High-precision low-dose 3D reconstruction of 2D materials via few-tilt electron ptychography

Abstract

From ripples to defects, edges and grain boundaries, the 3D atomic structure of 2D materials can drastically alter their properties. However, determining the 3D structure of 2D materials experimentally can be difficult. Conventional electron tomography requires images from a large number of tilt angles, necessitating many times the electron dose needed for a single micrograph. Such doses can alter structures before they can be determined. Here we provide an alternative route to 3D structure determination of 2D materials with a greatly reduced dose requirement. An event driven camera is used to enable rapid microsecond dwell time 4D scanning transmission electron microscopy (STEM), facilitating simultaneous annular dark field (ADF) and ptychographic imaging with minimal dose and drift. Crucially this combination of imaging modalities allows us to determine the projected positions of all the atoms in arbitrary 2D materials at each tilt angle. With this data, the 3D structures can be solved from very few tilts indeed. While as few as two tilts can be used, our low dose experimental setup and the efficiency of ptychography enables us to use several more tilt angles without exceeding our dose budget. This can be vital to providing picometer precision reconstructions of more complex structures. We demonstrate the technique with simulations for WS$_2$ and experimental data from a new 2D material, hexagonal CuI, both of which have complex layered structures containing both heavy and light elements.

Keywords: Electron ptychography · 2D materials · 3D reconstruction

1 Introduction

2D materials have garnered huge attention since the first isolation of graphene \[1\] thanks to the unusual properties that have emerged from their single atomic or a single unit cell thickness. These properties justify them as promising candidates for electronic and optoelectronic applications, energy storage devices, ultra-sensitive detectors and many other applications \[2, 3\]. However, they depend upon the precise 3D arrangement of their constituent atoms. For example, the long-range ripples in graphene have been shown to be important to its stability using electron diffraction \[4\]. Dopants in 2D materials also buckle the local environment, drastically changing their electronic properties \[5, 6, 7, 8, 9\]. However, 3D structure determination using conventional electron tomography \[10, 11\] can be difficult to apply to 2D materials which typically undergo structural changes during the large electron doses required for lengthy tilt series \[12, 13, 14\].

Recently, Hofer et. al demonstrated the 3D structure determination of defects in graphene using as few as two tilt angles with ADF STEM \[15\], representing a very large decrease in both the number of required tilt angles and electron dose. The method utilizes the fact that all the atoms in graphene remain visible after tilting, allowing a model to be iteratively updated such that image simulations based on it best match those from the experiment at each tilt angle. Many newer
2D materials such as the transition metal dichalcogenides (TMDs) are however more complicated in that they are both several atomic layers thick and contain elements which are more widely spaced in atomic number. The difference in the atomic number of the constituent elements represents a challenge because light elements are often obscured by the strong scattering of nearby heavy elements in ADF imaging. For such a 2D TMD, Tian et. al very recently demonstrated the 3D structure determination of Re doped MoS$_2$ with picometre precision using 13 tilt angles of ADF imaging [16]. While 13 projections is much reduced number of tilt angles compared to conventional tomography, it is still far larger than the two required for graphene and a method to further reduce the required number of tilts for arbitrary 2D materials has remained desirable.

Here we introduce a new form of few tilt tomography which is made applicable to arbitrary 2D materials with the addition of simultaneous 4D STEM imaging alongside the ADF imaging. The high dose efficiency of ptychographic methods based on 4D STEM, such as the single side band (SSB) method [17] used herein, complements the Z-contrast of the ADF imaging. The idea of using ptychographic-based tomography has been proposed before [18] and has the potential to significantly reduce the dose even when using a full tomographic series. While the Z-contrast remains useful for elemental identification, the ptychographic images provide clearer images of the atomic structure at low doses. The ptychographic signal is especially useful for locating light atoms next to heavy elements which can be impossible in low dose ADF images. We demonstrate that this enables a full 3D structure determination with as little as two tilt angles with simulations of defected WS$_2$ despite the greater difference in atomic number between W and S compared to Mo and S in MoS$_2$. We also show how the precision can be improved when using more tilt angles. Experimentally we demonstrate the method with its application to 2D CuI, a feat facilitated by the use of an event driven camera that provides 4D STEM data without any reduction of speed compared to normal fast microsecond dwell time ADF imaging, allowing us to minimize both the sample stage drift and electron dose simultaneously [19].
Figure 2: **Progress of the algorithm using one tilted projection as an example** (a) Quantitative SSB simulation of a tilted WS$_2$ model, which is the target image. Below is the model used for that. (b) Simulation of initialized model (model shown below). (c) Simulation of optimized model (model shown below). (d) Line profile of the target image, the simulation of the initial model and the optimized model, respectively. (e) Correlation as a function of iteration for three different projections. All images are at a dose of 4000 e$^-\text{Å}^{-2}$.

