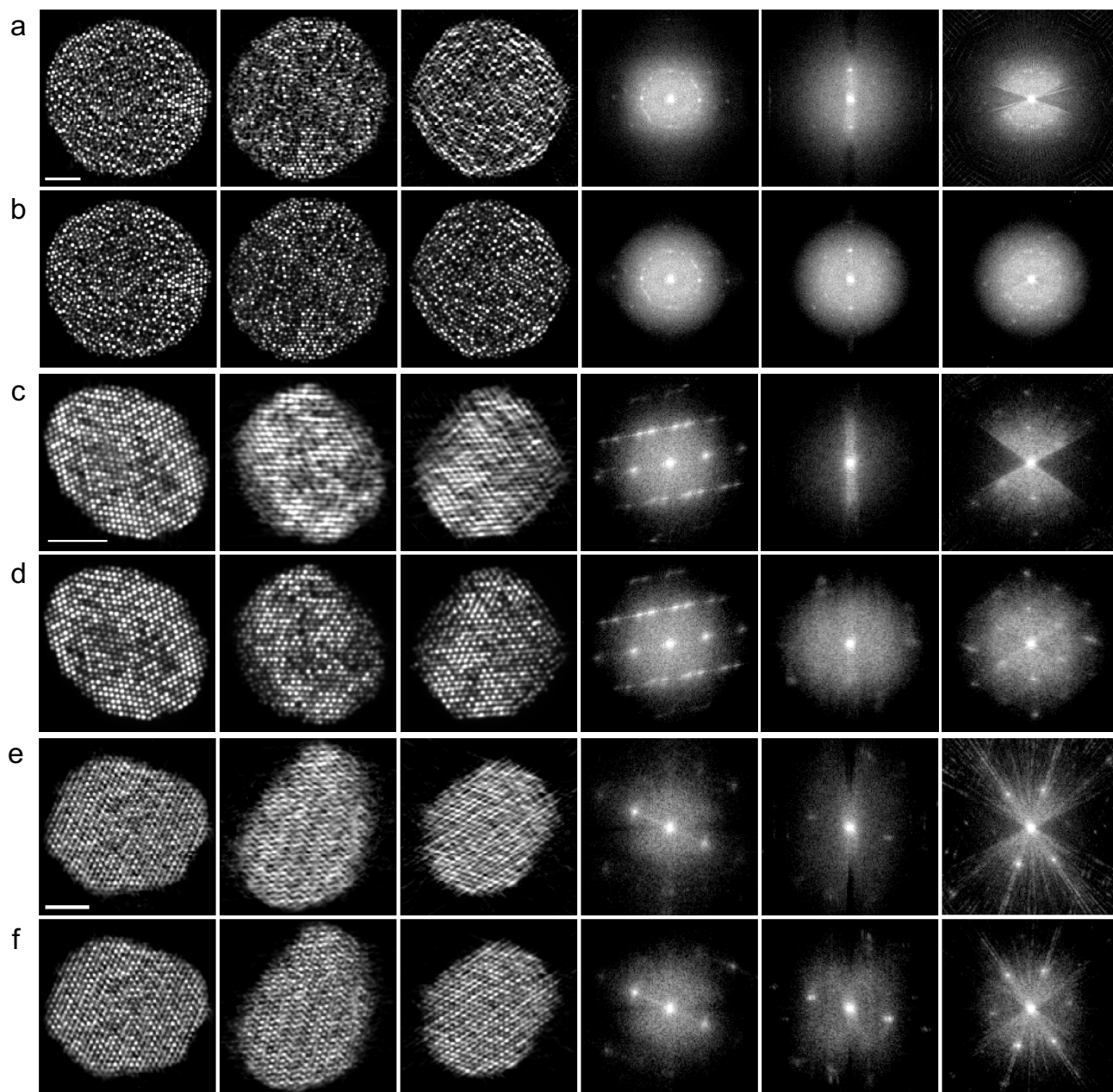
