

Understanding Magnetic Properties of Cd_{1-x}Sn_xCr₂Se₄ Using Local Structure Probes

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Abstract : Co-existence of long-range ferromagnetism and semi-conductivity with correlated behavior of structural, magnetic, optical and electrical properties in various sites doping at CdCr₂Se₄ makes it a most promising candidate for spin-based electronic applications and magnetic devices. It orders ferromagnetically below TC = 130 K with a direct band gap of ~ 1.5 eV. The magnetic ordering is believed to result from strong competition between the direct antiferromagnetic Cr-Cr spin couplings and the ferromagnetic Cr-Se-Cr exchange interactions. With an aim of understanding the influence of crystal structure on its magnetic properties without disturbing the magnetic site, we investigated four compositions with 3%, 5%, 7% and 10% of Sn-substitution at Cd-site. Partial substitution of Cd²⁺ (0.78Å) by small sized nonmagnetic ion, Sn⁴⁺ (0.55Å), is expected to bring about local lattice distortion as well as a change in electronic charge distribution. The structural disorder would affect the Cd/Sn - Se bonds thus affecting the Cr-Cr and Cr-Se-Cr bonds. Whereas, the charge imbalance created due to Sn⁴⁺ substitution at Cd²⁺ leads to the possibility of Cr mixed valence state. Our investigation of the local crystal structure using the EXAFS, Raman spectroscopy and magnetic properties using SQUID magnetometry of the Cd_{1-x}Sn_xCr₂Se₄ series reflects this premise. All compositions maintain the Fd3m cubic symmetry with tetrahedral distribution of Sn at Cd-site, as confirmed by XRD analysis. Lattice parameters were determined from the Rietveld refinement technique of the XRD data and further confirmed from the EXAFS spectra recorded at Cr K-edge. Presence of five Raman-active phonon vibrational modes viz. (T_{2g} (1), T_{2g} (2), T_{2g} (3), E_g, A_{1g}) in the Raman spectra further confirms the crystal symmetry. Temperature dependence of the Raman data provides interesting insight to the spin- phonon coupling, known to dominate the magneto-capacitive properties in the parent compound. Below the magnetic ordering temperature, the longitudinal damping of E_g mode associated with Se-Cd/Sn-Se bending and T_{2g} (2) mode associated to Cr-Se-Cr interaction, show interesting deviations with respect to increase in Sn substitution. Besides providing the estimate of TC, the magnetic measurements recorded as a function of field provide the values of total magnetic moment for all the studied compositions indicative of formation of multiple Cr valences.

Keywords : exchange interactions, EXAFS, ferromagnetism, Raman spectroscopy, spinel chalcogenides

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