PREFACE

Spin frustration or competing interactions between the interacting degree of freedom is one of the eternal and enlightened research areas in the condensed matter world. In this regard, disorder-driven spin frustration seeks immense attention in recent research. In these systems, spin frustration is generated by the disorder present in the system thus called disorder-driven spin frustration. Double perovskite exhibit disorder-driven spin frustration and have potential applications for multifunctional devices such as future generation four-state memory devices, spintronics, actuators, and transducers. The double perovskite has an $A_2BB'O_6$ type empirical formula i.e., $ABO_3.AB'O_3$. A double perovskite unit cell is formed by combining two-unit cells of single perovskite material (ABO_3). In general, the A-site is occupied by the alkaline or rare earth element, and B and B' sites are occupied by 3d, 4d, and 5d transition elements which makes an oxygen octahedral such that two types of BO_6 and $B'O_6$ sites are repeated in three directions to give the structure. These materials have a strong correlation among structural, electrical, electronic, and magnetic properties. Therefore, these materials led to huge attention from the scientific community for their profound physics and multifunctional behavior. Eventually, numerous dynamic properties and magnetic phenomena like the presence of high-temperature magnetic ordering, giant exchange bias, spin glass or cluster glass, Griffith phase, meta-magnetic transition, high dielectric constant with low loss, magnetodielectric coupling, and magnetoresistance frame the double perovskite systems more efficient and promising. Although A_2CoMnO_6 and A_2CoFeO_6 type double perovskites compounds are widely investigated, still there is a lack of studies exploring the correlation between electronic, Raman studies, and the magnetic study. Moreover, the Tb_2CoMnO_6 is relatively less explored as compared to other Co/Mn-based double perovskite systems. Thus, there is a large scope to flourish the characteristics

of the $Tb_2CoMnO6$ system. Moreover, to generate a suitable platform for giant exchange bias, the system whose end members have an exchange bias effect and lies in the different category of ordered and disordered family (such as Pr_2CoFeO_6 and Pr_2CoMnO_6) is one of the best ways to intensify such alluring and unusual properties along with the colossal exchange bias.

Along with the disordered-driven spin frustrated systems, the low-dimensional systems, in particular, 1D or quasi-1D magnetic systems where the magnetic ions interact preferentially along the single direction in space have attracted unprecedented attention in recent times. The interest is arsing by the fact that the 1D leads to a very rich physics as the quantum effects are enhanced by the low-dimensionality. Thus, among all the quantum manifestations of matter, the quantum phase transitions corresponding to the abrupt change of the ground state of much body system due to the quantum fluctuations by varying the parameters are particularly enhanced in the 1D antiferromagnetic system. Thus, the different experiments such as neutron scattering experiments coupled to the macroscopic measurements such as magnetic study, Raman study, electronic study along with the theoretical calculations allow a comprehensive study of both static and dynamical properties of these quantum spin chains, and therefore a better understanding of quantum phase transitions occurring in such systems.

This thesis aims to study the rigorous magnetic behavior of the double perovskite system and 1D quasi spin chain system and their correlation between electronic and Raman study. Our research work focused on the enhancement of the ravishing properties of double perovskite and the 1D spin chain system. Therefore, for a better understanding rich and correlated physics of the spin chain system, we study the $BaMn_2V_2O_8$ spin chain system since among all the other family members it has a magnetic transition temperature quite high that makes it feasible to unravel the different phenomena. Thus, to give a systematic

discussion, this thesis has been classified into seven chapters, and the outline of each chapter is given below.

In chapter 1, the introduction of magnetism along with the different models and the classification of the different types of magnetic ordering has been explained. With this, the theory and the concepts of spin frustration are explained. The disorder-driven spin frustration and related material i.e., double perovskite systems are addressed in this chapter. A brief introduction of the double perovskite and 1D spin chain system and their different magnetic phases, and the physics of these systems have been elaborated in this chapter. Here, we have also discussed various important phenomena like spin glass/cluster glass phase, exchange bias, Griffith phase, spin-phonon coupling, the correlation between electronic properties and magnetic properties, etc. Moreover, a brief motivation is also discussed in this chapter.

