CHAPTER 3

MAGNETIZATION AND ELECTRON SPIN RESONANCE STUDIES OF LaPbMnO₃ SINGLE CRYSTALS

3.1 INTRODUCTION

There has been a great interest in the properties of rare-earth manganite perovskite R₁₋ₓAₓMnO₃, where R is a rare-earth element such as La, Pr, Nd or Y and A is one of the divalent alkali earth ions including Sr, Ca, Ba and Pb (Helmolt et al 1993). The reason is not only the colossal magneto-resistance (CMR) that occurs in these systems, but also a rich variety of the physical phenomena including intrinsically inhomogeneous ground states, phase separation, charge/orbital ordering (Dagotto et al 2001). The competing interactions in the manganite showing CMR are mainly the Jahn–Teller interaction favoring insulating behavior and the double exchange (DE) favoring the ferromagnetic metallic state (Millis et al 1996). The properties of R₁₋ₓAₓMnO₃ are sensitive to the hole-doping level x and the average A-site radius, which determines the effective one electron bandwidth (W) or the equivalent Mn 3d e₈ electron transfer interaction (t).

It is widely recognized that both double exchange (DE) and the Jahn-Teller (JT) distortion play a major role in this colossal magneto-resistance phenomenon. However, various phase segregation models have been proposed recently that also seek to explain the CMR effect. In these models, the manganite is treated as a mixture of metallic FM clusters within
an insulating paramagnet (PM) matrix with the CMR as a percolation phenomenon driven by a magnetic field induced cluster concentration change. Experimental results on manganites with small A site cations, which have severe local distortion and a lower PM-FM transition temperature \( T_c \) have been attributed to the formation of magnetic clusters in support of the phase segregation models (Goodenough and Zhou 1997). The cluster formation might not be limited to these highly distorted manganites. Essentially conditions for cluster formation e.g. short-range magnetic interactions and strong carrier phonon coupling also exist in manganites with good size match and higher \( T_c \). In particular, a crossover with the applied magnetic field \( H \) might be expected if the clusters do exist. Such \( H \) induced crossover will lead to abnormalities of the resistivity and magnetization, which could be identified by analyzing the magnetization as function of temperature and field.

Nuclear magnetic resonance (NMR) and electron paramagnetic resonance (EPR) are excellent tools for detecting changes in the local electronic properties, especially the local magnetic anisotropy of various electronic sub-phases in colossal magneto-resistance manganites. The most common Mn ion that is measured via ESR is \( \text{Mn}^{2+} \) which is generally accepted as not being present in these compounds. Careful ESR measurements on doped manganites by Causa et al (1998) , Lofland et al (1997) show that both \( \text{Mn}^{3+} \) and \( \text{Mn}^{4+} \) contribute to the ESR signal. The bottleneck model used by Shengelaya et al (1996) shows that the ESR intensity is proportional to the total susceptibility of \( \text{Mn}^{4+} \) and \( \text{Mn}^{3+} \) spins. Composition dependent \( T_c \) and Mn valance of LPMO has been reported in the literature (Vladimirova et al 2001). \( \text{La}_{1-x}\text{Pb}_x\text{MnO}_3 \) \( (0.1 \leq x \leq 0.5) \) compounds with their \( T_c \) values around 300 K could be potential materials for room temperature applications and with relatively low doping level \( (x = 0.1) \)
possess large CMR value. Early ESR studies on the La$_{1-x}$Pb$_x$MnO$_3$ system were performed in single crystals (Searle and Wang 1969) and polycrystals (Ji et al 1998).

