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Authors

Chang, Dillan J

Kim, Dennis S

Rana, Arjun

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Ptychographic Atomic Electron Tomography:

Towards 3D Imaging of Individual Light Atoms in Materials

Dillan J. Chang¹, Dennis S. Kim¹, Arjun Rana¹, Xuezheng Tian¹, Jihan Zhou¹, Peter Ercius² and

Jianwei Miao¹

¹Department of Physics & Astronomy and California NanoSystems Institute, University of

California, Los Angeles, CA 90095, USA.

²National Center for Electron Microscopy, Molecular Foundry, Lawrence Berkeley National

Laboratory, Berkeley, CA 94720, USA.

Through numerical simulations, we demonstrate the combination of ptychography and atomic electron tomography as an effective method for low dose imaging of individual low-Z atoms in three-dimensions. After generating noisy diffraction patterns with multislice simulation of an aberration-corrected scanning transmission electron microscope through a 5 nm zinc oxide nanoparticle, we have achieved 3D imaging of individual zinc and oxygen atoms and their defects by performing tomography on ptychographic projections. The methodology has also been simulated in 2D materials, resolving individual sulfur atoms in vertical WS₂/WSe₂ van der Waals heterostructure with a low total electron dose where annular-dark-field images fail to resolve. We envision that the development of this method could be instrumental in studying the precise 3D atomic structures of radiation sensitive systems and low-Z atomic structures such as 2D heterostructures, catalysts, functional oxides and glasses.

Atomic Electron Tomography (AET) determines 3D coordinates of individual atoms in materials without assuming crystallinity by combining high resolution tomographic tilt series from

23 scanning transmission electron microscopes (STEM) with powerful iterative tomographic
24 algorithms [1]. Since its inception, AET has successfully imaged grain boundaries, dislocations,
25 point defects, bond distortion, strain tensors, chemical order/disorder and nucleation dynamics
26 with high 3D precision [2-10]. More recently, the method has been extended to 2D materials to
27 locate individual atoms in transition metal dichalcogenides with picometer precision and correlate
28 dopant induced local strain to electronic band structures [10].

29 However, all projection measurements in AET so far have been limited to *incoherent*
30 electron scattering methods such as (high angle) annular dark field (ADF/HAADF) [11-13],
31 whereas imaging methods from *coherent* beam sources have made significant strides in recent
32 decades [14-17]. One of these is ptychography, a powerful scanning coherent diffractive imaging
33 method that can solve for both the amplitude and phase of the specimen and probe by exploiting
34 sufficient redundancies from overlapped probes and using iterative algorithms [14, 18, 19].
35 Although ptychography was proposed in 1969 [20] and realized with STEM to image crystalline
36 silicon past the conventional information limit [21], the modern version of ptychography using
37 iterative algorithms to retrieve magnitude and phase of non-crystalline objects was demonstrated
38 with x-rays in 2017 [18], which was based on the first coherent diffractive imaging experiment in
39 1999 [22]. Ptychography has since been broadly applied to image a wide range of physical and
40 biological specimens in two and three dimensions using x-rays, electrons, high harmonic
41 generation and optical lasers [23-33].

42 Conventionally, ptychography is an imaging method in two dimensions, but it has also been
43 extended to recover 3D information from a single projection by modeling the object as multiple
44 slices of phase objects [34, 35]. However, the depth resolution of this method is limited by $D_{dep} =$

