Electronic Supplementary Material (ESI) for Nanoscale.

Supporting Information

Estimation of the Spatial Distribution of Frenkel Defects in $NiFe_2O_4$ by Simulation of HAADF-STEM Images

Haihong Bao,[†a] Jia-ao Wang,[† b] Wenjuan Yuan,*[a] and Jun Luo [a]

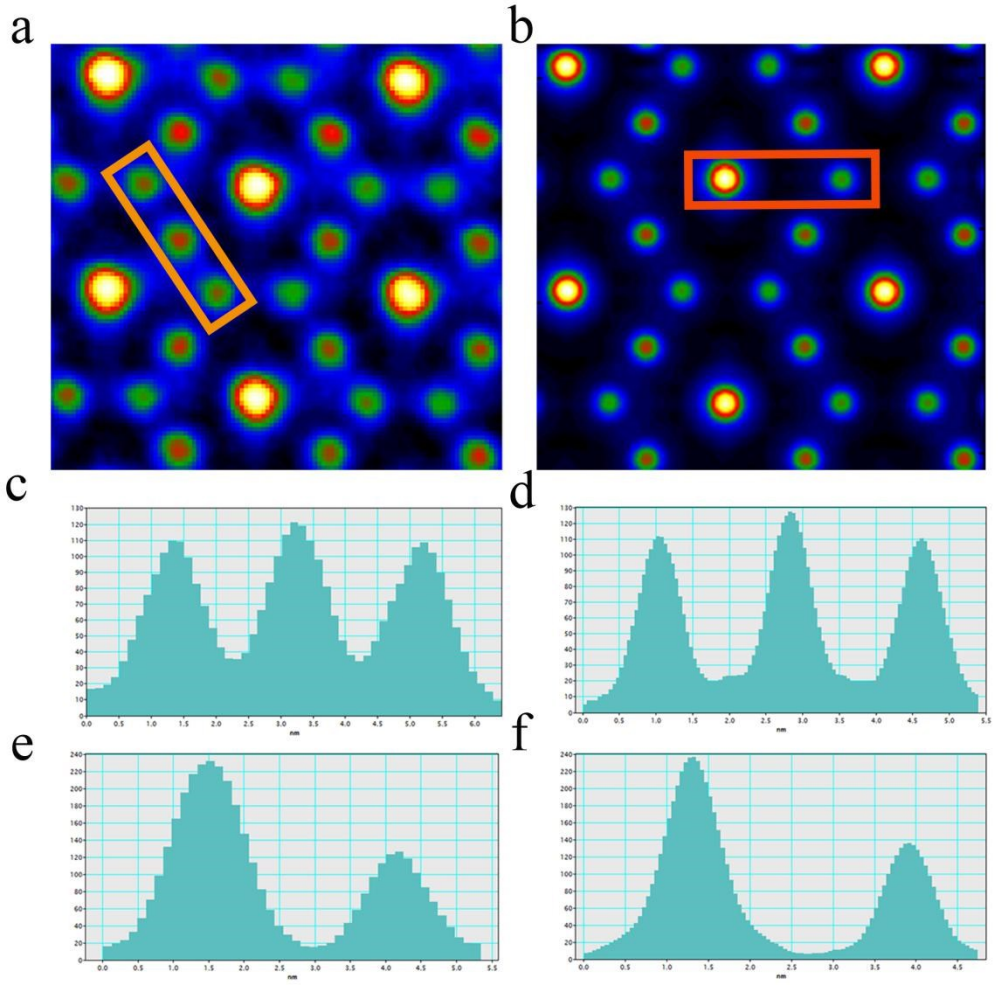


FIG. S1. Colored (a) experimental and (b) simulated images calculated along the (1 0 -1) direction and (c–f) intensity profiles corresponding to the regions represented by orange and red rectangles.