### 3.2 X-RAY PHOTOELECTRON SPECTROSCOPY

X-ray photoemission spectroscopy was carried out with the Mg K$_\alpha$ line (1253.6 eV) from a PHI Model 04-548 Dual Anode X-ray source. Energy distribution curves of the elemental core levels were acquired with a large hemispherical electron energy analyzer. The Fermi level was established from tantalum in electrical contact with the sample. The binding energy of core levels is reported with respect to this Fermi level. Sample surfaces were cleaned in ultrahigh vacuum by repeated annealing and exposure to low energy electrons to stimulate the desorption of contaminants. The XPS spectra of La 3d, Mn 2p, Pb 4f and O 1s core levels and the valence band region are shown in Figure 3.1. From the figure it is observed that the peaks of La 3d$_{3/2}$ and La 3d$_{5/2}$ each of which can be well fit by a double Gaussian. Two peaks of La 3d$_{3/2}$ are at 856.3 and 852.6 eV and those of La 3d$_{5/2}$ are at 838.6 and 835.5 eV, respectively. The spin-orbit splitting of La 3d is 17.15 eV. Here the binding energies of La 3d peaks are lower while the spin-orbit splitting is little larger than the corresponding values of La$_2$O$_3$ (Kowalczyk et al 1975). The oxygen core level can provide indications of the presence of defects in the manganese perovskites and a shoulder at higher binding energies.

The oxygen 1s core level of the perovskite manganese oxide La$_{0.9}$Pb$_{0.1}$MnO$_3$ exhibits one dominant peak at a binding energy of 529.7 eV. No satellite peak is observed at high energy and consistence with previous reports on LPMO (Borca et al 2000). In some perovskites, the higher binding energy O 1s peak observed at 531.3 eV has been attributed to either a different surface oxide or to oxygen associated with defects in the perovskite
structure (Choi et al 1999). However this has not been observed in this measurement. The peaks of Mn 2p\textsubscript{3/2} and Mn 2p\textsubscript{1/2} are located at 642.5 and 653.4 eV, respectively. Thus, the spin-orbit splitting of Mn 2p is 10.9 eV, which is very similar to that of MnO\textsubscript{2} (Kowalczyk et al 1975). However, the binding energies of Mn 2p lines are somewhat lower than those of MnO\textsubscript{2}. The full width at half maximum (FWHM) of the Mn 2p core level spectra is about 4.16 eV as compared to 4–4.5 eV for La\textsubscript{0.65}Ca\textsubscript{0.35}MnO\textsubscript{3} and La\textsubscript{0.65}Sr\textsubscript{0.35}MnO\textsubscript{3} (Choi et al 1999). The relatively narrow Mn 2p half width is consistent with the restriction of Mn to a single valence state and the absence of significant numbers of defects in the surface region. The Mn 2p spectra from the La\textsubscript{0.9}Pb\textsubscript{0.1}MnO\textsubscript{3} sample indicate that the disorder in the surface region is not as significant as in many other manganese perovskites. The Mn 3p\textsubscript{3/2} core level binding 642.5 eV for La\textsubscript{0.9}Pb\textsubscript{0.1}MnO\textsubscript{3} is very similar to that observed for Mn\textsuperscript{4+}O\textsubscript{2}~642.4 eV (Lam et al 1980, Kowalczyk et al 1975). The Mn 2p binding energies for LPMO are near to a Mn\textsuperscript{4+} state in the surface region and there is a possibility for mixing of Mn\textsuperscript{3+} and Mn\textsuperscript{4+}.

The binding energy of the Pb 4f\textsubscript{7/2} core level and Pb 4f\textsubscript{5/2} are at 135.4 eV and 140.2 eV respectively. The binding energy of 4f\textsubscript{7/2} is lower than the value observed for Pb\textsuperscript{2+} state in PbTe (137.3 eV), PbS (137.5 eV) and PbO (137.4 eV). The lower binding energy values indicate the lower oxidation state of Pb ion in the surface. As with the arguments put forward by Choi et al (1999), Taguchi and Shimada (1987), Pb is allowed higher oxidation states than the generally expected alkaline earth A\textsuperscript{2+} state (A=Ca, Sr) and this would act to lower the corresponding oxidation state of nearby Mn from 4+ to 3+. 
Figure 3.1  X-ray photoemission spectra of the La 3d, Pb 4f, O 1s, and Mn 2p core levels of La\(_{0.9}\)Pb\(_{0.1}\)MnO\(_3\)

