

Multislice hollow ptychography for simultaneous atomic-layer-resolved three-dimensional structural imaging and spectroscopy

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Electron-matter interactions produce elastic and inelastic scattering, forming the basis for imaging and spectroscopy in electron microscopy. However, integrating electron energy-loss spectroscopy (EELS) with 4D-STEM and ptychography remains challenging due to detector geometry conflicts. Hollow ptychography introduced by Song *et al.*, which uses a central aperture to allow low-angle electrons into the EELS spectrometer, suffers from multiple scattering in thicker samples. We propose multislice hollow ptychography (MHP), which incorporates a multislice model to address dynamical scattering and enables robust high-resolution imaging from hollow diffraction patterns while supporting simultaneous EELS acquisition. MHP achieves sub-ångström lateral resolution at intermediate doses (10^4 – 10^8 e[−]/Å²) and enables full three-dimensional (3D) atomic-layer reconstruction at ultrahigh doses ($\geq 10^8$ e[−]/Å²), limited only by lattice vibrations. Of note, up to 70% of electrons remain available for spectroscopy. This flexible and dose-efficient framework supports correlative 3D imaging and chemical mapping, opening new possibilities for analyzing complex energy and quantum materials.

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I. INTRODUCTION

The fundamental principle of electron microscopy depends on electron-material interactions, which result in elastic scattering that changes electron paths without energy loss or inelastic scattering that transfers energy to material excitations, including phonons, plasmons, and electronic transitions [1]. Scanning transmission electron microscopy (STEM), when integrated with electron energy loss spectroscopy (EELS), utilizes both scattering channels to enable atomic-scale characterization of complex materials [2–5]. STEM high-angle annular dark field (STEM-HAADF) imaging detects elastically scattered electrons at high angles, producing Z-contrast images that are particularly sensitive to heavy elements [6,7]. Simultaneously, inelastically scattered electrons are energy-filtered by a spectrometer to provide rich spectroscopic information on local composition, bonding, and oxidation states [8]. This dual-modality capability makes the combination of STEM-HAADF and EELS one of the most powerful and widely used characterization

approaches for probing the structural and electronic properties of technologically significant systems, such as functional oxides [4,5] and battery materials [9,10].

STEM-HAADF imaging provides great advantages because of its incoherent imaging properties, including intuitive Z-contrast and robustness to dynamic scattering, but it has several intrinsic limitations. Integration over a high-angle annular detector eliminates phase information, which reduces its sensitivity to image light elements and weak scatterers [6,7]. This technique has low electron dose efficiency because only a small percentage of incident electrons scattered at high angles effectively contribute to image contrast, which restricts its application for dose-sensitive materials. The depth resolution of HAADF optical sectioning depends on the depth of focus, which extends between 10 and 30 Å based on probe convergence angle, thus making true atomic-layer-resolved depth resolution challenging [11,12]. Three-dimensional (3D) atomic resolution imaging relies on tilt-series tomography [13], which also substantially increases the total electron dose.

The recent development of detector technology [14,15] has led to 4D-STEM [16], which records full diffraction pattern throughout a two-dimensional raster of probe positions. Through this mode, one can virtually reconstruct traditional imaging modes, including bright field, annular bright field, HAADF, and differential phase contrast imaging (DPC) [17], as well as access powerful computational imaging techniques such as electron

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ptychography [18–20]. Unlike HAADF, ptychography reconstructs both amplitude and phase of the specimen complex transmission function using the interference encoded in overlapping diffraction patterns. This method experimentally provides superior spatial resolution beyond the conventional diffraction limit of the lenses [21,22], enhanced sensitivity to both light and heavy elements [23–26], improved dose efficiency [27–29], and cryogenic conditions [28,30], which benefits wider applications in materials characterization [31,32] and biological samples [28,33]. Furthermore, multislice ptychography [34] offers an advanced approach by explicitly modeling the depth-dependent evolution of the electron probe and its interaction with the sample. A proof-of-principle demonstration of multislice electron ptychography [35] showed that by accounting for electron wave propagation through thick specimens, it can suppress the dynamical effects that limit single-slice approaches (ePIE [36], WDD [20,25]). Latest work by Chen *et al.* [37] demonstrated that it can achieve lateral atomic-resolution limits set by lattice vibrations and an improved depth resolution of 3.9 nm in thick, strong scattering specimens, facilitating 3D reconstruction of structural variations [38–42]. The dependence of depth resolution on the maximum diffraction or collection angle was experimentally verified in recent multislice ptychography studies, such as O’Leary *et al.* [43].

These capabilities of 4D-STEM ptychography make it an attractive alternative for traditional STEM-HAADF and EELS workflows, particularly in studies requiring dose efficiency or light-atom imaging. However, the main obstacle in this application is its incompatibility with EELS acquisition because conventional ptychographic phase retrieval requires capturing the full diffraction pattern, including the intense low angle scattered electrons. However, the same low-angle electrons are typically needed for EELS, as they carry the inelastically scattered signal into the spectrometer. In conventional configurations, the fast pixelated detector [14,15], which records the entire electron wave field, blocks the entrance to the EELS spectrometer, thus preventing simultaneous structural and spectroscopic data acquisition. This fundamental competition between imaging and spectroscopy for the same portion of the beam limits the integration of ptychography into STEM-EELS workflows.

To address this limitation, Song *et al.* [44] proposed a new 4D-STEM microscope configuration with a hollow-type pixelated detector [Fig. 1(a)], enabling hollow ptychography as a promising strategy. This hollow detector excludes the low-angle region of the bright field disk in the diffraction pattern and redirects low-angle scattered electrons to the EELS spectrometer, while retaining the high-angle regions for various other types of structural imaging modalities, such as HAADF and DPC. When combined with ptychography, they demonstrated that high-resolution phase reconstructions remain robust for 2D monolayer

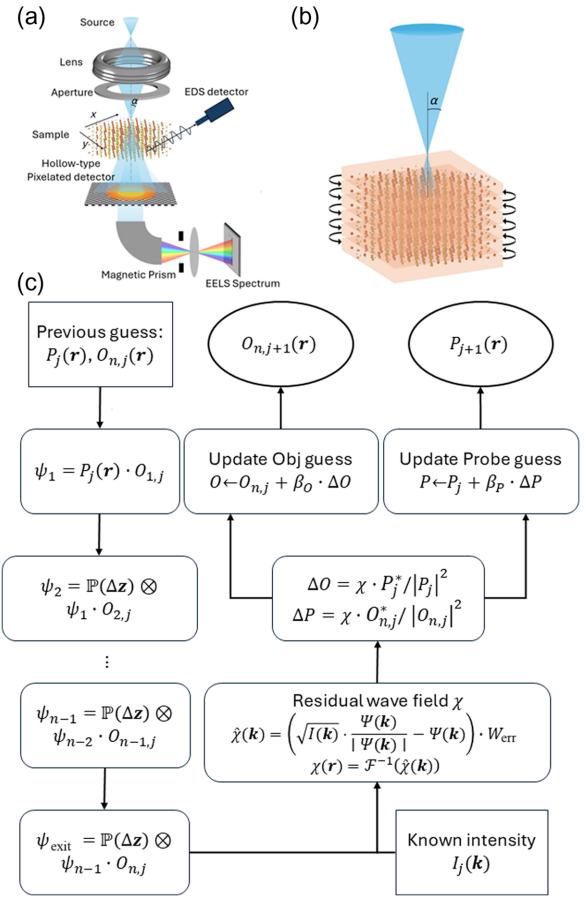


FIG. 1. Concept and algorithmic framework of MHP. (a) Schematic of the experimental configuration for MHP, (b) sectioning a thick sample into a set of thin slices using a multislice approach, and (c) flowchart of the multislice hollow ptychography reconstruction algorithm.