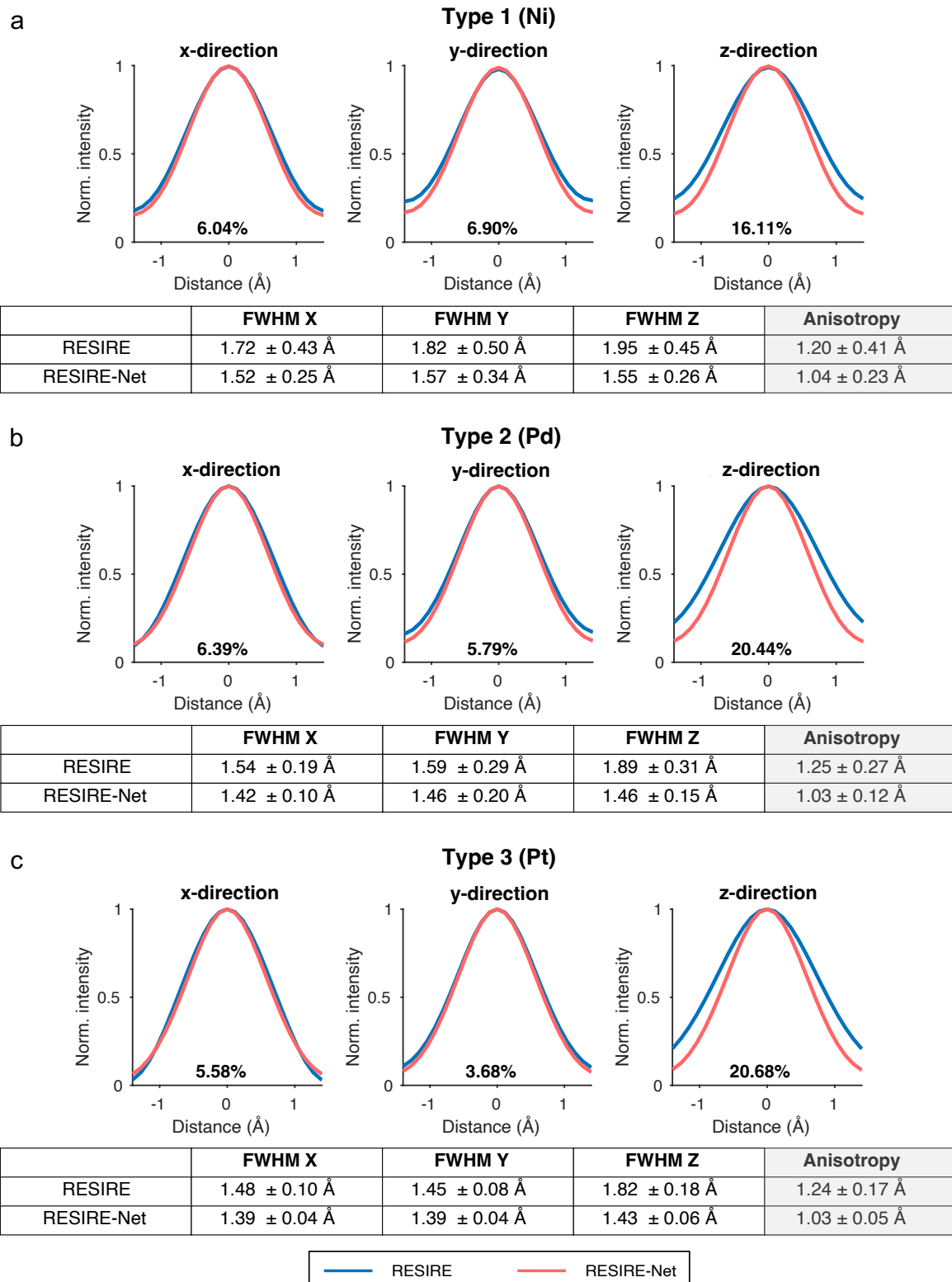


