ABSTRACT

Gigantic magnetoelectric coupling between ferroelectricity and magnetism has been demonstrated in a number of rare-earth perovskite oxides such as manganites, cobaltates, nickelates and ironates with either hexagonal or orthorhombic structures. In terms of applications, multiferroics have shown their distinction as, (i) special layer in a spintronic device to enable electric field control of magnetization and (ii) novel solid-state refrigerant due to their novel multicaloric cooling property. In this work, we have carried out a comprehensive study on selected rare-earth based double perovskites with Co and Mn occupied at the B-site to investigate the multiferroicity, phase separation and magnetic field induced phenomena like magnetodielectric, multicaloric, and exchange bias effects. Determining the intrinsic and extrinsic contributions to magnetodielectric effect to elucidate the authenticity on intrinsic ferroelectricity and magnetoelectric coupling is one of the central themes of the thesis. Such an analysis in parent B-site ordered La_2CoMnO_6 polycrystalline and nanoparticles sample has indicated the interplay of multiple sources and signify the effect of disorder on magnetodielectric effect. By replacing La by Y, a smaller but non-magnetic rare-earth ion, a systematic magnetization study on Y_2CoMnO_6 has revealed field induced metamagnetic phase transition from E*-type antiferromagnetic to ferromagnetic phase. In Y_2CoMnO_6, the critical field required for the sharp jump can be increased or decreased depending on the magnitude and direction of the cooling field; this is remarkably different from manganites where the critical field increases irrespective of the direction of the field cooling. A model has been proposed to account for the cooling field dependence on the sharp magnetization jumps. We have demonstrated magnetocaloric, electrocaloric and multicaloric effects in Y_2CoMnO_6 and discussed the significance of sign of magnetoelectric coupling on cooling efficiency. When replaced the La by magnetic rare-earth element Gd, we have found several complex magnetic interfaces at low temperatures below the ferromagnetic ordering (T_C ~112 K). Gd_2CoMnO_6 showed giant value of entropy changes (near to Gd spins ordering ~5 K) and exchange bias effect below the antiferromagnetic ordering of negative 3d-4f exchange interactions (T_N ~47 K) and field induced reentrant spin-glass behavior below T_C. We have doped Sr for La and investigated the role of disorder in $La_{2-x}Sr_xCoMnO_6$ ($0<x<1$) and found a spin disorder to ordering with Sr content. Samples with x=0.1 to 0.6 have exhibited spontaneous exchange bias and enhanced conventional exchange bias effects. A giant spontaneous exchange bias of ~0.65 T in x=0.5 sample is highest reported till date.

Keywords: Multiferroicity, phase separation, multicaloric effect, exchange bias effect, magnetodielectric effect, re-entrant spin glass and double perovskites.