MoS₂, even with the exclusion of low-angle electrons using a single-slice reconstruction algorithm. By varying the semiangle of the hollow region, they demonstrated that sub-ångström resolution could still be achieved, including the resolution of atomic features separated by 91 pm. This work established a practical dose-partitioning strategy between 4D-STEM imaging and spectroscopy. The contrast of the atomic column remains stable up to the sample thickness of 4.6 nm for MoS₂ [44] and 5 nm for SrTiO₃ [45] bulk materials. However, contrast reversal was observed in thicker MoS₂ samples (7.1 nm), which was attributed to multiple scattering [44]. This suggests that single-slice hollow ptychography (SHP) is inherently limited in its ability to account for wave propagation through the sample thickness, resulting in degraded resolution and contrast and lack of 3D structural information.

Here, we develop a new imaging modality and an associated algorithm, termed multislice hollow ptychography (MHP), based upon a multislice approach [35], which enables accurate phase reconstruction from hollow-type diffraction patterns with a large fraction of the central

beam excluded. We further investigate whether hollow ptychography can handle multiple scattering with this approach [Fig. 1(b)]. Using an experimental ptychographic dataset of a strongly scattering perovskite oxide, 21-nm-thick PrScO_3 (PSO) reported in the literature [37] together with our simulations, we systematically evaluate both achievable lateral and depth resolution under different hollow semiangles (HSAs), total electron dose, and thermal lattice vibration conditions. The simulation results show that MHP enables adjustable control over lateral resolution, depth sectioning, and compatibility with EELS, which makes MHP a powerful platform for correlative 3D atomic-layer-resolved imaging and spectroscopy of complex materials.

II. RECONSTRUCTION ALGORITHM FOR MULTISLICE HOLLOW PTYCHOGRAPHY

The reconstruction in this work is based on an iterative least squares maximum likelihood (LSQML) method, modified from the PtychoShelves framework shown in Fig. 1(c) [46–50]. The algorithm alternates between simulating the forward model and refining the object and probe estimates based on a masked likelihood-based error metric.

For each scan position j , an exit wave

$$\psi_1 = P_j(\mathbf{r}) \cdot O_{1,j}(\mathbf{r}), \quad (1)$$

is calculated from the current probe $P_j(\mathbf{r})$ and object estimates for slice $O_{1,j}(\mathbf{r})$, where $\mathbf{r}$ denotes the transverse real-space coordinate. The wave field is then propagated sequentially through the multislice object along the beam direction according to

$$\psi_n = \mathbb{P}(\Delta z) \otimes \psi_{n-1} \cdot O_{n,j}, \quad (2)$$

where $\mathbb{P}(\Delta z)$ is the Fresnel free-space propagation operator over a slice thickness Δz , O_n is the transmission function of the n th slice, and $\otimes$ denotes convolution.

After propagation through all slices, the exit wave at the object plane ψ_{exit} , is Fourier-transformed to obtain the predicted diffraction wave function $\Psi(\mathbf{k}) = \mathcal{F}(\psi_{\text{exit}})$, where $\mathbf{k}$ denotes the reciprocal-space coordinate. The residual wave field χ is computed as,

$$\chi(\mathbf{k}) = \left(\sqrt{I(\mathbf{k})} \cdot \frac{\Psi(\mathbf{k})}{|\Psi(\mathbf{k})|} - \Psi(\mathbf{k}) \right) \cdot W_{\text{err}}(\mathbf{k}), \quad (3)$$

$$\chi(\mathbf{r}) = \mathcal{F}^{-1}(\tilde{\chi}(\mathbf{k})), \quad (4)$$

where $\mathcal{F}^{-1}$ is the inverse Fourier transform and W_{err} is a reciprocal-space weight matrix used to exclude unmeasured regions of the diffraction pattern,

$$W_{\text{err}}(\mathbf{k}) = \begin{cases} 0, & |\mathbf{k}| < k_{\text{HSA}}, \\ 1, & |\mathbf{k}| > k_{\text{HSA}}, \end{cases} \quad (5)$$

where k_{HSA} corresponds to the HSA.

This ensures that the residual χ is computed only within the measured regions and that no updates are driven by unmeasured regions.

The residual wave field χ serves as the gradient signal for updating both the object and probe. The update directions in real space are given by

$$\Delta O = \chi \cdot P_j^* / |P_j|^2, \quad (6)$$

$$\Delta P = \chi \cdot O_{n,j}^* / |O_{n,j}|^2. \quad (7)$$

where $*$ denotes the complex conjugation. These directions are then refined using overlap-consistency constraints and a least-squares criterion to determine optimal update step sizes β_O and β_P . The refined step sizes can be obtained either by solving a local linear system or by a simplified projection method. The final updates are applied as

$$O \leftarrow O + \beta_O \cdot \Delta O, \quad (8)$$

$$P \leftarrow P + \beta_P \cdot \Delta P. \quad (9)$$

In the case of multilayer objects, the residual wave field is back-propagated layer by layer, and the object updates are applied separately for each layer. The probe is updated only from the first layer to maintain stability.

Optionally, the algorithm includes mechanisms for refining experimental parameters such as probe position, detector rotation, and Fourier space misalignment. Furthermore, a momentum acceleration scheme may be employed, where update directions from previous iterations are accumulated to improve convergence.

III. ROBUST HIGH-RESOLUTION IMAGING FOR STRONGLY SCATTERING MATERIALS

PSO, a complex perovskite oxide previously studied in the work [37], presents a challenging system for assessing the effectiveness of ptychography under conditions of strong multiple scattering. In this study, we analyzed the same 4D-STEM data [37] from a 20 nm [001]-oriented PSO crystal to test hollow ptychography by masking the central diffraction region. The HSA was systematically increased from 0 to 0.5α , 0.75α , and 0.95α [Figs. 2(c)–(f)] to assess the balance between structural imaging capability and the potential for simultaneous EELS mapping. The reconstruction method in this work is based on an iterative LSQML method, modified from the PtychoShelves framework [46]. Details of the dataset can be found in Sec. I in Supplemental Material [51].

