

## Towards Low-dose and Fast 4-D Scanning Transmission Electron Microscopy: New Sampling and Reconstruction Approaches

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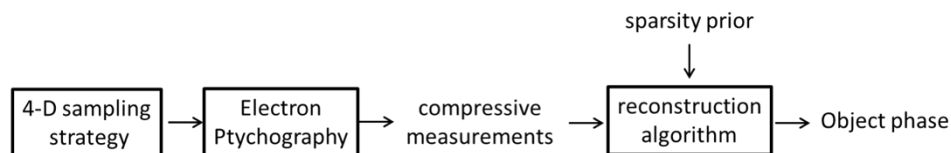
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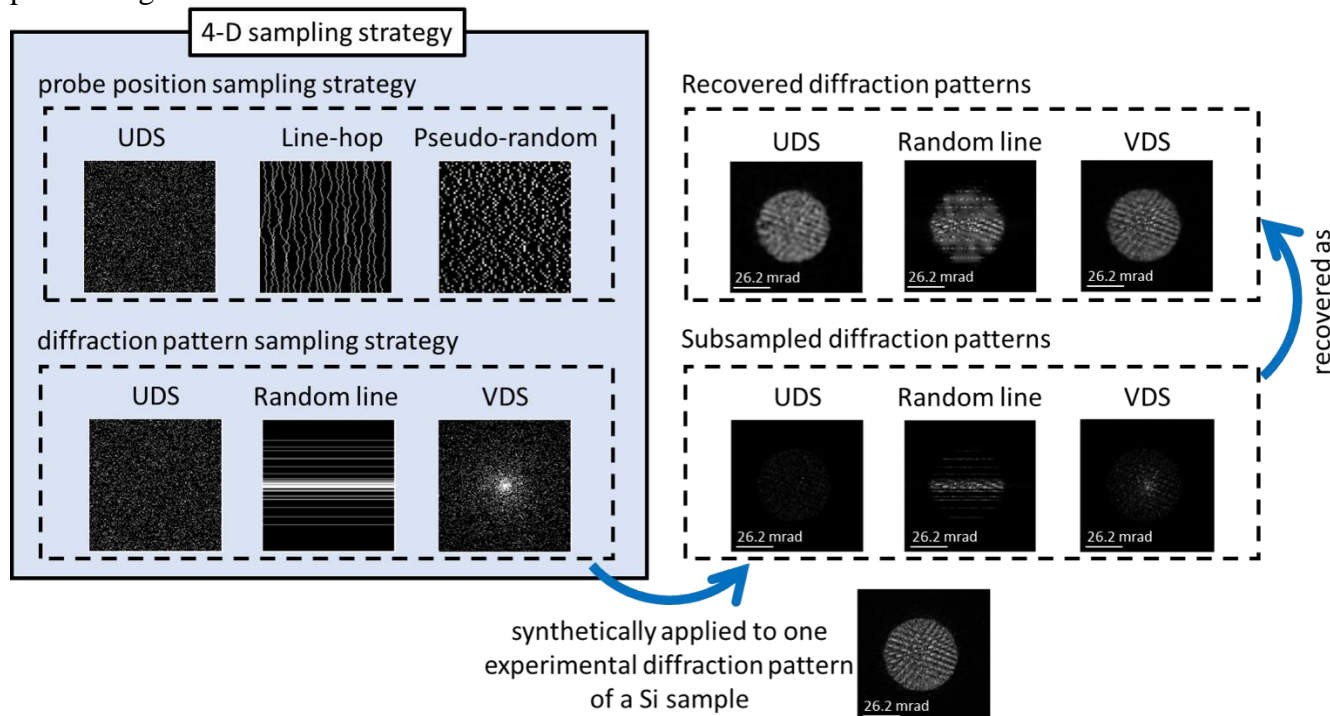
Four-dimensional Scanning Transmission Electron Microscopy (4-D STEM) relies on recording two-dimensional (2-D) diffraction patterns for every position of a convergent electron beam [1]. In combination with algorithms that solve the phase problem, a 4-D STEM dataset allows the reconstruction of a high-resolution object phase, by exploiting the high amount of redundancy in the data. However, the practical functionality of 4-D STEM is limited in many applications by the relatively slow scan speed imposed by the readout time of the detector (~1ms), longer than the typical dwell time (~10µs) in conventional STEM. One approach to circumvent this limitation is to bin the detector, at the cost of degrading the resolvable resolution in the reconstructed object phase. Another alternative is to increase the readout time of the detector and collect only binary counts [2]. A new strategy that followed from the successful application of the theory of Compressive Sensing (CS) to STEM [3], showed that a complete set of 4-D STEM measurements can be recovered from simulated incomplete data as if they were recorded by subsampling electron probe positions and the diffraction patterns [4]. A phase retrieval algorithm on the recovered 4-D STEM data can then be performed to obtain the object phase.

In this work, we use CS [5] to recover a 4-D STEM dataset by comparing efficient sampling strategies for joint subsampling of the probe positions and the diffraction patterns; and we directly convert these incomplete measurements to restore the object phase using a recovery algorithm. We highlight the importance of the sampling strategy [6] and show that the subsampling patterns generated following a Uniform Density Sampling (UDS) are unreliable. For the diffraction patterns we propose a Variable Density Sampling (VDS), inspired by the approaches in Magnetic resonance imaging [7] and Fourier transform interferometry [8], which is in favor of sampling low-frequency components; see an example in Figure 1. We introduce a pseudo-random sampling of the probe positions that prevents very short and long probe jumps to reduce issues of scanning coil hysteresis caused by large probe movements and potential beam damage due to densely populated probe positions [6]. For beam sensitive materials, this sampling strategy could help to reduce damage accumulation effects that are mediated by a diffusion process, which can be enhanced when neighboring positions are scanned [6]. Regarding the recovery algorithm for restoring the object from the 4-D STEM data, we leverage the sparsity of the phases (*i.e.*, an object phase image can be represented by only few elements of a proper dictionary, such as a dictionary of discrete Cosine basis or a learnable dictionary). Intuitively, at every iteration, the algorithm distinguishes the natural object phase from arbitrary noise-like phase shifts according to their sparsity level. This algorithm is robust to the decreased probe overlap quantity due to the subsampling.

In a nutshell, our results imply that the electron-dose in 4-D STEM can be directly reduced by subsampling the probe positions. Moreover, subsampling the diffraction patterns decreases the detector readout time and in turn further reduces the electron-dose [9].



**Figure 1. Block diagram of compressive 4-D STEM.** A 2-D sampling strategy controls the subsampling of the probe positions and another 2-D sampling strategy determines the subsampling of the diffraction patterns. Our reconstruction algorithm leverages, as a prior knowledge, the sparsity of the phase images.



**Figure 2.** Sampling patterns based on UDS results in poor recovered phase images, while a pseudo-random pattern for the probe position and a VDS of the diffraction patterns yields high quality images. A synthetic example on the right, advocates the importance of diffraction pattern subsampling method.

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