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Brownmillerite CaCoO_{2.5}: Synthesis, Re-entrant Structural Transitions and Magnetic properties¹ JUNJIE ZHANG, HONG ZHENG, CHRIS-TOS MALLIAKAS, JARED ALLRED, YANG REN, QING'AN LI, TIANHENG HAN, JOHN MITCHELL, Argonne Natl Lab — Cobalt oxides attract both fundamental and technological attention due to their physical properties including thermoelectricity, giant magnetoresistance, superconductivity and multiferroicity [1]. Here we report the first synthesis of $CaCoO_{2.5}$ single crystals using a high pressure opticalimage floating zone technique. We find that it is an ordered oxygen-deficient perovskite of the brownmillerite type, and it undergoes an unprecedented re-entrant structural phase transitions (Pcmb \rightarrow P2/c11 \rightarrow P12₁/m1 \rightarrow Pcmb) with decreasing temperature. We describe its temperature-dependent structural, thermal, and magnetic properties, including AFM ordering near 240 K, with a weakly spin canted ferromagnet ground state below 140 K. The magnetic response of CaCoO_{2.5} depends markedly on the cooling rate and field history. Magnetization data also imply the potential of a distinct, field-induced phase arising uniquely from the $P12_1/m1$ structure, revealed as kinetically trapped by a rapid-cooling protocol.

[1] Raveau, B.; Seikh, M. M. Cobalt Oxides: From Crystal Chemistry to Physics; Wiley-VCH: Weinheim, 2012.

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