Abstract Submitted for the MAR14 Meeting of The American Physical Society

Magnetostructural coupling in the magnetodielectric Mn_3O_4 MOUREEN KEMEI, JAYE HARADA, Materials Department and Materials Research Laboratory, University of California, Santa Barbara, California 93106, USA, MATTHEW SUCHOMEL, X-ray Science Division, Argonne National Laboratory, Argonne, Illinois 60439, USA, RAM SESHADRI, Materials Department and Materials Research Laboratory, University of California, Santa Barbara, California 93106, USA — At room temperature, the spinel Mn_3O_4 is distorted from cubic symmetry due to Jahn-Teller distortions of octahedral Mn³⁺ and is described by the tetragonal spacegroup $I4_1/amd$. It undergoes ferrimagnetic ordering near 43 K where anomalies in heat capacity and dielectric measurements are also observed. High-resolution variable-temperature synchrotron X-ray powder diffraction reveals a distortion of the long-range crystal structure at the onset of magnetic order. The structure of Mn_3O_4 below the magnetostructural ordering temperature is described by coexisting tetragonal and orthorhombic phases. A discontinuous change in lattice parameters at the magnetostructural ordering temperature illustrates that this system undergoes a first-order phase transition. Sharp changes in heat capacity at the magnetic ordering temperature are also consistent with a first-order phase transition. We present the complete crystallographic description of this important magnetodielectric spinel and suggest mechanisms behind the spin-driven lattice distortion.

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Date submitted: 14 Nov 2013

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