Depth resolution Ptychography reveals the presence of FeO on the surface of Fe_3O_4

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Introduction

Oxide materials exhibit complex surface and interface structures that strongly influence their physical and chemical properties. Advanced electron microscopy techniques, including HAADF-STEM, iDPC, and multislice electron ptychography, provide powerful approaches for atomic scale structural characterization.

In this work, Fe_3O_4 particles exsolved from $LuFeO_3$ thin films is investigated using various electron microscopy methods. Depth resolution ptychography directly reveals additional object phase localized near the upper and lower surfaces, observing the distribution of surface FeO structure along the electron beam direction.

Main Text

Atomic-resolution HAADF-STEM and iDPC images of Fe_3O_4 along the [110] zone axis are shown in Figure 1A-B. HAADF imaging resolved iron atom columns, while iDPC provided oxygen atom positions.

Figure 1C-D illustrates the principle of electron ptychography. A converged electron probe scans across the specimen while 4D diffraction patterns are recorded by a pixelated detector. It can obtain the phase of the object through redundant diffraction data iteration.

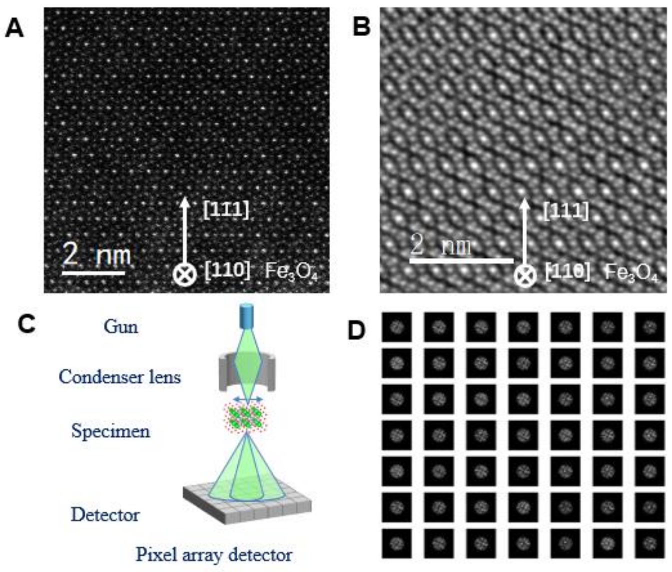

Figure 1. Structural characterization of Fe_3O_4 and ptychography principle (A) Atomic-resolution HAADF-STEM image (B) Corresponding iDPC image (C) Ptycho experimental optical path (D)4D diffraction patterns.

Multislice electron ptychography was employed to investigate depth-dependent structural variation inside Fe_3O_4. Reconstructed object phase images at different propagation depths are shown in Figure 2A, revealing clear evolution of sample phase contrast along the beam direction, additional intensity appear on the surface of the phase.

The depth-section profile in Figure 2B demonstrates additional object phase localized near the upper and lower surfaces of Fe_3O_4. The results showed that some phases only appeared in the two layers of the sample surface, indicating the presence of structurally distinct surface layers.

Figure 2C illustrates the multislice ptychography reconstruction process, where the sample is divided into thin layers of 1nm thickness to obtain slices of objects at different depths. This method enables the application of electron ptychography in thick samples.

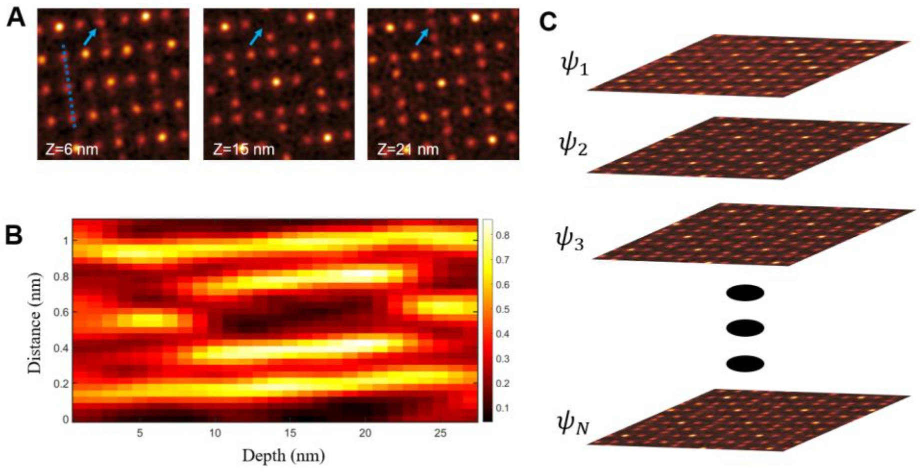

Figure 2. Depth resolution multislice electron ptychography of Fe_3O_4. (A) Phase image of sample at depth along beam direction, surface additional phase points correspond to FeO structure. (B) Depth profile of phase intensity along the blue dash-dotted line. (C) Schematic illustration of the multislice ptychography reconstruction process.

To identify the origin of the additional surface-localized phase contrast, FeO structural models and corresponding image simulations were constructed. Figure 3A shows the projected structure of FeO along the [100] direction, which is similar to the 6nm slice in Figure 2A.

Simulated HAADF image and simulation data ptychography reconstruction results shown in Figure 3B-C reproduce the experimentally observed surface contrast and phase localization. The agreement between experiment and simulation indicates that the upper and lower surfaces undergo FeO reconstruction induced by local transition from Fe^{3+} to Fe^{2+}.

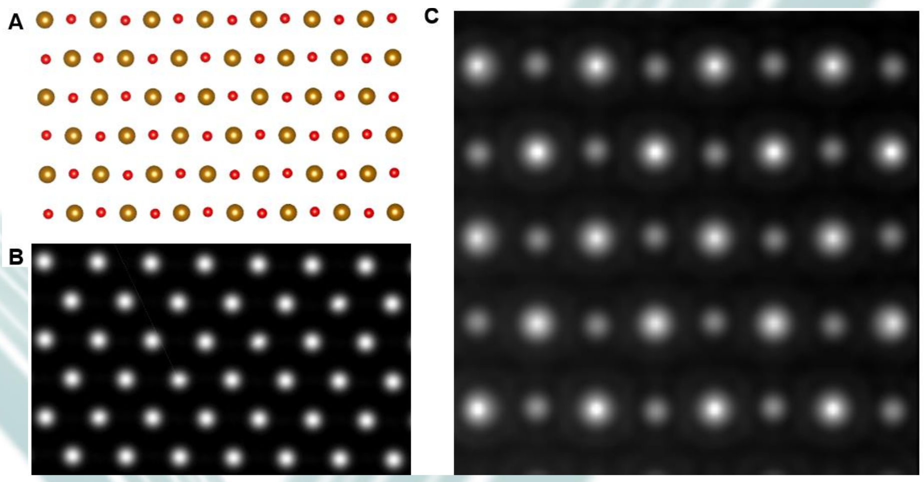

Figure 3. Structural model and image simulation. (A) The structural model of FeO along the [100] direction. (B) Simulated HAADF-STEM image of this model. (C) Simulated Ptychography phase image.

Conclusion

HAADF-STEM and iDPC imaging characterize Fe_3O_4 structure, while multislice electron ptychography directly visualizes depth-dependent object phase inside the epitaxial Fe_3O_4. Combined with structural modeling and image simulations, the reconstructed surface is identified as an FeO structure. These results highlight the significant potential of electron ptychography for oxide materials characterization, providing direct atomic-resolution access to depth-dependent structural reconstruction.

References:

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