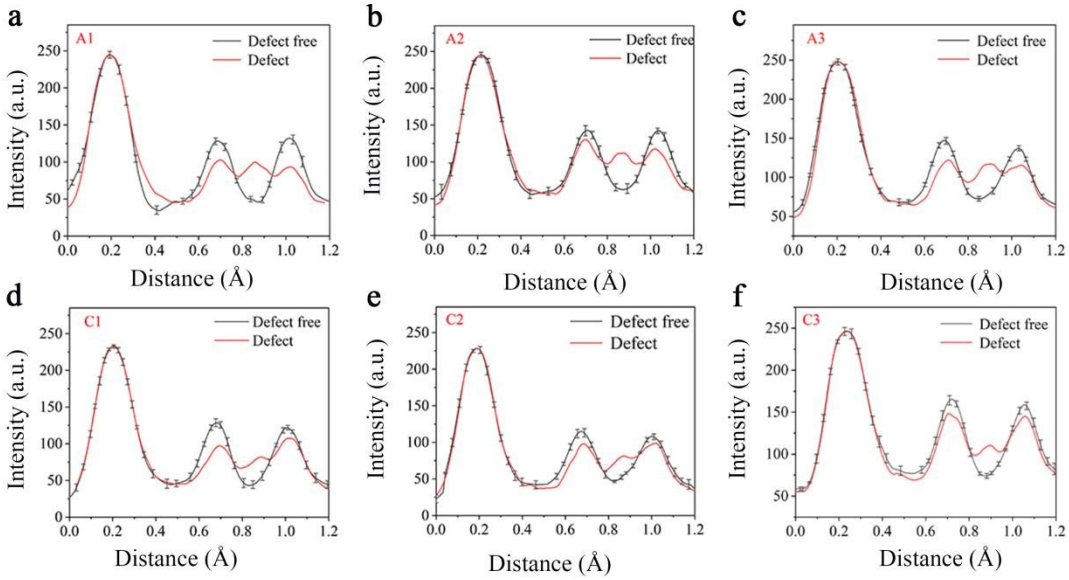


FIG. S2. Intensity profiles corresponding to the defects shown in Fig. 2(c).

