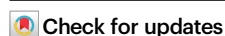


Atomically resolved imaging of radiation-sensitive metal-organic frameworks via electron ptychography

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Electron ptychography, recognized as an ideal technique for low-dose imaging, consistently achieves deep sub-angstrom resolution at electron doses of several thousand electrons per square angstrom ($e^-/\text{\AA}^2$) or higher. Despite its proven efficacy, the application of electron ptychography at even lower doses—necessary for materials highly sensitive to electron beams—raises questions regarding its feasibility and the attainable resolution under such stringent conditions. Herein, we demonstrate the implementation of near-atomic-resolution ($\sim 2 \text{\AA}$) electron ptychography reconstruction at electron doses as low as $\sim 100 e^-/\text{\AA}^2$, for metal-organic frameworks (MOFs), which are known for their extreme sensitivity. The reconstructed images clearly resolve organic linkers, metal clusters, and even atomic columns within these clusters, while unravelling various local structural features in MOFs, including missing linkers, extra clusters, and surface termination modes. By combining the findings from simulations and experiments, we have identified that employing a small convergence semi-angle during data acquisition is crucial for effective iterative ptychographic reconstruction under such low-dose conditions. This important insight advances our understanding of the rapidly evolving electron ptychography technique and provides a novel approach to high-resolution imaging of various sensitive materials.

Atomic-resolution scanning transmission electron microscopy (STEM) imaging of electron beam-sensitive materials presents a long-standing challenge^{1–4}. The challenge is multifold. First, the electron dose must be minimized to preserve the structural integrity of the sample,

necessitating the development of imaging mechanisms that maximize electron utilization efficiency. Second, to obtain interpretable images, the specimen needs to be ultrathin, a condition rarely met in practice. Third, achieving atomic resolution requires a precisely focused

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electron beam; however, structural damage likely occurs during the beam-focusing process if the specimen is particularly sensitive.

Electron ptychography^{5–7}, revitalized by advanced electron detector technologies that facilitate the efficient acquisition of four-dimensional STEM (4D-STEM) datasets, offers a promising solution to these challenges. Compared to conventional STEM, electron ptychography boasts notable advantages in several aspects^{8–16}. It has demonstrated superior resolution down to the deep-sub-angstrom level and greater tolerance to specimen thickness, establishing a new resolution record for real-space imaging^{17–19}. Moreover, it exhibits resolving power in the third dimension (i.e., depth resolution) at the nanometer level^{18,20–22}, which is difficult for conventional STEM to achieve. Additionally, electron ptychography does not require precise focusing and utilizes almost all the signals available in the electron diffraction patterns. This combination of straightforward data acquisition and high dose efficiency makes it an ideal low-dose imaging technique.

While extensive research endeavors have been undertaken to push the resolution limits of electron ptychography^{17–19}, its significant potential for low-dose imaging of beam-sensitive materials remains largely unexplored^{23–26}. Several attempts in this area were conducted with electron doses of thousands of electrons per square angstrom ($\text{e}^-/\text{\AA}^2$)^{20,27–29}. Although such dose levels are generally considered low for electron microscopy, they are still excessively high for numerous emerging functional materials, such as metal-organic frameworks (MOFs) and organic-inorganic hybrid halide perovskites, which exhibit electron dose thresholds of $<100 \text{ e}^-/\text{\AA}^2$ under STEM mode^{2,30}. High-resolution imaging of these materials through electron ptychography has yet to be accomplished due to the challenge of reconstruction with such low doses. Moreover, despite previous discussions on dose-limited resolution in general¹⁷, the limit of ptychographic image resolution under such extremely low electron dose conditions remains unclear. The only study to date on electron ptychography imaging of MOF exhibited a resolution of $\sim 4.8 \text{ \AA}$ (the d spacing of the (024) planes), which is insufficient to reveal structural details in the framework³¹. One possible reason for this limited resolution is that the relatively high electron dose ($>800 \text{ e}^-/\text{\AA}^2$) resulted in partial structural degradation to the MOF material, Hf-BTB. This underscores the significance of pushing forward electron ptychography into the realm of even lower doses. Although electron doses of tens of $\text{e}^-/\text{\AA}^2$ have been used for ptychography imaging of biological specimens, only nanometer-scale resolution has been achieved^{23,32}.

In this study, we demonstrate that electron ptychography can be effectively performed at dose levels of $\sim 100 \text{ e}^-/\text{\AA}^2$, yielding directly interpretable images for highly beam-sensitive MOF materials. Thanks to the phase-contrast characteristic of ptychographic images, both metal clusters and organic linkers are observed simultaneously, with adjacent atomic columns of metal within the clusters clearly separated, achieving a resolution of $\sim 2 \text{ \AA}$. This success is attributed to the optimal conditions for 4D-STEM data collection, identified through simulations, and the versatility of the employed reconstruction algorithm. The high resolution and contrast in the reconstructed images enable direct observation of various local structures in MOFs. Our study provides guidance and insights for imaging beam-sensitive materials using electron ptychography, by exploring its dose and resolution limits.

