Preface

Spin frustration or competing interactions between the interacting degree of freedom is one of the eternal and enlightened research area in condensed matter world. Geometrical Spin frustrated systems in which magnetic moments reside on corner sharing tetrahedra show multiple degenerate ground states. In such frustration mechanism, the systems do not maintain lowest energy state and switch to different magnetic ground states. Triangular lattice, kagome lattice and pyrochlore lattice are the name of some lattices which shows geometrical spin frustration. Particular, Pyrochlore material exhibits exotic physical and magnetic properties even at very low temperature. Generally, pyrochlore materials exhibit form $A_2B_2O_7$ type empherical formula in which A-site ion is rare earth elements and B-site elements are 3d transition elements. Among the Pyrochlore materials, Rare earth titanates ($R_2Ti_2O_7$) reveal different types of magnetic phases like $Tb_2Ti_2O_7$ as spin liquid, $Dy_2Ti_2O_7$ as spin ice, $Gd_2Ti_2O_7$ as long range ordered systems, $Eu_2Ti_2O_7$ as X-Y antiferromagnetically ordered systems and much more. Enrich in physics and utilization in real world make these materials important among geometrically frustrated systems.

Along with geometrical spin frustration, disorder driven spin frustration seeks unusual attention in recent times. When the disorder presents in a system causes the spin frustration among the structure, known as disorder driven spin frustration. The single perovskite and double perovskite which illustrates spin frustration due to presence of disorderness comes in the class of multifunctional materials. The double perovskite materials have an $A_2BB'O_6$ type empirical formula i.e. $ABO_3.AB'O_3$. One unit cell of double perovskite material consists of two unit cells of single perovskite materials. In this empirical formula A-site is generally alkaline or rare earth element and B and B'-site ions are 3d, 4d, 5d transition elements**.** The correlation between electrical and magnetic ordering at room temperature makes these materials more efficient and applicable in many devices such as memory devices, magnetic sensors, magnetic recording media etc. Eventually, numerous dynamic properties and magnetic processes like presence of high temperature magnetic ordering, exchange-bias, spin glass/cluster glass, Griffith like phase, high dielectric constant with low loss tangent, large magneto-dielectric frame the double perovskite systems more efficient and promising. Although, $A_2Co/Mn/NiO_6$ type double perovskite materials have been widely investigated. Still, there is a large scope to flourish the characteristics of La_2CoFeO_6 type double perovskite compounds. Moreover, Rare earth element and hole doping at La site is one of the best way to intensify such alluring and unusual properties and prepared them into use of different practical applications.

Therefore, our research work is focused to enhance the ravishing properties of rare earth titanates and La_2CoFeO_6 type double perovskite materials. In present thesis, the structural, magnetic, transport and magneto-dielectric properties of spin frustrated systems have been investigated. As to provide systematic discussion, this thesis has been classified into seven chapters. Outline of each seven chapter is given below-

Chapter 1, In chapter 1, the introduction of magnetism along with different models which classifies the different type of magnetic ordering has been explained. With this, the theory and concepts of spin frustration with their related materials has also been discussed. Two types of spin frustration i.e. geometrical spin frustration and disorder driven spin frustration are addressed in this chapter. Brief introduction of Pyrochlore material, their different magnetic phases, physics of perovskite and double perovskite materials with their literature review have been elaborated in this chapter. Here, we have also discussed various important magnetic phenomenons like spin relaxation process, exchange-bias phenomenon, spin glass/cluster glass, Dzyaloshinsky-Moriya (D-M) interaction etc. Moreover, different electrical transport models for conductivity are also discussed like variable range hopping model, Arrhenius model.

Chapter 2, This chapter contains the synthesis process involved in our research work. The basic principles and mechanism of different characterization tools which we have employed in our research work is also explained in this chapter. X-ray diffraction, Raman spectroscopy, neutron diffraction, X-ray absorption spectroscopy, X-ray photoemission spectroscope techniques has been described the structure, phase and electronic state identity. The description of Quantum Design MPMS magnetometer has been given for magnetic analysis. Dielectric and magneto dielectric analysis is also included in this chapter.