2 Results and Discussion

2.1 Simulation example: WS$_2$

We first introduce the method based on a monolayer of WS$_2$. As a numerical data set, we have simulated three different projections of ADF and CBED images with an electron dose of 4000 e$^-\text{Å}^{-2}$. We do not include aberrations as we expect that dose is the major limitation of the resolution rather than lens errors when using an aberration-corrected instrument and residual aberrations can also be corrected by electron ptychography [20, 21]. To increase the complexity of the system, we have inserted defects including different types of vacancies and substitutional C and Nb atoms into the WS$_2$ model as seen in Fig. 1. In addition, Ångstrom scale translations were used in the image simulations at each tilt angle, to imitate the shifts that occur when tilting experimentally. Images were simulated at zero tilt, at a 20$^\circ$ tilt in one tilt axis and at a 20$^\circ$ tilt in the tilt axis perpendicular to the first axis.

The first task is to initialize a model which contains all atoms in the experimental data set with the correct lateral (x,y) positions and the correct elemental identification. This model can be gathered from the non-tilted image, as the projected positional assignment is routine and the corresponding atomic intensities can be correlated to the atomic
elements. We modify the previously reported optimization algorithm for this task [22], but other methods can also be considered [23] [24]. Histograms of the extracted atomic scattering intensities and integrated cross-sections for the phase image can be found in Fig. 1b. The histograms show two peaks for the integrated scattering cross-sections (ISCS) and three peaks for the integrated phase cross-sections (IPCS). Since the phase image also contains a negative contribution, the squared phase could be considered for the analysis [25]. However, we abstain from taking the square since our analysis already reliably reproduces the chemical elements from the raw SSB image. The ptychographic phase and ADF signals compensate each others’ weaknesses for elemental characterization (cf. Fig. 1a). While the ADF easily distinguishes the W and the S$_2$ site, the signal is not sufficient to directly extract single S vacancies and other light elements from the S$_2$ site at this dose. The SSB image is capable of distinguishing the lighter elements such as S, S$_2$ and C, despite their neighboring heavy W sites. For this analysis, we consider the ISCS and IPSC as values which are capable of distinguishing the a priori known elements. Note that the exact element has to be known and Nb cannot be distinguished from S$_2$ either by ADF (due to the very similar Z contrast) or SSB, but still can be unambiguously assigned as it is positioned in the W-lattice site. Under very high doses, ADF imaging would in principle be able to distinguish these elements, but we want to avoid high electron exposures here as we want to spend the dose budget for different beam incident angle acquisitions and generally avoid damaging the structure.

Once the lateral positions, elements and occupations are assigned based on the ADF and SSB images at zero tilt angle, a model can be initialized. As at this point the z-coordinate is unknown to the reconstruction algorithm, such a model could be flat or use prior knowledge such as the expectation that the S atoms in the S$_2$ sites are displaced in z from each other such as the $z = 1.5$ Å offset we used here. The model is then optimized using the data from the other tilt angles. In detail, the correlation function to be maximized is

$$R_{\text{tot}} = \sum_{V} \sum_{m=\text{adf,ssb}} \frac{1}{N} \sum_{i=1}^{N} \frac{\mu_{\text{sim},m,V}^i - \mu_{\text{exp},m,V}^i}{\sigma_{\text{sim},m,V}^i} \frac{I_{\text{exp},m,V}^i - I_{\text{sim},m,V}^i}{N - 1},$$