45 $\frac{\lambda}{2\sin^2(\frac{\theta_{max}}{2})}$ compared to the transverse resolution limitation of $D_{tran} = \frac{\lambda}{\sin(\theta_{max})}$, where λ is the
46 wavelength of the probe and θ_{max} is the maximum detector collection angle [34, 36]. As an
47 example, a STEM experiment with an 80 keV electron probe and 80 mrad maximum collection
48 angle would yield a theoretical maximum depth resolution of 1.3 nm compared to its transverse
49 counterpart of 0.39 Å. Since atomic resolution typically requires near-ångstrom resolution, single-
50 projection multi-slice ptychography alone would not be able to recover signals from individual
51 atoms in three dimensions. Due to this limitation, ptychographic data has to be measured at
52 multiple tilt angles and combined with tomography to achieve high depth resolution in 3D phase
53 images [23, 33, 34]. As powerful single-electron pixel-array detectors that can achieve deep sub-
54 Ångstrom resolution [37, 38] become commercially available, ptychographic AET (pAET) has
55 evolved from a hypothetical idea to an experimental possibility. In this Letter, we use numerical
56 experiments to demonstrate pAET as a feasible method for low-dose 3D imaging of individual
57 light atoms by performing multislice simulations on a zinc oxide nanoparticle and a vertical
58 WS₂/WSe₂ van der Waals (vdW) heterostructure.

59 An atomic model of a spherical Wurtzite ZnO nanoparticle with a diameter of 5 nm was
60 generated with 1% randomly dispersed oxygen vacancies. This model was used to generate 0.2-
61 Å-thick projected potential slices for multislice simulation (probe sampling: 0.1 Å; sample
62 sampling: 0.1 Å; maximum detector angle: 104 mrad) [39, 40] using tabulated Hartree-Fock
63 approximations of atomic potentials [41, 42]. An aberration-corrected STEM probe was simulated
64 using the parameters of the TEAM 0.5 (electron energy: 80 keV; probe semi-convergence angle:
65 30 mrad; C1: 0 nm; C3: 900 nm; C5: -622 μm; probe size: 0.88 Å; probe step size: 0.4 Å). By
66 tilting the ZnO nanoparticle from -70° to +70°, we calculated 29 data sets of diffraction patterns

67 at 29 tilt angles. The reason that we chose a small tilt range was to demonstrate that pAET can
68 tolerate a large missing wedge. For each tilt angle, eight frozen phonon configurations at room
69 temperature were obtained and averaged for the data set. Unlike conventional AET tilt series where
70 each tilt image is a 2D ADF or HAADF image, a ptychographic tilt series consists of 4D data sets:
71 2 dimensions from scanning the sample in real space, and 2 from the diffraction in momentum
72 space. To represent real experimental conditions, the 4D data sets were corrupted with simulated
73 Poisson noise by sampling independent electron events from the ideal diffraction patterns. The
74 diffraction patterns were sampled to represent an effective dose per projection of $3.0 \times 10^4 e/\text{\AA}^2$,
75 thus simulating a total electron radiation dose of $8.7 \times 10^5 e/\text{\AA}^2$. Figure 1(a) shows a 2D logarithmic
76 heat map of the average diffraction pattern from one tilt series projection.

77 The overlap of adjacent probes (44%) gives enough redundancy to iteratively solve for
78 both object and probe functions using the extended ptychographic iterative engine (ePIE)
79 algorithm [43]. Due to high Poisson noise in individual diffraction patterns, we used a small update
80 parameter (0.01) for the object function to prevent overfitting the noise in reconstructions. Figure
81 1(b) shows an example of a ptychographic phase reconstruction on the left hand side, and the ADF
82 image (detector inner angle: 30 mrad; outer angle: 90 mrad) generated from the same diffraction
83 data set on the right hand side. 3D reconstructions of both ptychographic and ADF projections
84 were performed using REal Space Iterative Reconstruction (RESIRE) – a powerful tomographic
85 algorithm that iteratively minimizes the error between the measured and calculated projections
86 using the gradient descent [44]. Isosurface rendering of the 3D reconstruction performed with
87 ptychographic projections are shown in Figure 1(c), with a magnified isosurface rendering of the
88 volume's core in Figure 1(d). 3D rendering of individual oxygen atoms can be observed in the