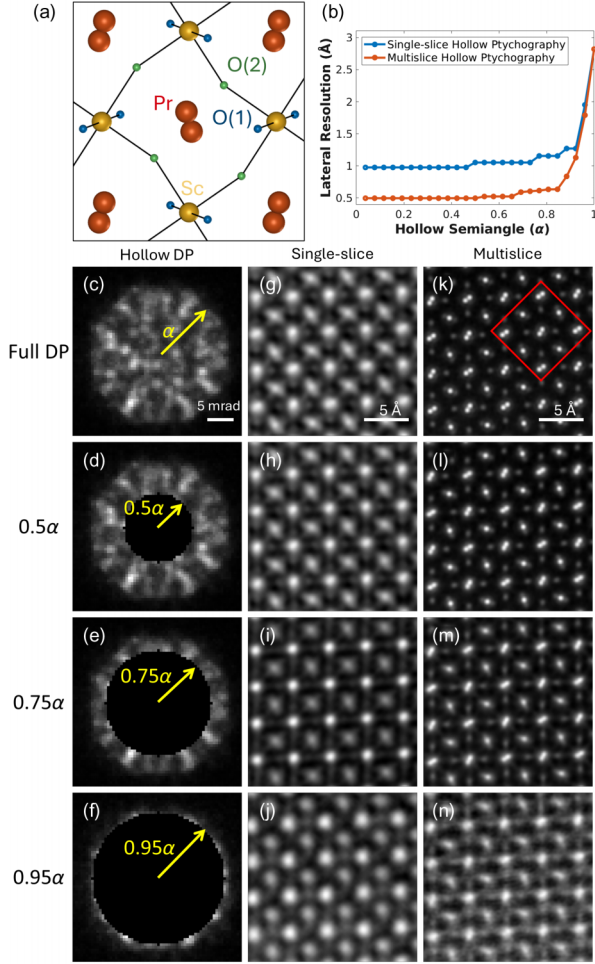


FIG. 2. Evaluation of lateral resolution under varying hollow semiangle (HSA) in single-slice (SHP) and multislice (MHP) hollow ptychography. (a) Projected atomic model of PrScO_3 along the $[001]$ direction. (b) Quantitative comparison of lateral resolution as a function of HSAs for SHP (blue dotted dashed lines) and MHP (red dotted dashed lines). (c) Experimental diffraction pattern acquired with a full detector (0% HSA). (d)–(f) Masked diffraction patterns generated from the experimental data in (c) by applying hollow masks corresponding to $\text{HSA} = 0.5\alpha$, 0.75α , and 0.95α , respectively, to emulate hollow detector geometries. Reconstructed phase images of PrScO_3 using SHP (g)–(j) and MHP (k)–(n) with corresponding diffraction patterns in (c)–(f).

SHP reconstructions revealed its limitations in these thicker specimens, consistent with previous work [44]. Notably, closely spaced Pr atomic pairs (Fig. 2(a) and Fig. S1 in Supplemental Material [51]) became indistinguishable and merged into single, large intensity peaks. As the HSA increased, the reconstructed phase images [Figs. 2(g)–(j)] and their corresponding power spectra (Fig. S2 in Supplemental Material [51]) displayed decreasing contrast, feature blurring, and inaccurate localization of atomic columns. Sc atoms could not be clearly resolved from surrounding oxygen atoms of configuration 2 labeled

as O(2) at HSAs beyond 0.5α . The oxygen-column signals of configuration 1, labeled as O(1) located between Pr and Sc sites became increasingly obscured due to the enlarged apparent radii and inaccurate intensities of the adjacent heavier atoms. The quantitative trends in lateral resolution, plotted in Fig. 2(b), show that beyond 0.5α hollow radius, the lateral resolution in SHP reconstructions rapidly deteriorates, ultimately exceeding $1 \text{ \AA}$.

In contrast, the MHP reconstructions maintained high spatial frequency information even as the HSA increased. The MHP reconstructions [Figs. 2(k)–(n)] and their corresponding power spectra (Fig. S3 in Supplemental Material [51]) showed that even at large HSAs (up to 0.75α), the atomic lattice of PSO remained well-resolved and accurately reconstructed. The atomic dumbbells of Pr, previously merged in the SHP reconstructions, were clearly resolved as distinct atoms. Sc atoms were separated from surrounding oxygen, O(1), and the O(2) columns located between Pr and Sc were better resolved, exhibiting enhanced contrast and more accurate localization. These images showed excellent agreement with the expected $x - y$ atomic structure model [Fig. 2(a)], resolving fine details that single-slice reconstructions failed to preserve. The resolution trend in Fig. 2(b) confirms that in the multislice approach, lateral resolution degrades only slightly up to approximately 0.75α , with more significant decline beyond approximately 0.90α . Notably, MHP consistently outperforms SHP across all HSAs, despite reduced data redundancy (Fig. S4(b) in Supplemental Material [51]) caused by the missing central region in the diffraction patterns (Figs. S3(a)–S3(d) in Supplemental Material [51]). These findings demonstrate for the first time that, despite the partial absence of central beam information, hollow ptychography can still be effectively integrated with the multislice approach, offering a powerful resolution to overcome resolution degradation caused by multiple scattering in thick specimens. The ability of the multislice algorithm to accommodate large HSAs up to 0.75 – 0.90α , while maintaining atomic resolution highlights its suitability for correlative structural and chemical mapping in complex materials systems like transition metal oxides [3–5,58], where simultaneous EELS mapping and high-resolution structural imaging are typically required.

IV. DOSE-DEPENDENT MHP RECONSTRUCTIONS

Understanding the influence of electron dose is significant when performing simultaneous EELS mapping, because the total dose must be divided between ptychographic imaging and spectroscopic acquisition. To investigate the influence of electron dose on MHP reconstructions, simulations of 4D-STEM datasets from a 20 nm PSO crystal slab were performed using abTEM [59] at 300 kV with a convergence semiangle (CSA) of 21.4 mrad , under total electron dose

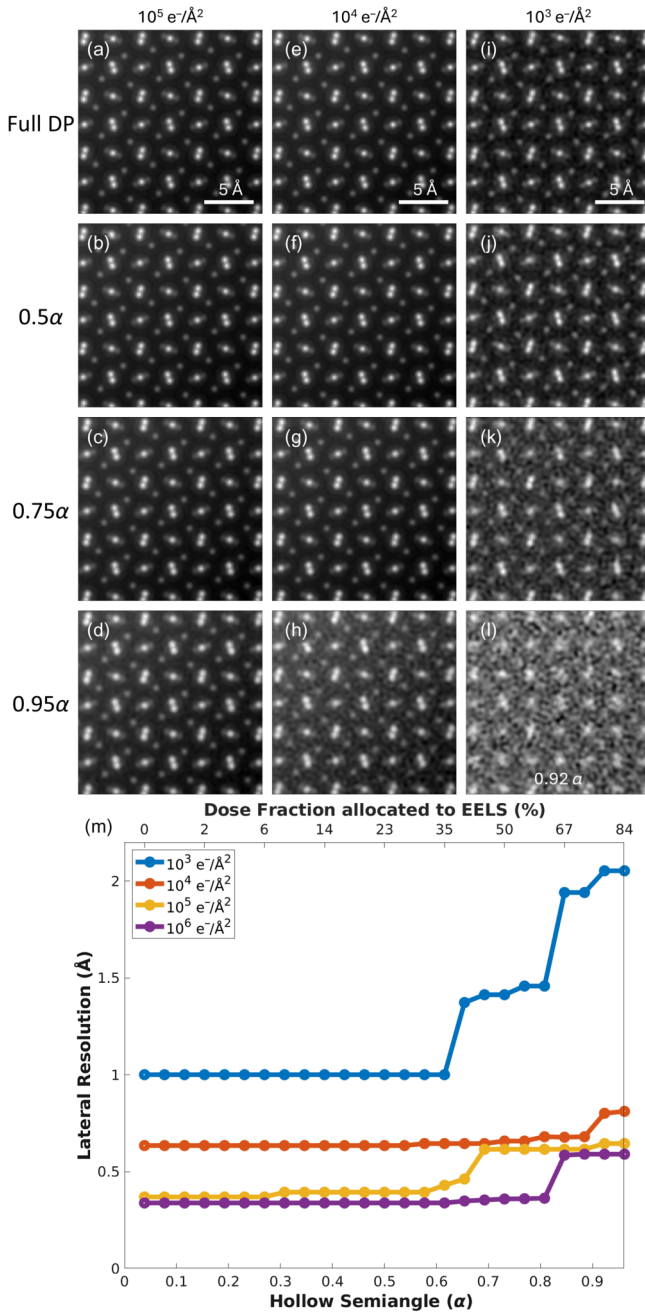


FIG. 3. Effect of dose and HSA on simulated MHP reconstructions. Phase reconstructions at electron doses of 10^5 (a)–(d), 10^4 (e)–(h), and 10^3 (i)–(l) $\text{e}^-/\text{\AA}^2$, with increasing HSAs from 0 to 0.95α for 10^5 and 10^4 $\text{e}^-/\text{\AA}^2$ and from 0 to 0.92α for 10^3 $\text{e}^-/\text{\AA}^2$. (m) Lateral resolution of the reconstructed phases as a function of HSA (bottom x axis) and the corresponding percentage of the total dose allocated to EELS acquisition (top x axis), for doses of 10^3 (blue dotted dashed lines), 10^4 (red dotted dashed lines), 10^5 (yellow dotted dashed lines), and 10^6 (purple dotted dashed lines) $\text{e}^-/\text{\AA}^2$.