Results

Figure 1a illustrates the optical configuration of a 4D-STEM setup, where an electron probe scans over the specimen in a 2D raster manner, while a far-field pixelated detector records the 2D convergent-beam electron diffraction (CBED) pattern at each scanning position, resulting in a 4D dataset. Our 4D-STEM experiments employed a 300-kV electron probe, a hybrid pixel array detector (EMPAD)³³, and 256×256 probe positions. Compared to our previous study on

zeolites²⁹, we opted to use a larger scan step of $1.05 \text{ \AA}$ in this research for MOFs to reduce the electron dose and accordingly used a defocused probe to ensure adequate overlap between adjacent illuminated areas. Using a defocused probe also eliminates the need for precise focus tuning, which, as previously discussed, poses a formidable challenge in conventional STEM imaging of sensitive materials. With these parameters preset, the electron dose consumed during 4D-STEM data acquisition can be lowered to $\sim 100 \text{ e}^-/\text{\AA}^2$ by reducing the beam current to $<0.02 \text{ pA}$, despite the EMPAD's limited frame rate of 1000 frames per second.

The remaining variable that may influence the ptychographic reconstruction is the convergence semi-angle (α) of the probe. While the effect of α on the contrast transfer properties of in-focus ptychography has been discussed³⁴, its impact on out-of-focus and iterative ptychography remains less explored. To investigate this, we conducted simulations using MOF Zr-BTB as a model material (see Fig. 1b). Specifically, we simulated 4D-STEM datasets of Zr-BTB with varying α values (21 mrad, 15 mrad, 10 mrad, and 5 mrad) at different electron dose levels (infinite dose, $1000 \text{ e}^-/\text{\AA}^2$, and $100 \text{ e}^-/\text{\AA}^2$), while maintaining a fixed probe defocus of 60 nm (see Supplementary Fig. 1). For each simulated 4D-STEM dataset, iterative ptychographic reconstruction was conducted using the least squares-maximum likelihood (LSQ-ML) algorithm^{35,36}, which incorporates multislice and mixed-state methods^{18,37,38}.

Using 4D-STEM datasets simulated with an infinite electron dose, iterative ptychography reconstruction readily converged, yielding phase images that match the projected structure of Zr-BTB, irrespective of the α values (Supplementary Fig. 2). The contrasts of light elements show slight variation with α , likely due to their relatively weak scattering ability. Similar outcomes were obtained when employing 4D-STEM datasets simulated with a dose of $1000 \text{ e}^-/\text{\AA}^2$ for reconstruction (Supplementary Fig. 2). These findings suggest that with sufficient electron doses, ptychographic reconstruction can proceed smoothly across a range of convergence semi-angles.

When the electron dose was lowered to $100 \text{ e}^-/\text{\AA}^2$, the CBED patterns exhibited extremely sparse and weak signals (Fig. 1c). On average, each CBED pattern contains only ~ 100 electrons, and the signals are predominantly single-electron events (Supplementary Fig. 3). Such a low-dose level presents a significant challenge for ptychography reconstruction, even with simulated data. As shown in Fig. 1d, the iterative processes failed to converge for data simulated at 15 mrad and 21 mrad, and terminated within 60 iteration cycles (Fig. 1d). The resultant images displayed poor contrast and resolution, lacking structural details (Fig. 1e). Interestingly, maintaining all the other conditions identical but using 4D-STEM data simulated at 10 mrad rendered a stable iteration process, leading to a reconstructed image that precisely matched the structural model at a high resolution (Fig. 1e and Supplementary Fig. 2). Further reduction of the α value to 5 mrad led to a decrease in resolution for the reconstructed image (Fig. 1e and Supplementary Fig. 2). This reduced resolution, associated with an excessively small α , corresponds to the width of the bright-field defined by diffraction limits³⁹, as nearly all signals usable for ptychographic reconstruction under such low-dose conditions are concentrated within the bright-field region (Supplementary Fig. 3). Reduced data redundancy in both real and reciprocal spaces due to smaller α is another unfavorable factor for high-precision reconstruction⁴⁰. We conducted additional simulations to examine the effects of convergence angle on reconstruction at a constant electron dose of $100 \text{ e}^-/\text{\AA}^2$. The results show that the unsuccessful reconstruction with a large α (e.g., 21 mrad) is not due to insufficient sampling in reciprocal space: the current simulation parameters meet Nyquist sampling requirements to avoid aliasing⁴¹, and increasing sampling with finer detector pixel sizes does not enhance reconstruction quality (Supplementary Fig. 4). Additionally, when the probe

