4.1:Introduction

Magnetic systems with frustrated spin interactions are known to exhibit very high sensitivity to external influences and can be changed even by small modifications of composition. Such frustrated systems possess unconventional magnetic states such as spin-liquid and spin-nematic [1,2]. ABO_2 type frustrated materials, in particular, $CuFeO_2$ and $CuCrO_2$ have been widely studied in terms of magneto elastic coupling and magnetoelectricmultiferroics, which are considered to be due to the spin frustration[3-6]. In these materials frustration can be lifted with lattice distortion [7,8]. But for the case of $CuMnO_2$ frustration can remain even after lattice distortion. $CuMnO_2$delafossite (a crednerite) is a quasi two dimensional frustrated anti-ferromagnet in which the magnetic phase transition is strengthened by spin lattice coupling [9]. The transition in $CuMnO_2$ is improper ferro-electrtic and the material displays spin driven ferroelectricity [10]. Moreover, Poienar et al. [11] from synchrotron and neutron powder diffraction study have shown that partial Cu for Mn substitution affects mainly the sign of the inter plane magnetic coupling (between MnO_2 layers) that changes from anti-ferromagnetic in $CuMnO_2$ to ferromagnetic in $Cu_{1.04}Mn_{0.96}O_2$. In their report Garlea et al. [12] have demonstrated that high temperature paramagnetic phase is transferred to the low temperature triclinic antiferromagnetic phase and upon substituting Cu for Mn, the exchange randomness tends to stabilize a 3-dimensional magnetic phase with ferromagnetic coupling. On the basis of*ab initio* band structure calculations Ushakov et al. [13] pointed out that in-plane magnetic ordering is mainly provided by the direct exchange interaction between the t_{2g}, while superexchange between e_g and t_{2g} orbitals on different sites is expected to be smaller. Theoretically they have also shown that substitution of a part of Mn^{3+} by Cu^{2+}, with corresponding creation of compensating Mn^{4+} ions, leads to the inversion of the interlayer coupling for very small doping.

Moreover, in recent years, multifunctional materials have attracted much attention due to the coexistence of more than two properties simultaneously e.g. multiferroicity, exchange Bias, Griffiths phase etc. The observation of a Griffith's phase (GP)is quite interesting. According to Griffith's theory, there is always a finite probability of finding ferromagnetic (FM) clusters in paramagnetic (PM) statein the temperature range T_C< T< T_G, where T_C and T_G are Curie-Weiss temperature and Griffith's temperature, respectively. The Griffith phase is characterized by the formation of FM clusters below T_G. These clusters are formed thermodynamically during cooling and nucleated by the intrinsic randomness, which cause inhomogeneous magnetic distribution in the sample. In fact, Griffith phase was characterized by the downturn deviation from Curie-Weiss law behavior in χ^{-1} as the temperature approaches to T_C from above.In this thesis we have seen that $CuMnO_2$shows the presence of a Griffiths-like phase (GP). The existence of GP indicates the short-range FMcorrelations in the AFM and PM regions at high fieldsora spin-flop transition in the AFM phase.

4.2:Experiment

The $CuMnO_2$sample was prepared by solid state reaction in an evacuated quartz tube. Powders CuO, MnO were mixed in appropriate ratio and pressed into pellets. The pellets were then placed in an alumina crucible, sealed in quartz tube under high vacuum (~10^{-6}m.bar) and heated at 950^0C for 12hrs. Powder X-ray diffraction (XRD) data were recorded using RigakuMiniflex II X-ray diffractometer. The observed XRD patterns were analyzed by Rietveld method using Fullprof program.The dc and ac magnetization were measured using a Quantum Design MPMS3 superconducting quantum interference device (SQUID) magnetometer. The magnetic data were all recorded during reheating. The temperature and frequency dependences of the ac susceptibilitywere measured in external dc magnetic fields 10 and 50Oe in the frequencies 5,50,500 Hz respectively.