3.3 MAGNETIC PROPERTIES OF La\(_{0.9}\)Pb\(_{0.1}\)MnO\(_3\) SINGLE CRYSTAL

Magnetic susceptibility is the magnetization of a material per unit applied field. It describes the magnetic response of a substance to an applied magnetic field. If \(M\) is the magnetization and \(H\) the applied magnetic field, then the magnetic susceptibility denoted by \(\chi\) is given by

\[
\chi = \frac{M}{H}
\]  

(3.1)

For crystalline material \(\chi\) may depend upon the direction of \(H\) with respect to the axis of the crystal because of anisotropic effects. The Curie
temperature $T_c$ is the temperature below, which there is a spontaneous magnetization ($M$) in the absence of an externally applied magnetic field and above which the materials are paramagnetic. A relation between magnetic or electric susceptibilities and the absolute temperature is followed by ferromagnets, antiferromagnets, nonpolar ferroelectrics, antiferroelectrics and some paramagnets. Ferromagnetic substances become paramagnetic when temperature rises above the Curie temperature ($T_c$), Curie-Weiss law is given by

$$\frac{M}{H} = \chi = \frac{C}{T - T_c}$$

(3.2)

where $\chi$ is the susceptibility, $C$ is a constant for material, $T$ is the absolute temperature and $T_c$ is called the Curie temperature, which marks the transitional boundary between the paramagnetic and ferromagnetic state (Jiles 1998). Above the Curie temperature ferromagnetic and antiferromagnetic materials become paramagnetic because the thermal motion is sufficient to randomize the orientation of magnetic dipoles. Thus, above $T_c$ there is no net magnetic moment in the absence of magnetic field. The Curie-Weiss law refers to magnetic and electric behavior above the transition temperature of the material in question. The Curie-Weiss behavior is usually a result of an interaction between neighboring atoms, which tends to make the permanent atomic magnetic dipoles point in the same direction. The Curie-Weiss law is in satisfactory agreement with the observed temperature dependence of susceptibility. Far above the $T_c$ the reciprocal susceptibility increases linearly with temperature, when extrapolated a paramagnetic Curie point $\Theta$ from the linear dependence of $1/\chi = 0$. 
3.3.1 Magnetic Hysteresis

Hysteresis is well known in ferromagnetic materials. When an external magnetic field is applied to a ferromagnet the atomic dipoles align themselves with the external field. Even when the external field is removed part of the alignment will be retained and the material becomes magnetized. The relationship between magnetic field strength ($H$) and magnetic flux density ($B$) is not linear in such materials. If the relationship between the two is plotted for increasing levels of field strength, it will follow a curve up to a point, where further increase in magnetic field strength will result in no further change in flux density. This condition is called magnetic saturation. If the magnetic field is reduced linearly, the plotted relationship will follow a different curve back towards zero field strength at which point it will be offset from the original curve by an amount called the remanent flux density or remanence. If this relationship is plotted for all strengths of applied magnetic field the result is a sort of S shaped loop. The 'thickness' of the middle bit of the S describes the amount of hysteresis, related to the coercivity of the material.

To study the ferromagnetic state of the samples, the hysteresis was measured at room temperature and 10 K with a maximum field of 10 kOe. At room temperature the hysteresis measurement showed a straight line up to 1 T field due to its paramagnetic nature. To calculate the easy axis of magnetization the magnetic hysteresis was measured at 10 K while the magnetic field was parallel and perpendicular to the ‘ab’ plane as shown in Figure 3.2. The magnetic field parallel to the ‘ab’ plane showed easy axis of magnetization and saturated around 3 kOe. The magnetic field perpendicular to the ‘ab’ plane showed hard axis of magnetization and saturated around 6 kOe field. The difference in saturation magnetization is due to magneto-crystalline anisotropy. While increasing the temperature, the hysteresis
follows the nature of the regular soft magnetic material and the coercivity of the compound depends on the temperature.

