Abstract Submitted
for the MAR12 Meeting of
The American Physical Society

The effects of increased Co-ion spin states on the Seebeck coefficient in thermoelectric $Ca_3Co_4O_9$[1] ROBERT KLIE, TADAS PAULAUSKAS, QIAO QIAO, ALEJANDRO REBOLA, SERDAR OGUT, University of Illinois at Chicago, DIPANJAN MAZUMDAR, ARUN GUPTA, Center of Materials for Information Technology, University of Alabama, STANISLAW KOLESNIK, Northern Illinois University, JUAN-CARLOS IDROBO, Materials Science and Technology Division, Oak Ridge National Laboratory — Thermoelectric oxides have attracted increasing attention due to their high thermal power and temperature stability. In particular, $Ca_3Co_4O_9$(CCO), a misfit layered structure consisting of single layer hole-doped CoO_2 sandwiched between insulating Ca_2CoO_3 rocksalt layers, exhibits a high Seebeck coefficient at 1,000 K. It was previously suggested that the Seebeck-coefficient can be further improved by stabilizing an increased Co-ion spin state in the CoO_2 layers. Here we report a significant increase in the room-temperature in-plane Seebeck coefficient of 40 nm thick CCO films grown by pulsed laser deposition on $SrTiO_3$ substrates. We combine aberration-corrected Z-contrast imaging, atomic-column resolved electron energy-loss spectroscopy, and density-functional calculations to show that the increase is caused by CoO_2 stacking faults with Co^{4+}-ions in a higher spin state compared to that of bulk CCO. The higher Seebeck coefficient makes the CCO system suitable for many high-temperature waste-heat-recovery applications. The role of dopants, such as Bi and Ti will also be explored.

[1]This work was supported by the National Science Foundation [DMR-0846748 (RFK), DMR-0938330 (J-CI)].

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Date submitted: 17 Nov 2011 Electronic form version 1.4