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Structural Inhomogeneities and Suppressed Magneto-Structural Coupling in Mn-Substituted GeCo2O4

A comprehensive study of Ge1-xMnxCo2O4 (GMCO) system was conducted using neutron powder diffraction (NPD), x-ray diffraction (XRD), Scanning electron microscopy, magnetometry, and heat capacity measurements. Comparative analysis with GeCo2O4 (GCO) highlights the influence of Mn substitution on the crystal and magnetic structure at low temperature. Surprisingly, phase separation is observed in GMCO with a targeted nominal composition of Ge0.5Mn0.5Co2O4. SEM/EDX analysis reveals that the sample predominantly consists of a Mn-rich primary phase with approximate stoichiometry Mn0.74Ge0.18Co2O4, along with a minor Ge-rich secondary phase of composition Ge0.91Mn0.19Co2O4. Although both GCO and GMCO crystallize in cubic symmetry at room temperature, a substantial difference in low-temperature structural properties has been observed. Magnetic and heat capacity data indicate ferrimagnetic ordering in the Mn-rich phase near TC = 108 K, while the Ge-rich phase exhibits antiferromagnetic order at TN = 22 K in GMCO. Analysis of heat capacity data reveals that the estimated magnetic entropy amounts to only 63% of the theoretical value expected in GMCO. A collinear ferrimagnetic arrangement is observed in the Mn rich phase below the magnetic ordering temperature, characterized by antiparallel spins of the Mn at A site and Co at B site along the c-direction. At 5 K, the refined magnetic moments are 2.31(3) for MnA and 1.82(3) uB for CoB in the Mn rich ferrimagnetic phase. The magnetic structure at 5 K in the Ge rich secondary phase is identical to the antiferromagnetic structure of the parent compound GeCo2O4. The refined value of the CoB moment in this phase at 5 K is 2.53(3) uB.

Topical Area

Hard matter: quantum, electronic, semiconducting materials

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