Figure 3.2  Magnetic hysteresis measured as a function of magnetic field parallel to ‘ab’ and ‘c’ plane at 10 K

3.3.2 Field Dependent DC Magnetization

In order to study the evolution of the magnetic properties as a function of the low magnetic field, magnetic measurements were carried out. The magnetization of the sample was measured at zero field cooled (ZFC) and at field cooled (FC) conditions with field H= 20 Oe and 500 Oe. All measurements were carried out along the easy axis of magnetization (‘ab’ plane) and magnetic field applied parallel to the ‘ab’ plane. The magnetization (M) was measured using a superconducting quantum interference device (SQUID) magnetometer (MPMS, Quantum design). In the case of ZFC measurements, the sample was cooled down to the desired temperature from well above the transition temperature in the zero magnetic field and
measurements were performed in the heating cycle with the magnetic field, while for the FC case the sample was cooled in the presence of the magnetic field and measured during heating like a ZFC measurement. The temperature dependence of DC magnetization (M) of $\text{La}_{0.9}\text{Pb}_{0.1}\text{MnO}_3$ single crystal was studied with the applied field of 500 Oe. Magnetization rise sharply up to $T_c$ (252 K) and showed saturation in the low temperature region due to the ferromagnetic ordering as shown in Figure 3.3. This shows that the samples exhibit sharp paramagnetic to ferromagnetic (PM-FM) transition (Curie temperature) calculated from the peak of $\frac{dM}{dT}$ in the M vs T curve. In ZFC measurements, the magnetization decreased continuously near $T_c$ and approaches to zero above $T_c$.

The measurement of low field ZFC–FC measurement is generally used to characterize spin-glass behavior. Paramagnetic to ferromagnetic transition occurred at 252 K in both cases and showed monotonic increase below $T_c$. Magnetization at 20 Oe field was identical up to $T_r$ and below this ZFC and FC curves started to deviate from each other. The irreversibility of the curve is due to the cluster glass (CG). ZFC results show that the $M_{ZFC}$ increases to a maximum value at $T_r$ then deviates from $M_{FC}$ with decreasing temperature. The ZFC and FC magnetization irreversibility suggests that the spontaneous magnetic state at low temperature is a magnetic cluster glass – like state (Anil Kumar et al 1998). The position of maximum $T_r$ of $M_{ZFC}$ marks the defreezing temperature of the cluster glass (Shi et al 2002) and above $T_r$ the system gradually approached the paramagnetic state after $T_c$. Fundamentally, a cluster glass is a type of spin glass. The magnitude of splitting and the $T_r$ depend on the magnetic field. Field of 500 Oe was applied to study the magnetic field dependence behavior. Irreversibility was not observed between ZFC and FC measurement for this field as shown in the Figure 3.4.
Figure 3.3 Magnetization as function of temperature measured in zero field cooled measurement with applied field 500 Oe

The field dependence in the irreversible region may be due to lack of complete saturation as it completely saturated in the higher field (500 Oe). This may be due to the existence of misalignment of spin in the low applied magnetic field. At lower fields, the magnetization followed mean-field-like fluctuations and the field was not sufficient to destroy the spin misalignment between adjacent Mn$^{3+}$/Mn$^{4+}$ ions (Heilman et al 2000). The short range behaviour is common in the double exchange ferromagnet because of the negligible interaction between well separated FM islands but these islands are believed to interact with each other when the magnetic field is applied (Murakami et al 2003). Above $T_c$, magnetic susceptibility can be fitted well using Curie-Weiss law. The fitting parameter Curie-Weiss temperatures ($\Theta$) were 261 K and 263 K for an applied field of 20 Oe and 500 Oe respectively.
Figure 3.4  Temperature dependent magnetization measured at zero field cooled and field cooled condition with applied field of 20 Oe