4.3:Results& Discussion

4.3.1: X-ray diffraction study

Structural characterization is performed using Rietveld refinement of the X-ray powder diffraction pattern. X-ray diffraction data are shown in Fig.4.1. The fitting performed using monoclinic C2/m symmetry. A satisfactory fit is obvious from the difference of plot which also demonstrates the absence of secondary phase in the compound. Since the valance states of the transition metal ions plays crucial role in deforming the ground state of magnetic oxides, it is very important to identify the same in the said oxides. We have already shown in our recent publication [14] from the X-ray photo-emission spectroscopy analysis that Cu is in the +1 state and Mn is in +3 state.

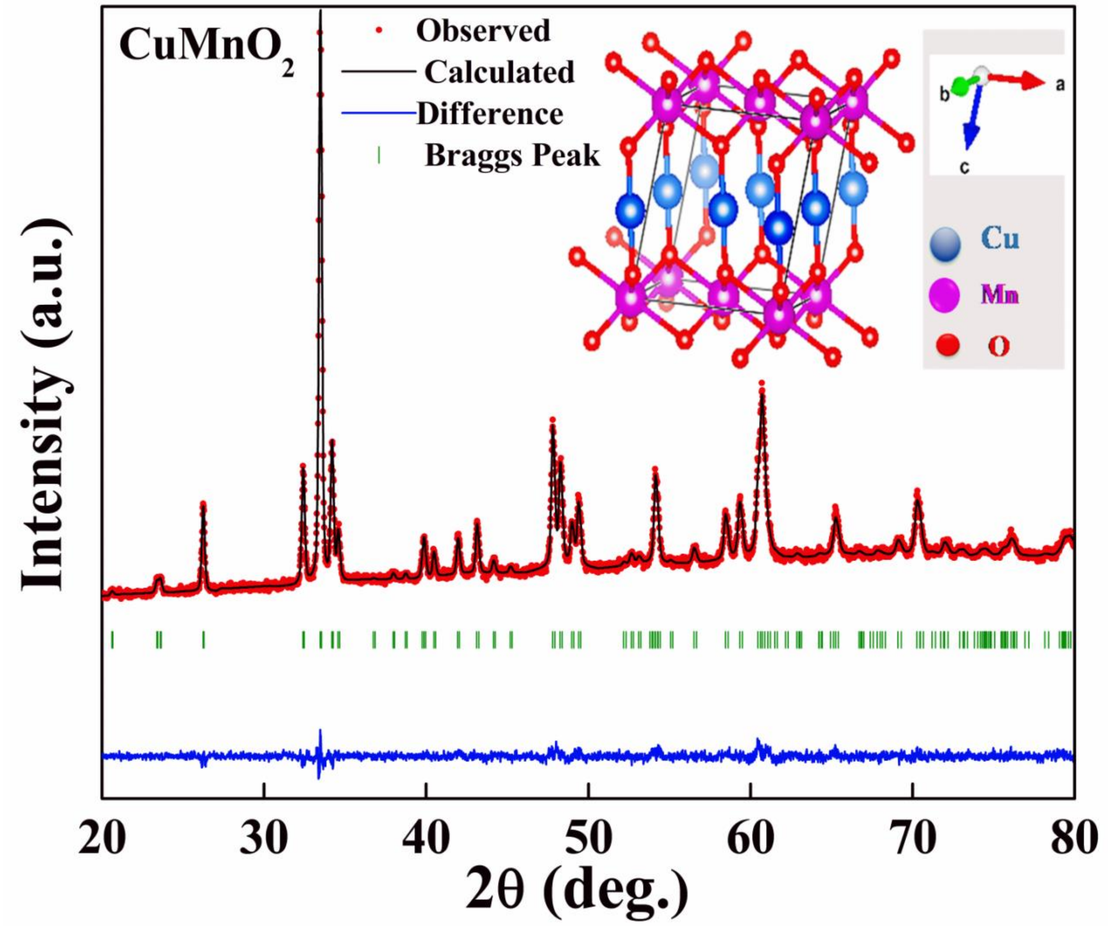

Fig.4.1. Reitveld refinement of the X-ray diffraction pattern of $CuMnO_2$. Refinement has been made with the C2/m space group.