where the sum runs over all $N$ probe positions in the scan, and $I_{\text{exp},m,V}^i$ and $I_{\text{sim},m,V}^i$ are the intensities of the $i$th pixel of the experimental data and the numerical simulation of a certain imaging method $m$ (ADF or SSB) in a certain view $V$, respectively. $\mu_{\text{exp},m,V}^i$ and $\mu_{\text{sim},m,V}^i$ are the corresponding mean values of the experimental and the simulated images, respectively and $\sigma_{\text{exp},m,V}^i$ and $\sigma_{\text{sim},m,V}^i$ are the corresponding standard deviations of the experimental and simulated images, respectively. This function is maximized based on a method using quadratic interpolation: Here, the gradients in each direction are calculated by a finite difference method and the minimum of a quadratic fit is estimated and used as the next iteration value. Other optimization methods such as gradient descent were also found to work, but are significantly slower.

The first sum, which must be maximized, can be regarded as the correlation between the simulated and experimental data of a specific method ($m$), either SSB or ADF, in a certain view ($V$). The whole $R_{\text{tot}}$ can be considered to be the sum of all correlations. We further find that only optimizing the 3D positions of the model is not sufficient to obtain the best match. Instead, source-size broadening, linear drift, tilt axis and the variation of atomic scattering factors must all be optimized.

For the simulations during the optimization procedure, the convolution method is chosen as it is computationally efficient [26]. This fast method works extremely well for the qualitative simulation of ADF images of very thin materials, but it has the major drawback that contrast reversals (e.g. negative phase at the center of the hexagons) as they appear in phase images cannot be reproduced as the convolution method always results in positive values. Nevertheless, it shows that the structural optimization indeed leads to a reliable match. This match is not expected to be perfect, as the difference image shows residual features rather than noise (see Supplementary Figure 1) but is still sufficient for a proper quantification of the chemical elements and assignment of the projected positions. We also want to point out that under these low-dose conditions, the SSB image is more suitable for the refinement of the atomic positions than the ADF image as it has a significantly better signal-to-noise ratio. The extracted positions are adopted for the ADF image, where only the atomic scattering factors were included in the optimization process.

To illustrate the principle of the method, we show its progression using a 20 degrees tilt angle in Fig. 2. Fig. 2a shows the target SSB image and the target model from which it is simulated, with the model viewed side on. This target SSB image is also input to the algorithm as the target to match and would normally be the experimental image from the microscope with the model unknown. Fig. 2b shows the SSB image based on the initialized model simulated at the same tilt angle. Again, the model has been initialized to match the lateral positions observed from zero tilt, but the z coordinates are set arbitrarily. The incorrect z coordinates result in a significant mismatch between the SSB image from the initialized model and that of the target. Fig. 2c shows the result of the optimization on the model and the SSB image simulated based on it. Line traces from the SSB images taken along the armchair direction of the lattice are plotted in
Few-tilt electron ptychography

Figure 3: Histogram of absolute distances between the corresponding atoms of the input model (WS$_2$) and the optimized model using two, three and 13 tilts, respectively. Each (tilted) projection is based on an electron dose of 4000 e$^{-}\text{Å}^{-2}$. The 13 tilts range from 5–30 degrees in both tilt axis.

part d of the figure, and the correlation as a function of iteration number is plotted in panel f. The correlation converges to a maximum value of 0.82 in the optimized state, demonstrating a good match between target and optimized image. The converged value of the correlation coefficient depends on the quality of the images, for instance how noisy they are. We suggest that the correlation coefficient of the non-tilted projection can be considered as a target value, since these projected positions and simulation parameters are already refined for the initialization of the structure (cf. Fig. 2).

Also note that all defects and substitutions have the correct assigned positions.

The pre-defined target model allows us to quantify the accuracy and precision of the method by calculating the difference between the target model and the optimized model. For this task, we aligned both models and calculated the absolute difference between the corresponding atoms. These values are summarized in Fig. 3 showing a histogram of the differences. The standard deviation is 23 pm, 16 pm and 3 pm using two, three and 13 tilts, respectively. This demonstrates that picometer precision is achievable with very few tilts with an aberration-corrected instrument with total electron doses below those typical for imaging 2D materials. We can also estimate the accuracy of the reconstruction. For this task, we calculated the z distance between the terminating sulfur planes. The values agree within 33 pm, 11 pm and 4 pm for two, three and 13 tilts, respectively.

