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Addressing Thickness Induced Contrast Reversals in Focused Probe Ptychography

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The annular dark field (ADF) signal in scanning transmission electron microscopy (STEM) has become an essential tool for materials science. Its atomic number contrast provides crucial information about which atoms are where. However light atoms near heavy elements can be hidden by the strong scattering of their neighbors. The need to locate light atoms with higher precision and reduced damage prompted the rise of annular bright field (ABF) imaging [1], and more recently methods such as integrated differential phase contrast (iDPC) and integrated center of mass (iCoM) imaging [2].

Compared to DPC segmented detectors, 4D STEM data collected on a pixelated detector can in principle provide a more precise measurement of the scattering center of mass. However the cameras used for 4D STEM have previously made it far slower than conventional detectors. Now, however, this bottleneck has been broken with the use of cameras such as the Timepix3 which provides 4D STEM at microsecond dwell times and beyond [3], removing the issue of drift for both iCoM and focused probe ptychography methods such as the single side band (SSB) [4] and Wigner distribution deconvolution (WDD) [5]. SSB and WDD are highly dose efficient and have been reported to clearly reveal light atoms, even when they are next to heavy elements [4-7]. However the theory used to explain the SSB and WDD methods usually relies on approximating the sample as thin and weak.

Here we investigate the ability of SSB and WDD ptychography to reach beyond their most simplistic theoretical descriptions and provide high quality images for typical materials science samples. Figure 1 displays experimental data from a $SrTiO_3$ sample well exceeding both the weak phase and the multiplicative approximations with an estimated thickness of 28 nm. Both the iCoM and the ptychography reveal the O columns significantly better than the ABF and show the complementary nature of the ADF and the 4D STEM signals. However, the ptychography simulated for such a sample in Figure 2 exhibits complex contrast reversals at zero defocus which could hamper interpretation. Fortunately applying a small defocus removes these reversals from the phase images while the ADF remains minimally affected. Such a small defocus could be present in the experimental data fortuitously and explain the clear appearance of the lattice in that data. The match between the experiment and the slightly defocused simulation is remarkable.

Figure 3 displays iCoM and SSB images of Fe_3O_4 extracted from a Timepix3 dataset with the focus optimized for ADF imaging. This data is of higher quality, displaying little if any drift compared to the Medipix3 data shown in Figure 1. Again, the ptychography shows the structure very clearly, including the light O columns next to the Fe columns. The ptychographic signal is also far clearer than the other STEM signals, including the iCoM, which we attribute to the much lower dose enabled by the Timepix3 and high dose efficiency of ptychography.

This illustrates an important point for the use of ptychography moving forward. Existing workflows for ADF imaging can continue and be combined with 4D STEM data taken simultaneously to obtain both Z-contrast and the locations of atoms hidden by either dose limitations or the strong scattering of nearby heavy atoms. With cameras such as the Timepix3 this can be performed without loss of scan speed, and if the defocus taken experimentally is not optimal for the ptychography, it can be adjusted via post collection defocus adjustment [5].

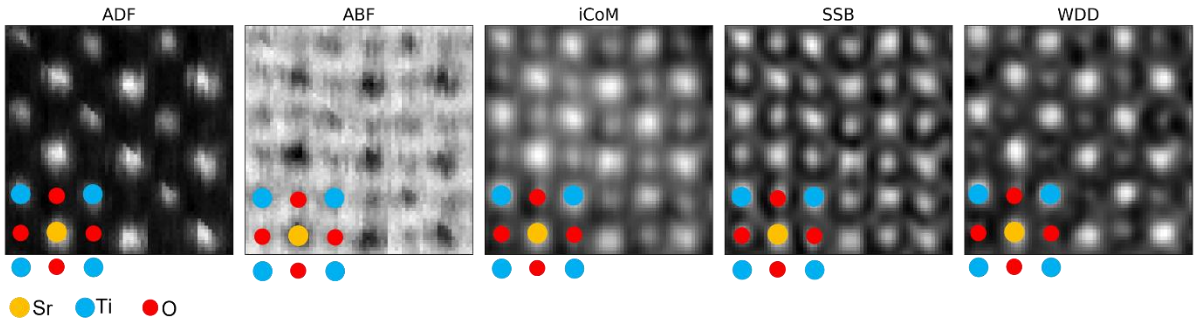

Figure 1. ADF and Medipix3 based 4D STEM data from $SrTiO_3$. iCoM and ptychography show the O columns more clearly than the ABF. However, this data shows some drift from the slow scan imposed by the camera.

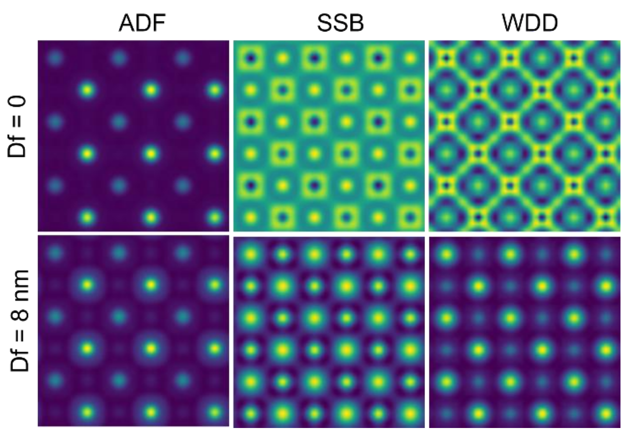

Figure 2. Multislice simulations for $SrTiO_3$ showing that although both SSB and WDD ptychography display complex contrast reversals at zero defocus for this 28 nm thick sample, by adjusting the defocus slightly the contrast reversals can be removed and all the atomic columns, including the O, clearly resolved.

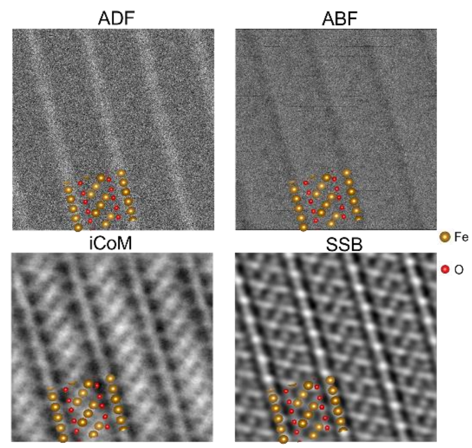

Figure 3. Comparison of ADF and Timepix3 based 4D STEM data of Fe_3O_4 showing the significantly stronger signal obtained from the ptychography, including for the O columns.

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