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Atomic-resolution study of cobalt valence and spin-state transitions in $Ca_3Co_4O_9$ using in-situ scanning transmission electron microscopy. GUANG YANG, YUAN ZHAO, ROBERT KLIE, Department of Physics, University of Illinois at Chicago — The misfit-layered $Ca_3Co_4O_9$(CCO) has been of great interest due to its high thermo-electric power and thermal stability. The CCO structure consists of five layers: three rock salt-type layers Ca_2CoO_3 are sandwiched between two CdI_2-type CoO_2 layers along the c-direction. The presence of different Co valence states is assumed to account for the thermal stability of CCO, and the abrupt changes of electrical resistivity at 420K is believed to be due to a Co spin-state transition. Here, we combine scanning transmission electron microscopy (STEM) with electron energy loss spectroscopy (EELS) to study the atomic and electronic structure of CCO. Using atomic-column resolved EELS, the Co valence states in the different layers are quantified and significant charge transfer from $CoO_2$to Ca_2CoO_3 is measured. The effects of the potential spin-state transition at 420K on the local structure will be studied by in-situ heating experiments. We will show how atomic-resolution Z-contrast imaging in combination with EELS and in-situ experiments can be utilized to understand the effects of interfacial charge transfer and spin-state transitions in complex oxide materials.

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