Table 4.1. Structural parameters of $CuMnO_2$ obtained by the Reitveld refinement of the X- ray diffraction pattern.

Samples /Parameters	**$CuMnO_2$**
Space group	C 2/m
a(Å)	5.5907(7)
b(Å)	2.8808(4)
c(Å)	5.8853(7)
Cell Volume (Å^3)	91.9930
α (deg)	90
β (deg)	103.940(4)
γ (deg)	90
Atomic Positions	
O(4i)x	0.42292(15)
y	0
z	0.18631 (13)
Occupancy	
Cu	1
Mn	1
O	2
Bond length	
Mn-Mn(Å)	2.8808(4)x 2 3.1446(3)x 4
Cu-O(Å)	1.792(7)x 3
Braggs R factor	
R_f	4.579
R_p	37.3
R_{wp}	18.3
χ^2	1.98

4.3.2: Magnetic Property Study:

4.3.2.1 DC Susceptibility:

DC magnetic behavior of $CuMnO_2$with temperature variation is shown in Fig.4.2.The zero-field cooled(ZFC) curve shows a maximum of ~40K whereas saturation is observed at lower temperature in the field cooled(FC) curve showing a bifurcation between ZFC and FC curves. The peak in ZFC curve is the indication of an antiferromagnetic (AFM)–like transition.

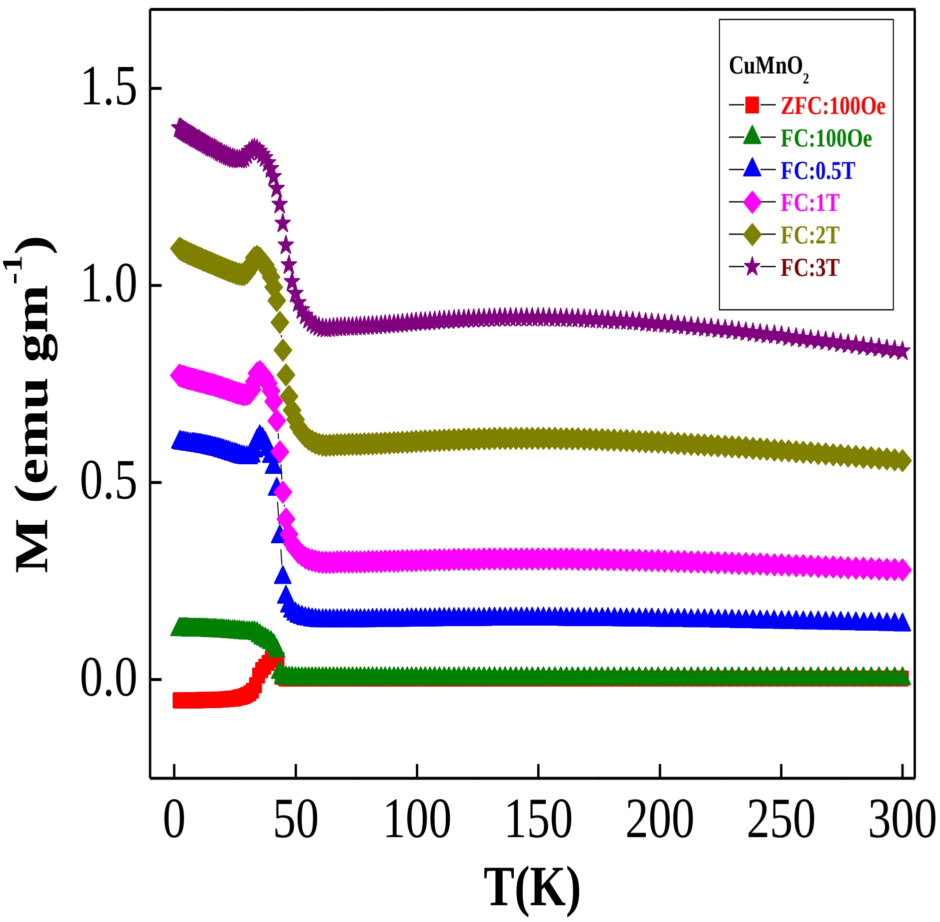

Fig.4.2. Magnetization of $CuMnO_2$ as a function of temperature at different field.