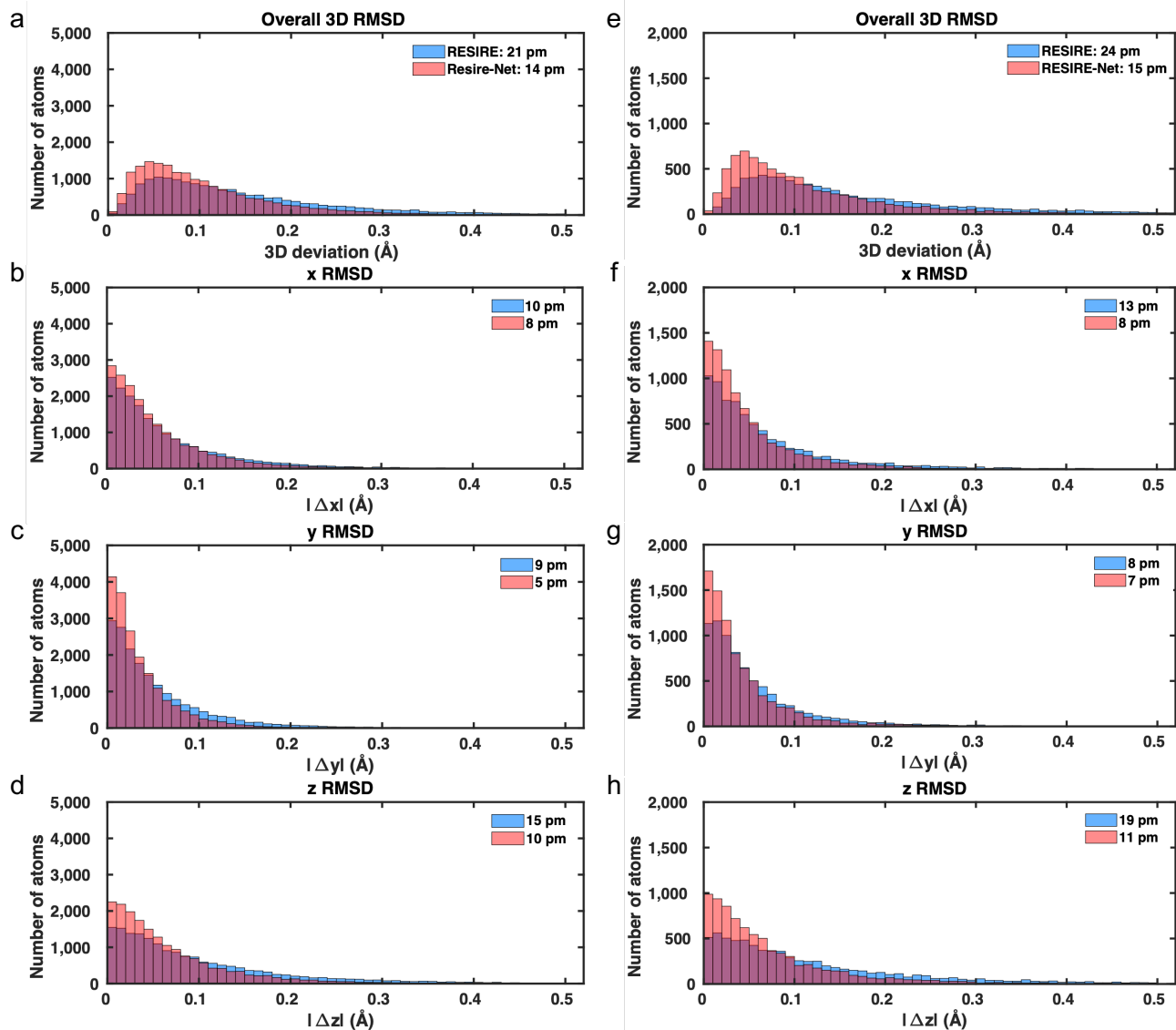
Supplementary Figures



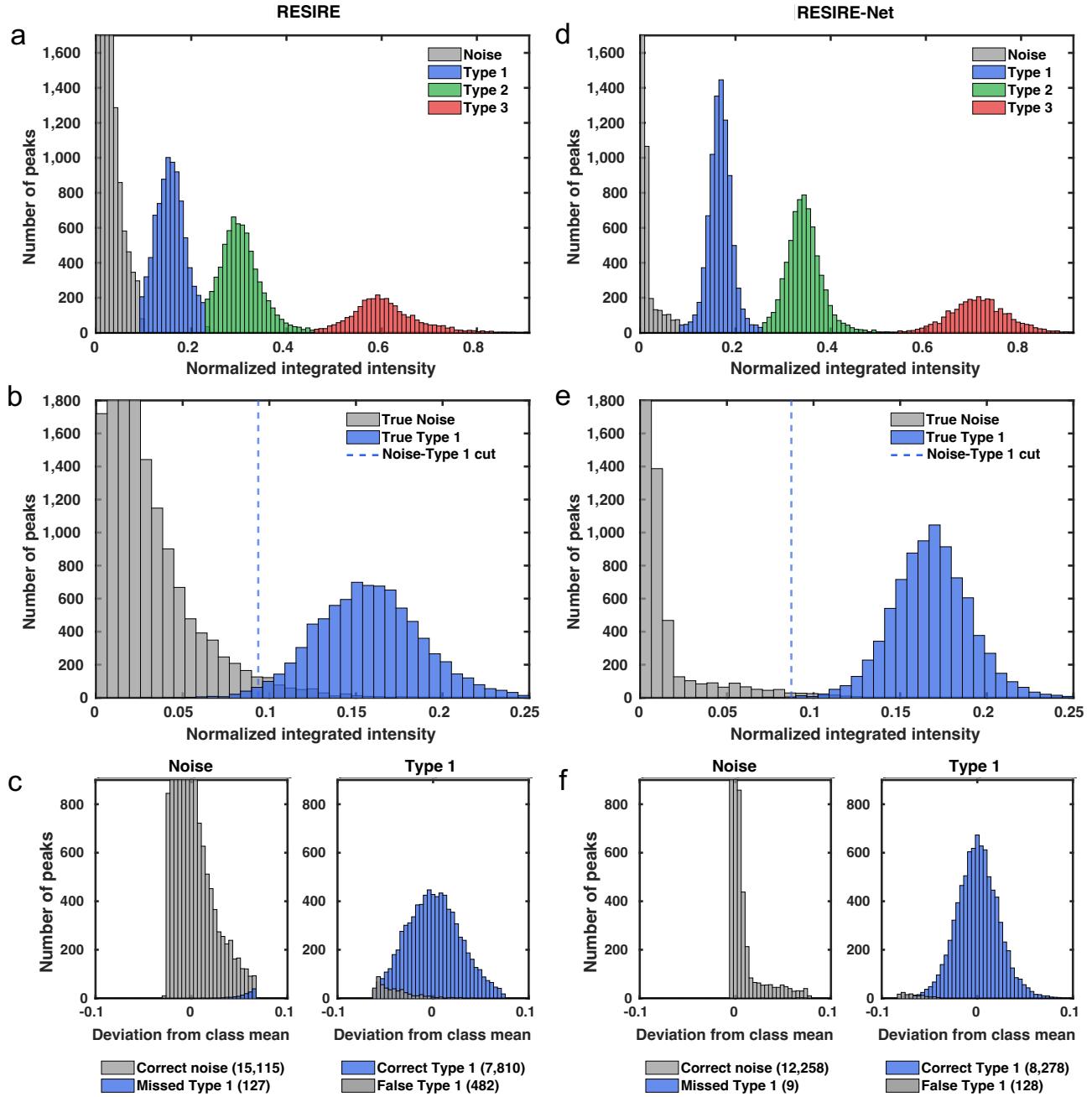
Supplementary Fig. 1. Comparison of real-space reconstruction slices and corresponding Fourier-space slices for RESIRE and RESIRE-Net reconstructions of multislice-simulated and experimental datasets. For each row, the first three panels show xy , yz , and xz real-space slices, and the last three panels show the corresponding central reciprocal-space slices from the 3D Fourier transform. **a, b**, Multislice-simulated metallic glass reconstructions using RESIRE (**a**) and RESIRE-Net (**b**), respectively (2.4-Å-thick slices). **c, d**, Multislice-simulated MEA-3 reconstructions using RESIRE and RESIRE-Net, respectively (2.6-Å-thick slices). **e, f**, Experimental MEA-2 reconstructions using RESIRE (**e**) and RESIRE-Net (**f**), respectively (2.5-Å-thick slices). Scale bars, 2 nm. Compared with RESIRE, RESIRE-Net produces sharper atomic features and partially restores Fourier amplitudes within the missing-wedge region, reducing elongation artifacts along the z -direction.