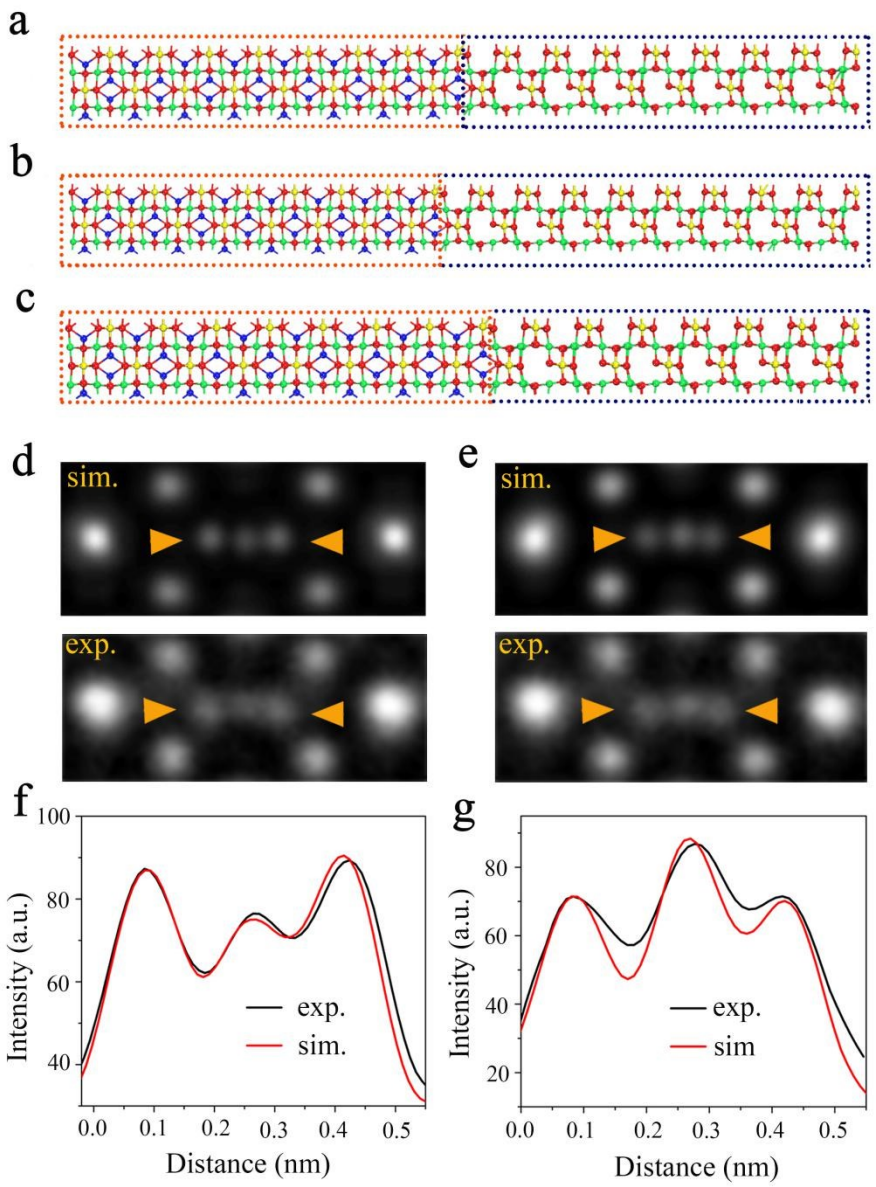


FIG. S3. (a–c) Atomic model corresponding to the simulated image shown in Fig. 3(c). The left region represented by a dark yellow dotted rectangle corresponds to the defect-free structure and the right region depicted by a dark blue dotted rectangle is the defect-containing structure. (d – e) Simulated and experimental images corresponding to the defect types B and C in Fig. 2(f – g) and their line-scanning intensity profiles along the direction indicated by the two yellow arrows.