Moreover, it is clearly observed that ZFC magnetization at 100 Oe is negative. This might be due to the formation of ferromagnetic (FM) clusters and domains in microscopic domain structure and induced antiphase boundary between clusters and domains where spins are divided by the anti-phase boundary with a 360° domain wall. As a matter of fact,it is very difficult to align the spins along field direction. Hence, the field of 100Oe is not sufficient to align all the frozen spins of clusters and domains along the direction of field. At lowering temperature the canted spinsare stabilized and resultant magnetic moment tends to be negative [18].

The inverse dc susceptibility χ^{-1}_{dc} is plotted as a function of temperature at differentmagnetic fieldsin Fig 4.3.The high T region shows linear behavior.The linear part has been fitted with the Curie Weiss (CW) law, $\chi_{dc}(T)=C/(T-Tc)$ where C is the Curie constant and Tc is the Curie Weiss temperature.The obtained Tc values from the fitting are 38K, 35K, 27K and 16K respectively for 0.5 T, 1.0 T, 2.0 T and 3.0 T magnetic fields.It may be noted that obtained Tcis far below the anti-ferromagnetic transition temperature T_N.

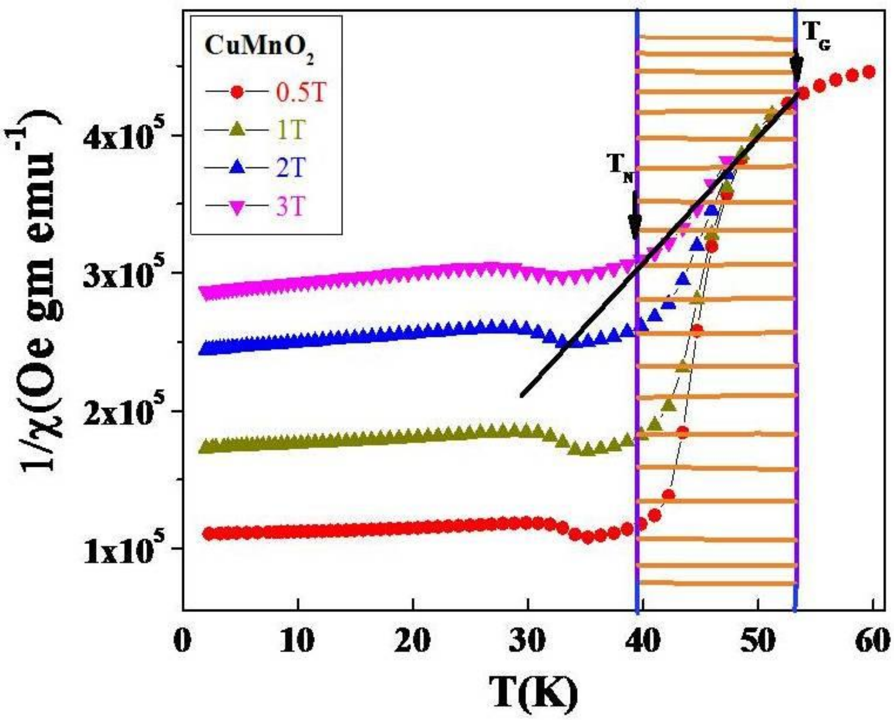

Fig.4.3. Inverse susceptibility of $CuMnO_2$ as a function of temperature at different field. The solid line shows the Curie-Weiss fitting. The T_N indicates the Neel temperature.