conditions of 10^5 , 10^4 , and 10^3 $\text{e}^-/\text{\AA}^2$. HSAs were set at 0, 0.5α , 0.75α , and 0.95α . Details of the simulation parameters are provided in Secs. II and III in Supplemental Material [51].

The reconstructed phases (Fig. 3) and corresponding power spectra (Fig. S5 in Supplemental Material [51]) show the performance of MHP reconstructions across these dose regimes. At a dose of 10^5 $\text{e}^-/\text{\AA}^2$ or above [Figs. 3(a)–3(d)], reconstructions exhibit strong, sharp phase contrast of all atomic columns, including challenging oxygen sites between Pr and Sc. High-order reflections were consistently recovered in power spectra (Figs. S5(a)–S5(d) in Supplemental Material [51]), and even at large HSAs up to 0.95α , the atomic lattice remains fully resolved. At 10^4 $\text{e}^-/\text{\AA}^2$ [Figs. 3(e)–3(h)], reconstructions maintain good phase contrast and resolution (Figs. S5(e)–S5(g) in Supplemental Material [51]) up to 0.75α HSA. Fine details such as closely spaced Pr atomic dumbbells and oxygen columns are clearly recovered, although slight degradation in contrast is observed beyond this HSA. These intermediate dose reconstructions highlight a practical balance point, delivering excellent structural resolution while minimizing total electron exposure. At the lowest dose of 10^3 $\text{e}^-/\text{\AA}^2$ [Figs. 3(i)–3(l)], reconstructions show reduced contrast and increased noise, particularly at HSAs beyond 0.5α , where heavier atomic columns become blurred and oxygen columns are difficult to resolve.

These dose-dependent trends in lateral resolution are quantitatively summarized in Fig. 3(m), which plots the resolution as a function of HSAs (bottom x axis) and the corresponding percentage of the total electrons contributing to EELS (top x axis). During the estimation of EELS fraction, the electrons scattered outside of the bright-field disc were considered. At the doses of 10^5 $\text{e}^-/\text{\AA}^2$ or above, lateral resolution remains consistently at sub-ångström levels, even up to an HSA of 0.95α , which corresponds to 70% of the total electrons contributing to EELS. At 10^4 $\text{e}^-/\text{\AA}^2$, the resolution remains around 0.7 Å up to approximately 0.9α HSA (corresponds to 75% of the electrons being directed to EELS), with only moderate degradation at larger HSA. However, at the low-dose condition of 10^3 $\text{e}^-/\text{\AA}^2$, lateral resolution of 1 Å deteriorates rapidly beyond 0.6α (approximately 35% of the total electrons to EELS), ultimately exceeding 2 Å at the largest hollow regions. These results highlight the flexibility of MHP, enabling tailored trade-offs between imaging resolution and EELS signal strength. The approach is thus well suited for general-purpose, dose-efficient 2D structural and chemical mapping across a broad spectrum of material systems, even beam-sensitive materials [60,61].

V. LATTICE-VIBRATION-DEPENDENT 3D MHP OPTICAL SECTIONING OF A CRYSTAL

The capability to resolve the 3D spatial arrangement of atoms within materials along the beam direction has been a long-standing goal for optical sectioning techniques in STEM, such as HAADF [11,12] and more recently, ptychography [35,37]. To evaluate the intrinsic depth resolution of MHP, we first conducted simulations using

a single Pr atom in vacuum without thermal lattice vibrations (TLV) under various CSAs and HSAs and evaluated the corresponding information transfer of MHP. (Details are provided in Sec. V in Supplemental Material and Figs. S6 and S7 in Supplemental Material [51]). The results show that MHP accesses substantially larger regions of spatial frequency domain than STEM-HAADF in both lateral and depth directions and that increasing CSA systematically extends the information transfer to high spatial frequencies in both directions. The results show that increasing CSA from 20 to 80 mrad significantly improves the depth resolution, from approximately 18.5 to approximately 2.8 Å. Increasing HSA up to approximately 0.75α has only a modest impact on the transfer function, while extremely large hollow angle ($\text{HSA} \gtrsim 0.95\alpha$) leads to a pronounced loss of the information transfer in both lateral and axial directions, resulting in the deterioration of attainable spatial resolution. These findings confirm that depth resolution in MHP is primarily determined by CSA and is largely independent of the hollow geometry under typical conditions, except in the large HSA regime.

The feasibility of atomic-layer 3D reconstruction of a crystalline sample in different TLV conditions was investigated using MHP at a large CSA. To evaluate the MHP performance under strong scattering conditions, simulations were conducted on a [001]-oriented PSO crystal. In this orientation, the distorted perovskite oxide has alternating layers of Pr, Sc, and O atoms along the beam direction (Fig. S1 in Supplemental Material [51]). The interplanar distances vary significantly: approximately 8 Å for Pr-Pr, approximately 4 Å for Sc-Sc, and both approximately 9 and approximately 3 Å for two distinct configurations of O-O layers, labeled O(1) and O(2), respectively (Figs. S1(d)–S1(f) in Supplemental Material [51]). These spacings and the type of atoms set critical thresholds for depth resolution. To meet this criterion, a large probe CSA of 80 mrad capable of achieving a theoretical depth resolution of 2.8 Å (Fig. S6(e) in Supplemental Material [51]) was used. Thermal atomic motion at 300 K (room temperature), 90 K (liquid nitrogen), and 4 K (liquid helium) was accounted for using the frozen phonon approach [62,63]. The MHP reconstruction itself was performed directly from the raw 4D-STEM data without imposing structural constraints from the known PSO atomic model. Details of simulation parameters are provided in Sec. VI in Supplemental Material [51].

