Novel low-dose imaging technique for characterizing atomic structures through scanning transmission electron microscope

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I. INTRODUCTION

Scanning transmission electron microscopy (STEM) is widely used for characterizing the atomic structures of and species in nanomaterials [1–9]. When the projection acquired through STEM is approximately linear—that is, the projection shows the monotonic distribution related to atomic thickness only—the three-dimensional (3D) distribution of both the structure and density of atoms can be obtained through tomographic reconstruction [4,10–21]. A tilt series of two-dimensional (2D) projections with subangstrom resolution and a high signal-to-noise ratio (SNR) are the most critical data-acquisition factors in atomic-resolution 3D tomographic reconstruction. For yielding high-quality images, despite the significant progress in instrumentation, a long exposure time—which induces three major problems, namely sample drifting, carbon contamination, and radiation damage—remains unavoidable.

Sample drifting is caused by the time delay in the data acquisition as the probe scans across individual pixels within the region of interest, and distortion occurs when the rate of drift is higher than the total dwell time of the acquired image. This distortion intensifies at high magnifications, making interpretation of atomic structures difficult; specifically, distortion artifacts might be misinterpreted as structural dislocations. Sample drift can arise from various conditions, for example, mechanical problems in the specimen stage or a temperature gradient in the sample. Ideally, researchers must acquire images only after the sample has stabilized; however this is impractical when apparatus use time is limited and more so when projections must be obtained from multiple orientations. A typical solution is to acquire several images at the same area captured from different scanning orientations. This distortion intensifies at high magnifications, making interpretation of atomic structures difficult; specifically, distortion artifacts might be misinterpreted as structural dislocations. Sample drift can arise from various conditions, for example, mechanical problems in the specimen stage or a temperature gradient in the sample. Ideally, researchers must acquire images only after the sample has stabilized; however this is impractical when apparatus use time is limited and more so when projections must be obtained from multiple orientations. A typical solution is to acquire several images at the same area captured from different scanning orientations.

Reducing exposure time is a potential solution for solving sample drifting and for reducing the total dose, but this approach usually yields poor-quality images whose SNR is too low to reveal the lattice structures of interest. This problem could be overcome by performing denoising during data analysis; to this end, the Wiener filter, which reduces noise by minimizing the least-square error, is the most commonly used...
approach [24]. Applying this filter in the reciprocal space of an image significantly enhances atomic structures [25]. Without further knowledge of the sample, using the rotational average of Fourier intensities to estimate the background spectrum often results in the retention of the structural information but loss of the density distribution information because the low-frequency region is almost screened by the reciprocal space Wiener filter. Consequently, as the requirement of linearity is no longer satisfied, 3D reconstruction becomes problematic. Several denoising methods have been successfully applied to low-noise images [19,26–28] but none to high-noise images.

The approach developed in this study improves on the concept of the Wiener filter by using additional experimental input rather than performing denoising. The reciprocal space Wiener filter modifies the Fourier amplitudes of the target image; the noise barely affects the phases, especially those around Bragg peaks, which indicate the positions of the lattice features [25]. Rather than using the rotational average to generate a filter, if these amplitudes can be replaced with those with better SNRs, the density distribution of the image would improve significantly. In contrast to the phases, amplitudes around the origin are much more informative than are those in the high-frequency region. Accordingly, the correct phases can be obtained from a high-magnification image captured at a short dwell time, following which the amplitudes of the image can be replaced with those of a low-magnification image captured at a longer dwell time. This approach has two main advantages: The longer dwell time significantly improves the SNR, and low-magnification imaging is less susceptible to sample drift. Thus, we hypothesized that combining the respective merits of high- and low-magnification respective images would generate a high-quality image. Furthermore, the shorter exposure time reduces the required electron dose.

In the following sections, we first examined our approach by combining a noisy high magnification STEM projection and a low magnification one generated from multislice calculation. We then applied our approach to a tilt series of noisy electron micrographs and performed tomographic reconstruction to demonstrate that 3D local structures are consistent with the original model. Two experiments were conducted to verify the feasibility of our method in terms of alleviating the issues of sample drifting and carbon contamination under dose-reduced conditions.