In $CuMnO_2$ competing components of AFM and FM superexchange interactions, the sum of which actually determines the value of Tc[15,16]. The positive value of Tc bringing out the importance of *ab* plane ferromagnetic (FM) interactions [17]. Moreover, a sharp downturn is observed well above Curie-Weiss temperature. As the magnetic field increases the downturn decreases and χ^{-1}_{dc} (T) curve obeys the Curie Weiss law.This behavior is due to the existence of Griffith Phase (GP) [19-22].

Generally, in the observed system with FM ground state the observed GP is due to the short range ferromagneticallycorrelated clusters above the long range ordering temperature in the paramagnetic matrix. At low magnetic field the moment of the FM dominates over the back groundparamagnetic (PM) matrix and produces the downturn in χ^{-1}_{dc}. On the other hand, at high field a considerable increase in PM matrix moment is occurred and as a matter of fact, the deviation from Curie-Weiss behavior is decreased [23].The feature in the present case even is observed for 3T magnetic field.

The existence of GP can also be confirmed by analyzing magnetic susceptibility which for a Griffith Phase, should be characterized by an exponent less than unity, that is $\chi(T) \propto (T/T_C^R-1)^{-(1-\lambda)}$where $0\leq\lambda\leq1$ [24].The T_C^R is defined to lie above the actual ordering temperature but below the highest ordering temperature allowed by the exchange bond distribution (T_G) [25-27]. The modified CW form described above reproduces the behavior in many GP systems [28-33]. For high-magnetic field, with $\lambda= 0$, above equation reduces to the CW equation. The value of λ ($0\leq\lambda\leq1$) signifies deviation from CW behavior.To evaluate the correctT_C^R is a serious problem in analysing data. One may chooseT_C^R =Tc so that it leads to $\lambda= 0$ in the PM region [34,35]. In the present investigation, values of Tc are much below the long range ordering temperature, T_N. In fact, as already mentioned Tc is determined by the sum of AFM and FM interactions. These coexisting interactions decrease the value of Tc much below T_N. To overcome this we considerT_C^R =T_N. In Fig. 4.4 we have shown the fitting of χ^{-1}_{dc} (T) and the fitted λ value is found to be 0.37, 0.41, 0.56 and 0.73 respectively for 0.5T, 1.0T, 2.0T and 3.0T, which lie in the range, $0\leq\lambda\leq1$.