To investigate the influence of TLV on MHP reconstructions, we compared the reconstructed phase at three Z sections from a PSO crystal at temperatures of 300 K (room temperature), 90 K (liquid nitrogen), and 4 K (liquid helium) using the full diffraction pattern ($\text{HSA} = 0\alpha$) at a dose of $10^8 \text{ e}^-/\text{\AA}^2$ [Figs. 4(a)–(i)]. At 300 K, only atomic columns with a large distance of 8–9 Å, such as Pr [Fig. 4(a)] and O(1) [Fig. 4(b)] columns, can be clearly

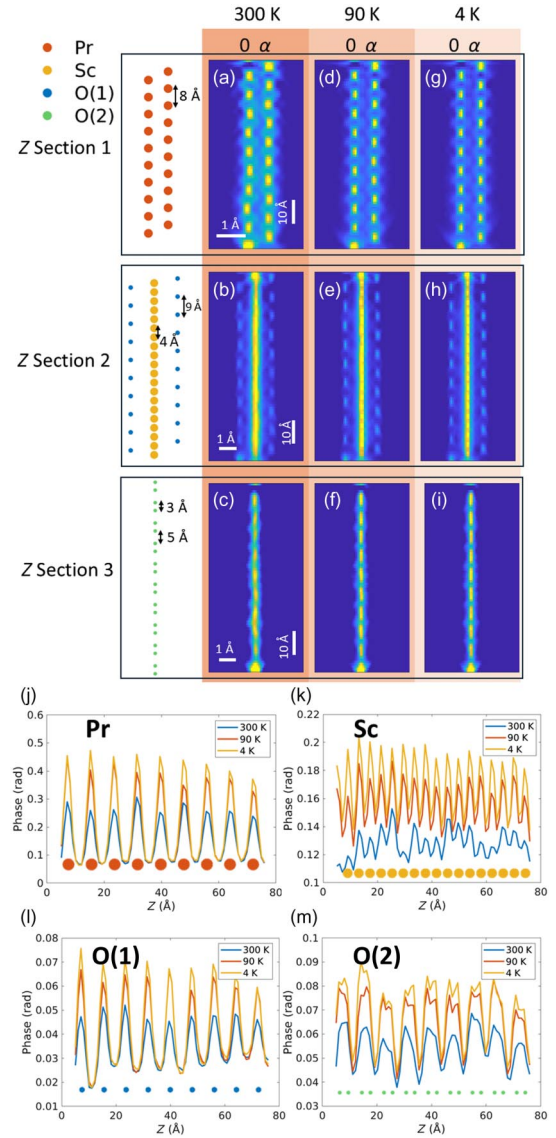


FIG. 4. Thermal-lattice-vibration-dependent 3D MHP optical sectioning of a PrScO_3 crystal with a CSA of 80 mrad using full simulated diffraction patterns ($\text{HSA} = 0\alpha$) at a dose of $10^8 \text{ e}^-/\text{\AA}^2$. (a)–(i) Z sections extracted from three representative positions (labeled 1–3 in a unit cell, as shown in Fig. S1(b) in Supplemental Material [51]) in the 3D reconstructions of PrScO_3 at 300, 90, and 4 K, respectively. (j)–(m) Depth profiles extracted along atomic columns of Pr, Sc, O(1), and O(2), respectively, under different temperature conditions as shown in (a)–(i).

resolved. The heavier Sc columns with a 4 Å spacing become indistinguishable, as indicated by the 300 K profile in Fig. 4(k). For the O(2) columns, the O-O dumbbell pairs separated by approximately 5 Å are visible, whereas the individual O atoms within dumbbells are not fully separated under the dose condition used here [Figs. 4(c) and 4(m)].

At 90 K [Figs. 4(d)–(f)] and 4 K [Figs. 4(g)–(i)], the Pr, Sc, and O(1) atomic layers become clearly resolved along

the depth direction. For the O(2) configuration, the O-O dumbbell pairs are clearly distinguishable, although the individual O atoms within some dumbbells are not fully separated under the slice sampling and dose conditions used in Fig. 4. Additional high-dose simulations ($10^{10} \text{ e}^-/\text{\AA}^2$) and corresponding depth line profiles are provided in Fig. S8 in Supplemental Material [51], demonstrating that the individual O atoms within the O(2) dumbbells can in principle be resolved when sufficient signal-to-noise ratio is available. The line profiles in Figs. 4(j)–(m) show that as temperature decreases, reduced TLV strongly enhances ptychographic phase signal and depth sectioning performance. This improvement can be attributed to the fact that atomic vibrations are suppressed at lower temperatures, leading to a sharper and tighter atomic potential. This greatly enhances the coherence of electron scattering and results in stronger, more localized phase signals. Therefore, reduced TLV at cryogenic temperatures directly improves the quality of 3D optical sectioning and enables atomic-layer 3D reconstruction of all atomic species including light atoms such as oxygen, using multislice ptychography.

VI. DOSE-DEPENDENT 3D MHP OPTICAL SECTIONING OF A PrScO_3 CRYSTAL UNDER CRYOGENIC CONDITIONS

In this section, we investigate the atomic-species-dependent phase contrast and depth localization behavior in MHP by quantitatively analyzing the reconstructed peak intensities and atomic-position accuracies as functions of HSA and temperature. Figure 5 summarizes the phase-intensity and localization behavior of different atomic species as functions of HSA, while Figs. S9–S15 in Supplemental Material [51] present additional reconstruction examples and supporting quantitative analyses.

Reconstructions were carried out by varying the HSA from 0 to 0.95α and examining the effects of four different electron doses: 10^8 , 10^7 , 10^6 , and $10^5 \text{ e}^-/\text{\AA}^2$ (Figs. S11 and S12 in Supplemental Material [51]) at 90 K as an example, although slightly better performance can be expected under 4 K conditions for PSO (Figs. S13 and S14 in Supplemental Material [51]). The corresponding results at 300 K show the same overall trends but with somewhat degraded reconstruction quality, particularly at lower doses (Figs. S9 and S10 in Supplemental Material [51]). At the highest dose of $10^8 \text{ e}^-/\text{\AA}^2$ or above, reconstructions remained robust across the full HSA range (Figs. S11(a)–S11(l) in Supplemental Material [51]). All atomic layers, Pr, Sc, and both configurations of O(1) and O(2), were clearly resolved along the depth direction, even when the central beam was excluded up to $\text{HSA} = 0.97\alpha$ [Figs. 5(d)–5(f)]. The Pr and Sc columns showed strong peaks, while both oxygen configurations showed distinguishable depth features despite their lower scattering strengths (Figs. S15(a)–S15(d) in Supplemental