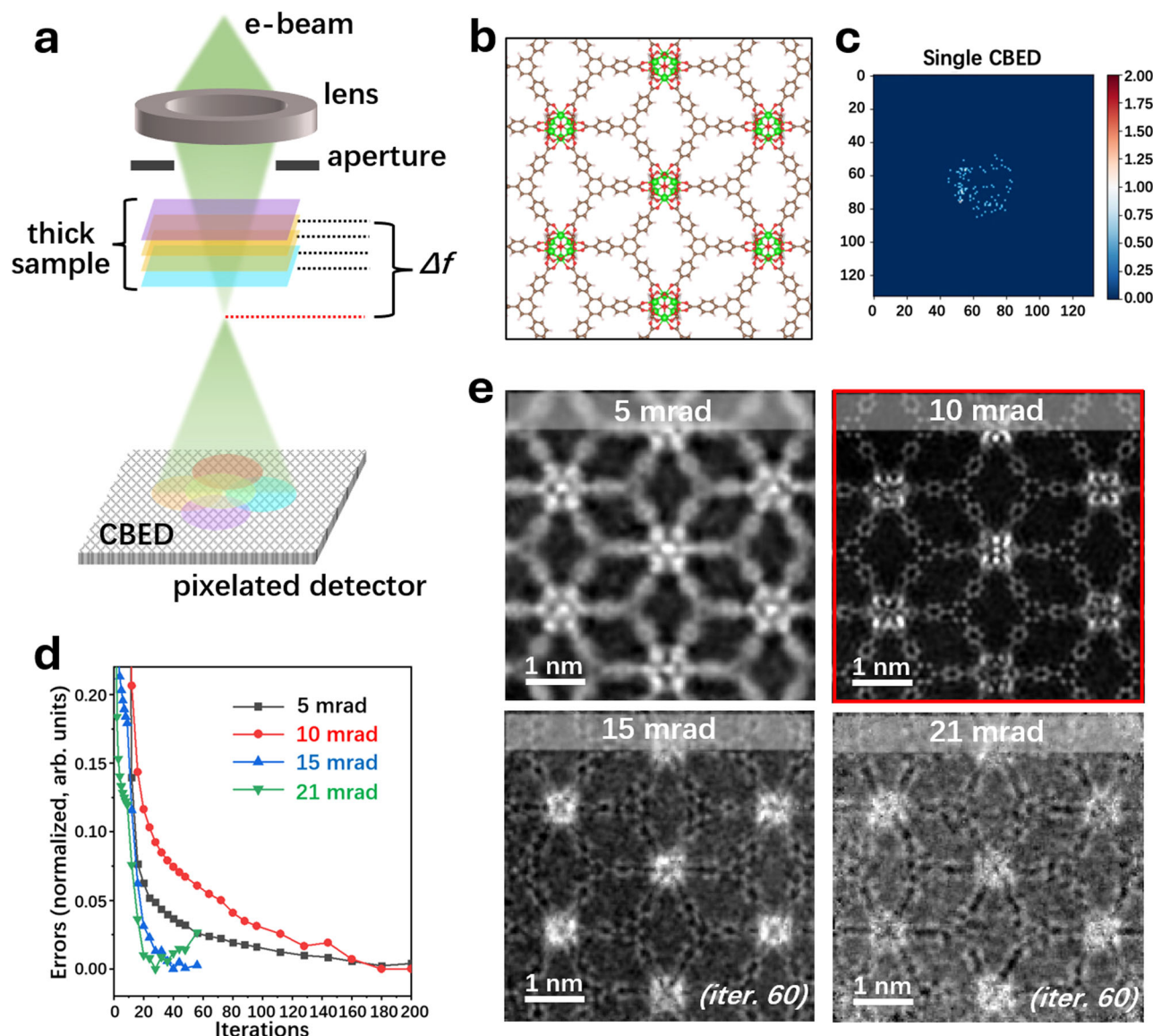


Fig. 1 | Analysis of the impact of 4D-STEM conditions on low-dose ptychography through simulations. **a** Schematic representation of 4D-STEM utilized for multislice electron ptychography on a thick specimen. **b** Structural model of MOF Zr-BTB employed in 4D-STEM simulations, projected along the [100] axis. Green Zr, red O, gray C, pink H. **c** A single convergent-beam electron diffraction (CBED)

pattern from a 4D-STEM dataset simulated with a total electron dose of $100 \text{ e}^-/\text{\AA}^2$. **d, e** Iterative reconstruction error curves (**d**) and reconstructed ptychography phase images (**e**) using 4D-STEM datasets simulated at varying convergence semi-angles with a consistent electron dose of $100 \text{ e}^-/\text{\AA}^2$. The 10-mrad case shows the best reconstruction, as marked by the red square.

size is fixed at $\sim 7.8 \text{ \AA}$ across various convergence angles by adjusting the defocus, the reconstruction using 10 mrad data consistently outperforms those using 15 mrad and 21 mrad data (Supplementary Fig. 5).