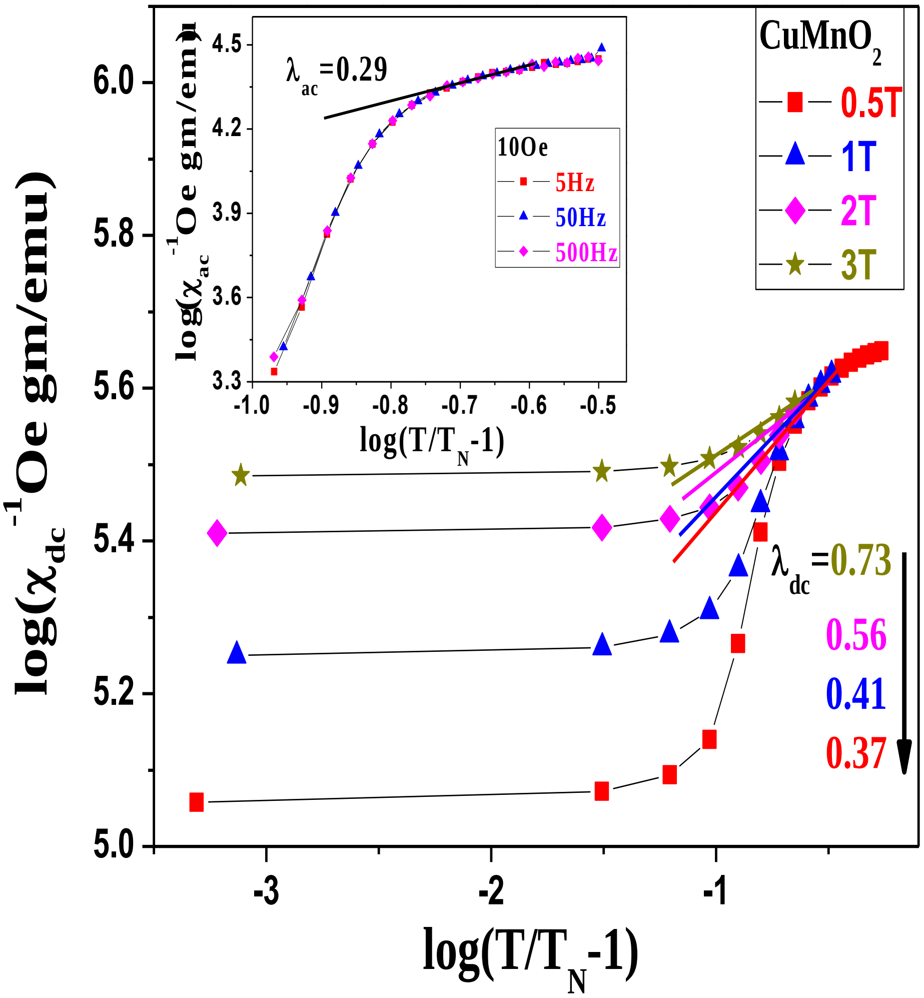

Fig.4.4. Plot of χ_{dc}^{-1} as a function of $(T/T_C-1)^{(1-\lambda)}$ to estimate the value of λ. Inset: χ'^{-1} as a function of $(T/T_C-1)^{(1-\lambda)}$.

Table 2: Obtained fitting parameters T_c and λ.

Magnetic Field (T)	$T_C(K)$ from $\chi_{dc}(T)=C/(T-T_C)$	λ_{dc} from $\chi_{dc}(T) \propto (T/T_N-1)^{-(1-\lambda)}$ where $0\leq\lambda\leq 1$
0.5	38	0.37
1	35	0.41
2	27	0.56
3	16	0.73

4.3.2.2 AC Susceptibility:

The ac susceptibility is a very powerful tool for probing metastable state [33,36,37]. Fig. 4.5 and 4.6 show the plots of χ' and χ'' respectively.

