This thesis concerns and combines the results of experimental studies on magnetization and magnetotransport properties of electron–doped manganites using PPMS and SQUID magnetometry as the main characterization techniques. In addition, the dielectric properties of the ferroelectric–ferromagnetic composites material are investigated. All the compounds are prepared by standard solid state reaction method. A special attention has been paid to several experimental techniques such as x-ray diffraction, Rietveld analysis, scanning electron microscopy (SEM), impedance spectroscopy to characterize the compounds. This work is a systematic approach towards synthesis of electron–doped manganites followed by measurements of electrical conductivity as a function of temperature and magnetic field. These diverse measurements allow a comprehensive understanding of electron transport in the framework of hopping theory.

A representative class of such materials, which has attracted considerable attention, is of doped manganese oxides with a perovskite related structure, commonly known as manganites. In these systems the charge, spin, orbital and lattice degrees of freedom are not only very active but strongly entangled as well. These correlations manifest themselves in a variety of conducting and insulating phases with different types of structural distortions, electronic properties and magnetic ordering. The degeneracy not only allows the different phases to coexist under certain conditions but also makes these systems extremely susceptible to a wide variety of external perturbations like temperature, magnetic fields and internal parameters like doping level and electronic bandwidth.

The physical mechanisms and empirical relationships found among the magnetism, resistivity and magnetoresistance are studied in greater depth and compared.
Synopsis

with theoretical predictions The basic manganite oxide CaMnO₃ is a paramagnetic insulator at the room temperatures. It exhibits paramagnetic to antiferromagnetic transition around 125 K. Any substitution at divalent A-site by different trivalent rare-earth Re³⁺ having chemical formula of type (Ca₁₋ₓReₓ)MnO₃ drives Mn to B site to exist in mixed valent (Mn³⁺/Mn⁴⁺) state which is responsible for most of the magnetic and transport properties of the manganites. Both ferromagnetic and antiferromagnetic interaction is mediated by the charge carriers along the Mn–O–Mn bonds of the perovskite structure. The optimum trivalent cation doping at A site drives the resulting material to exhibit properties such as coincident paramagnetic–ferromagnetic/antiferromagnetic (PM–FM/AFM) transition and insulator–metal (I–M) transition. Due to this mixed valent state of Mn, these compounds are also called as mixed valent manganite oxides. The partial substitution by rare-earth ions affects the filling of 3d Mn, Mn–O–Mn bond angle and the tolerance factor. Consequently, these aspects in manganite-compound research significantly impinge on the magnetic and transport properties of the manganites.

In semiconducting region main three types of model have been found to be ruling the conduction mechanism such as (i) thermal activation (TA) model, (ii) small polaron hopping (SPH) model and (iii) variable range hopping (VRH) model. In TA model, if the thermal energy is sufficient to overcome the band gap, the electron becomes free to conduct and mostly applicable for high temperature. In SPH model, the thermal energy is sufficient to allow the electrons to hop to their nearest neighbors by multiphonon assisted process. In low-Τ region, the thermal energy is not enough to allow the electrons to hop to their nearest neighbors. In that case, it is more favorable for the electrons to hop further to a site with lower energy difference and the conduction mechanism is mostly of variable range hopping (VRH) type. In this scenario, the hopping distances can be different depending on the temperature.

The phenomenon of magneto resistance (MR) which is measure of resistivity drop when subjected to magnetic field and its potentiality in application in magnetic memory devices has spurred an interest in scientific community to synthesize new materials exhibiting large magneto resistance. The $ABO_3$ types of perovskite manganites exhibit negative magnetoresistance with many interesting inter related phenomena.
intrinsic factors such as the grain and grain boundaries structure, spin-polarized tunneling between the adjacent grains governs the magnetoresistance. This analysis provides a useful method for predicting the magnetoresistance as a function of temperature and magnetic field. The interplay with size of the cations to alter the MR properties of the manganite perovskites evolved plenty of scope in further research in related materials.

In the electron-doped region, the best transport and magnetic properties appear for $\text{Ca}_{1-x}\text{Re}_x\text{Mn}_2\text{O}_4$, $x = 0.15$ substituted system. The application of magnetic field forces the parallel alignment of spins causing drop in resistivity to favor Double exchange interaction and hence maximum MR effect. The lattice electron coupling or the polaron formation localizes the $e_g$ electron to show insulating behavior. The $A$-site cation radius distorts the structure of the perovskite that controls the buckling of the Mn-O bonds. The smaller size cation buckles Mn-O-Mn bond angle from its ideal 180-degree geometry. The various inter-related phenomena can be summarized as follows.

Manganite–ferrite composites are of interest due to its unusual magnetic and electrical properties. On the basis of technological importance, this composite can be used as magnetic tapes, sensing devices, read heads for hard disc & magnetic storage. The Dielectric properties such as relative permittivity, loss factor, impedance value of these composites are measured. By varying with frequency of the electric field, the change in dielectric properties and microstructural behaviour are observed. The relative permittivity values and loss factor were found to be decrease with increase in frequency. The impedance value was also found to be decrease with increase in frequency. The electric modulus data have been analyzed by invoking the decay relaxation function. All the formalisms describe a non-Debye relaxation involved in the composite. In the scaled coordinate, the dielectric permittivity and electrical modulus fall on a single master curve, indicating the existence of a general scaling formalism. The layout of this thesis is as follows.

**Chapter 1** gives an introduction and basic principles about of the doped rare earth manganites. Here an overview of the structural, magnetic and electronic properties of manganite is provided.

**Chapter 2** deals with the different theoretical models used to explain the transport and magnetic properties of mixed valence manganites. The concept of phase and the types of
phase transitions are addressed Fundamental concepts and terms used within this thesis are introduced.

**Chapter 3** essentially describes the synthesis procedure mainly the solid state reaction with high temperature programmable furnace followed by an overview of the x-ray diffraction measurements and electron imaging techniques, magnetization, resistivity, magneto-resistance, dc conductivity, dielectric measurement as a main characterization of the compounds Finally, a description of the instrumentation used to measure magnetization and resistivity is presented.

**Chapter 4** described different structural and morphological characterization the manganite samples using rietveld analysis of the x-ray diffraction data by fullprof suite and scanning electron microscope

**Chapter 5** discusses the results obtained from the magnetic field and temperature dependence of electrical resistivity and magneto-transport studies of the electron-doped manganites \((\text{Ca}_{0.85}\text{Re}_{0.15}\text{MnO}_3 \; \text{Re} = \text{La, Pr, Nd, Sm, Eu, Gd and Dy})\). The charge-ordered state and different types of hopping conduction depending on the temperature region are described. Magneto-resistance is also investigated considering the spin-polarized tunneling between the adjacent grains

**Chapter 6** deals with temperature-dependent magnetization, magnetic susceptibility \((\chi)\) of the electron-doped compounds \((\text{Ca}_{0.85}\text{Re}_{0.15}\text{MnO}_3 \; \text{Re} = \text{Pr, Eu and Dy})\). Nature of the magnetic phase transition in addition to the order of the transition is discussed here Thermoelectric power and specific heat measurements on few compounds are also explained in this chapter.

**Chapter 7** discusses the results obtained from dielectric spectroscopic and impedance spectroscopic studies of ferroelectric-ferromagnetic composite materials \(\text{BaTiO}_3-\text{La}_0.2\text{A}_0.8\text{MnO}_3 \; A = \text{Ca, Pb and Sr})\). All results and conclusions derived from capacitance and conductance measurements are discussed

The thesis concludes with a general remark on the results and discussions from the present investigations. Finally a list of publications and reprint has been presented