These simulation results suggest that at very low electron doses, a relatively small α is preferable for achieving successful ptychography reconstruction for an MOF material. This appears to contradict the conventional wisdom in STEM imaging, where a larger α is generally associated with enhanced resolution. We speculate that this unusual outcome stems from a confluence of factors. Firstly, the MOFs, characterized by large unit cells and open frameworks, do not require information transfer at very high spatial frequencies to effectively characterize their key structural features. For a given limited total electron dose, a larger α value would allocate more signal to the high spatial frequency realm, at the expense of low frequencies, resulting in a weakened signal for reconstruction (Supplementary Fig. 3d). Consequently, a smaller α value is preferred in this context. Secondly, although ptychography can correct probe aberrations when sufficient

dose is available⁴², our practical experience is that at very low dose, solving for both an unknown object and probe becomes numerically unstable. Instead, a probe with minimal aberrations is essential for successful reconstruction as it can be more accurately measured and modeled. Therefore, a probe with a smaller α is advantageous because it involves a lower degree of aberrations. Thirdly, the convergence angle directly affects the probe size and depth of field. Using a small convergence semi-angle can prevent the generation of an overly large probe size under defocused conditions while maintaining a large depth of field, again reducing sensitivity to the initial estimate of probe shape. Additionally, with a large convergence semi-angle, minor defocusing can result in a significantly enlarged probe size (Supplementary Fig. 6, Supplementary Table 1), risking exceeding the limits of the reconstruction box size (Supplementary Fig. 7) and potentially leading to wrap-around artifacts or failed reconstruction^{41,43}. This issue in practical experiments can also be effectively avoided by employing a small convergence semi-angle.

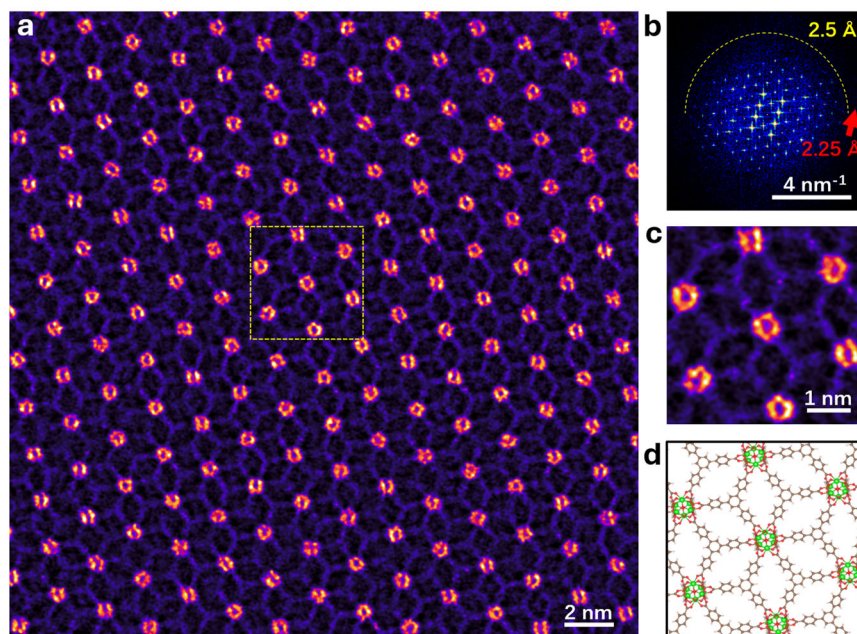


Fig. 2 | Experimental electron ptychography imaging of MOF Zr-BTB.

a Ptychography phase image reconstructed from an experimental 4D-STEM dataset acquired at conditions of 10-mrad convergence semi-angle, 1.05 Å scan step, and an electron dose of 115 $\text{e}^-/\text{\AA}^2$. **b** The power spectrum of (a), demonstrating information

transfer up to 2.25 Å. **c** Enlarged image from the square-marked area in (a).

d Structural model of MOF Zr-BTB, projected along the [100] axis. Green Zr, red O, gray C, pink H. The high consistency between (c) and (d) indicates the accuracy of ptychography reconstruction at high resolution.