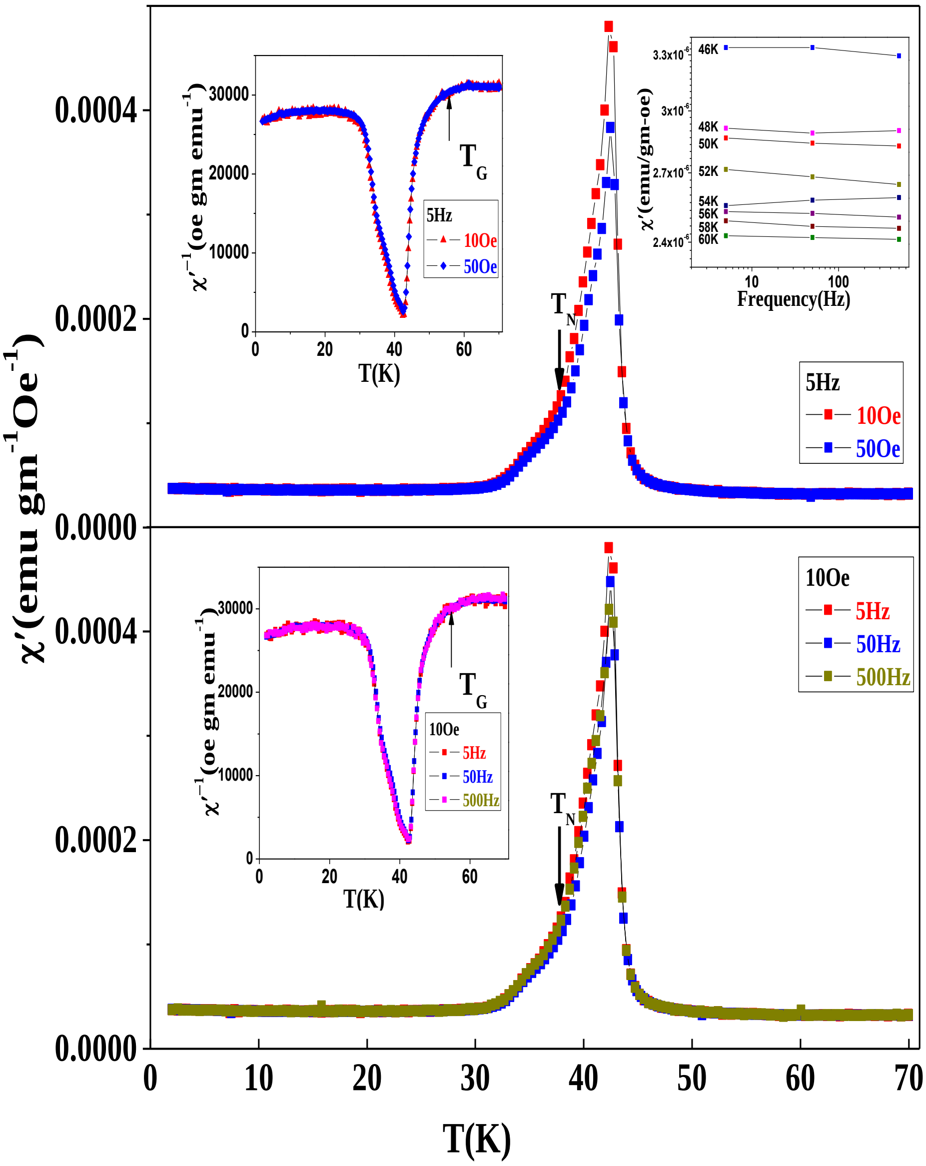

Fig.4.5. Variation of χ' as a function of temperature. Upper Panel: Plotted at different magnetic field. Lower Panel: Plotted at different frequencies. Inset Upper panel: χ'^{-1} as a function of temperature and linear region of χ'^{-1} as a function of frequency at different temperature. T_G indicates the onset of the Griffith like phase.

The anomaly observed in χ' and χ''is attributed to the onset of GP. The GP fully develops between 55K and 40K. The dependence of frequencies of χ'in the GP is shown in the inset of Fig.5. A steady decay is observed in the frequency range 5Hz to

500Hz except the onset temperature, 55K where an increase in slope is observed which is consistent with the report by Perezetetal. [38]. As the frequency increases, the intensity in both χ'(T) and χ''(T) decrease. Also with increase of frequency, the anomaly shifts towards higher temperature. The peak observed in χ''_{ac}(Fig. 4.6) shifts from 41.95 K to 42.40K with variation of frequency from 5Hz to 500Hz.

Generally the dynamics of critical phenomena in correlated and uncorrelated systems is characterized by the shifts of the critical temperature per frequency decades [39] which is expressed as $\Delta Tg/\mathrm{Tg}\ \Delta\log(\omega)$where ω is thefrequency.