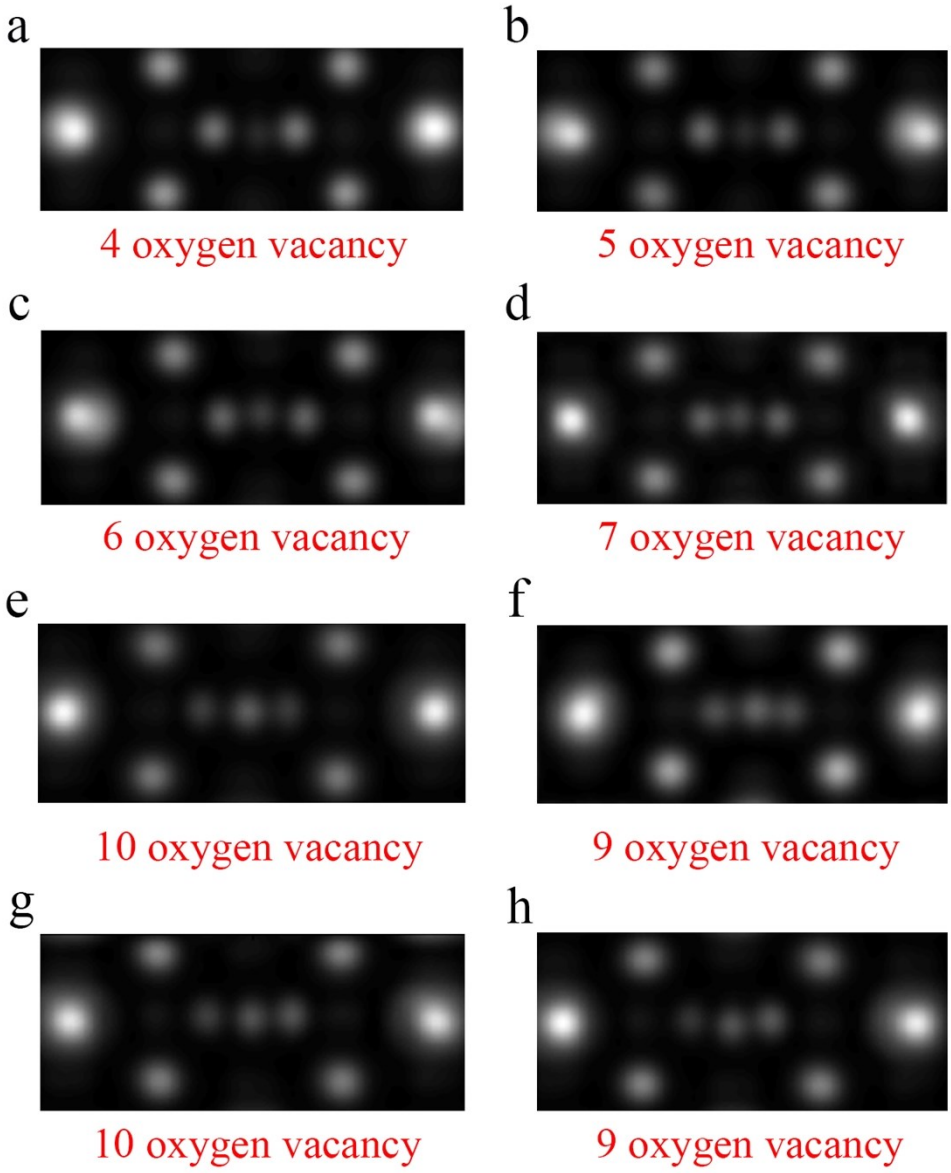


FIG. S4 Simulated images of the different Fe occupancies in the defects structure. For the images in S4a-d, each tetrahedral site Fe columns contain 8 defect-free Fe atoms and 4-7 Fe vacancies and octahedral site interstitial columns contain 4-7 Fe atoms. For the images in S4e(f), each tetrahedral site Fe columns contain 8 (9) defect-free Fe atoms and 10 (10) Fe vacancies. The Fe occupancy in octahedral site interstitial columns is the same with in vacancy columns. In S4g-h, the Fe occupancy in two neighboring tetrahedral is unequal.

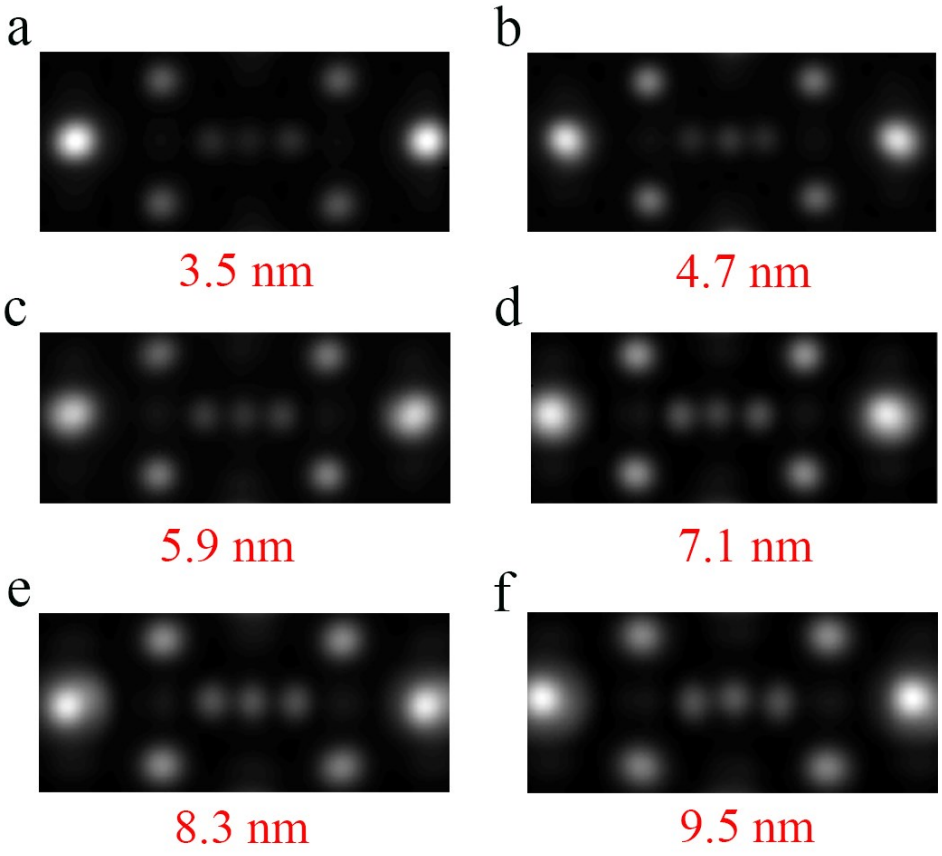


FIG. S5. Simulated images of the different thicknesses corresponding the defects atomic structure at position A in Fig. 2c (in main text).