Following the simulation finding that a 10-mrad α is advantageous for achieving robust ptychography reconstruction for MOFs under low-dose conditions, we experimentally acquired 4D-STEM data for Zr-BTB along the [100] zone axis, at conditions of 10-mrad convergence semi-angle, 1.05 Å scan step, and an electron dose of 115 $\text{e}^-/\text{\AA}^2$. An unfocused probe was used for the experiment, which was later determined through ptychography reconstruction to have a defocus value of 45 nm. The low-magnification TEM and HAADF-STEM images of the investigated Zr-BTB crystal are displayed in Supplementary Fig. 8.

A hybrid approach incorporating *J*-matrix and *Fourier* methods was used for the experimental 4D-STEM datasets, which allows for corrections for scan position and sample misorientation prior to commencing the iterative calculations¹⁵. These corrections prove to be crucial, significantly enhancing the convergence in the subsequent ptychography reconstruction processes for data collected at such low doses with poor signal-to-noise ratios.

Because of the use of an unfocused probe aimed at reducing structural damage, conventional STEM modes including bright-field (BF), annular bright-field (ABF), annular dark-field (ADF), and integrated center-of-mass (iCOM) all showed restricted image resolution, thus providing minimal structural insights (Supplementary Fig. 9). In stark contrast, ptychographic reconstruction from the same 4D-STEM dataset yielded an image with nearly atomic resolution and distinct contrast. Within this image, the fundamental structural features of Zr-BTB along the [100] projection, such as Zr_6 clusters, three-arm organic linkers, their interconnections, and the resulting rhombic channels, are clearly discernible (Fig. 2a). The power spectrum of the image revealed an information transfer reaching up to 2.25 Å (Fig. 2b), far surpassing the resolution previously attained for this MOF³¹ and rivaling that achieved by electron ptychography for a zeolite at much higher electron dose (1000 $\text{e}^-/\text{\AA}^2$)²⁷. While the six Zr atoms within each cluster cannot be completely distinguished at this resolution, the overall shape of the cluster and the pair of Zr atoms that exhibit longer projected interatomic distances are identifiable (Fig. 2c).

Ptychography reconstruction also revealed a specimen thickness of ~40 nm for the examined area. At this thickness, conventional low-

dose STEM, such as iDPC-STEM fails to attain the same resolution, despite achieving precise focusing through additional efforts (see Supplementary Fig. 10). These findings affirm that electron ptychography offers advantages in both data acquisition simplicity and tolerance to specimen thickness. While a few studies in the literature have demonstrated near-atomic resolution imaging of MOFs using low-dose iDPC-STEM^{44–46}, simulation results indicate that the specimens in these cases must be exceptionally thin, likely <20 nm (see Supplementary Fig. 11). In contrast, electron ptychography effectively transcends this stringent limitation on specimen thickness.

To further verify the general feasibility of low-dose electron ptychography, we acquired 4D-STEM data for another MOF, termed MOSS-6⁴⁷, employing nearly identical optimal conditions as identified previously. The defocus of the probe and electron dose used for this dataset were determined to be 100 nm and 98 $\text{e}^-/\text{\AA}^2$, respectively. As a MOF solid solution, MOSS-6 incorporates two MOF phases, NU-1000 and NU-901, within plate-like crystals that are hundreds of nanometers wide and tens of nanometers thick (Supplementary Fig. 12)⁴⁷.

Consistent with the findings for Zr-BTB, conventional STEM modes yielded virtually no structural information owing to the use of a defocused probe (Supplementary Fig. 13), whereas ptychographic reconstruction produced an atomic-resolution image with crisp contrast (Fig. 3a). For this dataset, ptychography attained an exceptionally high resolution (information transfer) of 1.96 Å, as determined from the power spectrum of the reconstructed image (Fig. 3b). The examined area displays the typical NU-1000 structure, projected along the [001] direction. The combination of high resolution and excellent image contrast enables the direct identification of the characteristic Kagome lattice of NU-1000, along with the Zr clusters and organic linkers forming this lattice. Remarkably, within many clusters, the four Zr atomic columns—two with strong intensity and two with weak—are distinctly resolved, and their orientations align perfectly with the structural model (Fig. 3c, d). This exciting outcome is attributed to the enhanced image resolution and a slightly longer Zr-Zr distance in this projection compared to the previously discussed case of Zr-BTB. Supplementary Fig. 14 compares the achievable resolution under the

