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Signature:

(Supervisor)

Prof. Sandip Chatterjee
Department of Physics
Indian Institute of Technology
(Banaras Hindu University)
Varanasi - 221005, India

DECLARATION BY THE CANDIDATE

I, "***Mohd Alam***", certify that the work embodied in this thesis is my own bona fide work and carried out by me under the supervision of "***Prof. Sandip Chatterjee***" from "***July 2017***" to "***January 2022***", at the "***Department of Physics***", Indian Institute of Technology (BHU), Varanasi. The matter embodied in this thesis has not been submitted for the award of any other degree/diploma. I declare that, I have faithfully acknowledged and given credits to the research workers wherever their works have been cited in my work. I further declare that I have not willfully copied any other's work, paragraphs, text, data, results, etc., reported in journals, books, magazines, reports, dissertations, theses, etc., or available at websites and have not included them in this thesis and have not cited as my own work.

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(***Prof. Sandip Chatterjee***)
Supervisor

(***Prof. Sandip Chatterjee***)
Head of the Department

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Sincerely

(*Mohd Alam*)

DEDICATED

To

MY FAMILY

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Preface

Magnetically frustrated materials that possess two or more than two degenerate ground magnetic states in a single phase have much attracted peoples since past few times. The presence of magnetic frustration leads the to the several interesting physical states due to the different competing magnetic interactions. The magnetic frustration arises mainly owing to the disorder present in the system and the special geometry of the system, where the magnetic spins are unable to seek a single stable ground state. The perovskite (ABO_3) and double perovskite ($A_2BB'O_6$; where at A-site are occupied by alkaline/rare earth ions & B and B'-site with 3d/4d/5d transition metal ions) are the examples of materials where presence of disorder drive magnetic frustrations. In earlier literature, it is reported that the Co/Mn based rare earth double perovskites (R_2CoMnO_6; R = rare earth elements), presence of magnetic frustration leads emergence of different magnetic singularities viz. low-temperature spin glass/re-entrant spin glass/cluster glass, Griffiths-like-phase, exchange bias, multifunctionality, meta-magnetism, etc. The interesting magnetic state in these double perovskites mainly arises owing to different Co-O-Mn/Co-O-Co/Mo-O-Mn exchange interactions as well their interaction with rare-earth ions. So, they can be exploring for different technological devices especially spintronic devices.

However, the Kagome lattice, triangular lattice & pyrochlore oxide lattices are the example of geometrically driven magnetic frustrated materials. Among them, the pyrochlore oxide ($A_2B_2O_7$; having A-site rare earth ions & B-site 3d transition metal ions) materials are famous particularly for their very low temperature mysterious magnetic states. Among them, the rare earth-based titanates ($R_2Ti_2O_7$) pyrochlore make known for different magnetic phases like spin liquid, spin ice, X-Y anti-ferromagnetic ordering and many more rich physics make these materials to be utilize in real life in technological application as well as for understanding and correlating different physical phenomena.

In the assessment of the above statements, we focussed on the investigation of the "Magnetic and Electric properties of Some Magnetically Frustrated Materials" for our Ph.D. thesis. Our detailed investigations on these materials have resulted in several new important findings not reported by earlier authors. The outlines of different findings resulting from investigation on these systems in this thesis work is briefly mentioned below:

In **Chapter 1,** the broad area of present investigation has been introduced. The detail introduction of magnetism and classification of magnetic materials depending on their attraction/repulsion in external magnetic field have been described. Then, we have discussed the frustration and spin/magnetic frustration and reason of arising the same. Further, the different type of magnetically frustrated materials has been introduced and main focus has been given to perovskites/double perovskite (which have disorder driven magnetic frustration) and pyrochlore oxides (which have geometrical driven magnetic frustration). A brief literature review on double perovskite and pyrochlore have also been carried out in this chapter. We have discussed many interesting phenomena, which usually possess by above mentioned the magnetically frustrated oxides. Finally, we have discussed the motivations and objective, why we have started working on the particular system.