II. VERIFICATION THROUGH MULTISLICE SIMULATION

In this section, we discuss in detail the proposed algorithm by using a fivefold (decahedral) Pt nanoparticle with a size of \( \sim 7.3 \times 7.0 \times 4.5 \, \text{nm}^3 \) as the test model, which includes edge and screw dislocations so that truncated distortions during 3D reconstruction can be examined. Two 0 deg projections with pixel resolution of 1 Å and 0.25 Å (\( R_{\text{LowMag}} \) and \( R_{\text{HighMag}} \), respectively) are calculated along the fivefold direction through the multislice method [29] with the following parameters: energy, 200 keV; spherical aberration, 1.2 mm; illumination semimangle, 10.7 mrad; inner and outer angle of the high-angle annular dark-field detector, 35.2 and 212.3 mrad, respectively. We impose a typical high noise level of 20% Poisson noise on \( R_{\text{HighMag}} \) to simulate the low-SNR image, \( R_{\text{noise}}^{\text{HighMag}} \), with a short dwell time; \( R_{\text{LowMag}} \) represents the low-magnification image with a long dwell time.

Because we have the perfect image (i.e., \( R_{\text{HighMag}} \)) of the model, we can calculate the errors of phases and amplitudes in the reciprocal space due to the application of Poisson noise. The reciprocal space of \( R_{\text{noise}}^{\text{HighMag}} \) is denoted as \( A_{\text{noise}}^{\text{HighMag}} \) and the phases and amplitudes of \( F^{\text{noise}}_{\text{HighMag}} \) are denoted as \( P^{\text{noise}}_{\text{HighMag}} \) and \( A_{\text{noise}}^{\text{HighMag}} \), respectively. The error metric of the phase can be calculated using

\[
\text{error}_P = \frac{\sum_k |P^{\text{noise}}_{\text{HighMag}}(k) - P_{\text{HighMag}}(k)|}{\sum_k P_{\text{HighMag}}(k)},
\]

where \( k \) denotes the selected \( k \) points in the reciprocal space. The projections contain ten Bragg peaks (i.e., five [111] and five [200] peaks) in total; we calculate the error for each of these peaks in a circular region with a diameter of 5 pixels. The average error of the phases is only 3.26%, whereas that of the amplitudes is 22.96%, which means that the phase information is accurate enough to reveal the positions of the lattice structures even in a low-SNR image.

We interpolate the pixel of \( R_{\text{LowMag}} \) four times to generate \( F_{\text{LowMag}} \) by Fourier transform in order to replace \( A_{\text{noise}}^{\text{HighMag}} \) with \( A_{\text{LowMag}} \). Because \( A_{\text{LowMag}} \) contains meaningful information only in the low-frequency region, we generated the initial reciprocal space by combining \( P^{\text{noise}}_{\text{HighMag}} \) and \( A_{\text{noise}}^{\text{HighMag}} \) for amplitudes larger than a threshold value, \( V_{th} \):

\[
F_{\text{initial}}^{\text{HighMag}}(k) = \begin{cases} A_{\text{LowMag}}(k) \exp (i \cdot P^{\text{noise}}_{\text{HighMag}}(k)) & \forall k \in D : A_{\text{noise}}^{\text{HighMag}}(k) > V_{th} \\ 0 & \text{otherwise} \end{cases}
\]

where \( D \) denotes the entire reciprocal space. \( V_{th} \) is determined by minimizing the variance within the region without the specimen when converting \( F_{\text{initial}}^{\text{HighMag}} \) to real space. Because some Fourier components of \( F_{\text{initial}}^{\text{HighMag}} \) are removed, the edge effect is produced by the remaining components. The density fluctuations in real space due to this effect can be monitored by observing the oscillations in the zero-density region; hence, selecting the optimal \( V_{th} \) (i.e., the \( V_{th} \) with the smallest variance) preserves the strong Fourier components and reduces the artifacts.