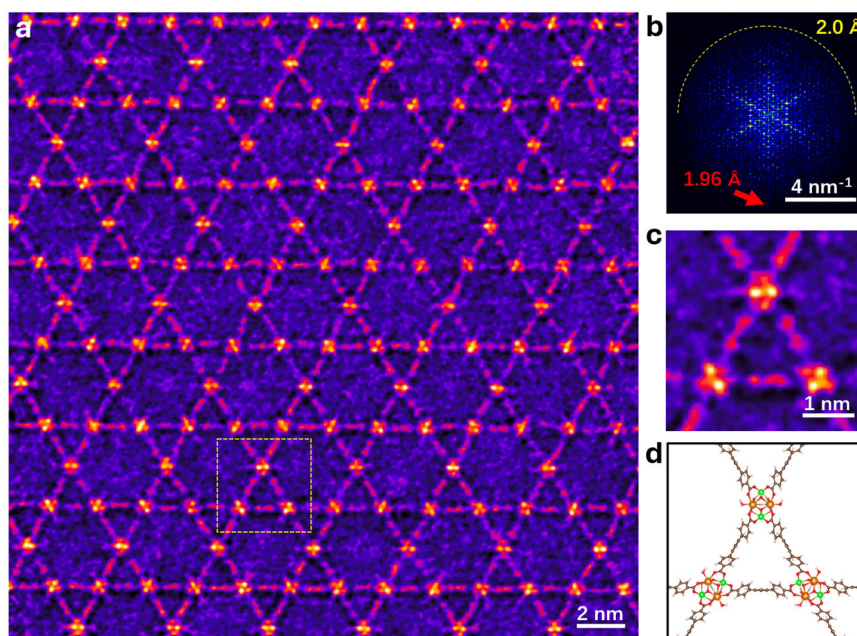


Fig. 3 | Experimental electron ptychography imaging of MOF MOSS-6.

a Ptychography phase image reconstructed from an experimental 4D-STEM dataset acquired at conditions of 10-mrad convergence semi-angle, 1.05 Å scan step, and an electron dose of $98 \text{ e}^-/\text{Å}^2$. **b** The power spectrum of (**a**), demonstrating information transfer up to 1.96 Å. **c** Enlarged image from the square-marked area in (**a**). **d** Local region of the [001]-projected structure of NU-1000, a component of MOSS-6.

Green/orange: Zr; red: O; gray: C; pink: H. Within each Zr cluster, the Zr pairs with strong and weak projected electrostatic potential intensity are labeled in orange and green color, respectively. The comparison between (**c**) and (**d**) indicates that the reconstructed ptychography phase image accurately reflects the MOF's structural features, including the cluster orientation.

employed low-dose conditions with the resolutions required to identify various structural features in NU-1000.

Figure 4a shows the ptychography phase image reconstructed from a 4D-STEM dataset from another area, revealing the structural inhomogeneity of MOSS-6. The domain marked by red dashed lines features 'extra clusters' at the centers of hexagonal channels, while the remaining area displays the ideal NU-1000 structure, characterized by vacant hexagonal channels. The enlarged images of a representative vacant channel and a channel containing an 'extra cluster' are shown in Fig. 4b, c, respectively. The contrast from the organic linkers connecting the extra cluster with adjacent clusters is also visible (Fig. 4c). Given that MOSS-6 is a solid solution of NU-1000 and NU-901, we infer that the observed 'extra cluster' domain is associated with the vertical intergrowth of these two MOF phases. We have developed a structural model for the intergrowth of NU-1000 and NU-901 to explain the observed image contrast (refer to the Supplementary PDB File). However, the chemical validity of this model requires further verification, as it includes clusters with low connection numbers at the interface. Additionally, the ptychography image reveals that the MOSS-6 crystal surface alternates between complete triangular channels (marked by blue arrows) and open hexagonal channels (Fig. 4d). Notably, in local areas lacking a protruding triangular channel, a 'missing linker' defect is observed (highlighted by orange arrows), leading to the formation of two dangling triangular channels (Fig. 4d). These findings suggest that the crystal surface is predominantly terminated by triangular channels, indicating a preference over complete hexagonal channels as a surface termination mode.

Additional simulations were conducted to investigate the feasibility and optimal conditions for low-dose high-resolution ptychography imaging of other sensitive materials with diverse structural characteristics (see Supplementary Table 2). The results reveal that for MOFs ZIF-8⁴⁸ and In-sod-ZMOF-102⁴⁹, the 10-mrad convergence semi-angle remains the best choice under low-dose conditions