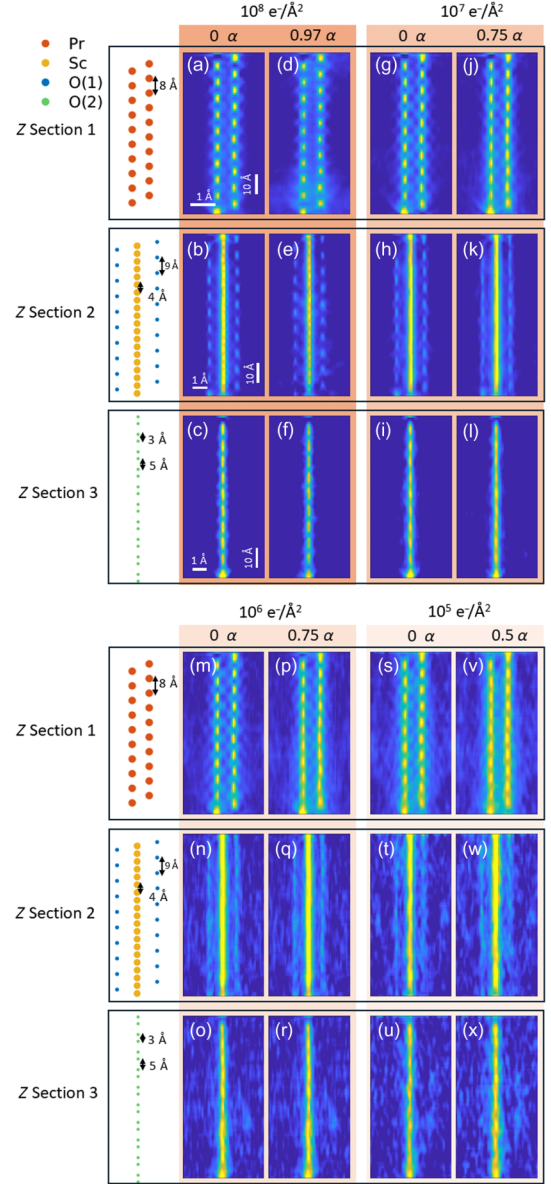


FIG. 5. Simulated 3D MHP optical sectioning of a PrScO_3 crystal with a CSA of 80 mrad at 90 K under varying electron dose and HSA conditions. (a)–(f) Z sections extracted from three representative positions (labeled 1–3 in a unit cell, as shown in Fig. S1(b) in Supplemental Material [51]) in the 3D reconstruction of PrScO_3 at a dose of $10^8 \text{ e}^-/\text{\AA}^2$ with HSAs of 0α (a)–(c) and 0.97α (d)–(f). Reconstructions at lower doses: $10^7 \text{ e}^-/\text{\AA}^2$ with HSAs of 0α (g)–(i) and 0.75α (j)–(l); $10^6 \text{ e}^-/\text{\AA}^2$ with HSAs of 0α (m)–(o) and 0.75α (p)–(r); and $10^5 \text{ e}^-/\text{\AA}^2$ with HSAs of 0α (s)–(u) and 0.5α (v)–(x), respectively.

Material [51]). At $10^7 \text{ e}^-/\text{\AA}^2$ (Figs. S11(m)–S11(x) in Supplemental Material [51]), reconstructions of the Pr columns, as well as O(1) with its wide 9 Å spacing, remained stable for HSAs up to 0.75α [Figs. 5(j) and 5(k)]. However, the Sc and O(2) columns [Figs. 5(k) and 5(l)], with a much narrower spacing of approximately 3–4 Å, became increasingly difficult to resolve. Even with the full

diffraction pattern ($HSA = 0$, as shown in Fig. 4), the weak scattering signal and tight depth packing prevented these layers from being recovered as distinct maxima (Figs. S15(e)–S15(h) in Supplemental Material [51]). The loss of resolution in this case suggests the limits imposed by both signal-to-noise ratio and the intrinsic depth response width of the reconstruction. The Pr and O(1) layers remained resolvable up to $HSA = 0.75\alpha$ at $10^6 \text{ e}^-/\text{\AA}^2$ [Figs. 5(p) and 5(q)], due to their large $8 \text{ \AA}$ separation. When the dose was reduced to $10^5 \text{ e}^-/\text{\AA}^2$, only the Pr atomic layers remained resolvable for HSAs up to 0.5α [Fig. 5(v)]. Their strong scattering signal and wide interplanar spacing allowed them to be sufficiently distinguishable along the beam direction, even under dose-limited and hollow conditions. Sc and both O configurations were no longer distinguishable [Figs. 5(w) and 5(x)]. Phase retrieval under these conditions produced noisy and elongated features, with no clear maxima corresponding to atomic columns in the depth profiles (Figs. S15(n)–S15(p) in Supplemental Material [51]). The results show that atomic-layer depth sectioning with MHP at low temperature depends on multiple factors: probe CSA, HSA, total electron dose, atomic number, and interplanar spacing. At a sufficiently large CSA (e.g., 80 mrad), all atomic species in PSO can be resolved in 3D at doses $\geq 10^8 \text{ e}^-/\text{\AA}^2$ with $HSA \leq 0.75\alpha$. For light atoms in closely spaced layers, resolvability is more dose-sensitive and HSA needs to be minimized.

It is significant to note that the phase value of reconstructed atomic peaks varies between different atomic species, as previously shown in the literature such as LaB_6 [23] and MoS_2 [44]. In PSO, Pr columns exhibit the strongest phase contrast, followed by Sc, while O columns produce the weakest signal. This is consistent with trends observed in previous work [37]. This relationship remains consistent throughout the entire range of HSAs examined [Fig. 6(a)], reflecting the dependence of phase contrast on atomic number and scattering strength. Further details of the standard deviation calculations are provided in Sec. VII in Supplemental Material [51].

Therefore, peak detection algorithms can be adapted to account for species-specific contrast. As shown in Fig. 6(b), the 3D localization of Pr, Sc, and O atoms can be achieved by applying element-specific detection thresholds to the reconstruction at the dose of $10^8 \text{ e}^-/\text{\AA}^2$, without the use of reference atomic positions from the structural model. Nearly all 100 Pr atoms and 80 O(1) atoms are successfully identified, as shown in Fig. S17 in Supplemental Material [51]. For Sc and O(2), approximately 4–5 atoms out of 80 are not detected because of the small interatomic spacing between neighboring atomic columns. Most of the missed atoms are located near the crystal surfaces, where the reconstruction quality is lower than in the interior of the specimen. Further details of the peak-detection procedure are provided in Sec. VIII in Supplemental Material [51].

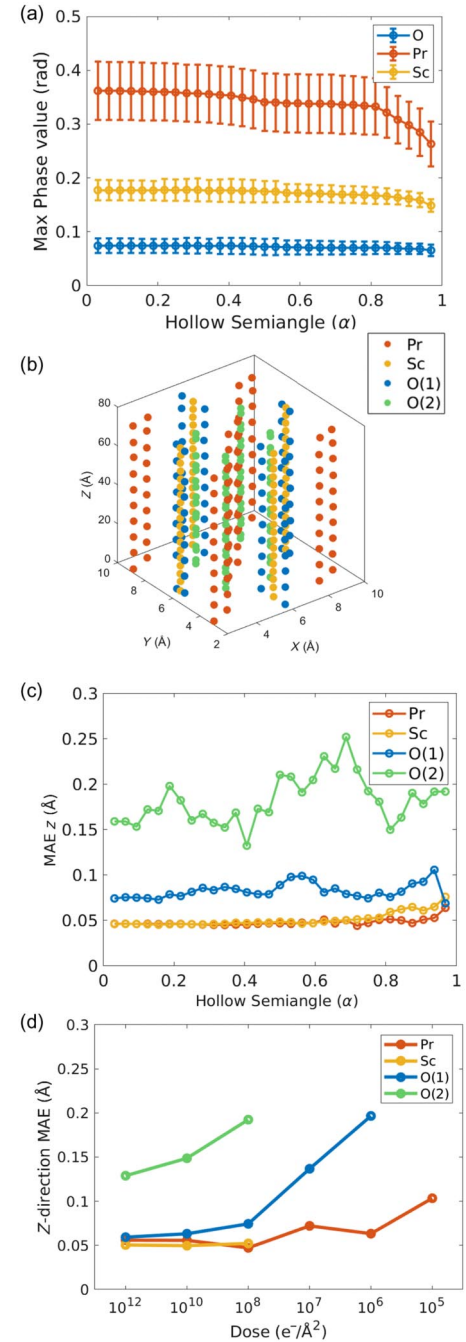


FIG. 6. Quantitative evaluation of phase signal strength and full 3D atomic positions of PrScO_3 from ptychographic reconstruction with a CSA of 80 mrad at 90 K. (a) Maximum phase values of Pr (red dotted dashed lines), Sc (yellow dotted dashed lines), and O (blue dotted dashed lines) atoms as a function of HSA (α). Error bars indicate the standard deviations of the maximum reconstructed phase intensities measured from multiple atomic columns of the same species within a single reconstructed volume. (b) Reconstructed full 3D atomic positions of PrScO_3 visualized along x , y , and z axes at a dose of $10^8 \text{ e}^-/\text{\AA}^2$ ($HSA = 0.75\alpha$). (c) Mean absolute error (MAE) in the Z direction for different atomic species as a function of HSA at a dose of $10^8 \text{ e}^-/\text{\AA}^2$, and (d) Z -direction MAE as a function of electron dose at an HSA of 0.75α .