In chapter 2, the synthesis process involved in our research is discussed in detail. The basic principles and mechanisms of different characterization tools employed in our research work are also elaborated in this chapter. This chapter contains X-ray diffraction, Raman spectroscopy, neutron diffraction, X-ray absorption, and X-ray photoemission spectroscope techniques that have been described to study structure, phase, and electronic state identification. The X-ray magnetic circular dichroism is also included for the magnetic nature of the individual elements. The description of the Quantum Design MPMS magnetometer has also been provided for the magnetic properties analysis.

In chapter 3, we have investigated the structural, electronic, and magnetic properties of double perovskite Tb_2CoMnO_6. The structural study by X-ray diffraction and neutron diffraction are well supported by each other, although the neutron diffraction analysis suggested the presence of anti-site disorder in the system. As a matter of fact, the anti-site

disorder is further supported by the electronic structure analysis through an X-ray photoemission spectroscopy study which reveals the presence of a mixed oxidation state (Mn^{4+}/Mn^{3+} and Co^{2+}/Co^{3+}) of B-site ions. It has been demonstrated that the presence of inherent anti-site disorder along with mixed valence states of B-site ions and Jahn-Teller active ions are the most important ingredients for the evolution of interesting phenomena such as the emergence of the Griffith phase, the appearance of Hopkinson-like peak, and also unusual slow relaxation. Each phase is confirmed by the various magnetic measurements and models.

In chapter 4, we have further explored the double perovskite compound Tb_2CoMnO_6 using X-ray absorption spectroscopy, Raman spectroscopy, magnetic measurements, and *ab* intio band structure calculations. It is observed that both the anti-ferromagnetic and ferromagnetic phase coexists in this material. We have observed a prominent metamagnetic transition in the M(H) behavior that has been explained by the drastic reorientation of the pinned domain which is aligned antiparallel by the antiphase boundaries at zero fields. The further study of the ac susceptibility suggested spin frustration at low temperature which leads to the re-entrant cluster glass ~ 33 K. The coupling between the phononic degree of freedom and spin in the system has also been demonstrated. It is observed that the theoretical calculation is consistent with that of the experimentally observed behavior.

In chapter 5, we have studied the antiferromagnetic material with a giant exchange bias effect that can be used for high-density spintronic devices. In this chapter, we investigated structural (geometrical and electronic) and magnetic studies in the polycrystalline $Pr_2CoFe_{0.5}Mn_{0.5}O_6$ double perovskite system. The observed lack of training effect suggests the existence of robust exchange bias. In addition, the detailed magnetic studies and Raman studies unravel the Griffith phase along with the spin-phonon coupling in the present system. The XPS analysis supports the more than valence state of B-site elements which

generates competition between ferromagnetic and antiferromagnetic interactions along with the anti-site disorder in the system. The neutron measurement confirms the G-type antiferromagnetic spin arrangement which is well supported by the DFT calculation. The magnetic studies have been well correlated with the electronic structure, neutron study, and theoretical first principle calculations.

In chapter 6, we have investigated the spin-chain compounds which are known to exhibit fascinating magnetic properties depending on the competing intrachain and interchain exchange. Our neutron diffraction and magnetization study unravelled a collinear AFM long-range ordering at a relatively higher temperature T_N~38 K for the present quasi-one-dimensional spin-chain material $BaMn_2V_2O_8$, system, wherein the nearest-neighbor spins have antiferromagnetic coupling along the spin-chain, i.e., along *c*-axis. The study also reveals the existence of the short-range magnetic ordering prevailing at considerably elevated temperatures above its transition temperature. Furthermore, theoretical calculations based on a classical Heisenberg model demonstrate that the nearest-neighbor intrachain exchange interactions govern the magnetic ground state in this system, while the interchain interactions are much weaker. Temperature-dependent Raman spectroscopy study demonstrates an occurrence of spin-phonon coupling below transition temperature at least for two modes, whereas the study also shows an unusual thermal evolution of the Raman modes above transition temperature which can be presumably attributed to the short-range magnetic ordering.

In Chapter 7, the summary of the present thesis with a brief glimpse of future studies is contained.