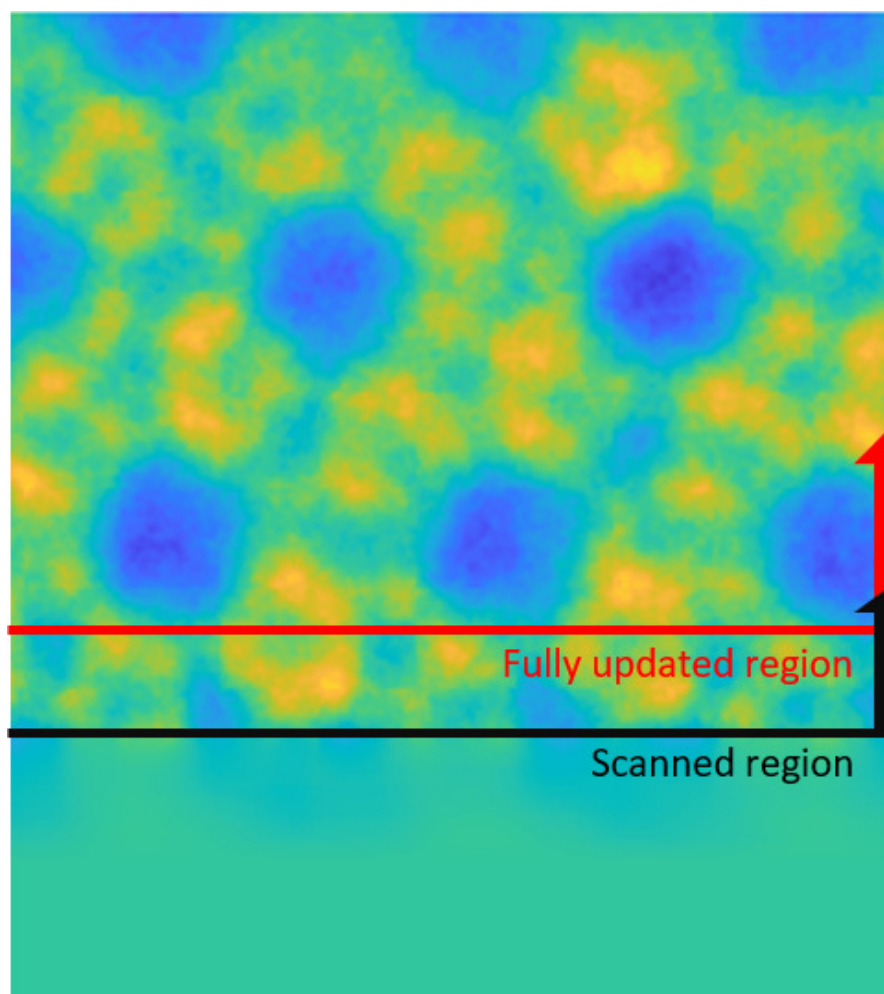
By putting  $i\hat{x}$  together with function C, and make use of the derivative rule of fourier transform, we have

$$\begin{aligned} \int \int \bar{B}(\hat{x}, \hat{y}) (i\hat{x} C(\hat{x}, \hat{y})) e^{ix\hat{x}} e^{iy\hat{y}} d\hat{x}d\hat{y} &= \int \int \bar{B}(\hat{x}, \hat{y}) F\left\{\frac{\partial c(x, y)}{\partial x}\right\} e^{ix\hat{x}} e^{iy\hat{y}} d\hat{x}d\hat{y} \\ \frac{\partial}{\partial x} F^{-1}\{\bar{B}(\hat{x}, \hat{y}) \times C(\hat{x}, \hat{y})\} &= F^{-1}\{\bar{B}(\hat{x}, \hat{y}) \times F\left\{\frac{\partial c(\hat{x}, \hat{y})}{\partial x}\right\}\} \\ \frac{\partial}{\partial x} (b \star c) &= b \star \frac{\partial c}{\partial x} \end{aligned}$$