89 magnified figure. Figure 1(e) shows 2.0-Å-thick central slice of the 3D volume reconstructed with
90 ptychographic phase projections on the left hand side, and with ADF-STEM projections on the
91 right hand side, with a magnified image shown in Fig. 1(f). The smaller blobs found in the
92 ePIE/RESIRE reconstruction correspond to individual oxygen atoms, which were not resolved
93 using ADF/RESIRE. Because RESIRE does not assume any periodicity while performing 3D
94 reconstruction, individual oxygen atom defects placed in the original model were also able to be
95 resolved in the pAET reconstruction as indicated by the red arrow in Fig. 1(f). Furthermore, the
96 improved quality in pAET reconstructions is especially evident when looking at a slice through
97 the missing wedge direction, shown in Fig. 1(g), and magnified in Fig. 1(h).

98 To better understand the nature of individual low-Z atom contrast with pAET, we
99 performed multislice simulations of individual atoms with varying atomic number to measure their
100 relative contrast. Similar calculations have been performed analytically for phase contrast [45],
101 and numerically for ADF-STEM and bright field STEM contrast [41]. Multislice simulations of
102 individual atoms ranging from C ($Z = 6$) to Xe ($Z = 54$) were performed with an aberration-
103 corrected probe (semi-convergence angle: 24 mrad) of varying energies (60 keV, 120 keV, and 200
104 keV) and probe step size of 0.4 Å. Ptychographic phase projections were reconstructed with ePIE
105 and ADF-STEM projections were reconstructed by integrating the diffraction patterns from 24
106 mrad to 120 mrad. Contrast per atom was defined as the height of the fitted 2D Gaussian function
107 and were normalized by setting the atomic contrast of Xe to unity. Finally, projected atomic
108 potentials were calculated and fitted as 2D Gaussians to measure ideal relative atomic contrasts.
109 The results of these calculations are shown in Fig. 2. Note that the contrasts from the potentials
110 are not strictly monotonic as a function of atomic number, as variations in the filling of electron

111 shells create fluctuations reflected by the Hartree-Fock approximations. We found that the contrast
112 from ADF-STEM was monotonic as a function of the atomic number, supporting previous studies
113 that report a power relation (roughly proportional to $Z^{1.8}$) [11-13]. Such a relation can be tolerated
114 while conducting tomography in metallic samples, but this makes it more difficult to
115 simultaneously image individual low and high Z atoms, such as in metallic oxides, due to a large
116 ratio of the relative signal ($S_{Zn}/S_O = 7.75$). In contrast, ptychography was not only able to recover
117 a higher relative signal for low- Z atoms compared to ADF-STEM ($S_{Zn}/S_O = 2.48$), but also was
118 sensitive enough to recover the aforementioned fluctuations in atomic potentials for all three beam
119 energies. This advantage of using phase signals for tomography has also been numerically
120 demonstrated with multi-slice simulation of high-resolution transmission-electron microscopy
121 images as input projections [46].

122 Successful tomography requires that the input projections need to be a sum of some
123 monotonic response to a physical property along the direction of projection – a requirement aptly
124 named as the *projection requirement* [11, 47]. Although perfect linearity is ideal, power law atomic
125 contrast in ADF/HAADF-STEM projections is sufficient to locate and identify individual atoms
126 in materials, but it requires a relatively high electron dose [1-10]. In contrast, ptychography
127 reconstructs the phase induced in the transmitted beam by the Coulomb potential, which acts as a
128 linear response to atoms. Therefore, we should expect a higher degree of linearity in AET
129 projections reconstructed with ptychography. Figure 3(a) shows the graphical representation of the
130 multislice simulation used to test linearity in ADF/HAADF-STEM images. N Si atoms separated
131 by a distance $d = 3 \text{ \AA}$ were placed co-linearly in the path of the electron beam (energy: 80 keV;
132 semi-convergence angle α : 24 mrad), and was sampled with probe step size of 0.4 \AA . Three

