Preface

Greetings from the fascinating world of multiferroic materials, where researchers are working to solve the puzzle of simultaneous ferromagnetic and ferroelectric capabilities. Recent years have seen a tremendous amount of interest and several scientific inquiries generated by discovering and studying these fascinating materials. Ferroelectric and ferromagnetic materials were once considered separate things with their characteristics and behaviors. This traditional knowledge was disproved by the development of multiferroic materials, which introduced an entirely novel category of compounds capable of simultaneously showing ferroelectric and ferromagnetic properties. Unusual opportunities for essential study and practical improvements have been made possible by the amazing coexistence of electric and magnetic orderings. The investigation of multiferroics has uncovered a wide range of materials with various structural and functional characteristics. Researchers have delved into several areas of material science to find and comprehend these interesting systems, from oxides of perovskite to organic molecules and much more. Through their research, several well-known multiferroic substances were uncovered, each with unique features and possible uses. For a vast array of technological applications, the special qualities of multiferroics offer enormous potential. Significant developments in fields including storage of data, detection, energy generation, and multimedia processing are made possible by the ability to regulate both the electric and magnetic characteristics of a single material. Technology can undergo a revolution by using the potential of multiferroic materials, opening the door for cutting-edge products that were previously unthinkable. Nevertheless, despite of the advancements achieved in this area, there are still many obstacles to overcome.

Researchers throughout the world are still searching for novel multiferroic materials with improved characteristics and stability. Furthermore, it remains a challenging undertaking that needs the cooperation of specialists from other fields to fully understand the complex mechanisms behind the coexistence and coupling of ferroelectric and ferromagnetic regimes. Bismuth Ferrite, which concurrently exhibits exceptional ferroelectric and ferromagnetic characteristics, has recently emerged as one of the most fascinating and promising multiferroic compounds. Bismuth Ferrite is a perovskite crystal structure consisting of successive layers of bismuth (Bi) and iron (Fe) atoms arranged according to the crystallographic axis. By occupying

the interstitial spaces between these layers, the oxygen (O) atoms create a three-dimensional structure. For Bismuth Ferrite, this configuration results in a rhombohedral crystal structure. $BiFeO_3$'s ferroelectricity appears to have a source in the stereochemical activity of the Bi^{+3} $6s^2$ lone pair electrons, whereas the canted G-type antiferromagnetism with a periodicity of 62 nm is caused by the modulated cycloidal spin structure of the transition metal ion Fe^{+3} (d^5) on the B-site. Still, due to numerous problems in synthesizing, characterization, and achieving the appropriate degree of magnetoelectric coupling, the potentials of this material have yet to be achieved.

Structure, electricity, electronics, and magnetism are all closely correlated in these materials. Therefore, due to their fundamental physics and multifunctional behavior, these materials attracted a lot of interest within the scientific community. In the long run, a variety of dynamic characteristics and magnetic phenomena, including the presence of high-temperature magnetic ordering, exchange bias, spin glass or cluster glass, high dielectric constant with low loss, magnetodielectric coupling, and magnetoresistance, frame the double perovskite systems to make them more effective and promising.

The goal of the current thesis is to establish improved magnetoelectric multiferroic ability by fabricating a system with solid state method using the co-doping method at A-site (Tb, La) and B-site (Mn, Al).

In Chapter 1 we have introduced the ferroic materials and then discussed different types of magnetic and ferroelectric materials. Further we have discussed different types of coupling between ferroic materials, multiferroic materials, and magnetoelectric coupling. There are two types of multiferroic materials and further, also they are categorized into different types. Then we did a structural and our materials of interest historical review study. At last, we have discussed some interesting phenomena such as exchange bias and spin glass behavior.

In Chapter 2 we have discussed the sample preparation techniques and experimental tools of characterization. To explore structure, phase, and electronic state identification, X-ray diffraction, Raman spectroscopy, neutron diffraction, X-ray absorption, and X-ray photoemission spectroscopy techniques have been discussed in this chapter. For information of the magnetic properties of the different elements, X-ray magnetic circular dichroism is also introduced. For the investigation of magnetic characteristics, a description of the Quantum design MPMS magnetometer has also been given.

In Chapter 3 the structural, dielectric, and magnetic properties of $Bi_{0.8}Tb_{0.2}Fe_{0.8}Mn_{0.2}O_3$ have been described. A structural shift from rhombohedral (space group R3c) to orthorhombic (space group Pn21a + Pnma) phase is observed with doping. The substitution of Mn and Tb gives a very large value of the dielectric constant. It has been demonstrated that the observed spontaneous exchange bias is larger than the conventional exchange bias at room temperature. Moreover, doping reduces the Neel temperature from 643 to 521 K, and magnetization increases. Thermoremanent magnetization studies at room temperature show that the system is composed of an interfacial layer of an antiferromagnetic core and a two-dimensional diluted antiferromagnet shell with a net magnetization under the field.

In Chapter 4 the structural, electrical and magnetic properties of $Bi_{0.90}Tb_{0.1}Fe_{0.90}Mn_{0.1}O_3$ system have been explored. We carried out UV-visible measurements and obtained a smaller band gap (i.e., semiconductor-type behavior). We analyzed the structural phase of the present system with X-ray diffraction and Neutron diffraction measurement. Structural phase analysis revealed the system contains two nuclear phases (rhombohedral structure (R3c space group) with orthorhombic (Pn21a space group)). Moreover, we also found more bending in the bond angle, and the existence of a magnetic phase with a nuclear phase for the $Bi_{0.90}Tb_{0.1}Fe_{0.90}Mn_{0.1}O_3$ system is also confirmed by Neutron diffraction. The magnetic moment versus temperature (M-T) curve demonstrates that our system's Neel transition temperature is at 568 K. The magnetization data shows enhancement in the magnetic property by showing the weak ferromagnetic-type behavior at room temperature in the magnetic field vs. magnetic moment (M-H) curve as compared to the parent compound. From dielectric measurement, the dielectric constant increases while the loss decreases.

In Chapter 5 we have reported the magnetic, dielectric, and structural properties of the perovskite $Bi_{0.5}La_{0.5}Fe_{0.4}Al_{0.1}Mn_{0.5}O_3$ which crystallizes in disordered orthorhombic phase with the space group *Pnma*. We found two consecutive magnetic transitions at 42 K and 147 K which have been suggested to be associated with the spin-glass and long-range ordering transitions respectively. A spin-phonon coupling has also been demonstrated. The dielectric findings suggest the unusual frequency-dependent step-like trend with a high dielectric constant and relaxor behavior at ambient temperature. The discovered characteristics suggest that such material is suitable for spintronic devices and high dielectric applications.

In Chapter 6 we explored the structural, magnetic, and dielectric properties of erbium (Er)

doped polycrystalline $GaFeO_3$. Solid state reaction was used to synthesize polycrystalline $Ga_{0.75}Er_{0.25}FeO_3$ sample. We have found transition temperatures near room temperature i.e. 293 K. There is a pinched type behavior in the M-H loop (i.e. also known as wasp-waisted hysteresis) at 2.2 and 10 K after doping of Er by 25% at the Ga site. We found Debye-like dipolar relaxation behavior from dielectric measurement and found a low loss value.

Chapter 7 This chapter includes an overview of the current thesis and a summary of forthcoming research.