Abstract

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A detailed electrical and magnetic investigation is carried out on the polycrystalline $Pr_{0.50}Ca_{0.50}MnO_3$, $La_{0.50}Ca_{0.50}MnO_3$, $Nd_{0.50}Ca_{0.50}MnO_3$ and $Sm_{0.50}Ca_{0.50}MnO_3$ and $Pr_{0.50-x}La_xCa_{0.50}MnO_3$ (where x = 0.00 - 0.50) compounds. These materials are freshly synthesized through solid state reactions method and room temperature X-ray diffraction measurements are employed for the determination of phase purity. Impedance spectroscopy is explicitly employed to understand; firstly, the effect of metal electrode on these materials. Secondly, using relaxation processes information of different phases from the bulk of the material at low temperatures is our major findings. Thirdly, the interplay between electrical and magnetic properties of low bandwidth $Pr_{0.50}Ca_{0.50}MnO_3$ and intermediate band width $La_{0.50}Ca_{0.50}MnO_3$ compounds, able us to confer about the important phenomena like metal to insulator transition, melting and collapse of charge and orbital ordering and conduction mechanism of charge carriers.

The nature of electrode for these heterogeneous polycrystalline $Pr_{0.50}Ca_{0.50}MnO_3$, $Nd_{0.50}Ca_{0.50}MnO_3$ and $Sm_{0.50}Ca_{0.50}MnO_3$ ceramics are explored by using In-Ga and sputtered Au metal electrodes. At selected low temperatures, dc biasing measurements are carried out to substantiate the extrinsic Schottky barrier i.e. electrode effect at different voltages. The dc biasing measurements show sensitivity of low frequency data which is associated with electrode effect for all investigated samples. Mott-Schottky plot of $1/C_e^2$ versus dc bias shows linear response for these ceramics. A detailed impedance spectroscopy measurement of $Pr_{0.50}Ca_{0.50}MnO_3$ sample is carried out to identify different electro-active regions associated with bulk, grain boundary and electrode response ceramic in the temperature range of 300K to 10K. The main features are the emergence of three different relaxations in the impedance plane plots associated with electrodes, grain boundaries and grain effects. The coexistence of two phases are demonstrated using combined impedance and modulus spectroscopic plots and the origin of these LF-phase and HF-phase are discussed due to grain relaxations. Both phases are thermally activated and the activation energies (Ea) calculated for HF-phase and LF-phase are 4.21meV and 6.1meV, respectively. The activation energies of other thermally active, i.e., grain boundary and electrode responses are calculated to be 75meV and 115meV, respectively.

The mixing of intermediate band width and short bandwidth i.e., $Pr_{0.50-x}La_xCa_{0.50}MnO_3$ (where x = 0.0 to 0.50) results in variation in the electrical and magnetic properties at low temperatures. The doping of La at Pr site is discussed in term of promoting conducting channels and at $Pr_{0.30}La_{0.20}Ca_{0.50}MnO_3$ concentration a metal to insulator transition is shown and discussed. The conduction mechanism of charge carriers are analyzed by employing Mott's variable range and spin polaronic hopping models. Localization length is calculated and the decrease of overall resistance values with La doping is explained in terms of increase in the localization length. Tan□ shows thermally activated relaxation peaks for the x=0.0 and 0.1 samples and for dopant x=0.2, temperature independent peaks correspond to MIT region. Relaxation time calculated from the multiplication of resistance and capacitance values (derived from equivalent circuit model) and from the peak frequency of tan□ showed similar activation energies of different electro active regions. Same type of carriers is responsible for the tan□ relaxation peaks and the electrical properties of these manganites. The magnetic results of $Pr_{0.30}La_{0.20}Ca_{0.50}MnO_3$ sample shows main magnetization peak around 245K (T_{CO}) and at higher magnetic field the emergence of peak at around 150K is shown which is in accordance with our impedance results. Magnetic results of La addition at Pr site, shows a strong competition and correlation of charge ordering (T_{CO}) and Neel temperature (T_N) peaks. A strong melting and collapse of AFM domains is observed for $La_{0.50}Ca_{0.50}MnO_3$ compound and is discussed into FM domains having Mn^{3+}-O-Mn^{4+} networks. $La_{0.50}Ca_{0.50}MnO_3$ sample shows a relatively minor peak at T_C and major peak at T_N. M(H) loop results show strong melting and collapse of charge and orbital ordering around 155K for $La_{0.50}Ca_{0.50}MnO_3$ sample.