Since we now have partial information in both reciprocal and real spaces, an iterative algorithm can be used to perform optimization by satisfying the given constraints [30,31]. The reciprocal space is then produced by combining the Fourier
The obtained 3D image can be further processed to alleviate the noise and artifacts introduced by the missing wedge. The algorithm is terminated when the difference between \( R_n \) and \( R_{n+1} \) is smaller than a certain threshold. The final image tends to converge to a single solution because some critical phases are fixed. In our numerical simulations, \( \beta \) is less sensitive to the uniqueness and the convergence of restored images within the range of 0.5 ~ 2.5. However, the restored images become deteriorated when \( \beta \) is larger than 3.0.

Figures 1(a) and 1(b) depict \( R_{\text{LowMag}} \) and \( R_{\text{HighMag}} \) of the test model, respectively, and Fig. 1(c) presents the reconstructed image obtained using the proposed algorithm. The error of the reconstructed image, measured against the perfect image \( R_{\text{HighMag}} \) [Fig. 1(d)], improved from 20% to 10%, indicating that the proposed method enhances the image quality of low-SNR data. Because the aforementioned error is an average value, it does not reflect the error distribution. Therefore, we plotted line scans [Fig. 1(e)] to compare the atomic profiles of the images with that of \( R_{\text{HighMag}} \); the accuracy of the peak position in the profile of \( R_{\text{LowMag}} \) obtained through interpolation, is low, and so is the density distribution in the profile of \( R_{\text{HighMag}} \), because of noise. By contrast, the image obtained through the proposed method is accurate in terms of both peak position and density. Throughout our simulations, the algorithm is capable of revealing lattice structures even when the Poisson noise is much higher than 20%. In Fig. 2, we successfully restored images (the right column) from the original images (the left column) with Poisson noise 30%, 40%, and 50% (from up to down).

A more intricate comparison can be performed through examining local atomic arrangements in 3D reconstruction. Through multislice simulation with the same parameters, \( R_{\text{LowMag}} \) and \( R_{\text{HighMag}} \) are calculated at 53 equally sloped angles ranging from \(-70^\circ\) to \(70^\circ\). In each \( R_{\text{HighMag}} \), 20% Poisson noise is imposed to simulate the image with a short dwell time. \( R_{\text{noise}} \), the proposed algorithm is then applied to the 53 projections, following which the projections are subject to a novel tomographic algorithm, equally sloped tomography, in order to reconstruct the 3D image [15]. When the algorithm is terminated, the cores of the edge and screw dislocations are observed to verify the robustness of the proposed approach. The obtained 3D image can be further processed to alleviate the noise and artifacts introduced by the missing wedge during the 3D reconstruction [28]. However, to highlight the effectiveness of the proposed approach, only those 3D images from projections obtained with and without our algorithm are compared in this section.

Figure 3 illustrates the 3D atomic structures of the cores of the edge and screw dislocations. Panels (a) and (b) show the 3D reconstruction of the 53 \( R_{\text{noise}} \) images. Because of the 20% Poisson noise, no clear structures can be observed in these images. In panels (c) and (d), the 3D reconstruction of the 53 noisy projections generated by increasing 10 times of the electron dose is performed for addressing the equivalence of our algorithm. By contrast, the quality of 3D reconstruction of the images processed using the proposed algorithm is substantially higher than Figs. 3(a) and 3(b) and is compatible to Figs. 3(c) and 3(d), as evidenced by a comparison of the generated [Figs. 3(e) and 3(f)] and original images [Figs. 3(g) and 3(h)]. Although the contrast in the generated images is not as high as that in the original images, the results
FIG. 2. The performance of our algorithm demonstrated by using simulated images with Poisson noise 30%, 40%, and 50%. (a), (c), (e) Simulated images with 30%, 40%, and 50% Poisson noise. (b), (d), (f) The restored images using our algorithm from given noisy images (a), (c), and (e). The lattice structures are clearly visible even those cannot be observed in the raw images.

demonstrate that the proposed approach can delineate local atomic structures without requiring sophisticated denoising processes and at nearly one tenth the electron dose is required otherwise.