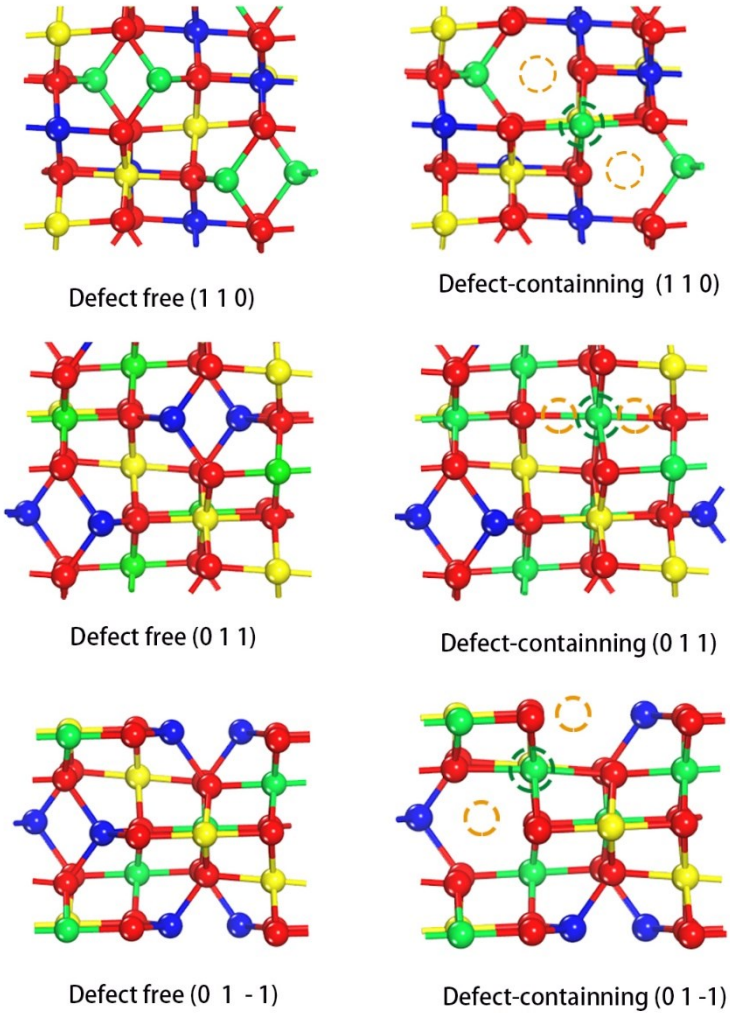
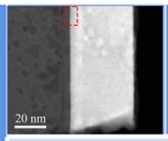
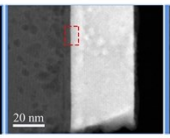
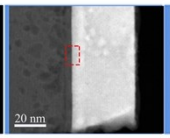
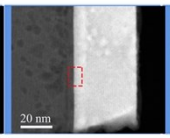
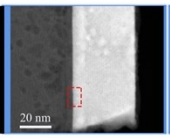
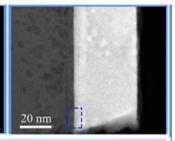


FIG. S6. View of the defect-containing atomic structures along different projections of the crystal. The red, yellow, blue, and green balls represent the O, Ni, tetrahedral site Fe, and octahedral site Fe, respectively. The yellow dotted circles represent the tetrahedral site Fe vacancy and the green dotted circles surrounding single atoms represent the interstitial octahedral site Fe atom.

Table S1. Ni, Fe, O, Al, Mg, F contents of the composite film shown in Fig. 1a.

Element	Mean Atomic Fraction (at. %)	Mean Square Error (%)
Fe	17.05	1.15
Ni	8.74	0.64
O	49.88	0.97
C	22.71	0.38
Al	0.88	0.06
Mg	0.73	0.06

Table S2. Composition distribution of different sample areas. The red and blue dotted rectangles in the HAADF-STEM images represent the regions where the corresponding atomic fraction was obtained.

element						
	Atomic Fraction (at. %)					
Fe	9.65	10.94	11.21	9.73	9.04	8.39
Ni	5.53	6.04	5.98	5.54	4.69	4.59
O	57.18	57.64	56.92	55.91	57.93	56.97
C	9.97	9.91	11.61	14.04	12.89	12.75
Al	10.46	10.78	9.87	10.45	10.92	12.25
Mg	5.21	4.66	4.41	4.33	4.52	5.05