(Supplementary Figs. 15 and 16). However, a slightly higher dose of $110 \text{ e}^-/\text{Å}^2$ was found necessary for successful reconstruction of ZIF-8, likely due to the complex electron scattering behavior of its relatively dense structure. As material density increases, the optimal conditions diverge from those identified for MOFs. For the hybrid perovskite $\text{CH}_3\text{NH}_3\text{PbI}_3$ ^{50,51}, for instance, thinner ($\sim 20.4 \text{ nm}$) specimens and higher electron doses ($\sim 150 \text{ e}^-/\text{Å}^2$) are required for effective ptychographic imaging (Supplementary Fig. 17). Since this dose exceeds the previously determined electron dose threshold for $\text{CH}_3\text{NH}_3\text{PbI}_3$ ⁵², achieving ptychographic reconstruction for this material experimentally under current conditions would be challenging. Moreover, the image reconstructed from the 10-mrad data exhibits donut-shaped artifacts on heavy atoms. While using larger convergence angles can mitigate this issue, it results in reduced contrast for light elements due to the stronger Z-dependence effect⁵³ (Supplementary Fig. 17). These findings demonstrate that, while a defocused probe and small convergence angle are generally advantageous for ptychographic imaging of sensitive materials, the optimal conditions are material-dependent and require careful refinement to achieve.

Discussion

In this study, we have demonstrated the feasibility of effectively conducting 4D-STEM ptychography for highly beam-sensitive materials, exemplified by two MOFs, at low electron doses of $\sim 100 \text{ e}^-/\text{Å}^2$ necessary to preserve their structural integrity. The reconstructed phase-contrast images exhibited a $\sim 2 \text{ Å}$ resolution, clearly discerning organic linkers, metal clusters, and even atomic columns within clusters, while revealing previously unknown local structural features in MOFs. We have determined that the combined utilization of a large scan step, a properly defocused probe, and a relatively small convergence angle during 4D-STEM data acquisition is pivotal for the success of the subsequent iterative ptychography reconstruction. The advantageous effects of employing a small convergence angle on

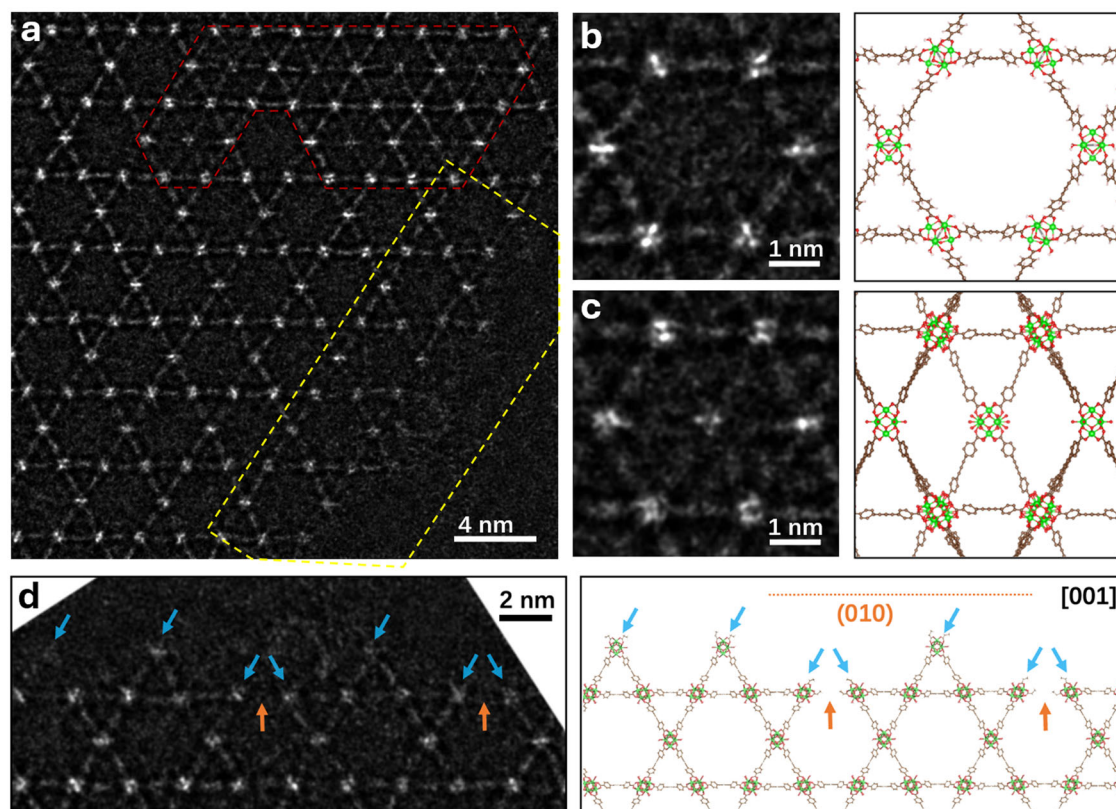


Fig. 4 | Local structures in MOF MOSS-6 identified in a ptychography image. **a** Reconstructed ptychography phase image of an area with structural inhomogeneity: the majority of the imaged area corresponds to the structure of NU-1000, while the region marked with red dashed lines shows the presence of ‘extra clusters’ in hexagonal channels, relative to NU-1000. **b** Enlarged image from (a), showing a typical hexagonal channel of NU-1000, and the corresponding structural model. **c** Enlarged image from the region marked with red dashed lines in (a), revealing the

