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Signature of Griffith Singularity in Half Doped $\text{LaMn}_{0.5}\text{Cu}_{0.5}\text{O}_3$

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Abstract. $\text{LaMn}_{0.5}\text{Cu}_{0.5}\text{O}_3$ synthesized via sol-gel auto combustion method is found to having ferromagnetic ground state. The sample crystallizes in orthorhombic $Pbnm$ space group with lattice parameters (in Angstrom) $a = 5.4811(4)$, $b = 5.5236(4)$, $c = 7.7668(6)$. Evidence of Griffith's singularity is observed in magnetic susceptibility data. The Griffith temperature (T_G) and the exponent (λ_{GP}) of modified Curie – Weiss law are found to be 36K and 0.984 respectively.

Keywords: phase inhomogeneity, Griffith phase, manganite

PACS: 77.80.B, 75.47.Lx, 75.50.Dd

INTRODUCTION

Phase inhomogeneity and quenched disorder in manganites have drawn considerable attention among researchers. Inhomogeneity involves preformation of clusters of ferromagnetic/metallic phase above ferromagnetic long range ordering temperature in a matrix of paramagnetic/resistive phase. Quenched disorder is introduced due to the random distribution of substituted divalent ions and local lattice distortion by John – Teller active ion Mn^{3+} . Phase inhomogeneity and quenched disorder leads to Griffith singularity¹. Griffith singularity was initially proposed for randomly diluted Ising ferromagnets. In manganites, the Griffith singularity is commonly observed in R – site substituted compounds like $\text{R}_{1-x}\text{A}_x\text{MnO}_3$ (where R = La^{+3} , Pr^{+3} and A = Ca^{+2} , Sr^{+2} , Ba^{+2}). In these compounds both ferromagnetism (due to double exchange mechanism) and Griffith singularity (due to inhomogeneity and quenched disorder) is introduced by the substitution of divalent cations at La site. Besides La site, substitutions at Mn site by other magnetic ions such as Co, Ni, Cr etc.² were also carried out but reports of Griffith phase in such compounds are rare. We have chosen a non-magnetic ion Cu^{2+} for substitution at Mn site. Since these elements have ionic radii similar, a quenched disorder or phase inhomogeneity is not intrinsic to the Mn site. Moreover $S = 1/2$ of Cu^{2+} may bring disorder in magnetic moment ordering, so ferromagnetism with evolution of Griffith singularity is expected.

EXPERIMENTAL

The sample $\text{LaCu}_{0.5}\text{Mn}_{0.5}\text{O}_3$ (LCM) is prepared by sol-gel auto-combustion method. Aqueous clear solution of $\text{La}(\text{NO}_3)_3 \cdot 4\text{H}_2\text{O}$, $\text{Cu}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$ and $\text{Mn}(\text{NO}_3)_2 \cdot \text{H}_2\text{O}$ with glycine (as the fuel to carry out combustion), is heated under continuous stirring. This ultimately turns to gel and burns vigorously to result blackish powder. The blackish powder so obtained is grinded, pressed into pellet and sintered at 1000°C for 12hrs. One of the sintered pellets in powdered form is used for XRD measurement in Burker D8 Advance system. The XRD data is analyzed by Rietveld

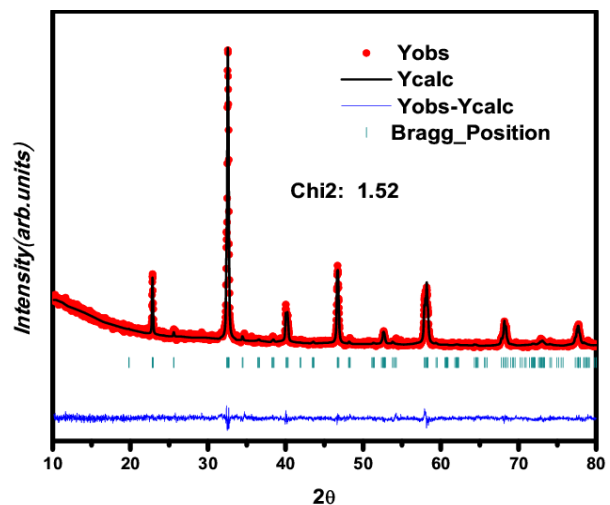


FIGURE 1. Rietveld refined XRD data of $\text{LaMn}_{0.5}\text{Cu}_{0.5}\text{O}_3$.

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refinement method using FULLPROF program. DC - magnetization data at different magnetic fields (0.1 kOe, 1 kOe & 10 kOe) is collected in a temperature range 2K to 300K using Quantum Design make 9T PPMS-VSM.

DATA AND ANALYSIS

Figure 1 shows the Rietveld refinement plot of XRD data taken at room temperature. No secondary phases are observed in the XRD reflection. The refinement was carried out with space group $Pbnm^3$ (orthorhombic crystal structure) with an agreement factor $\chi^2 = 1.52$. The lattice parameters obtained from the refinement are $a = 5.4811(4)$ Å, $b = 5.5236(4)$ Å, $c = 7.7668(6)$ Å.