III. FEASIBILITY DEMONSTRATION THROUGH EXPERIMENTS

Given the favorable simulation results, we demonstrated the feasibility of the proposed method using experimental images. In the first demonstration, lattice structures along the zone-axis direction of sputtered AuPd bimetallic nanostructures were carefully examined. To further detail the procedures, we provide a schematic layout in Fig. 4 and practical guidelines as follows. (i) In the STEM mode, optimize the focus and Ronchi-gram around the region of interest. (ii) Set the magnification at 7.2 M and then acquire the image at an appropriate dwell time determined by observing the speed of drifting. The drifting has to be negligible comparing to the speed of scanning. (iii) Set the magnification at 1.8 M and then acquire the image at a longer dwell time. This procedure guarantees the consistency of the fast-scan high-magnification image and the slow-scan low-magnification one. (iv) Interpolate the low-magnification

FIG. 3. Comparison of the cores of edge and screw dislocations in 3D reconstructions obtained using raw and reconstructed images. We select the Z axis as the beam direction, and the Y axis as the rotation axis. (a), (c), (e), (g) Four 1.25-Å-thick ZX slices of 3D reconstructions from raw projections (with 20% Poisson noise), projections generated by increasing ten times of the electron dose (with 6.6% Poisson noise), restored projections, and the original model, respectively. The core of the edge dislocation is located at the lower grain of the nanoparticle. (b), (d), (f), (h) Four 3.25-Å-thick 

threshold values of isosurface rendering inside the closeups are adjusted to clearly visualize the typical zigzag patterns. The comparisons indicate that the proposed approach reveals the atomic structures of the cores of the dislocations with significantly reduced electron dose.
FIG. 4. The schematic layout of proposed iterative algorithm for experimental data. We first combine the amplitudes of a slow-scan low-magnification and the phases of a fast-scan high-magnification image to generate the initial reciprocal space [Eq. (2)]. The nonzero pixels in the initial reciprocal space are treated as referenced pixels. The initial reciprocal space is then subject into the iteration cycle. For each cycle, the reciprocal space is updated with the referenced pixels, and the remaining pixels are unchanged [Eq. (3)]. In real space, the negative-valued pixels inside the support and the pixels outside the support are adjusted [Eq. (4)].

image four times. Use the cross-correlation method to find out and crop the corresponding region in the interpolated image.

(v) Apply the iterative algorithm to obtain the restored image.

Figure 5 displays the raw images acquired using an FEI Titan electron microscope at 200 keV and the indicated magnifications and dwell times per pixel, respectively: (a) 1.8 M and 64 μs; (b) 7.2 M and 4 μs; (d) 7.2 M and 64 μs. The pixel resolutions of the 1.8- and 7.2-M images were 1 Å and 0.25 Å, respectively. Accordingly, Figs. 5(a), 5(b), and 5(d) can be considered $R_{\text{LowMag}}$, $R_{\text{noise}}$, and $R_{\text{HighMag}}$, respectively. The reconstructed image is presented in Fig. 5(c). Closeup images of the areas enclosed by the red, blue, and yellow squares in panel (a), which show the atomic structures at the grain boundary and the surface, are presented in panels (e), (f), and (g), respectively. In these areas, clear atomic structures are evident in neither the $R_{\text{LowMag}}$ nor the $R_{\text{noise}}$, whereas distinct and consistent structures can be seen in the images generated using the proposed approach, which required only one-eighth the electron dose required for $R_{\text{HighMag}}$.

FIG. 5. Experimental demonstration of the proposed method by obtaining images of Au-Pd nanostructures at different pixel resolutions and dwell times. (a) Raw image with a pixel resolution of 1 Å and a dwell time of 64 μs, $R_{\text{LowMag}}$. (b) Raw image with a pixel resolution of 0.25 Å and a dwell time of 4 μs, $R_{\text{noise}}$. (c) Reconstructed image obtained using the proposed iterative algorithm. (d) Raw image with a pixel resolution of 0.25 Å and a dwell time of 64 μs, $R_{\text{HighMag}}$. Although experimental images always contain noise, images with long exposure times can be considered high-SNR images and are the only feasible reference images. (e)–(g) Selected closeup images of (a)–(d). Red, blue, and yellow squares in (a) indicate the rows, from top to bottom, in (e)–(g), respectively. The images in each row, from the left to right, correspond to (a)–(d), respectively. The consistent atomic structures are obtained at only one eighth of the total dose used for (d).