The parameters can be adjusted based on expected signal strength for each atomic species to localize full 3D atomic positions under imaging conditions of $\geq 10^8 \text{ e}^-/\text{\AA}^2$. To quantify the depth accuracy, the MAE in the Z direction was calculated for different atomic species as a function of HSAs [Fig. 6(c)] and the total electron dose [Fig. 6(d)]. In Fig. 6(c), at a high dose of $10^8 \text{ e}^-/\text{\AA}^2$, heavier Sc and Pr columns exhibit consistently lower and more stable MAE values (approximately equal to 0.05–0.07 Å) over a wide range of HSAs. For the lighter element columns, O(1) remains relatively stable at approximately 0.08–0.1 Å over the low-HSA range, but increases slightly beyond about 0.5α . In contrast, O(2) exhibits consistently larger MAEs, ranging from approximately 0.12 to 0.25 Å. This reduced localization accuracy is attributed to the weaker scattering contrast of O atoms, the small separation between neighboring O(2) sites, and the asymmetric spacing of adjacent O(2) layers along the depth direction, which make peak localization more challenging. This behavior suggests a higher robustness of depth localization for heavy elements.

Figure 6(d) shows the evolution of the Z -direction MAE as the total electron dose decreases from 10^{12} to $10^5 \text{ e}^-/\text{\AA}^2$ at a fixed HSA of 0.75α . For all atomic species, the MAE increases gradually as the dose is reduced. Pr atoms remain identifiable over the entire simulated dose range from 10^{12} down to $10^5 \text{ e}^-/\text{\AA}^2$. This superior performance arises from both their larger interatomic spacing (approximately 8 Å) and their stronger scattering power, which together yield the best attainable depth resolution and precision. O(1) columns can be accurately localized at doses down to approximately $10^8 \text{ e}^-/\text{\AA}^2$, with MAEs remaining below approximately 0.08 Å. As the dose is reduced further, the localization error increases substantially, reflecting the decreasing signal-to-noise ratio. The O(2) and Sc columns can be robustly localized only at doses above approximately $10^8 \text{ e}^-/\text{\AA}^2$. For the oxygen columns, this limitation primarily arises from their weak scattering strength and correspondingly lower signal-to-noise ratio. In the case of Sc, the neighboring atomic layers are separated by only approximately 4 Å along the depth direction, leading to overlapping axial line profiles and preventing reliable peak localization at lower doses.

For all species, whenever atomic columns can be reliably identified, the corresponding MAE remains relatively stable. These observations indicate that the identifiability of atomic columns is jointly determined by atomic number and interatomic spacing. Taken together, these results demonstrate that both the electron dose and the hollow-cone semiangle jointly determine the attainable lateral and depth precision in multislice hollow ptychography. MHP therefore provides a favorable balance between atomic-layer-resolved three-dimensional imaging and high EELS signal collection, enabling dose-efficient

correlative analysis for practical applications requiring both structural imaging and spectroscopy.

VII. DISCUSSION

The ability to simultaneously achieve sub-ångström lateral resolution and atomic-layer-resolved depth sectioning is central to the development of next-generation 3D electron imaging techniques [64]. Our results demonstrate that MHP offers a flexible, dose-dependent framework for correlative structural and chemical mapping, particularly in thick, strongly scattering materials like PSO. By iteratively applying this masked error-driven update process, the LSQML algorithm effectively recovers both the object and the probe function, even in the presence of missing hollow information in the diffraction patterns.

In the low dose regime ($< 10^3 \text{ e}^-/\text{\AA}^2$), which is relevant for beam-sensitive materials, such as zeolites, solid electrolytes, and organic-inorganic perovskites, MHP reconstructions remain feasible and lateral resolution is around 1 Å up to approximately 0.5α , while allowing approximately 50% of the dose to be directed to EELS. However, beyond this HSA, lateral resolution degrades rapidly ($> 2 \text{ Å}$), and noise dominates lighter atomic columns. In the intermediate dose regime (10^4 – $10^6 \text{ e}^-/\text{\AA}^2$), applicable to systems such as lithium battery cathodes and in the high dose regime (10^7 – $10^8 \text{ e}^-/\text{\AA}^2$) typical for functional oxides or semiconductors, lateral resolution remains better than 0.7 Å up to approximately 0.75 – 0.95α , enabling over 70% of total dose to be used for EELS. This capability supports high-resolution, high-contrast 2D structural imaging with ptychography together with correlative chemical analysis, such as elemental identification and electronic structure studies using core-loss EELS or energy-loss near-edge structure (ELNES) [3–5,8]. However, atomic-layer-by-layer 3D reconstruction remains challenging at these dose levels. To resolve features such as atomic layers, dopants, and defects in depth, their separation must exceed the theoretical depth resolution, typically by at least a factor of 2, as demonstrated in the case of PSO.

In the ultrahigh dose regime ($\geq 10^8 \text{ e}^-/\text{\AA}^2$), permitted in highly stable, radiation-tolerant samples such as oxides and metals, full atomic-layer-resolved reconstructions become achievable with the ultimate limit set by TLV. It is worth noting that the total electron doses required for multislice ptychography to achieve true 3D atomic resolution (typically $\geq 10^7$ – $10^8 \text{ e}^-/\text{\AA}^2$) remain higher than those used in AET and ptychographic AET. For instance, Miao *et al.* achieved 3D atomic reconstructions at approximately $1.3 \times 10^6 \text{ e}^-/\text{\AA}^2$ [13], while Chang *et al.* reported pAET results at around $8.7 \times 10^5 \text{ e}^-/\text{\AA}^2$ [65]. However, unlike tilt-based tomography, MHP does not require multiple high-precision tilt acquisitions and long reconstruction times, yet achieves comparable or improved lateral and depth resolutions using a single dataset. This

enables faster data turnover and a simpler experimental implementation, especially beneficial for *in situ* and correlative spectroscopic studies. Most significantly, MHP enables correlative atomic-resolution EELS to be acquired from the same orientation, whereas in HAADF-AET tilt tomography the per-tilt EELS dose is typically too low to yield atomic-scale spectroscopic information.

Under cryogenic conditions, both lateral and depth resolutions reach their theoretical limits of approximately 0.5 Å laterally and approximately 2.9 Å in depth across a wide HSA range up to 0.95α . In this regime, all atomic planes, including closely spaced light-element layers, can be accurately recovered without compromising compatibility with EELS. Apart from the aforementioned use in core-loss or ELNES, this regime also could provide optimal electron counts for high-quality extended energy-loss fine structure analysis, which enables high signal-to-noise ratio measurements of bonding environments and coordination shells [66,67].

Therefore, MHP can be adjusted to match diverse experimental goals by tuning dose, convergence angle, HSA, and operating temperature. Its ability to provide atomic-layer-resolved imaging makes it particularly well-suited for investigating structurally and chemically complex materials, including functional oxides, heterostructures, and other advanced material systems. These include local chemical and structural disorders associated with point defects, dopants, and dislocation cores. The method is also applicable to amorphous regions, strain fields, and interfacial atomic configurations in heterostructures, superlattices, and grain boundaries with compositional modulation.

