ABSTRACT

The thesis is mainly concerned with experimental investigation of cooperative phenomena in strongly correlated electron systems, namely, perovskite-type doped manganites. This is an interesting topic in modern solid state physics for both fundamental study and technological applications.

One of the main goals of this thesis is to investigate the effect of nano-meteric grain size modulation of polycrystalline Pr- and La- based manganites, namely, Pr_{1-x}Sr_xMnO₃ (PSMO) (0.2 $\leq x \leq 0.4$), LaMnO_{3+ δ} (LMO), and polymer coated La_{1-x}Sm_xSr_{0.1}MnO₃ (LSSMO) (0.1 $\leq x \leq 0.5$) compounds on their microstructural, magnetic-, electronic- and magneto-transport properties synthesized by chemical "pyrophoric" reaction process. A detailed characterization of the micro structural and magnetic properties as well as electronic and magnetic transport properties of these materials has been carried out in order to obtain better understanding of the physics of CMR manganites and to elucidate the role of polaronic effects and exchange interaction mechanism. Interestingly, in Pr_{0.8}Sr_{0.2}MnO₃ nano-manganite, a metallic ferromagnetic state is observed whereas its bulk counterpart remains in a ferromagnetic insulating state. The observed results have been explained by a phenomenological surface polaron-breaking model originating from the enhanced grain surface disorder. In the case of LMO series, we have found that the oxygen content in LMO compounds studied decreases with increase of La vacancies in parent site and a parasitic Mn_3O_4 phase evolved in the range $0.9 \ge La/Mn \ge 0.7$. A second magnetic cluster (or spin glass) phase is found below 50 K for the bulk sample. However, the glassy phase is suppressed in nano-metric particle size. Disorder, induced by doping at the La site, via increased Sm doping in nano-metric particles of LSSMO compound is also investigated in view of the fielddependent electronic transport and magnetic properties. We found that the metallic behavior is suppressed by increasing Sm³⁺ content. This is possibly due to the fact that magnetic Sm³⁺ doping at nonmagnetic La^{3+} site induces an extra magnetic coupling in the system. A comparative experimental investigation on physical properties of the polymer-coated LSSMO nano-manganite with its bare compound has also been carried out. We have interpreted the observed electronic, magneto-transport, and magnetic properties by introducing a core-shell type model where an intermediate exchange coupling between the shell (surrounding) and antiferromagnetic core (uncompensated surface spins) plays a crucial role. To get a deeper insight of the magneto-transport behaviour of these doped manganites, the experimental data obtained from field dependent magneto-transport measurements at different temperatures have been analyzed for an entire series of samples using spin-polarized tunnelling and spindependent scattering mechanisms. Appearance of semiconducting resistive nature above T_C has been explained using small polaron hopping and variable-range hopping models. The observed low temperature resistivity up-turn anomalies (< 50 K) in these samples are discussed using a common framework of electron-electron interaction and weak localization. The observed enhancement of $T_{\rm C}$ with nano-metric grain size modulations compared to their bulk counterpart is explained through the increase of cationic vacancies in nano-metric particles along with their increase of surface to volume ratio.

The second part of the thesis primarily deals the pressure and thickness dependent investigations on oxygen-rich LaMnO₃ (LMO) manganite thin films that are ferromagnetic and insulating in their ground state. High quality *c*-axis oriented thin films of the nominal composition of LMO on LSAT (001) substrates were grown by using Pulsed Laser Deposition technique employing a very high vacuum thin film deposition chamber attached with a RHEED gun assembly along with an extremely controlled atmosphere-monitoring

system. The study of various epitaxial complex manganite thin films is still of great interest for fundamental study as well as for technological applications including spintronics. This is in the context of possible applications in magneto-resistive devices by enhancing ferromagnetic Curie temperature, magneto-resistance properties etc.. We found that, depending on the oxygen background pressure during growth, the LMO films contain sizeable amounts of La and/or Mn vacancies that strongly influence their electronic and magnetic properties. Specifically, we show here the role of cationic vacancies which originate from various growth pressures, having markedly different effects in their transport, magnetic and optical properties. Depending upon the oxygen annealing, the most La-deficient films develop a metallic behavior with a larger ferromagnetic saturation moment of 3.8 $\mu_{\rm B}$ / Mn ion. In contrast, in the oxygenated Mn-deficient films, the ferromagnetic order is strongly suppressed having saturation moment less than 0.5 $\mu_{\rm B}$ / per Mn ion and the transport remains insulator-like. Here, we compare our results with the ones that were previously obtained on bulk samples and present an interpretation in terms of the much stronger disruption of the electronic and magnetic structure by the Mn vacancies as compared to the La vacancies. In the thickness dependent study, we have found that $T_{\rm C}$ value is reduced with decrease of film thickness and an extra antiferromagnetic phase appears in low temperature. Observed results have been discussed in the common framework of self-doped cationic vacancies and thickness dependent strain. From the field-dependent magneto-resistance and electronic transport behavior we confirm the existence of metamagnetic/ spin-canting minority phase in the observed films where the phases are embedded in a ferromagnetic metallic background.

Keywords:Manganite, Nanoparticle, Thin film, CMR, Ferromagnetism, Antiferromagnetism, Spin glass, Magnetoresistance, Surface disorder, Cationic vacancy, Oxygen vacancy, Optical conductivity.