Figure 3.5  Temperature dependent magnetization measured at zero field cooled and field cooled condition with applied field of 500 Oe
The deviation of $\Theta$ from the Curie-Weiss law, that is generally associated with ferromagnetic spin clustering effect in the paramagnetic region (Phan et al 2007) and starts at the temperature near to 1.03 $T_c$ and 1.04 $T_c$ and these values are lower than the reported value (Volkov et al 2007). Inverse susceptibility of 20 Oe applied field shows splitting between ZFC and FC condition in the inverse paramagnetic susceptibility region as shown in the Figure 3.6. In the case of 500 Oe field, the inverse susceptibility did not show any difference between ZFC and FC in the paramagnetic region as shown in the inset of Figure 3.6. Application of low field is not enough to destroy the cluster glass property of the sample as confirmed by ZFC and FC measurements and the moderate field of 500 Oe is sufficient to destroy the clusters.

Figure 3.6 Inverse susceptibility as a function of temperature with applied field of 20 Oe. Inset shows inverse susceptibility with 500 Oe field
It is observed that $T_{MI}$ is lower than $T_c$ because $T_{MI}$ is sensitive to the homogeneity, strain distribution, and local chemistry of the material, whereas spin alignment plays a major role in the magnetic properties of the manganites. Two different mechanisms are used to explain inhomogeneity in manganites, electronic phase separation for nanometer scale inhomogeneity and the effects of disorder for micrometer-scale inhomogeneity. Micro meter inhomogeneity is due to the random distribution of rare earth/alkaline earth ions (Ahn et al 2004). It is believed that the magnetization of FM clusters freeze in a random fashion below the glass transition and the magnetization of each cluster acts like an individual spin in a spin-glass. The magnetization of each cluster is free to rotate above the glass temperature. Application of the magnetic field removes inhomogeneity and produces large colossal Magneto-resistance (CMR) effect due to the formation of ferromagnetic long range order. Short range order behaviour could be due to the misalignment of A site ion rather than superparamagnetic or anti-ferromagnetic contribution and it produces large CMR value near $T_c$. Low field magnetization measurements show cluster glass property and this is consistent with the reported value of La$_{1-x}$Pb$_x$MnO$_3$ bulk materials (Shi et al 2002, Chau et al 2003).

### 3.3.3 Frequency Dependence of AC Magnetization

AC susceptometer is a unique tool among the magnetic measurement techniques, where it is capable of measuring susceptibility under very small ac magnetic fields with or without a dc bias field. It can also measure the magnetic susceptibility as a function of frequency and temperature, while it is capable to separate the real component $\chi'$, and the imaginary component $\chi''$ of the complex susceptibility. Since the actual response of the sample to an applied ac field is measured, the magneto dynamics can be studied through the complex susceptibility ($\chi' + i\chi''$). The real component $\chi'$ represents the component of the susceptibility that is in phase
with the applied ac field, while $\chi''$ represent the component that is out of phase. The imaginary component $\chi''$ is related to the energy losses, or in other words, the energy absorbed by the sample from the ac field. The coexistence of different competing factors in manganites commonly results in different kinds of glassy behaviours with unusual relaxation dynamics and frequency-dependent phenomena. The experiments prove only the existence of a sort of collective relaxation behaviour, which does not provide any definitive evidence of a true spin-glass (SG). At this moment the fundamental question whether the mixed valent manganites with the unusual glassy phenomena can be classified in a classical SG still remains open.

