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Electrical and magnetic transport in Strontium doped Europium Ferrimanganites



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ABSTRACT

Eu_{0.65}Sr_{0.35}Fe_xMn_{1-x}O₃ (x=0.1, 0.3 and 0.5) has been prepared using a standard solid state reaction method. The under-investigation compounds is found to crystallize in a single-phase orthorhombic structure in the P_{bnm} space group (62). The adiabatic polaron electronic transfer was obtained for all samples and the activation energy of x=0.1 sample is equal to 1.013 meV and slightly increase at x=0.3 (1.289 meV) while is doubled for x=0.5 to be 2.1065 meV. The magnetization–temperature dependence measurements of Eu_{0.65}Sr_{0.35}Fe_xMn_{1-x}O₃ show the ferromagnetic ordering at low iron concentration x=0.1 and when iron concentration increase to x=0.5 the noncollinear magnetic ordering (the canted antiferromagnetic) is obtained. The magnetic phase transition (paramagnetic-ferromagnetic transition) in the Eu_{0.65}Sr_{0.35}Fe_{0.1}Mn_{0.9}O₃ is observed at T_c of 150 K. For Eu_{0.65}Sr_{0.35}Fe_{0.5}Mn_{0.5}O₃ the multi-magnetic phase transition is observed at T_c of 200K and T_N of 430 K. The resistivity at low temperature is measured. Theoretical Calculations using Monte Carlo code have been done. The magnetization as function of temperature has been calculated using Monte Carlo simulations for Eu_{0.65}Sr_{0.35}Fe_xMn_{1-x}O₃ (x=0.0, 0.1, 0.2, 0.3, 0.4 and 0.5). Ising model is a suitable model to study the magnetization for our compounds. The internal energy for x=0 is the highest value compared with the other x values which have nearly a ground state value equal to 2.7 J

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1. Introduction

Over the last few decades, the rare earth ferrimanganite within perovskite-like structure has attracted scientific attention due to its remarkable properties and promising applications. Among doped rare-earth ferrimanganites materials, the $R_{\nu}A_{1-\nu}Mn_xFe_{1-x}O_3$ have been extensively investigated by several researchers [1-4]. The electrical, magnetic, dielectric, and optical properties exhibited by these perovskite-type oxide materials are technologically useful. The underlying physics of those phenomena has been interpreted in terms of the double exchange (DE) model, in which a strong exchange interaction occurs between Mn³⁺ and Mn⁴⁺ ions through intervening filled oxygen 2p states [5], the Jahn-Teller (JT) polaron [6]. In particular, these systems exhibit the novel phenomena of magnetocaloric effect metal-insulation (MI), colossal magnetoresistance (CMR) and charge ordering (CO), in manganites [7-11]. The bipolar control of coexisting phases in a nanostructured film of manganite, La_{0.85}Ca_{0.15}MnO₃ was reported by Rajib Nath and A.K. Raychaudhuri using an applied gate bias in a field effect (FE) device configuration. The difference in the electronic phases is attributed to the low hole doping of La_{0.85}Ca_{0.15}MnO₃ manganites [12]. Spintronic devices are based on the spin of electron to sense magnetic fields [13], store information, or perform logical operations. Colossal magnetoresistive (CMR) manganites are a new class of materials under investigations for future spintronic applications such as nonvolatile magnetic computer memory (MRAM). The formation of polarons could be described as follows; electrons whose interaction with the lattice creates a deformation that traps the electron, as a pocket on a pool table traps a billiard ball. Polaron formation in a CMR material attributed to the electron localization as polarons is a defining characteristic of all CMR materials. The theoretical polaron-hopping and the localized electronic states are studied by Christov [14] and are described by using the adiabatic approximation and both "large" polarons in ionic crystals and "small" polarons in molecular crystals are considered. According to Manjunatha et al. [15] at low temperatures, electron-magnon scattering play very important role in thermal transport in

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 $La_{0.8}Ba_xCa_{0.2-x}MnO_3$ (0 $\leq x \leq$ 0.2). The adiabatic small polaron hopping (ASPH) model was observed in the high-temperature insulating range viz. $T > T_{MI}$. Analyzing the electrical transport data in metallic region i.e. $T < T_{MI}$ indicated that the electrical transport is governed predominantly by electron-electron scattering process. Crystal structure and the chemical bonding of these compounds are of a great importance to the understanding of the peculiar electronic and magnetic properties in these perovskites. The A-site substitution primarily changes the carrier density of manganese in this compound and strongly affects the Mn³⁺-O-Mn⁴⁺ angle (lattice distortion), thus transforming the parent compound RMnO from an insulating antiferromagnet into a metallic ferromagnet [16]. Alternatively, the B-site substitution by iron directly modifies the crucial Mn³⁺-O-Mn⁴⁺ network. The important correlation between the crystal structure of the materials and their transport properties are reported in [17,18]. The effect of the tilt in the octahedra plays an important role in defining the magnetic and electric exchange interaction between transition metal element e_g orbital and oxygen 2p orbital. The dependence of magnetic ordering in cobalt doped La_{0.7}Te_{0.3}MnO₃ on the Mn-O-Mn angle is clear as showed in Ref. [19] where this angle changed with the substitution of manganese by cobalt. The structural transition due to the disordered distribution of Mn²⁺/ Mn³⁺ and Co²⁺/Co³⁺ ions was found from the X-ray photoelectron spectroscopy (XPS).