As the depth resolution of multislice ptychography is largely sensitive to lattice vibrations, the use of highly stable, low-drift *in situ* cryo-holders [68–72] operating at liquid-nitrogen, liquid-helium, or variable-temperature provides essential advantages, including phonon suppression and radiation damage reduction. In addition, given the demonstrated sensitivity of ptychographic phase contrast to spin [73,74] and charge [75,76], multislice ptychography, including the MHP approach, has the potential to emerge as a powerful new method to probe the lattice, spin, and charge degrees of freedom in three dimensions at the atomic level. Combined with simultaneous spectroscopic capabilities, this approach offers a powerful characterization platform for studying quantum phenomena [77] in condensed matter systems at low temperatures, including ferroic ordering, charge-order transitions, and superconductivity.

Although multislice ptychography shows strong potential for atomic-scale 3D characterization, the practical realization of atomic-layer-resolved imaging under cryogenic conditions remains constrained by current experimental hardware. Currently, even for fast-scanning HAADF imaging, the attainable lateral and depth resolution at cryogenic temperatures remains limited by the residual instabilities of *in situ* cryo-holders. State-of-the-art

low-drift cryogenic platforms, including H-bar [78] and CondensZero [79], have enabled a recent demonstration of atomic-resolution ptychography at liquid-helium temperatures [30]. Nevertheless, these systems still exhibit residual mechanical and thermal drift on the order of approximately equal to 0.25 Å/s or higher. For a typical 4D-STEM acquisition comprising 176×176 probe positions, a detector operating at the current practical speed of approximately equal to 7500 frames/s (e.g., PN detectors) [80] requires several seconds to complete a single scan, during which such drift accumulates to ångström-scale probe-sample motion. This accumulated drift blurs the interference fringes in the diffraction patterns, degrades phase stability, and ultimately limits the achievable depth precision, particularly for atomic-layer-resolved reconstructions. Additional constraints arise from vibrations induced by liquid-helium flow, limited mechanical stiffness of the cold stage, and thermal gradients across the specimen, all of which introduce scan distortions and defocus variations. As a result, the experimentally attainable depth resolution is expected to fall short of the simulated limit predicted for ideal, drift-free conditions in this work. Looking forward, two complementary hardware advances could bridge this gap. First, further reductions in cryo-holder drift through improved stage stability and thermal isolation would directly suppress probe-sample motion. Second, substantially faster pixelated detectors would dramatically shorten the total acquisition time. For example, a detector operating at approximately 120 000 frames/s [81] would reduce the acquisition time of a 176×176 4D-STEM dataset to approximately 0.26 s, lowering the cumulative drift to well below 0.1 Å even at current cryogenic drift rates. Under such conditions, the depth precision predicted by MHP simulations would become experimentally achievable. Together, advances in detector speed and cryogenic stage stability are therefore key to fully realizing the ultimate potential of MHP under cryogenic conditions.

In our simulations, the CSA was set to a large value of 80 mrad. Experimentally realizing such large CSAs remains challenging because it requires highly corrected probe optics with minimal residual aberrations and extremely stable alignment. However, recent advances in CEOS-corrected systems have demonstrated convergence angles up to 70–80 mrad at 200–300 kV accelerating voltages, with stable phase plates and probe-forming apertures [82]. These developments indicate that the parameters adopted in this work are approaching experimental feasibility.

In addition, temporal incoherence can also influence the achievable axial resolution in high-convergence-angle ptychographic optical sectioning through chromatic defocus broadening. To evaluate this effect, we performed additional simulations incorporating temporal incoherence using a chromatic aberration coefficient of $C_c = 1$ mm and energy spreads ranging from 0 to 0.2 eV, representative of modern monochromated STEM instruments [83]

(Fig. S18 in Supplemental Material [51]). At an energy spread of $\Delta E = 0.05$ eV, the depth profiles remain nearly identical to those obtained under fully coherent illumination. As the energy spread increases further, gradual degradation of axial localization and reconstruction stability is observed, particularly for weakly scattering atomic columns and closely spaced atomic layers. These results indicate that moderate temporal incoherence has only a limited impact on the achievable depth resolution of multislice ptychography. However, operation under highly monochromated conditions generally leads to reduced beam current, thereby prolonging the acquisition time. An alternative strategy is to reduce chromatic broadening through the Cc-corrected STEM. For example, a chromatic aberration coefficient below approximately 0.1 mm combined with a cold field-emission gun would be sufficient for the applications considered here. Further details are provided in Sec. IX in Supplemental Material [51].

The recent development of hollow-type fast pixelated detectors provides a direct experimental route to implement MHP. PN detector has introduced a practical design with dimensions of 264×264 pixels and a physical central hole with a radius of 25 pixels (Fig. S16(a) in Supplemental Material [51]) [80]. This geometry allows the fraction of low-angle electrons to be collected by the EELS spectrometer. However, the presence of an approximately 15-pixel-wide inactive ring at the edge of the central hole results in the loss of a small fraction of the diffraction signal that would otherwise be recorded by the detector. Taking this inactive ring into account, Fig. S16(b) in Supplemental Material [51] shows that the effective percentage of the total dose allocated to ptychography and EELS as a function of the size of the bright-field disc on the detector. For a bright-field disc of 60 pixels, corresponding to an HSA of 0.7α , the electron dose is distributed such that 50.6% is allocated to ptychography and 19.2% to EELS. Although the PN-type hollow detector design is not ideal due to the inactive region, it nevertheless provides a feasible path for simultaneous ptychographic and EELS acquisition. Future improvements in hybrid sensor design and EELS coupling optics are expected to further minimize the inactive region and enhance collection efficiency for fully integrated correlative 4D-STEM and EELS experiments.

Overall, further improvements in corrector design, source coherence, and fast-readout pixelated detectors are expected to enable routine operation in this high-CSA regime, which will be critical for achieving ultimate depth and lateral resolution in multislice hollow ptychography.

VIII. CONCLUSIONS

In this work, we introduce a new imaging modality and reconstruction algorithm, termed MHP, and validate its ability to overcome the limitations of SHP by addressing multiple scattering in thick specimens, achieving enhanced

lateral and depth resolution from hollow diffraction patterns. Our theoretical analyses of probe CSA, HSA, electron dose, and lattice vibrations show that MHP provides an adjustable balance between lateral resolution, depth sectioning, and EELS compatibility across various dose ranges. With up to 70% of the total dose available for EELS analysis, it can achieve sub-ångström lateral resolution at intermediate doses (10^4 – 10^8 e $^-$ /Å 2) and perform full 3D atomic-layer reconstruction including light-element layers at ultrahigh doses ($\geq 10^8$ e $^-$ /Å 2) at the resolution limit of thermal lattice vibrations. Therefore, MHP has the potential to provide a powerful and tunable framework which enables correlative 3D imaging and chemical mapping in complex materials, such as energy and quantum materials. Future development of detector geometry and speed, aberration correctors, algorithm design, artificial intelligence, and mechanical and thermal stability of cooling holders will further enhance the robustness and efficiency of hollow ptychography to achieve more accurate and comprehensive 3D materials analysis in electron microscopy.

IX. CODE AVAILABILITY

The MultiHollowPtycho source code is available at the GitHub repository [84].

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DATA AVAILABILITY

The experimental data used in this study are publicly available through the dataset associated with Ref. [37]. The simulated datasets can be reproduced using the abTEM simulation package with the simulation parameters provided in Tables S2–S4 in Supplemental Material [51].

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