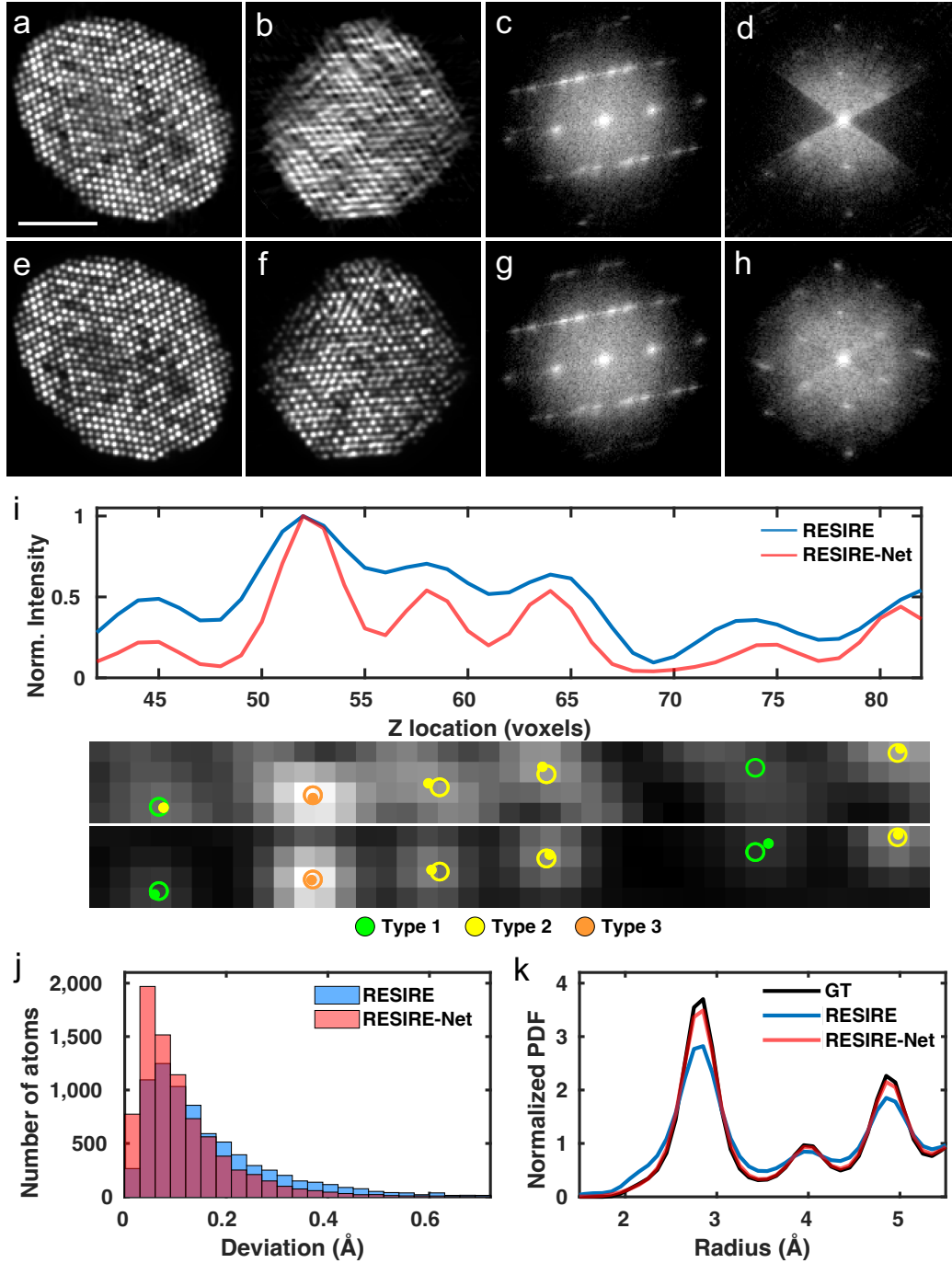
Supplementary Fig. 2. Comparison of per-type reconstruction anisotropy for atoms in the simulated metallic glass reconstructions (ms sim MG¹). **a-c**, Normalized average intensity profiles along the x, y, and z-directions are shown for Type 1 (**a**), Type 2 (**b**), and Type 3 (**c**) atoms in the final RESIRE (blue) and RESIRE-Net (red) models. Reported percentages indicate the reduction in FWHM achieved by RESIRE-Net relative to RESIRE along the corresponding direction. The tables below each set of profiles summarize the mean FWHM values ($\pm$ standard deviation) computed from individual atomic profiles, together with the anisotropy ratio (defined as $\text{FWHM}_z/\text{FWHM}_x$). RESIRE-Net consistently produces narrower and more isotropic atomic profiles, with the largest improvements observed along the z-direction, corresponding to the missing-wedge axis.