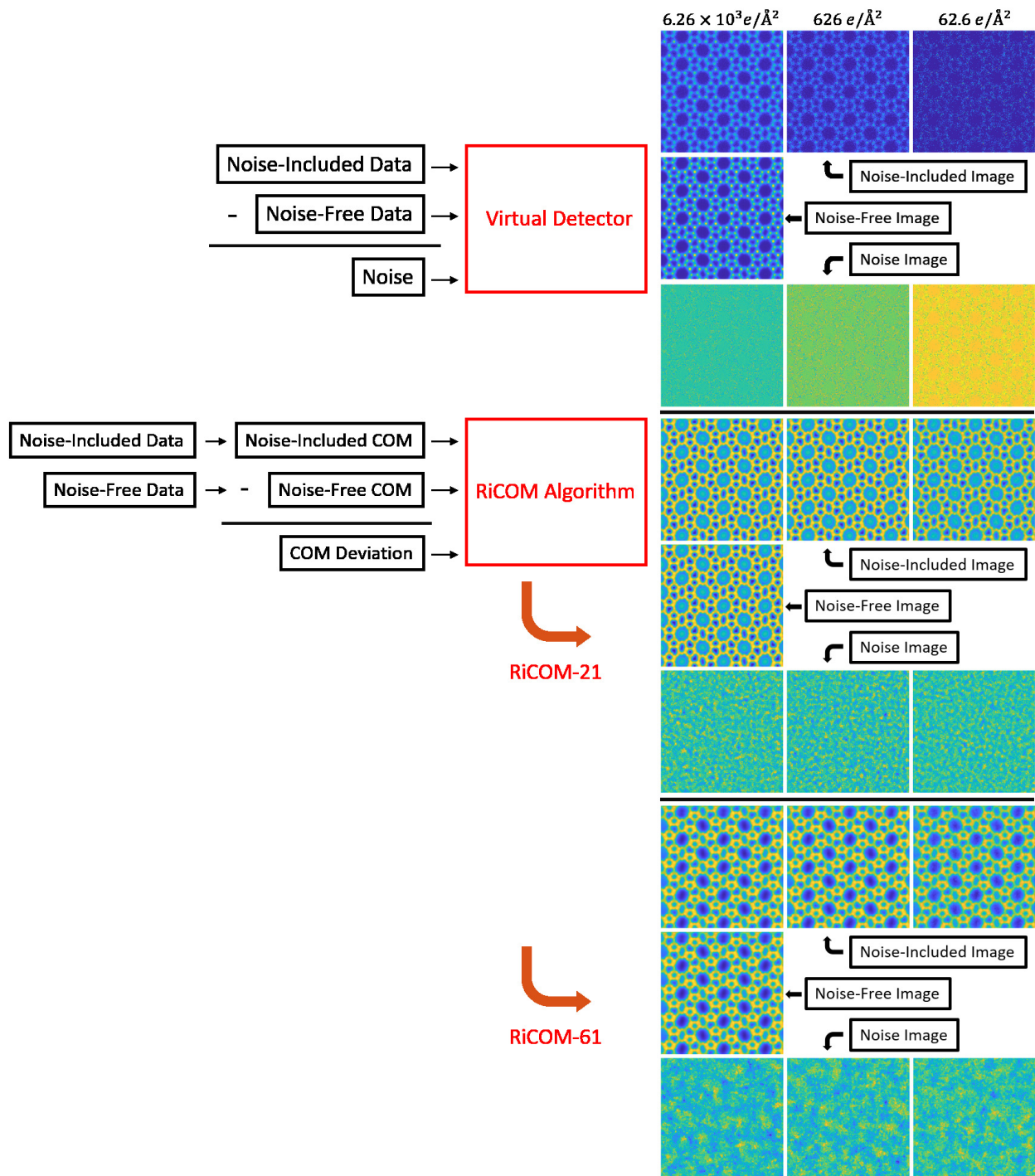
The gradient is represented as  $\nabla = \frac{\partial}{\partial x} \cdot \mathbf{n}_x + \frac{\partial}{\partial y} \cdot \mathbf{n}_y$ , where  $\mathbf{n}_x$  and  $\mathbf{n}_y$  are the unit vectors in both directions. This shows that we simply need to take another partial derivative with y to acquire the gradient of the function  $b \star c$ , and due to the associative property of cross correlation, this applies to the right side of the equation as well. So in the end we have

$$\nabla(b \star c) = b \star \nabla c.$$

*Additional Figures*



**Fig. S1.** A halfway reconstructed image. Scanning leads the full update of iCOM image by half of the kernel size, but the result shows that atomic features are already visible before the image is fully updated.



**Fig. S2.** A schematic of how noise is decomposed from the data and from the COM shift map. The resulting images from noise-included, noise-free data / COM are shown, as well as images reconstructed from noise / COM deviation.