The temperature dependence of inverse dc susceptibility, χ^{-1} is shown in Figure 2. A linear behaviour χ^{-1} with temperature, following Curie-Weiss (CW) law is found for 300K to 100K. Below this, at 85K, the behaviour deviates from linearity and shows a rapid downfall till 75K and then becomes constant. The effective paramagnetic Curie temperature (θ_p) obtained by extrapolating the high temperature linear region on the temperature scale turns out to be $\theta_p = 26$ K.

On increasing magnetic field, the non-linearity in the range $25\text{K} < T < 100\text{K}$ weakens at $H = 1\text{kOe}$ and disappears completely at $H = 10\text{kOe}$. Such a non-linear behaviour and its disappearance with magnetic field is an indication of Griffith like singularity⁴. The downturn temperature is referred as Griffith temperature (T_G). Appearance of Griffith's singularity is due to the formation of ferromagnetic clusters in the paramagnetic state, much before the long range ferromagnetic order sets in. These ferromagnetic clusters align themselves in the magnetic fields leading to deviation from CW behavior much before the actual ferromagnetic Curie temperature. Griffith's singularity is very often characterized by power law behavior¹

$$\chi^{-1} = (T - T_C^R)^{1-\lambda} \quad (1)$$

This power law behavior is a modified CW law, where T_C^R is the critical temperature of ferromagnetic cluster in paramagnetic matrix and the parameter $0 \leq \lambda \leq 1$ marks the deviation from CW law. For CW law, $\lambda = 0$ and $\lambda > 0$ implies occurrence of Griffith phase. According to eq. (1), λ can be obtained from the slope of χ^{-1} vs. $(T/T_C^R - 1)$ plotted in log - log scale (see fig 2b). The slope, obtained by straight line fitting to the linear portion of the curve in the Griffith phase and the paramagnetic phase gives λ_{GP} , λ_{PM} respectively.

Accurate determination of λ depends highly on the precision of T_C^R values so obtained. A special

procedure (see ref 1) is followed to correctly estimate the values of T_C^R . Initially the effective paramagnetic Curie temperature is set equal to T_C^R , i.e., $T_C^R = \theta_p$. Then the T_C^R value is so adjusted that the value of λ_{PM} becomes close to zero.

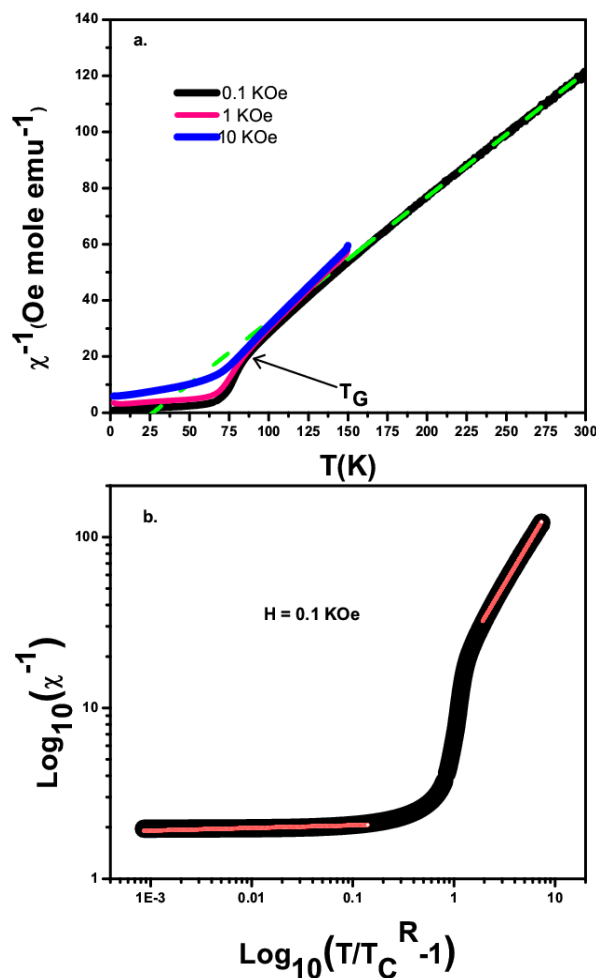


FIGURE 2. (a) Temperature dependent inverse dc susceptibility at different magnetic fields. (b) The inverse susceptibility data plotted in double logarithm scale.

This was done by considering the fact that $\lambda \sim 0$ in paramagnetic region where eq. (1) is valid. With this value of T_C^R , λ_{GP} is calculated by linear fitting in the Griffith phase region. The θ_p , T_G , T_C^R , λ_{GP} , λ_{PM} values are given in Table.1. The high value of λ_{GP} (close to unity), signifies that Griffith singularity is reasonably strong in LCM.

TABLE 1. Characteristic temperatures (θ_p , T_G , T_c^R), and Griffith singularity parameters (λ_{GP} , λ_{PM}) for LCM.

Parameters	Values
θ_p	26K
T_G	81K
T_c^R	36K
λ_{GP}	0.984(4)
λ_{PM}	0.002(2)

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