In **Chapter 2,** we have described the methodology of sample preparation and different experimental techniques and their basic principles, which was used for different characterization & measurements in our study. The X-ray diffraction measurement, Raman spectroscopy and X-ray photoemission spectroscopy have been emplyoed for structural, phase and elemental chemical state identification. The detail introduction of MPMS3 magnetometer used for different type of magnetization measurements with its working principle have been described. We have also added other techniques viz. UV-Vis. spectroscopy, dielectric measurement, electrical resistivity measurement, magneto-transport measurement, etc. in details in this chapter.

In **Chapter 3,** some novel magnetic behaviors of the double perovskite Eu_2CoMnO_6 (ECMO) have been described. The X-ray diffraction analysis confirms that it crystallized in single pure orthorhombic phase with space symmetry Pnma. The XPS study shows the presence of mixed valence states of transition metal ions. The UV-Visible absorption spectroscopic study suggests that the ECMO has a direct wide band gap. A second-order magnetic phase transition as a sudden jump in the magnetization curve has been observed around 124.5 K. The large bifurcation between the ZFC and FC, suggests existence of strong spin frustration in the system. The inverse DC susceptibility confirms the presence of the Griffiths like phase. Sharp steps in magnetization have been observed in M-H curve at 2 K, which vanishes on increasing temperature. The AC susceptibility study demonstrates the Hopkinson like effect as well as presence of volume spin-glass-like behavior. The temperature dependent Raman spectrum shows the presence of spin-phonon coupling.

In **Chapter 4,** transport, dielectric and magnetic behaviors of polycrystalline double perovskite $Eu_{2-x}Tb_xCoMnO_6$ (x = 0.0 and 1.0) have been investigated. Temperature-dependent resistivity follows the variable range and small polaron hopping mechanism. The temperature-dependent dielectric property shows usual frequency-dependent step-like behavior along with thermally activated relaxor peak in loss curves with colossal dielectric constant near room temperature. The temperature-dependent magnetization for the both samples demonstrate a second order phase transition around 125 K (x = 0) and 113 K (x = 1) respectively. The critical behavior near the magnetic transition were systematically investigated. The reliability of critical parameters was verified by the Widom scaling relation and magnetic entropy change calculation. The estimated values of critical parameters are close to the value of the theoretical mean-field model.

In **Chapter 5,** we have reported the structural, magnetic, transport, dielectric and complex impedance of polycrystalline double perovskite $EuPrCoMnO_6$ which crystallizes in disordered orthorhombic phase with space group Pnma. The DC magnetization shows two successive ferromagnetic transitions around 146 K and 138 K. The temperature and magnetic field variation of DC-susceptibility suggest the existence of Griffith phase and spontaneous exchange bias. AC susceptibility measurement shows a glassy dynamic behavior near ferromagnetic transition. Further a re-entrant glassy dynamic state is seen at low temperature around 40 K. Temperature-dependent resistivity shows semiconducting/insulating nature, which gets increased under application of magnetic field, showing positive magnetoresistance. The dielectric study shows usual frequency-dependent step-like behavior with a colossal dielectric constant near room temperature. The complex impedance study shows both grain and grain boundary contribute in electrical properties. The observed properties suggest the material can be used for spintronic device and high dielectric applications.

In **Chapter 6,** the temperature-dependent Raman spectroscopy and dielectric property of pyrochlore $Eu_2Ti_2O_7$ have been investigated. Phase purity of the sample was confirmed using the structural analysis. The appearance of additional phonon modes below 200 K is observed suggesting a local structural change. The anomalous softening of the phonon modes and existence of crystal field induced short range magnetic ordering below 150 K unveiled the possible spin-phonon coupling. The signature behavior of a dipolar glassy-relaxor state and super-paraelectric nature have been demonstrated. Additional dielectric anomaly at lower temperature, below 40K, related to spin-spin correlation has also been reported.

In **Chapter 7**, a brief conclusion and glimpse/possibility of future scope in this research area have been discussed.