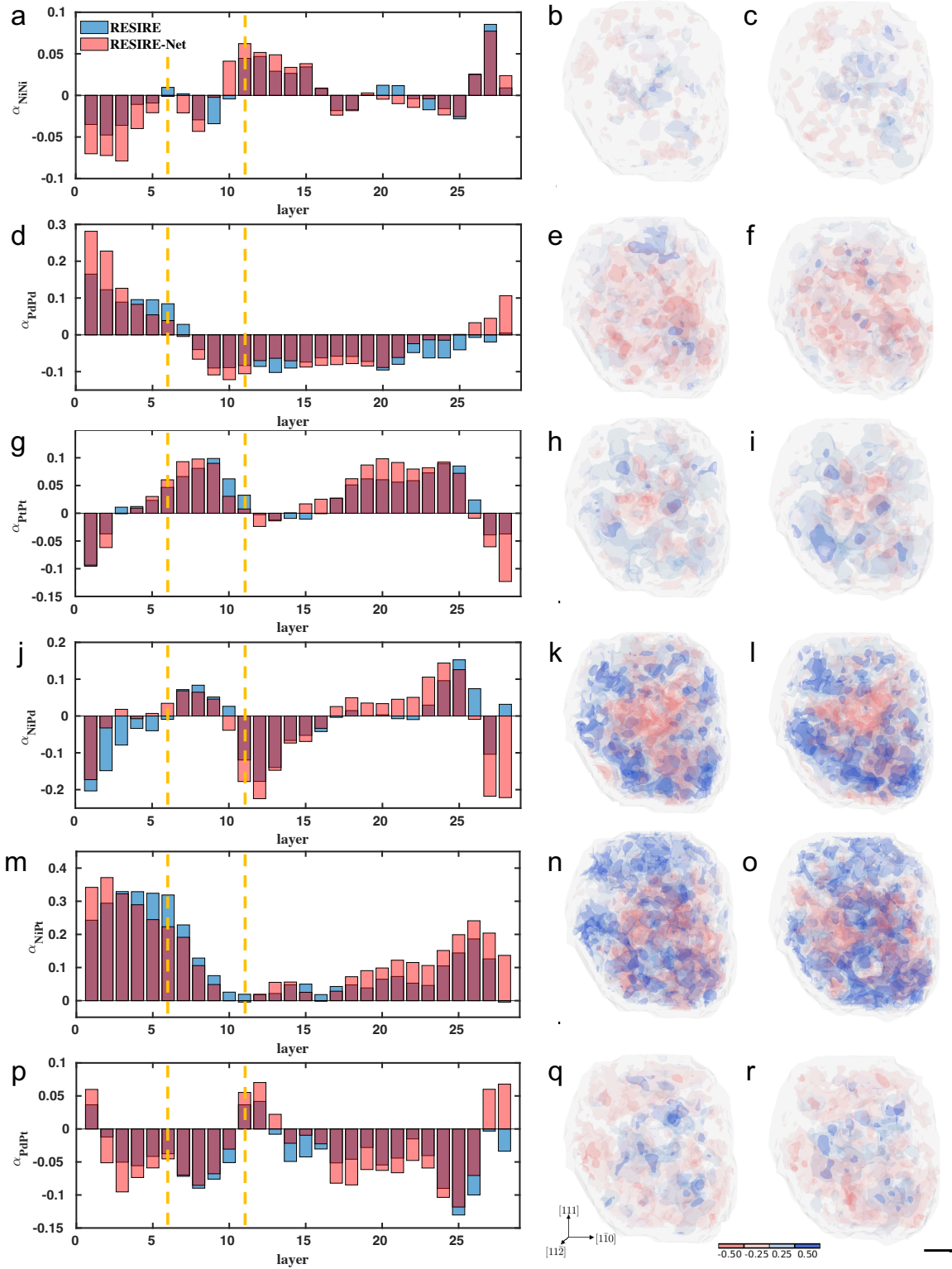
Supplementary Fig. 3. Average 3D RMSD, and x, y, and z deviations between model atoms and matched ground truth atoms in simulated datasets (ms sim MG¹ and ms sim MEA³). **a**, Distribution of 3D RMSD values calculated between atoms in each simulated metallic glass model and their matched ground truth atoms. Legends show the average value for each model. **b-d**, Distribution of x (**b**), y (**c**), and z (**d**) deviations of simulated metallic glass model atoms from their ground truth matches. **e-h**, Corresponding plots of the 3D RMSD (**e**), x (**f**), y (**g**), and z (**h**) deviations between atoms in the simulated “MEA-3” models and their matched ground truth atoms. In both datasets, RESIRE-Net offered the greatest improvement in the z (depth) direction, as expected with accurate filling of the missing wedge of Fourier-space information.



Supplementary Fig. 4. Comparison of initial k-means classification step on all peaks surviving polynomial tracing for simulated metallic glass data¹. **a, d**, Histograms of integrated peak intensities for all traced peaks surviving polynomial tracing, colored by k-means-assigned class (noise, Type 1, Type 2, Type 3), for the RESIRE (**a**) and RESIRE-Net (**d**) reconstructions. Integrated intensity values represent the sum of the intensity in the subvolume immediately surrounding each traced peak. **b, e**, Close up of the low-intensity regime showing ground-truth noise and Type 1 peaks, with a dashed line marking the cut determined by k-means, used to separate noise from true atoms, for the RESIRE (**b**) and RESIRE-Net (**e**) reconstructions. All peaks to the left of the cut were rejected as noise and all peaks to the right were accepted as Type 1 atoms. The peaks are colored by “true” type, as determined through matching with the ground truth model. **c, f** Diagram of the atoms rejected as noise and accepted as Type 1 atoms in the RESIRE (**c**) and RESIRE-Net reconstructions (**f**), colored by “true classification” (determined by the ground truth model). RESIRE-Net rejects fewer true atoms as noise and accepts fewer nonatoms into the model as compared to RESIRE. This difference is due to the improved separation between the peaks that is offered by RESIRE-Net.