133 different projections of atomic columns were calculated using five different inner and outer ADF
134 detector angle combinations. After fitting each projection of atom columns to a 2D Gaussian, the
135 contrast per atom was measured as the height of the 2D Gaussian divided by the number of atoms.
136 The result of this calculation is shown in Fig. 3(b), along with a horizontal line at 1 indicating
137 contrast per atom from ideal linear projections. We observed significant deviation from the linear
138 contrast when the collection angles were smaller than 6α as channeling effects due to low electron
139 beam energy distorted the images of columns. Furthermore, the measured atomic contrast was
140 highly sensitive to the collection angles in this regime due to multiple scattering. Only when the
141 measured inner angle was higher than 6α did we observe monotonicity in the contrast per atom.
142 This might suggest HAADF-STEM as a more suitable method than ADF-STEM when performing
143 AET using low energy beams, but the electron dose in real HAADF-STEM experiments required
144 to sufficiently overcome Poisson noise makes it a less reliable choice when imaging dose sensitive
145 materials. For a comparison, we performed a similar numerical experiment by replacing the ADF
146 detector with a pixel array detector for ptychography, shown in Fig. 3(c), and plotted results from
147 contrast calculations in Fig. 3(d). Ptychography was able to maintain a more linear contrast per
148 atom than ADF/HAADF-STEM.

149 Lastly, the method of pAET can also be used to image individual low-Z atoms in
150 geometries other than nanoparticles such as thin films and vdW heterostructures. The vdW
151 interaction between the top and bottom layers mediates various types of coupling across the
152 interface in vdW heterostructures. It has been reported that the quantum properties of vdW
153 heterostructures are highly tunable with vertical stacking through moiré potentials, and that
154 different stacking alignments can cause drastic changes in exciton excitation as well as other

155 quantum properties such as superconductivity and correlated insulator states [48-52]. The
156 capability to precisely determine the 3D coordinates and chemical species of individual atoms
157 combined with *ab initio* calculations is anticipated to reveal unprecedented details about the
158 correlation between the atomic structure of vdW heterostructures and their exotic quantum
159 properties. Using identical electron probe parameters as those used in Fig. 1, multislice simulation
160 was performed on an atomic model of vertical WS₂/WSe₂ van der Waals heterostructure, with its
161 two tungsten layers separated by 6.54 Å and tiled by 12.5°. Figure 4(a) shows 2.0-Å-thick slices
162 of every atomic layer of the heterostructure when reconstructed with ePIE and RESIRE. Despite
163 severely corrupting the diffraction patterns by simulating a total electron dose of $5.1 \times 10^4 e/\text{Å}^2$,
164 every atomic layer including the two sulfur layers are resolved. However, when tomography was
165 performed on ADF-STEM projections with an equivalent electron dose, as shown in Fig. 4(b), the
166 signals from the sulfur layers attenuate below a level at which individual atoms are traceable.

167 In summary, by leveraging improved signal from low-Z atoms and linearity in atomic
168 projections, ptychographic reconstructions from 4D-STEM data can offer significant advantages
169 when performing electron tomography at the atomic scale. To demonstrate these advantages, we
170 simulated a ptychographic tomography tilt series of 29 projections of a 5 nm Wurtzite zinc oxide
171 nanoparticle using the multislice simulation, reconstructed each projection using ePIE, and
172 performed tomography with RESIRE to resolve individual oxygen atoms and their defects. We
173 also believe that pAET serves as a possible alternative to conventional AET methods for low dose
174 3D atomic imaging of 2D materials. After simulating pAET on vertical WS₂/WSe₂ van der Waals
175 heterostructure with a low electron dose of $5.1 \times 10^4 e/\text{Å}^2$, we were able to resolve every layer
176 including the lightest sulfur layer to the atomic resolution. As high quality pixel array electron

177 detectors become more commercially viable and data storage and management become more
178 streamlined, we envision pAET as a powerful method for studying the 3D atomic structures of
179 low-*Z* and radiation sensitive materials such as transition metal oxides, functional 2D
180 heterostructures and glasses.