2.2 Experimental example: 2D CuI

We now apply the method to an experimental data set from a monolayer of hexagonal CuI. The 2D form of this material has been very recently discovered and is stabilized between two graphene layers. A detailed report about this novel material will be published in the near future by Mustonen and Hofer et. al. Structurally, it is equivalent to a single layer of $\beta$-CuI, in which both Cu and I occupy both sublattice sites [27]. Therefore, ADF STEM imaging reveals the honeycomb lattice with equal intensities in both sublattice sites as shown in Fig. 4a at a dose of approx. 1 $\times$ 10$^6$ e$^{-}\text{Å}^{-2}$.

In order to reconstruct the full 3D model, the method as described above is applied. To reduce the dose imposed on the region of CuI used for the reconstruction we performed the few tilt series in a new unirradiated region. We first acquired a series of 10 scans at 0 tilt at a 2 microsecond dwell time at a dose of 5 $\times$ 10$^3$ e$^{-}\text{Å}^{-2}$ per scan. The simultaneous recording of ADF and 4D STEM data at this speed was made possible by our event driven Timepix3 detector. Importantly, the rapid scan speed greatly facilitates the low dose operation and greatly reduces the effects of drift in each scan. We then build up signal by summing the drift corrected scans to obtain ADF and SSB images at 5 $\times$ 10$^4$ e$^{-}\text{Å}^{-2}$. The lateral positions for the initial model were determined from the SSB image, and are shown in the
Figure 4: **Characterization of 2D CuI.** a) A high, $1 \times 10^6 \, \text{e}^- \, \text{Å}^{-2}$, dose ADF STEM image of 2D CuI taken at zero tilt angle with the initial positions used in the model input to the algorithm overlayed with I in purple and Cu in copper. The model is initially flat. Simultaneous b) ADF and c) SSB images taken from a 20° tilt angle using a drift corrected series of 10 rapid 2 microsecond dwell time 4D STEM scans with a total dose of $5 \times 10^4 \, \text{e}^- \, \text{Å}^{-2}$). The model obtained from the 4 tilt tomography using both ADF and SSB inputs is shown at the same tilt in panel d, illustrating how the tilt makes all the atoms visible in projection. Line profiles along the red lines overlaid on the tilted ADF and SSB images are shown in e) and f). g) the 3D structures from 4-tilt tomography using just the ADF signal and from the combined ADF and SSB data are visualized side on alongside the results of independent DFT relaxation.

Despite the much lower dose of the SSB image, these positions can be seen to fit the lattice observed in the high dose ADF image in the figure.

The model is initialized flat, without any height variation in the atomic positions. To obtain the out of plane positions we used data from the same region at specimen tilts of $\alpha = 20^\circ$, $\alpha = 30^\circ$, and $\beta = 15^\circ$ using the same 2 microsecond dwell time scan series plus drift correction method. Tilting shifts the atoms that are aligned in columns in the untilted projection laterally in the tilted projections according to their relative heights. Tracking these shifts as a function of tilt angle is what allows us to determine the 3D geometry. However due to the high atomic number of iodine compared to Cu, it is not possible to clearly reveal the Cu atoms in the tilted ADF projections at this dose. An example is shown in Fig. 4b. Without clear Cu positions the 3D reconstruction cannot be accurate. The ptychography however provides a much stronger signal from which the projected positions of all the atoms can be more reliably extracted, as illustrated in the SSB image shown in Fig. 4c.
The optimized model is overlaid on the same SSB image in 4d, illustrating how the tilt shifts the projected positions of the atoms laterally. Despite the signal from the Cu atoms not being fully separated from that of the I in the images, the well resolved elongated shape of the tilted columns is nonetheless well put to use by the few tilt algorithm because it maximizes the correlation between the simulated and experimental images which does not require a strict resolution of all atoms in all projections. However the strength of the signal from the atoms is paramount. Line traces from along the red lines in Fig. 4b and c are shown in e and f. The strong smooth signal from the SSB ptychography is far easier to fit the atomic positions to than the noisy ADF image, and allows the algorithm to correctly identify the projected positions of even partially overlapping atomic positions.

