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Direct measurement of charge transfer and spin stat transitions in thermoelectric $Ca_3Co_4O_9$ GUANG YANG, University of Illinois at Chicago, QUENTIN RAMASSE, National Center for Electron Microscopy, Lawrence Berkeley National Laboratory, ROBERT KLIE, University of Illinois at Chicago — The misfit-layered thermoelectric material $Ca_3Co_4O_9$ has been the focus of many recent studies due to its high thermal power and good high temperature stability. In particular, it has been suggested that the presence of a mixed valence state in the strongly correlated CoO_2 layer is essential for the high p-type thermoelectric properties in $Ca_3Co_4O_9$. In this study, we combine aberration-corrected scanning transmission electron microscopy (STEM) with electron energy loss spectroscopy (EELS) to study the atomic and electronic structures of $Ca_3Co_4O_9$. We will show that the position of the O atomic columns in the CoO_2 layers are highly ordered and can therefore be directly imaged, while the CoO columns in the Ca_2CoO_3 rocksalt layer exhibit a strong modulation in the (010) direction. Further, we measure the local Co valence and find significant hole transfer from the rocksalt CoO to the strongly correlated CoO_2 layers. In addition, we will present the results of our in-situ heating experiments of $Ca_3Co_4O_9$ [010] at 500 K, which show that the phase transition at 420 K is not accompanied by a structural transition but rather a transition of the Co-ion spin states.

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