The second experimental demonstration focused on sample drifting and density preservation of the restored image. The FeAu alloyed nanoparticles were synthesized and coated with PEGylated mesoporous silica nanoshells [32,33]. The magnetic and optical properties are strongly related to their atomic structures; thus delineating their atomic structures has practical applications, such as in their use as biomaterials in magnetic resonance imaging [32]. Because the synthesis is performed in aqueous media, the contamination buildup rate is significantly high, and some drift and vibration is inevitable. These problems make long acquisition times difficult for these specimens.
FIG. 6. Second experimental demonstration of our method wherein images of Fe-Au nanoparticles were obtained at different pixel resolutions and dwell times. (a) Raw image with a pixel resolution of 1 Å and a dwell time of 32 μs, R_{LowMag}. (b) Raw image with a pixel resolution of 0.25 Å and a dwell time of 4 μs, R_{HighMag}. (c) Reconstructed image using the proposed iterative algorithm. (d) Image stitched from 32 images, with a resolution of 0.25 Å and a dwell time of 4 μs per frame, R_{HighMag}. (e) Raw image with a pixel resolution of 0.25 Å and a dwell time of 128 μs. The image is completely distorted because of drifting. White arrows indicate the iron-gold overlapping region. The lattice structures can be observed only in the restored image, (c). The total dose of (c) is less than one-twentieth that of (d). (f) and (g) Line-scan comparisons of (c) and (d) in gold-rich and iron-rich regions. The density distribution of dark (i.e., iron-rich) and bright (i.e., gold-rich) areas are compared using dot and solid lines. The restored image (red lines) shows a higher contrast than does the stitched image (green lines).

As explained earlier, the sample drifting problem can be solved by acquiring multiple images and adding all the images to enhance the SNR. However, this approach cannot reduce the required dose or solve the contamination problem. Hence, we applied the proposed method to the obtained FeAu data, as follows. We acquired a 1.8-M image with a dwell time 32 μs, and a 7.2-M image with a dwell time of 4 μs, R_{LowMag} and R_{noise_{HighMag}}, depicted in Figs. 6(a) and 6(b), respectively. To obtain the referenced image R_{HighMag}, we acquired 32 7.2-M images with a dwell time of 4 μs in each frame. The final image obtained by adding all 32 images can be considered equivalent to a 7.2-M image with a dwell time of 128 μs [Fig. 6(d)].

To highlight the sample drifting problem, we also acquired a 7.2-M image with a dwell time of 128 μs [Fig. 6(e)]; as evident, no atomic structures can be observed in this image because of sample drifting.

The restored image is presented in Fig. 6(c), in which the atomic structures at the boundary of gold-rich and iron-rich regions are clear. Despite the much lower electron dose (less than one-twentieth that of the reference image), the distribution is consistent in the region of weak density [Fig. 6(f)] and strong density [Fig. 6(g)] regions. The contrast in the restored image (red lines) is higher than that in the stitched image (green line) because of chemical buildup on the surface of the specimen during the data acquisition of the latter. In addition, only the restored image clearly shows the lattice structures within the FeAu overlapping region (white arrows).

IV. CONCLUSION

We developed an approach to obtain a high-quality image at a much reduced electron dose. The developed approach overcomes the low-SNR problem by combining a low-magnification image with a noisy high-magnification image rather than performing denoising or filtering. The feasibility of the method was demonstrated both through simulation and experiments. This method is expected to improve the imaging of specimens that are radiation and contamination sensitive. We believe this approach will also intrigue the scientists of a broad spectrum of fields such as radiation-sensitive and time-resolved imaging.

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