The experimental investigations of the critical behavior of manganites near the PM–FM phase transition using a variety of techniques provided a wide range of values for the magnetization critical exponent b which embraces the mean field, short-range isotropic 3D–Heisenberg, and 3DIsing model estimates [20–23]. Chen et al. [24] when they studied the ferroelectric control of magnetization in La_{1-x}Sr_xMnO₃ manganites they construct an Ising-like model, which is based on the interaction between nearest-neighbor Mn magnetic moments in order to more quantitatively describe the energy sequence. Ising model with two exchange interaction to simulate the magnetic bonds in a perovskite manganites of the type (PrCa)MnO₃ to study the multiferroicity due to charge ordering [25]. The random-field Ising models proposed for micrometer-scale domains do not capture long-range elastic interactions that are known to be crucial for manganites [26].

We considered a new dynamic version of the Ising model where the exchange interactions fluctuate in time between two exchange interaction according to a probability distribution magnetically doped insulating oxides, particularly ZnO, there are free spins associated with transition metal)TM) ions, such as manganese $\rm Mn^{2+}$ or $\rm Co^{2+}$, that have been substituted on to the Zn sites at a concentration of typically ~5% [27].

The present work deals with the effect of iron–manganese substitution on the electrical and the magnetic transport in Strontium doped Europium Ferrimanganites experimentally and theoretically.

2. Experimental details

The sample preparation was described in details elsewhere [28,29]. Phase purity and crystallographic information of the synthesized samples were characterized using powder X- ray diffraction (XRD). The XRD analysis was performed using a PA-Nalytical X'PertPRO diffractometer at room temperature with Cu (K_{α}) radiation (λ =1.5406 Å). The DC resistivity–temperature dependence measurements were carried out using four points probe technique in the temperature range from room temperature down to 30 K. Magnetization temperature dependence measurements were carried out using Vibrating Sample magnetometer VSM in

the temperature range from 75 to 973 K.

3. Results and discussion

Crystal structure of ABO₃ may be modified by heat treatment during synthesis of these compounds, moreover doping in the A site or the B site or by simultaneous doping to tailor the properties to specific applications [30-35]. The ABO₃ cubic perovskite structure consists of a BO₆ array of corner-shared octahedral with a large cation A at the body-centered position. Geometric stability of the perovskite structure is determined by the Goldschidt tolerance factor t. X-ray powder diffraction investigations at room temperature indicated that the samples were in a single phase with no detectable secondary phases. Fig. 1 shows XRD pattern of x=0.1and x=0.5 and as well as the 3-dimensional representation of the unit cells. All the measured reflection peaks in the XRD pattern describes the orthorhombic symmetry of the space group Pbnm (No. 62). The substitution of Eu³⁺ ions by Sr²⁺ and Mn³⁺ by Fe³⁺ leads to variation of the tolerance factor [36]. The tolerance factor takes the value t < 1 in the CMR manganese perovskites where the structure adjusted to have these t's by a cooperative tilting of the corner-shared octahedral leading to the formation of the so-called O-type orthorhombic structure with $a \le c/\sqrt{2} \le b$. To describe the distortion in this type of compound, a geometrical quantity, the tolerance factor t, is usually introduced; t is defined as [11]:

$$t = \frac{(\left\langle r_{Eu/Sr} \right\rangle + \left\langle r_{O} \right\rangle)}{\sqrt{2} \left(\left\langle r_{Fe/Mn} \right\rangle + \left\langle r_{O} \right\rangle\right)}$$

where $< r_{Eu/Sr} >$ is the average ionic radius of the A-site ions $(Eu^{3+} \text{ and } Sr^{2+})$ and $< r_{Fe/Mn} >$ is the average ionic radius of the B-site ions (Mn and Fe) in our studied samples. In this case, the average ionic radius of the A-site is fixed and the change is only in B-site. The observed lattice distortion, in our case, is caused by the partial replacement of Mn by Fe. Therefore, the structural studies of manganites doped by Fe with systematic compositional variation can be advantageous for gaining important insight into the structure-property relationships for these compounds. The tolerance factor calculated from the refinement of XRD data is shown in Table 1. It is clear that the tolerance factor of the compound under investigation does not change if the valency of the manganese and the iron ions are trivalent because the ionic radius of Fe³⁺ and Mn³⁺ both are equal 0.645 Å. At low temperature the tetravalent manganese appears. The tolerance factor obtained for the samples with iron concentration less than x=0.35 is t=86.30 while t starts to change for the samples beginning with x > 0.35 (t at x = 0.5 is 85.26). The distortion factor of the ideal perovskite structure, D, was calculated according to the following formula [36]:

$$D = \frac{1}{3} \sum_{i=1}^{3} \left| \frac{a_i - \overline{a}}{a_1} \right|,$$

where
$$a_1 = a$$
, $a_2 = b$, $a_3 = \frac{c}{\sqrt{2}}$ and $\overline{a} = \left[\frac{a.b.c}{\sqrt{2}}\right]^{\frac{1}{3}}$
The numerical values of the distortion factor of the ideal per-

The numerical values of the distortion factor of the ideal perovskite structure D is given in Table 1. The less value is obtained at the low iron concentration x=0.1 (D=1.78 × 10⁻³) where D increase with increasing the iron concentration. The scattering density inside the unit cell of a crystal is calculated using the Fast Fourier Transform (FFT) as a subprogram in Fullprof [37]. The electron density, $\rho(r)$, is calculated according to the following expression:

$$\rho(r) = \frac{1}{V} \sum_{H} F(H) e^{-2\pi \cdot (H.r)}$$

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