Supplementary Fig. 5. Demonstration of RESIRE-Net on previously-published multislice-simulated medium entropy alloy (MEA) nanoparticle data (simulated “MEA-3”²). **a, b**, 2.6-Ångstrom-thick slices through the xy (**a**) and xz (**b**) planes of the RESIRE reconstruction. Scale bar, 2 nm. **c, d**, FFTs corresponding to the real-space slices of the RESIRE reconstruction (**a, b**). **e-h**, Corresponding real space slices and FFTs of the RESIRE-Net reconstruction. **i**, Normalized intensity along the z-direction for a representative region of each reconstruction (top), computed by summing over a 4-voxel-wide strip in x and a single-voxel slice in y. The corresponding xz regions are shown below (RESIRE, middle; RESIRE-Net, bottom). Overlaid markers indicate atom positions: colors represent atom types (green: Type 1, yellow: Type 2, orange: Type 3). Open circles denote ground truth atoms, and filled circles denote atoms identified by each model. **j**, Histograms of the 3D deviations of each model's atoms from their corresponding ground truth atoms. The peak, mean, and root-mean-square-deviation of the RESIRE histogram are 7 pm, 18 pm, and 24 pm, respectively. For the RESIRE-Net histogram we find those values to be 5 pm, 11 pm, and 15 pm. **k**, Comparison of the normalized pair distribution function (PDF) of each model. The RESIRE-Net PDF shows closer agreement with the ground truth, indicating improved structural fidelity. RESIRE results are reproduced from Ref.² for comparison.



Supplementary Tables

Supplementary Table 1. Acquisition and simulation parameters for all datasets.

	Simulated Metallic glass ^a	Simulated MEA ^b	Experimental MEA ^b
Name in original publication	Particle 1	MEA-3	MEA-2
Data collection and processing			
Voltage (kV)	200	300	300
Convergence semi-angle (mrad)	25	30	30
Probe size (Å)	0.8	0.8	0.8
Detector inner angle (mrad)	38	38	38
Detector outer angle (mrad)	190	190	190
Depth of focus (nm)	8	8	8
Pixel size (Å)	0.347	0.434	0.434
# of images	55	53	55
# of frames/angle		3	3
Tilt range (°)	−69.4° to +72.6°	−58.5° to +62.2°	−58.5° to +62.2°
Total electron dose ($10^5 e^-/\text{Å}^2$)	9.5	4.7	4.9

^a Parameters extracted from Supplementary Table 1 in Ref.¹.

^b Parameters extracted from Supplementary Table 2 in Ref.².

References

1. Yang, Y. *et al.* Determining the three-dimensional atomic structure of an amorphous solid. *Nature* **592**, 60–64 (2021).
2. Moniri, S. *et al.* Three-dimensional atomic structure and local chemical order of medium-and high-entropy nanoalloys. *Nature* **624**, 564–569 (2023).
3. Yang, Y. *et al.* Deciphering chemical order/disorder and material properties at the single-atom level. *Nature* **542**, 75–79 (2017).

Supplementary Table 2. Comparison of atom-level reconstruction accuracy and error statistics for models of a simulated metallic glass nanoparticle.

	GT ^a	RESIRE ^a	RESIRE-Net
Final model			
# Type 1 atoms	8,322	8,438	8,444
# Type 2 atoms	6,896	6,905	6,890
# Type 3 atoms	3,138	3,138	3,138
# Total model atoms	18,356	18,481	18,472
Refinement			
Error (R_1) ^b		4.76%	4.19%
Position accuracy			
X-RMSD (pm)		10	8
Y-RMSD (pm)		9	5
Z-RMSD (pm)		15	10
3D RMSD (pm)		21	14
# Position matches to GT		17,987	18,340
# Unmatched GT atoms (FN)		369	16
# Unmatched model atoms (FP)		494	132
% GT model matched		97.99%	99.91%
% Test model matched		97.32%	99.29%
Classification accuracy			
Type 1 accuracy (%)		94.87%	99.74%
Type 2 accuracy (%)		99.14%	99.83%
Type 3 accuracy (%)		100.00%	100.00%
Overall classification accuracy (%)		97.35%	99.81%
# Type 1 matches		7,895	8,300
# Type 2 matches		6,837	6,884
# Type 3 matches		3,138	3,138
# Total accurate matches		17,870	18,322
Raw error metrics			
# Correct atoms (N_{TP})		17,870	18,322
# Missed atoms (N_{FN})		369	16
# Misclassified atoms ($N_{misclas}$)		117	18
# Invented atoms (N_{FP})		494	132
Errors			
# Total error atoms (N_{error})		980	166
Total error rate (E_{atom})		5.34%	0.90%
Recall (R)		97.35%	99.81%
Precision (P)		97.31%	99.29%