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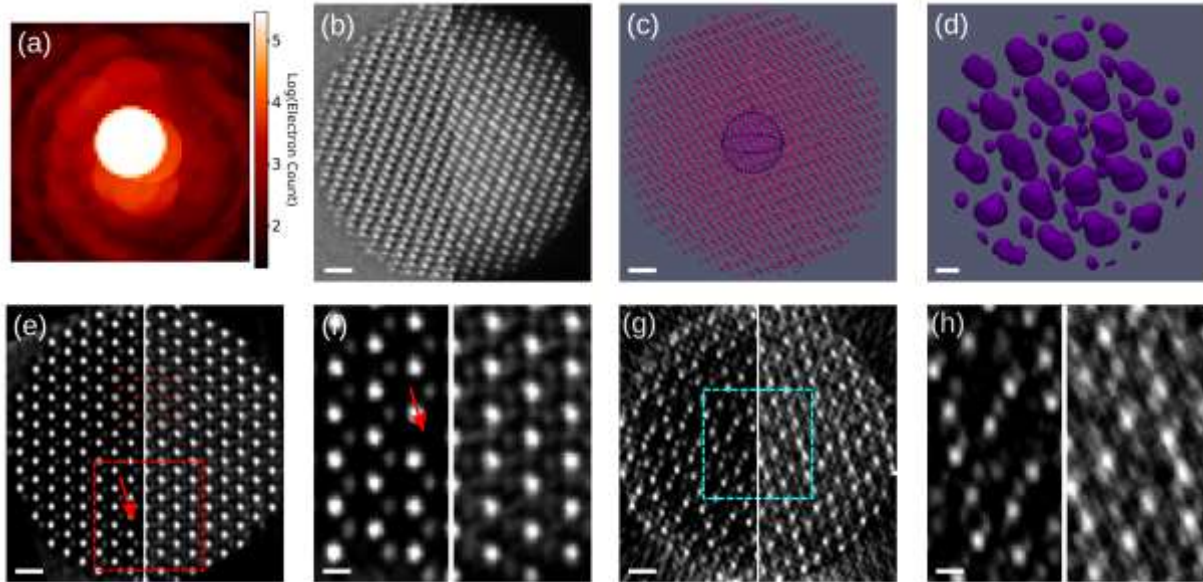
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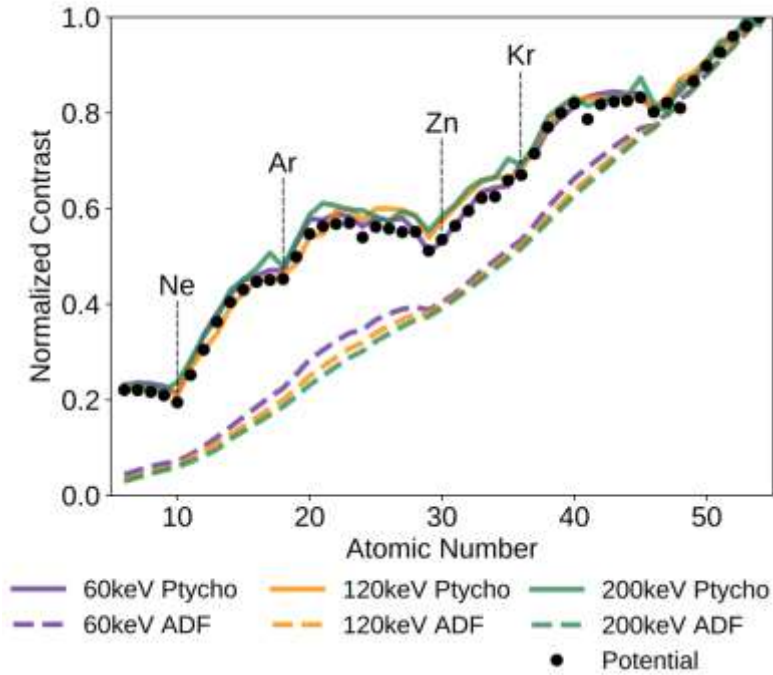
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264 **Figures and Figure Captions**



