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**Ab Initio Study on Na Ordering and its Relation to the Electronic/Magnetic Properties of P2-NaCoO2 Thermoelectrics** YING SHIRLEY MENG, Massachusetts Institute of Technology, ANTON VAN DER VEN, University of Michigan, MARIA CHAN, M.I.T., GERBRAND CEDER, M.I.T. — The unusual electronic properties of NaxCoO2 make it a material of considerable interest. The system displays an unusual combination of high Seebeck coefficient and low metallic resistance. NaxCoO2 is also an interesting material on which to test our fundamental knowledge of mixed valence transition metals. NaxCoO2 is a mixed valence system with a fraction of x Co4+ and (1- x) Co3+ ions. Because of the high mobility of Na and large vacancy concentration, Na-vacancy ordering is likely in NaxCoO2. This ordering breaks the symmetry on the Co sublattice and may assist in charge ordering of Co3+ and Co4+. Mixed Co3+/Co4+ systems tend to display rich physics as they are often close to spin transitions and metal insulator transitions. In this study, we use both standard Density Functional Theory (DFT) in the Generalized Gradient Approximation (GGA) as well as GGA+U calculations to investigate the possible Na-vacancy and charge-ordered structure of P2-NaxCoO2 throughout the sodium composition range. We have identified new ground state structures at Na concentrations 0.11, 0.75 and 0.80. We have also found a strong coupling between the Na-vacancy ordering and Co3+/Co4+ charge ordering in the system. Such knowledge is crucial for understanding the thermoelectric properties of this material and similar mixed valence oxides.

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