^a Model originally published in Ref.¹.

^b The R_1 factor is defined as in equation 5 of Ref.³.

Supplementary Table 3. Comparison of atom-level model accuracy for RESIRE-Net models of a simulated metallic glass nanoparticle with and without the manual adjustment step, alongside the final published RESIRE model.

	GT ^a	RESIRE ^a	RESIRE-Net	
Model version				
Inclusion of manual tracing step		Yes	No	Yes
Final model				
Type 1 atoms	8,322	8,438	8,426	8,444
Type 2 atoms	6,896	6,905	6,889	6,890
Type 3 atoms	3,138	3,138	3,137	3,138
Total model atoms	18,356	18,481	18,452	18,472
Refinement				
Error (R_1) ^b		4.76%	4.20%	4.19%
Position accuracy				
X-RMSD (pm)		10	8	8
Y-RMSD (pm)		9	5	5
Z-RMSD (pm)		15	10	10
3D RMSD (pm)		21	14	14
# Position matches to GT		17,987	18,324	18,340
# Unmatched GT atoms (FN)		369	32	16
# Unmatched model atoms (FP)		494	128	132
% GT model matched		97.99%	99.83%	99.91%
% Test model matched		97.32%	99.31%	99.29%
Classification accuracy				
Type 1 accuracy (%)		94.87%	99.57%	99.74%
Type 2 accuracy (%)		99.14%	99.80%	99.83%
Type 3 accuracy (%)		100.00%	99.97%	100.00%
Overall classification accuracy (%)		97.35%	99.72%	99.81%
# Type 1 matches		7,895	8,286	8,300
# Type 2 matches		6,837	6,882	6,884
# Type 3 matches		3,138	3,137	3,138
# Total accurate matches		17,870	18,305	18,322

^a Model originally published in Ref.¹.

^b The R_1 factor is defined as in equation 5 of Ref.³.

Supplementary Table 4. Comparison of atom-level reconstruction accuracy and error statistics for models of a simulated medium-entropy alloy (MEA) nanoparticle.

	GT ^a	RESIRE ^a	RESIRE-Net
Final model			
# Type 1 atoms	2,399	2,334	2,413
# Type 2 atoms	3,476	3,561	3,453
# Type 3 atoms	2,266	2,270	2,266
# Total model atoms	8,141	8,165	8,132
Refinement			
Error (R_1) ^b		5.28%	3.90%
Position accuracy			
X-RMSD (pm)		13	8
Y-RMSD (pm)		8	7
Z-RMSD (pm)		19	11
3D RMSD (pm)		24	15
# Position matches to GT		7,677	8,101
# Unmatched GT atoms (FN)		464	40
# Unmatched model atoms (FP)		488	31
% GT model matched		94.30%	99.51%
% Test model matched		94.02%	99.62%
Classification accuracy			
Type 1 accuracy (%)		77.03%	98.08%
Type 2 accuracy (%)		98.68%	99.17%
Type 3 accuracy (%)		100.00%	100.00%
Overall classification accuracy (%)		92.67%	99.08%
# Type 1 matches		1,848	2,353
# Type 2 matches		3,430	3,447
# Type 3 matches		2,266	2,266
# Total accurate matches		7,544	8,066
Raw error metrics			
# Correct atoms (N_{TP})		7,544	8,066
# Missed atoms (N_{FN})		464	40
# Misclassified atoms ($N_{misclas}$)		133	35
# Invented atoms (N_{FP})		488	31
Errors			
# Total error atoms (N_{error})		1,085	106
Total error rate (E_{atom})		13.33%	1.30%
Recall (R)		92.67%	99.08%
Precision (P)		93.92%	99.62%

^a Model originally published in Ref.².

^b The R_1 factor is defined as in equation 5 of Ref.³.