Abstract

Designing new functional materials, or finding new functional properties of existing ones is necessitated by new technological challenges. In this regard, the discovery of perovskites in 1839 was a landmark. The discovery of double perovskite (DP) materials has led to an explosion of theoretical and experimental activities over the last two decades due to their fascinating structural, electronic, magnetic, and optical properties and their potential applications. The physical properties of the DP materials can be tuned by varying different parameters leading to potential applications. In this thesis, we first try to unravel the role of defects and disorder in determining the structural, electronic, magnetic, and optical properties of La_2CoMnO_6 (LCMO) DP material. Defects are introduced in the structure by hole and electron doping at the La site and O-site, respectively. Our study shows that Co and Mn ions alter their valence states to maintain the overall charge neutrality of the system while structural changes accommodate the strain, caused by doping. It is found that doping helps to magnify anti-site disorder at the Co/Mn site. The electronic ground state of anti-site disordered LCMO and ordered/disordered $LaSrCoMnO_6$ are found to be half-metallic with hole doping. However, electron doping brings it back to its insulating state. Also, the Curie temperature of the system reduces enormously with doping. We observe that the defects introduce a significant amount of anisotropy in the system with high birefringence value, which has immense technological importance for optical devices. Next, we investigate the physical properties of the Y_2CoMnO_6 DP compound with defects. Magnetic Gd atom with a larger atomic radius is partially substituted at the Y site in order to introduce structural defects. Our results show that Co and Mn ions are present in mixed-valence states to compensate for the distortion caused by Gd doping. Multi-magnetic phases appear due to frustration and $3d$-$4f$ interactions evolve with doping. An appreciable value (38%) of ASD is found on Gd doping. Our measurements show a conventional exchange bias effect, with a high value of $H_{EB} = 1.07$ kOe and $H_C = 8.85$ kOe is observed below T_N, resulting from FM/AFM interactions. This makes the DP materials a promising candidate for application in spintronic devices. It is always interesting to unveil the microscopic mechanism behind these novel properties. Spin-phonon coupling (SPC) offers a fundamental context for comprehending a variety of fascinating phenomena. Towards the end, we address the issue of the correlation of observed SPC with defects in the $YGdCMO_6$ DP compound. The stretching and anti-stretching modes show softening below FM transition temperature T_c with a discontinuity near T_N. This is a clear manifestation of the SPC present in this sample and its coefficient value (λ) is found to be 0.29 cm^{-1}. Such a lower value of SPC coefficient probably arises due to ASD.

Defects can modify the oxidation state of transition metal oxides and exchange pathways hence altering the existing magnetic order. It can also improve other physical properties to a great extent. Therefore, disorder can be a very useful handle for added or modified functionality in perovskite oxides.

Key words: Double perovskite, antisite-disorder, Density functional theory, Magnetism, AC-susceptibility, Exchange-bias, Spin-phonon coupling, Multiferroicity.