doi:10.1017/S1431927622009655
Microsc. Microanal. 28 (Suppl 1), 2022

Few-tilt Electron Ptychotomography: A New Method to Determine the 3D Structure of 2D Materials with High-precision and Low-dose

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Although atomic resolution imaging in scanning transmission electron microscopy (STEM) has become standard, greater effort is required when a three-dimensional (3D) structure of the specimen is desired. Tomography typically uses tilt-series with more than a hundred projections to extract a 3D structure. This requires the sample to be stable over long time periods and large doses which is problematic for beam sensitive samples such as 2D materials. These are often only stable for a couple of scans before structural changes occur, especially when one is interested in their defects [4].

Here, we present a new method which is capable of precisely determining the 3D structure of 2D materials with far lower doses [5]. As few as two tilts can provide a 3D structure at better than 30 picometer precision. This is possible because the position of each atom can be reliably tracked when the sample is sufficiently thin. We include simultaneously acquired single side band electron ptychography (SSB) in addition to the ADF signal (Fig. 1). To enable truly rapid scans, a microsecond dwell time capable pixelated camera is employed [6]. The ptychography delivers much higher contrast images, especially at low doses, which enables it to retrieve high quality images with less damage inflicted on the material. Crucially, it also enables accurate location of light elements even next to heavy elements as often occurs in newer 2D materials.

The method iteratively optimizes a 3D model of the structure so that image simulations based upon it best match the experimental data set across the whole few-tilt series. We show using simulations and experiments that the method reliably reconstructs different 2D materials with picometer precision. As a benchmarking material, we have used monolayer defective WS_2, which is difficult to analyse with ADF imaging alone because of the low intensity of S compared to W (Fig. 1). A quantification of the error of the optimized model reveals that the best precision reported previously [7] can be matched at a dose 30 times lower with the new method. Any remaining dose budget available before damage can be used to collect more projections and further increase the accuracy of the reconstruction. We applied the method to determine the 3D structure of the recently discovered monolayer hexagonal copper iodine (Fig. 2) [8]. The ptychography provides a clear signal of the Cu when the sample is tilted, which is not possible with the ADF alone, because the Cu signal is hidden in the dominant iodine signal. Therefore, using the ADF only in the reconstruction scheme leads to an unrealistic structure, but when using ADF and SSB together the resulting model excellently agrees with density functional theory (DFT) calculations [9].

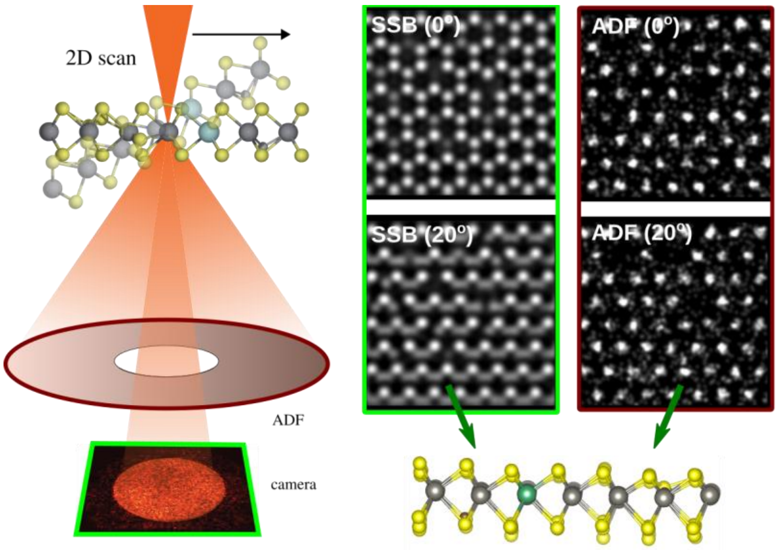

Figure 1. The experimental setup and benchmarking using simulated SSB and ADF data of WS_2 and the resulting picometer precision 3D structure.

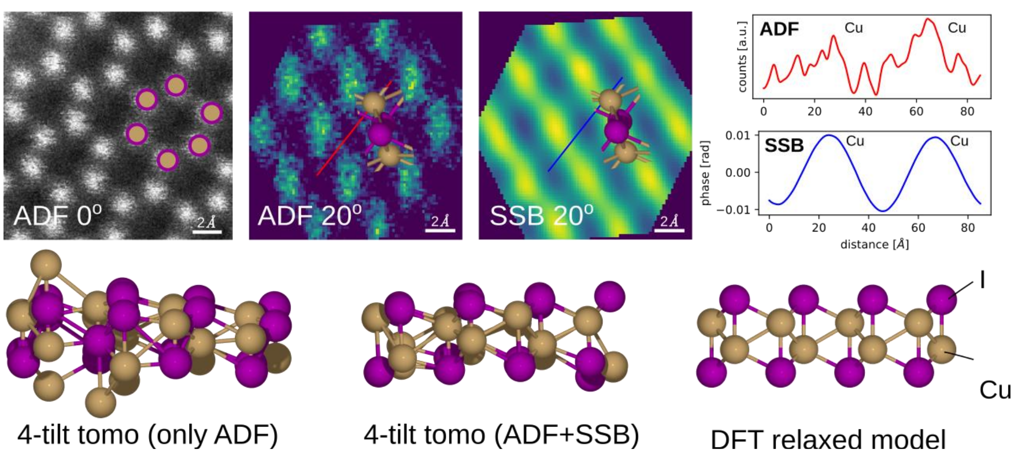

Figure 2. Experimental reconstruction of 2D CuI. Here, in total four tilt angles are used to determine the 3D structure. A line profile reveals that the Cu positions can be indeed resolved in the tilted projections.

References:

[1] O. Krivanek et al., Nature **464** (2010), p. 571-4. doi: 10.1038/nature08879.
[2] Q.M. Ramasse et al., Nano Letters **13** (2012), 10, p. 4989–4995, doi : 10.1021/nl304187e
[3] W. Zhou et al., Physical Review Letters **109** (2012), p. 206803, doi: 10.1103/PhysRevLett.109.206803
[4] S. de Graaf and B.J. Kooi, 2D Materials **9** (2022), p. 015009, doi: 10.1088/2053-1583/ac3377
[5] C. Hofer et al., arXiv:2108.04625 (2021)
[6] D. Jannis et al., Ultramicroscopy **233** (2022), p. 113434, doi: 10.1016/j.ultramic.2021.113423
[7] X. Tian et al., Nat. Mater. **19** (2020) p. 867–873. doi: 10.1038/s41563-020-0636-5
[8] K. Mustonen et al., Adv. Materials (2022) p. 2106922. doi: 10.1002/adma.202106922
[9] Research funded by European Research Council Grant no. 802123-HDEM.