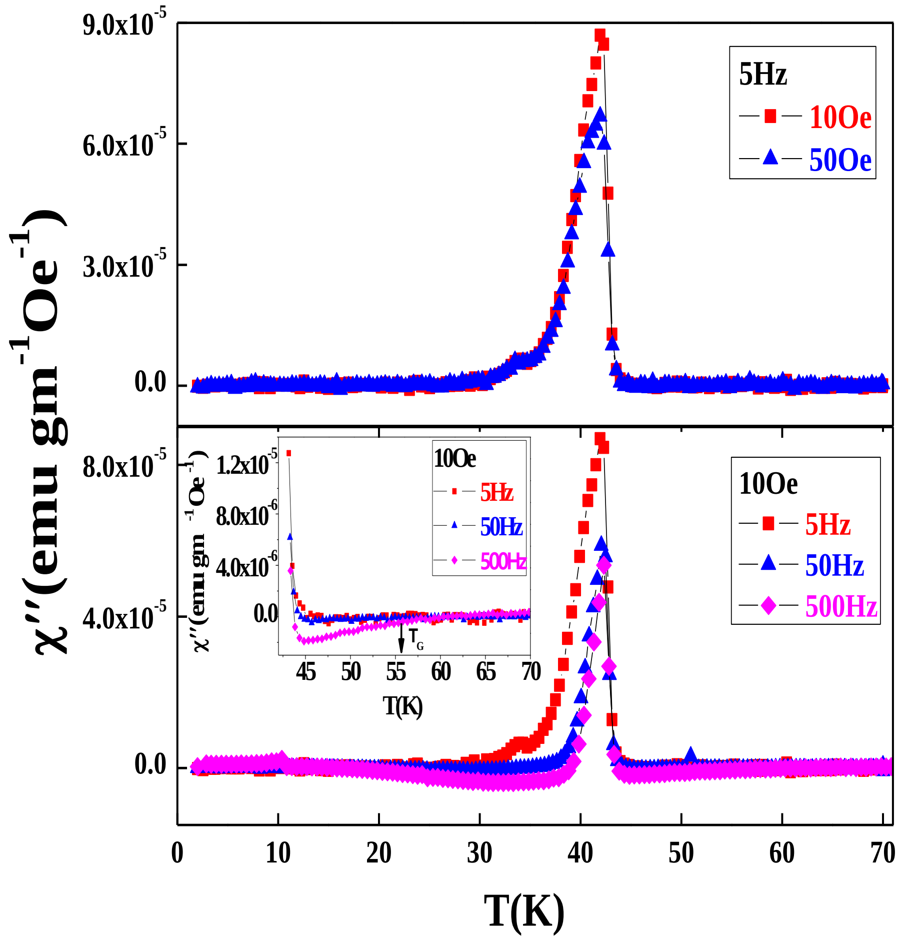

Fig.4.6. Variation of χ'' as a function of temperature. Upper Panel: Plotted at different magnetic field. Lower Panel: Plotted at different frequencies. Inset Lower panel: χ''as a function of temperature at different frequencies. T_G indicates the onset of the Griffith like phase

It is observed that in the present case the value is about 0.003, which is about two orders of magnitude lower than those found in systems with high magnetic frustration such as, spin glass [39]. For the Gd_5Ge_4 this value is around ~0.001 (Ref. 28) and for $ErCo_2$ frequency dependence is significant [40]. For the case of $ErCo_2$, the clusters of Co spins coherently rotate [41] whereas, in Gd_5Ge_4 the onset of GP is due to the effective critical slowing down which occurs because of the existence of the strong magnetic correlations yielding collective excitation [38].In the present case also it might be the case that the onset of GP is due to the collective excitations.

4.4:CONCLUSION

The ac and dc magnetization measurements have been performed on $CuMnO_2$ system. A sharp downturn is observed well above Curie-Weiss temperature in dc magnetization. As the magnetic field increases the downturn decreases and χ^{-1}_{dc} (T) curve obeys the Curie Weiss law. The existence of GP can also be confirmed by analyzing magnetic susceptibility which for a Griffith Phase, should be characterized by an exponent less than unity, that is $\chi(T) \propto (T/T_N-1)^{-(1-\lambda)}$where $0\leq\lambda \leq 1$. The anamoly observed in χ' and χ'' is attributed to the onset of GP. A steady decay in χ'(T) is observed in the frequency range 5Hz to 500Hz except the onset temperature, 54K where an increase in slope is observed which is consistent with the onset of GP. As the frequency increases, the intensity in both χ'(T) and χ'' (T) decreases. Also with increase of frequency, the anomaly shifts towards higher temperature. The GP anamoly in χ' gradually diminishes with the application of small dc field, in accordance with the extreme sensitivity shown by the GP in other compounds. This behavior of χ_{ac} with the applied magnetic field also supports the existence of Griffith like phase above the AFM order in the present system. In this system the exchange interaction at short bonds is stronger, which, with the uniaxial magnetic anisotropy of Mn^{3+}, leaves the system frustrated. This frustration may induce the Griffith-like phase.

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