presence of an ‘extra cluster’ in the hexagonal channel. The ‘extra cluster’ is inferred to be associated with the intergrowth of NU-1000 and NU-901. On the right is the proposed structural model. **d** The region marked with yellow dashed lines in (a) after orientation adjustment, illustrating missing linker defects (orange arrows) and the resultant triangular channel-terminated crystal surface (blue arrows), along with the corresponding structural model. Green Zr, red O, gray C, pink H.

low-dose, thick-specimen electron ptychography have not been recognized in previous studies. Our results have demonstrated that electron ptychography can achieve a further dose reduction of approximately an order of magnitude, reaching a level of ~ 100 electrons per square angstrom while maintaining atomic resolution. This breakthrough enhances the field of low-dose imaging and opens up new possibilities for the characterization of 4D-STEM ptychography in materials science.

Methods

Materials synthesis

The MOFs Zr-BTB and MOSS-6 investigated in this study were synthesized by strictly following the previously reported methods^{47,54}. The as-synthesized MOF samples were dispersed in ethanol, and the resulting suspensions were dropped onto Lacey carbon Cu TEM grids for subsequent 4D-STEM experiments.

4D-STEM simulations

Simulations of the 4D-STEM dataset were conducted using the abTEM software package⁵⁵ in a GPU-accelerated Python environment. Electron probes were generated at an accelerating voltage of 300 kV, using convergence semi-angles of 21 mrad, 15 mrad, 10 mrad, and 5 mrad, each with a defocus value as specified in Supplementary Table 3. Other key parameters used in the simulations, including sample thickness, scanning step size, and pixel size in reciprocal space, are also detailed in Supplementary Table 3. Upon generating the 4D-STEM datasets,

varying dose levels were introduced using the Poisson model to simulate realistic electron exposure.

4D-STEM experiments

The 4D-STEM experiments were performed using a double Cs-corrected FEI Titan Cubed Themis Z TEM, operated at an accelerating voltage of 300 kV. A convergence semi-angle of 10 mrad was utilized, with a proper camera length ensuring that the bright-field disk occupies roughly one-third of the detector. The electron microscopy pixel array detector (EMPAD)³³ was employed to CBED patterns across a scanning grid of 256×256 , with a scanning step size of 1.05 Å. The beam current was finely tuned via the TEM monochromator to achieve a low electron dose level of $\sim 100 \text{ e}^-/\text{Å}^2$. At such low doses, the signals received by the detector were extremely sparse and weak, predominantly consisting of single-electron events, as illustrated in Supplementary Fig. 18 and Supplementary Table 4.

4D-STEM data processing

BF, ABF, ADF, and iCOM images were reconstructed using the py4D-STEM software package⁵⁶ and custom Python scripts. The probe defocus values were initially estimated using Parallax in py4DSTEM and then refined through the iterative process of ptychographic reconstructions. Prior to the reconstructions, scan position corrections were applied using published codes¹⁵. The iterative ptychography reconstructions were carried out based on LSQ-ML methods^{35,36}, incorporating both the multislice and mixed-state approaches. These reconstructions were implemented in MATLAB and custom-developed software.

Data availability

All data supporting the findings of this study are available within the paper and its Supplementary Information files. The raw data have been deposited and are available at *Zenodo* under the accession code <https://doi.org/10.5281/zenodo.13958144>.

Code availability

The correction methods/codes used in this paper prior to ptychography reconstruction can be found in GitHub repositories: <https://github.com/ningustc/4DSTEM-Calibration>; the `fold_slice` codes for mixed-state and multislice ptychography reconstructions are from previous publications^{17,18}. All of these are available from the corresponding authors on request.

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Author contributions

Y.H. initiated and supervised the project. Y.H., H.Z., and S.N. coordinated the research. G.L. performed the 4D-STEM simulations and experiments. S.N. and G.L. performed ptychographic reconstruction. M.X., W.-Q.T., and Z.-Y.G. provided the MOF samples. Y.L., L.L., and D.Z. helped with the data collection. D.A.M. hypothesized and verified the effect of convergence semi-angles on ptychographic reconstruction under low-dose conditions. G.L. and Y.H. wrote the paper. C.C. and Z.L. gave precious suggestions on the projects. All authors contributed to the manuscript writing and the proofreading of the paper.

Competing interests

The authors declare no competing interests.

Additional information

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