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Characterisation of LCMO perovskites employing ultrasonic studies

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Abstract: Ultrasonic measurement is a vital non-destructive tool used to explore the characteristics of the perovskite manganite materials. $La_{1-x}Ca_xMnO_3$ perovskite samples with the composition of x= 0.70 and 0.75 were prepared using the solid state reaction technique. The crystalline nature of the samples was confirmed by the XRD patterns. The samples were found to have orthorhombic structure with no secondary phases. The relative variation in longitudinal velocity and its attenuation was measured out in the temperature range from 500 to 700 K using the through transmission method at the fundamental frequency of 5 MHz. The temperature-dependent observations showed anomalous behaviour at the transition temperatures. The anomalies observed at the temperatures 640 and 560 K for the samples for the composition *x*=0.70 and 0.75 were used to explain the high temperature metal-insulator transition existing in the prepared samples. Further, the investigation reveals that the magnetic entropy increases with a decrease in the doping concentration of calcium in the samples.
Keywords: perovskite; ultrasonic measurements; metal-insulator transition.

Introduction and Experimental:

Investigations on the mixed valence perovskite manganites $R_{1-x}A_xMnO_3$ (R- Rare earth element and A-Alkaline element) assume significant importance. This is because of the special properties which includes colossal magneto resistance (CMR), charge ordering (CO), ferromagnetic (FM) to paramagnetic (PM) phase transition, metal-insulator (MI)phase transition etc [1-2]. They find various applications such as magnetic storage devices, switching devices and sensor technology. The dopant and its concentration has strong impacts on the transport properties, Jahn-Teller (JT) induced strains and orthorhombic to rhombohedral structural transformation [3-4]. The interesting electrical and magnetic characteristics revealed by the perovskite manganite materials are attributed to the double exchange (DE) interaction which involves charge and spin exchanges between Mn^{3+} and Mn^{4+} ions via O^{2-} ions. Hence on the basis of DE theory qualitative explanation may be given for the magnetic and charge transport properties of LCMO [5]. However, the DE theory alone could not explain the phase diagram of these materials. Because besides DE interaction phenomena like Jahn-Teller (JT) interaction, charge orbital (CO) ordering, electron-lattice and spin-lattice coupling also play a vital role on their properties [6]. Thus, a systematic study is essential to understand their properties.

In the present investigation, it is aimed to reveal the high temperature phase transition in $La_{1-x}Ca_xMnO_3$ perovskite manganite samples with the composition of x = 0.70 and 0.75, here after will be called Calcium70 and Calcium75. The samples were prepared employing solid state reaction technique [7]. The crystalline nature of the samples were confirmed by the X-ray diffraction pattern (XRD) patterns obtained employing the diffractometer (X'PERT PRO PANalytical, the Netherland). Longitudinal ultrasonic velocity and attenuation were measuredover a wide range of temperature from 300 to 700 K using a high power ultrasonic pulse receiver (Olympus NDT, 5900PR, USA) [7].

Result and Discussion:

Fig. 1 shows the XRD pattern of prepared perovskites and shows the sharp peaks which confirm the crystalline nature of the prepared samples. The samples are found to have orthorhombic structure with no secondary phases as they are in close agreement with the JCPDS file 89-8091.The temperature-dependent longitudinal ultrasonic velocity and its attenuation was carried out and shown in Figs. 2. In order to understand the ultrasonic measurements, the temperature range is classified into three zones as shown in the Table 1. In the zones I and III, there is a monotonic decrease in velocity/ increase in attenuation occurs. In region II, an anomalyis observed in both velocity and its attenuation.

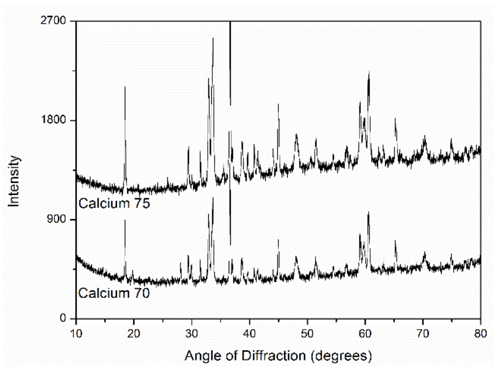

Fig.1: XRD spectrum of $La_{1-x}Ca_xMnO_3$ perovskite samples

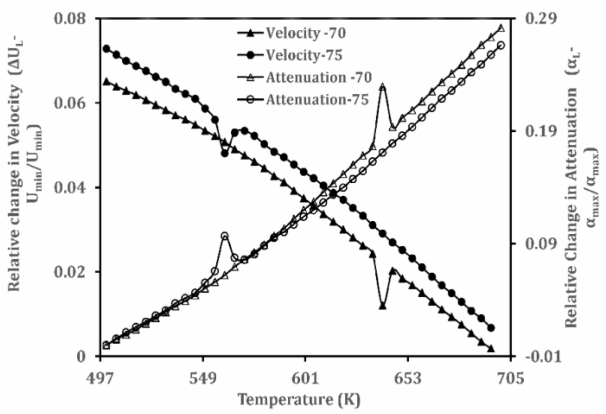

Fig.2: Relative variation of longitudinal velocity (U_L) and longitudinal attenuation (α_L) with the temperature in$La_{1-x}Ca_xMnO_3$ samples

Table1: Temperature range and its dependence of relative variation of ultrasonic parameters in $La_{1-x}Ca_xMnO_3$ perovskite samples

Temperature Zones		Calcium70	Calcium75
Temperature K	Zone I	500 - 635	500 - 555
	Zone II	635 - 645	555 - 570
	Zone III	645 - 700	570 - 700
	Peak/Dip	640	560
Relative variation	$(U_L - U_{Lmin})/U_{min}$	0.010199	0.0066404
	$(\alpha_{Lmax} - \alpha_L)/\alpha_{Lmax}$	0.044112	0.0259310

In Fig.2 in the sample Calcium70, this anomalous behaviour starts at 635 K. The velocity reaches a minimum value at 640 K followed by a steep increase up to 645 K. A reverse trend is observed in the case of attenuation. The minimum in velocity/maximum in attenuation for Calcium75 sampleoccurs at 560 K. The anomaly revealed at a particular temperature is correlated with the phase diagram of $La_{1-x}Ca_xMnO_3$ perovskite manganite samples [7]. From the correlation, it is concluded that the temperature at which is the velocity is minimum (and the attenuation is maximum) is the metal-insulator transition temperature (T_{MI}) of the prepared perovskite samples [8].

It is worth to mention that in the observed measurements T_{MI} decrease when the calcium content in the sample increases from 70 to 75%. This trend is attributed to the fact that in the increase in calcium content causes the distortion of $Mn^{3+}O_6$octahedran due to JT effect will come into play. Hence MI phase transition temperature reduces [9]. This discussion is similar to the earlier reports [6-7]. The change in relative percentage variation of velocity ($\Delta(U_L-U_{Lmin}/U_{Lmin})$) and attenuation ($\Delta(\alpha_L-\alpha_{Lmax}/\alpha_{Lmax})$) at transition region (zone II) was measured from Fig. 2. The same is given in Table 2. It is found that $\Delta(U_L-U_{Lmin}/U_{Lmin})$ for Calcium 70 is much higher than calcium 75. It indicates that the pronounced in Calcium70 is ascribed to the effect of larger disorder [10]. Hence, the measurement of ultrasonic parameters confirms that the magnetic entropy increases with the decrease in calcium content. Thus, it is conclude that ultrasonic velocity/attenuation measurement is an effective tool for the characterization of the perovskites.

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