265 **Figure 1.** Numerical experiment on ptychography- and ADF-STEM-based atomic electron
 266 tomography of a 5 nm ZnO nanoparticle. (a) 2D logarithmic heat map of the average of 15625
 267 diffraction patterns from a tilt series. The diffraction patterns simulate a dose per projection of 3.0
 268 $\times 10^4 e/\text{\AA}^2$. (b) A representative ptychographic phase projection (left) and an ADF-STEM
 269 projection (right). Both projections were reconstructed with the same diffraction patterns seen in
 270 (a). (c) 3D isosurface rendering of the volume after tomographic reconstruction of ptychographic
 271 phase projections. (d) Magnified isosurface rendering of the core of the volume in (c). Individual
 272 oxygen atoms rendered as smaller spheres are observed. Note that the zinc atoms look
 273 disproportionate large due to the isosurface rendering effect. (e) A 2.0-Å-thick central slice of the
 274 volume in the [0001] direction reconstructed with ptychographic phase projections (left) and with
 275 ADF-STEM projections (right). (f) A magnified image of (e), indicating an oxygen defect with a
 276 red arrow. (g) A 2.0-Å-thick slice through the missing wedge direction of the reconstruction
 277 performed with ptychographic projections (left) and with ADF projections (right). (h) A magnified

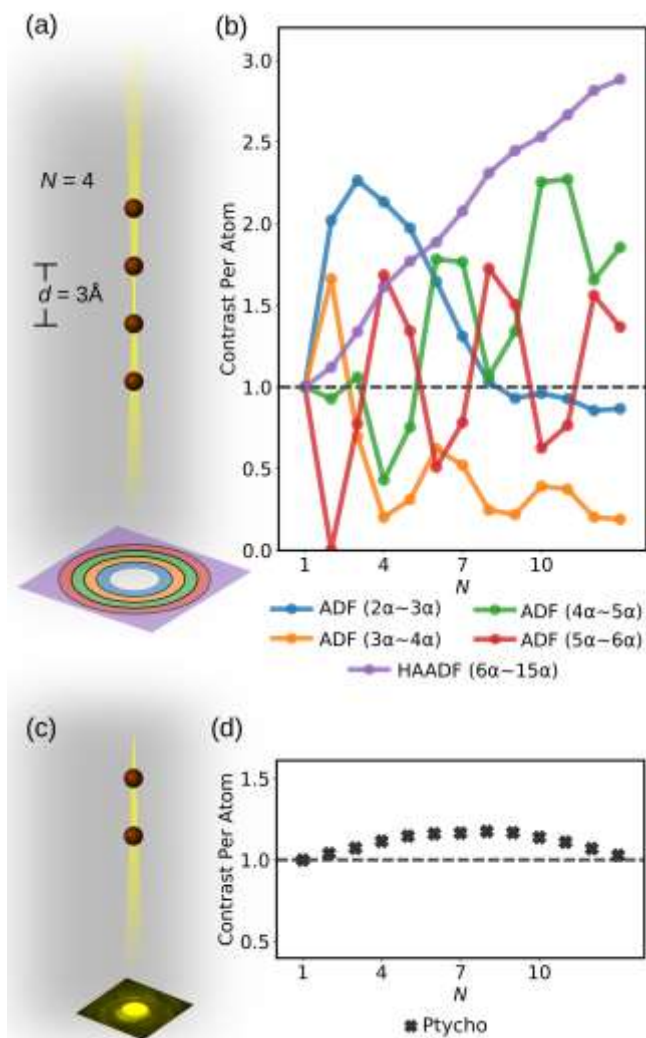
278 image of (g), demonstrating greater missing wedge artifacts in the reconstruction when performing
279 tomography with ADF-STEM projections. Scale bars in (b), (c), (e) and (g) indicate 5 Å, scale bar
280 in (e) indicates 1 Å, and scale bars in (f) and (h) indicate 2 Å.



