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Understanding Magnetic Properties of Cd1-xSnxCr2Se4 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 Cd1-xSnxCr2Se4 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. (T2g (1), T2g (2), T2g (3), Eg, A1g) 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 Eg mode associated with Se-Cd/Sn-Se bending and T2g (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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