The value of the ptychographic imaging to the few tilt reconstruction is highlighted by comparing the models produced by the algorithm using four tilts with only the ADF images and with including both the ADF and SSB images to the DFT relaxed model. These are shown viewed from the same side on vantage point in Fig. 4g. Because of its relatively poor signal the ADF only reconstruction is far more disordered than that of the combined SSB and ADF reconstruction, which is a much closer match to the DFT model even though the two reconstructions used exactly the same dose, being from exactly the same set of scans.

Figure 5: Analysing the precision of the reconstruction of CuI. a) Histogram of optimized z positions of iodine and copper (left and right, respectively) using both ADF and SSB and only ADF (top and bottom, respectively). The corresponding atomic models are schematically shown as insets to the plots. b) Histogram of the distances between iodine and Cu in the same lattice site using both ADF and SSB images for each projection (left) and only the ADF images (right) respectively.

We now further analyse the precision of the experimental reconstructions and quantitatively demonstrate the utility of electron ptychography for accurate reconstructions at low doses. Because the models are purely based on experimental data, the real structure is not known. However, the z distribution of atomic heights reveal two separate peaks for the iodine and copper atoms representing the average height of both sublattices as seen in Fig. 5. Their mean values excellently agree with DFT calculations (dashed lines in Fig. 5) when using both ADF and SSB together for the reconstruction. The standard deviations of the distribution of absolute differences are calculated to be 22 pm and 21 pm for iodine and copper, respectively. The (x,y) coordinates are neglected by this method, justified by the much higher precision of determining the projected (x,y) positions using STEM. The precision of the experimental reconstruction is slightly less than would seem indicated by the WS₂ simulations. This discrepancy can be explained by the presence of residual non-linear drift, scan distortions, the finite accuracy of the tilting holder and out-of-plane atomic vibrations.

When excluding the SSB images, the reconstructed iodine positions not only have a broader distribution with a standard deviation of 37 pm, but the distance between both planes is also smaller than expected from DFT calculations. We assume that this error comes from the convolution with the incorrectly assigned Cu intensities. The precision of the Cu positions is, as expected, much poorer still with a standard deviation of 62 pm.
We also analysed the distances between the Cu and I of the same lattice sites of both optimized models (using only ADF and ADF and SSB, respectively). Also here, the combination of SSB and ADF images produces the most reliable reconstruction with a precision of 27 pm which agrees with our previous analysis. Note that the precision is slightly poorer than that calculated for single atoms as a result of the propagation of uncertainty when calculating the distance between two atoms. Excluding the SSB in the optimization drastically degrades the precision to 73 pm.

A very similar algorithm in which only ADF images were used has been successfully applied to atomically thin materials to reveal out-of-plane distortions of graphene defects and graphene distortions in a heterostructure [15, 28, 29]. However, since CuI is far from being only a single-atomic layer thick, it can be speculated that the number of ADF images can be increased in order to get a more reliable reconstruction. This, however, has the major drawback of a higher dose requirement and secondly, a very high signal-to-noise ratio is required in order to reveal the contrast of Cu. As we have shown, electron ptychography is an excellent method to overcome those issues: The quality of the reconstructed images are significantly improved and it reveals more elements such as the projected positions of the light elements.

The analysis can be further extended to the refinement of the 3D atomic positions by only using the phase image, since the SSB image has usually a significantly better signal-to-noise ratio than ADF images. Therefore, one might expect that including the latter might lead to a less accurate reconstruction. Our empirical attempts, however, show that the exclusion of ADF leads to a poorer structural refinement. We assume that the high intensity of I over-compensates the noisy Cu signal in the ADF images as noise does have a high contribution to the change of the correlation function.