Chapter 3, This chapter investigated the structural and magnetic properties of $(Tb_{1-x}Eu_x)_2Ti_2O_7$ pyrochlore compounds. All the ordered samples have cubic crystal structure with Fd−3m space group. High series expansion fit on temperature dependent DC magnetic data reveals the presence of dipolar and exchange interactions in Eu rich samples. Spin liquid $Tb_2Ti_2O_7$ compound shows a partial spin freezing at ~ 33 K in presence of zero magnetic field. This partial spin freezing of $Tb_2Ti_2O_7$ compound is single ion spin freezing in nature. The AC magnetic susceptibility data observed this partial single ion spin freezing for all compounds. A single moment saturated field induced transition is observed for all compounds at low temperature in presence of 10 kOe magnetic field.

Chapter 4, This chapter presented the systematic structural, raman and magnetic study of $Dy_{2-x}Eu_xTi_2O_7$ pyrochlore materials. A systematic shift in all phonon modes with higher Eu content in $Dy_{2-x}Eu_xTi_2O_7$ observed in Raman spectra confirms the successful substitution of Dy^{3+} ion by Eu^{3+} ions. High concentration of Eu induces the exchange interactions between atoms and alters the crystal-field interactions in $Dy_{2-x}Eu_xTi_2O_7$. Rich Eu content samples (x= 1.5, 1.8 and 1.9) show the existence of wasp-waisted hysteresis loop which is explained on the basis of dipolar field, anisotropy exchange interaction and spin frustration present in the samples. Interestingly, AC susceptibility shows two single ion spin freezing transitions corresponding to Dy^{3+} and Eu^{3+} ions respectively in Eu rich samples only.

Chapter 5, This chapter deals with the effect of spin frustration due to disorder present in $La_{1.8}Pr_{0.2}CoFeO_6$ double perovskite sample. The compound was investigated by the X-ray diffraction, X-ray absorption spectroscopy, magnetic, dielectric (zero-field and in-field) and Raman spectroscopy techniques. The same oxidation state (+3) for Co and Fe ion is identified through XAS spectra. Above room temperature magnetic ordering with existence of re-entrant cluster glass state is analyzed by magnetic data. This Double perovskite sample is also renowned for magneto-dielectric coupling (MDC) which is observed in two temperature regions (25-80 K and 125-275 K). Presence of cluster glass at low temperature and spin lattice interaction at high temperature are the possible explanation of magneto-dielectric coupling. The resistivity measurement reveals its semiconducting nature which was explained by different conduction models. Eventually, huge dielectric constant and the low tangent loss make $La_{1.8}Pr_{0.2}CoFeO_6$ sample promising candidate from the application point of view.

Chapter 6, This chapter includes the , X-ray diffraction, X-ray photoemission spectroscopy (XPS), Neutron diffraction, magnetization measurements and density function theory calculations of double perovskite $La_{1.5}Ca_{0.5}CoFeO_6$ sample. A ferrimagnetic transition is observed at T_C ~ 167 K. The ferrimagnetic ground state is also confirmed by DFT and neutron powder diffraction. The presence of anti-site disorder (ASD) has also been demonstrated. Double re-entrant cluster glass transitions (T_1~ 11 K and T_S~35 K) were observed which has been attributed to the ASD effect. Both conventional and spontaneous exchange bias are observed in this double perovskite system. The observed values of large H_{SEB} ~ 2.106 kOe and giant H_{CEB} ~ 1.56 T at 5 K might be due to the coexistence of long range magnetic ordering and glassy state. The presence of double glassy states, large exchange bias effect and different magnetic phases make this system a potential candidate for spintronics applications.

Chapter 7, This chapter keeps the details of conclusion of our research work presented in this thesis. The brief glimpse of future research work has also added in this chapter.