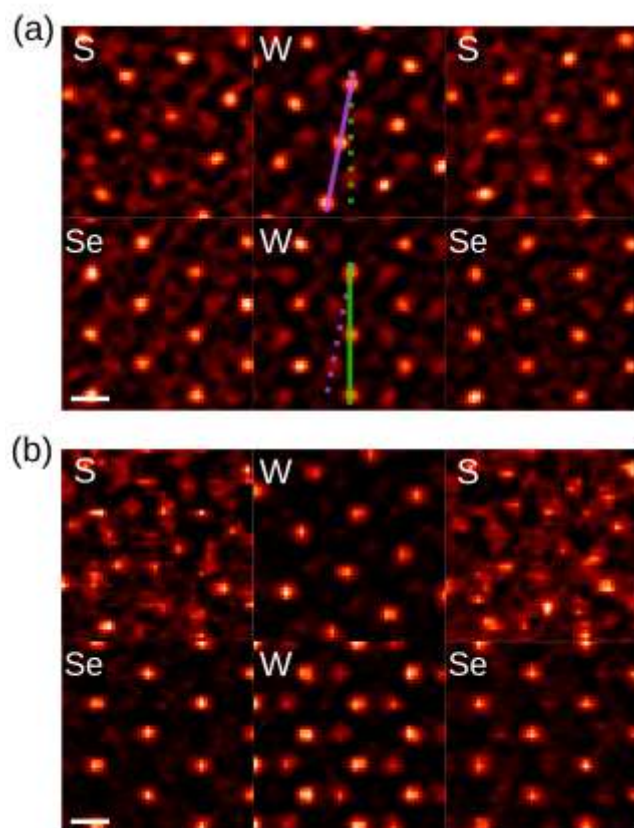
281

282 **Figure 2.** Quantitative comparison of normalized atomic contrast from C to Xe between
283 ptychography and ADF for three different electron beam energies (60 keV, 120 keV, and 240 keV).
284 Normalized peak heights from Hartree-Fock potential estimations are also plotted. Atoms where
285 valence shells become fully filled (Ne, Ar, Zn, and Kr) are indicated to explain fluctuations in the
286 potential peaks.

287



288 **Figure 3.** Contrast per atom when imaging along the zone-axis with ADF/HAADF and
 289 ptychography. (a) An experimental schematic of multislice simulation of an 80 keV electron beam
 290 probe and 24 mrad semi-convergence angle imaging a column of N Si atoms separated by distance
 291 of 3 Å. Five different combinations of inner and outer angles were simulated. (b) Contrast per atom
 292 in atomic columns when measured with various ADF angles. The horizontal gray line plotted at 1
 293 indicates the ideal linear projection. (c) Similar experimental schematic as (a) except the
 294 substitution of ADF detectors with a pixel array detector, allowing for ptychographic
 295 reconstruction. (d) Contrast per atom in columns when reconstructed with ptychography.



296 **Figure 4.** Numerical experiment on ptychography- and ADF-STEM-based AET of a vertical
 297 WS_2/WSe_2 van der Waals heterostructure. (a) 2.0-Å-thick slices of each atomic layer when
 298 simulated with a total electron dose of $5.1 \times 10^4 e/\text{Å}^2$ and reconstructed with ePIE and RESIRE.
 299 Tilt angle of 12.5° between the two tungsten layers was recovered, indicated by colored lines. (b)
 300 2.0-Å-thick slices of the same experiment as (a) when reconstructed with ADF and RESIRE. Scale
 301 bars, 2 Å.