3 Conclusion

We have developed and experimentally demonstrated a new few-tilt tomographic approach that combines ADF and ptychographic imaging to enhance our ability to solve the 3D structure of general 2D materials. The addition of the SSB ptychography provides greater dose efficiency images that allow a more accurate and precise determination of atomic positions at significantly lower doses than with ADF imaging alone. Importantly, the ptychography also provides far greater sensitivity to light atoms near heavy elements that would be hidden in the ADF signal. With the greater ability to detect all the atoms more efficiently, we require fewer tilt angles to accurately determine the height of each atom. From simulations, it is clear that accurate results can be obtained from very few tilt angles indeed with this method, even for 2D materials that are several atomic layers thick, further facilitating the use of minimal dose. We further showed that although as few as two tilt angles can provide an accurate reconstruction for TMDs such as WS2, increasing the number of tilt angles can increase the precision if the material can withstand the additional dose. Experimentally we demonstrated the technique with the just discovered 2D hCuI which has an even more complicated four atomic layer thick structure. With a total of four tilt angles we achieve a 3D reconstruction that is in excellent agreement with the results of DFT with our new method, which we also show was not possible with ADF imaging alone. Crucial to the experimental precision was our use of an event driven camera which enabled us to perform multiple scans at microsecond dwell time which were then drift corrected and summed at each tilt to minimize drift. The results of the work opens up a new route for precise and accurate determination of the complete 3D atomic network of thin and beam sensitive materials. The method will be especially useful when imaging structures such as defects in 2D materials, which are usually only stable for a few scans until structural changes occur.

Methods

Electron microscopy

Microsecond dwell time electron ptychography was conducted using a fast, event-driven Timepix3 camera in a probe-corrected FEI Themis Z instrument with a probe convergence angle of 30 mrad and a beam current of ca. 1 pA. 10 scan frames of $1024 \times 1024$ probe positions were acquired sequentially at each tilt angle ($0^\circ, \alpha = 20^\circ, \alpha = 30^\circ$ and $\beta = 15^\circ$) with a dwell time of 2 μs. All 10 data sets were processed by the SSB method and the final images were aligned and averaged using non-rigid registration. The averaged images where further sequenced into smaller images and registered in order to improve the signal-to-noise and reduce the size of the reconstructed model. The algorithms for the SSB and the 3D reconstruction written in python can be found on gitlab.

STEM simulations

STEM image multi-slice simulation were carried out by the PyQSTEM Package. The electron energy was chosen to be 60 kV, the convergence angle 30 mrad and the step size approx. 18 pm per pixel. The number of slices is 3 for the normal incident image, but increased up to 30 for the tilted structures.
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4 Supplementary information

4.1 Match between convolution simulation and SSB image

A convolution between the intensity profile of an electron probe and the potential of the structure can excellently reproduce STEM images of 2D materials as described in the main text. The contrast mechanism of phase images, as the SSB electron ptychography produces, are more complicated to simulate efficiently. However, the atomic positions and intensities are still well recovered by the optimization as SFig.1 shows. Here, the difference between the target image and the optimized simulation still shows a pattern as a result of contrast reversals of the phase image. More accurate simulations methods will be investigated in a future work.

Supplementary Figure 1: Target image of WS$_2$, simulation of optimized model and pixel-by-pixel difference.

4.2 Complete few-tilt series

SFig.2 shows the simulated ADF and SSB images used for the 3D reconstruction of WS$_2$. The dose for each image is $4000 \text{ e}^\text{-} \text{Å}^{-2}$.

SFig.3 shows the ADF and SSB images used for the 3D reconstruction of CuI. The dose of each image is estimated to be approx. $20 000 \text{ e}^\text{-} \text{Å}^{-2}$. This is indeed sufficient to obtain a decent signal-to-noise image of both methods.
Supplementary Figure 2: **Three-tilt tomographic series of WS$_2$.** Simulated ADF (top) and SSB phase images (bottom) for different tilt angles. The simulation parameters are described in the methods part of the main text of the manuscript.

Supplementary Figure 3: **Four-tilt tomographic series of CuI.** ADF (top) and SSB phase images (bottom) for different tilt angles which are used for the reconstruction of the model. The acquisition parameters are described in the methods part of the main text of the manuscript.