Supplementary Information:

1. FFT Images Including Plane-Spacing

Figure S1 shows FFT images from several individual NPs. The patterns in the FFT images (dots, not the circular patterns) are characteristic of single-crystalline NPs. Various d-spacings are from different planes in these single-crystalline NPs having either Mn_2O_3 or Mn_3O_4 phases, as the spacings match these two phases.

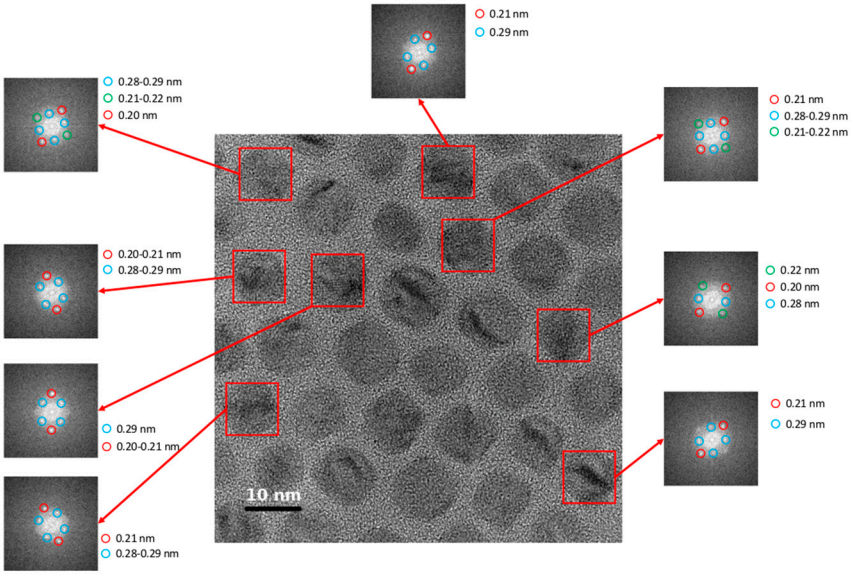

Figure S1. FFT images for several individual NPs including plane-spacing calculations. The dot patterns in the FFT images are characteristic of single-crystalline NPs. Various d-spacings are from different planes in these single-crystalline NPs.

2. A 57 Atom Cluster Model Fit

It is possible to obtain a good fit to the experimental G(r) by using a cluster model based on a distorted rock salt MnO structure. In particular, the relative intensities of the peaks can be greatly improved by using a cluster instead of periodic model structures. Figure S2 shows G(r) for a hypothetical 57 atom cluster. In this model, MnO_6 octahedra become significantly distorted and some of the MnO_6 octahedral complexes share oxygen atoms with the neighboring units resulting in new MnO_x entities. This model, which is too simplified to be correct, may nevertheless suggest the formation of distorted MnO_6 structures when an MnO thin film is deposited on a silica substrate.

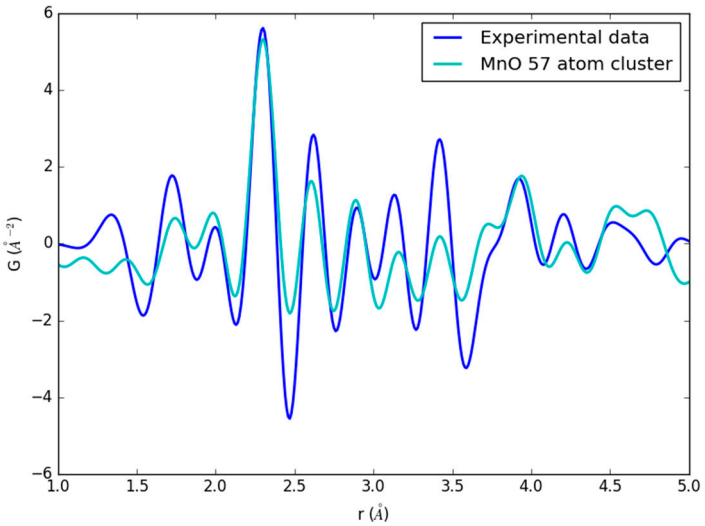

Figure S2. Comparison of experimental G(r) with the corresponding simulated results for a hypothetical 57 atom (relaxed) MnO cluster.