AC susceptibility was measured with an alternating field of $h_{ac} = 10$ Oe with different frequencies viz. 500, 1000 and 10,000 Hz. Figures 3.7 and 3.8 shows the temperature dependence of the ZFC in phase ($\chi'$) and out of phase ($\chi''$) susceptibility for single crystals of $La_{0.9}Pb_{0.1}MnO_3$. The ferromagnetic transition temperature or $T_c$ is defined as the inflection point of the susceptibility or peak in $dM/dT$ and the $\chi$ curves display a transition from paramagnetic (PM) to ferromagnetic (FM) state. At about 254 K, both susceptibilities abruptly decrease to zero, exhibiting the ferromagnetic-paramagnetic phase transition. The frequency dependent maxima in the $\chi''$ and $\chi'$ together with cups in the low field DC magnetization are completely associated with the FM state. One can see that the peak temperature in the out of phase susceptibility increases with the frequency as expected in a conventional SG system (Chen et al 1996). The various applied frequency values like 500, 1000 and 10,000 Hz changed the peak temperatures to 250 K, 253 K and 255 K respectively as shown in Figure 3.8. With lowering temperature, a spin glass behavior is possibly exposed by a shoulder in $\chi'$ and hump in $\chi''$ and these are frequency dependent.
Figure 3.7  Temperature variation of in-phase ac susceptibility ($\chi'$) of LPMO single crystal

Figure 3.8  Temperature variation of out of phase ac susceptibility ($\chi''$) of LPMO single crystal
The shoulder considerably shifts to lower temperature with lowering frequency (Shi et al 2002). We observed frequency dependent hump in the $\chi''$ susceptibility at low temperature and this may be the defreezing temperature $T_f$ of the cluster glass state. At the temperature below $T_c$ the material shows the mixing of ferromagnetism with glassy magnetism. The signature of the defreezing temperature was not clearly seen in the imaginary part and it may be removed with magnetic field. Spin glass or cluster material showed decrease in defreezing temperature with magnetic field (De et al 2007). Removal of cluster behavior with magnetic field was confirmed by magnetic field dependent field cooled and zero field cooled measurements. The linear $\chi''(T)$ and $\chi'(T)$ AC susceptibility shows frequency dependence in the ferromagnetic region and may be due to cluster glass property. Unusual dynamics and frequency-dependant phenomena are common features of this magnetically inhomogeneous phase, which is normally quoted as a cluster-glass or SG like behaviour.

3.4 TEMPERATURE DEPENDENT ELECTRON SPIN RESONANCE

The magnetic state of rare earth manganite materials is complex and is known to be strongly influenced by both divalent substitution and oxygen composition. Clearly, inhomogeneity and nonuniformity are serious concerns. It seems crucial to obtain a prototypical manganite sample whose magnetic homogeneity is unequivocal. The electron spin resonance (ESR) studies were carried out below and above the Curie temperature of the LPMO single crystal at 9.5 GHz (X band) using in a JEOL spectrometer.
Figure 3.9 Temperature dependence of electron spin resonance spectrum below and above the Curie temperature
Electron spin resonance (ESR) has been proved to be useful for studying spin dynamics of Mn$^{3+}$ and Mn$^{4+}$ ions, interaction mechanisms, and dynamical Jahn-Teller distortions in perovskite manganites around $T_c$, where the CMR effect is often observed. Figure 3.9 shows asymmetric ESR signals for the samples at some representative temperatures in the ferromagnetic and paramagnetic phases. A single asymmetric line was observed in the paramagnetic state. The parameters were derived from the ESR measurements such as line width ($\Delta H_{pp}$), $g_{eff}$, resonance field ($H_{res}$), double integrated intensity (I) and asymmetry parameter (AP). The ESR line started to appear with a shoulder around 270 K due to the ferromagnetic cluster in the paramagnetic matrix. Near $T_c$ the spectrum is splitted into prominent low field and high field ferromagnetic resonance lines. The lines are very well separated and the low field side started to increase with the decreasing temperature. Below $T_c$ high field ferromagnetic resonance line moved towards high field region. This may be due to the influence of the localized magnetic moment (Shengelaya et al 1996).

The temperature dependence of the peak-to-peak ESR line width prominently increases on decreasing the temperature below $T_c$ as shown in the Figure 3.10, which corresponds to the more inhomogeneous local magnetic field distribution. The g-value of LPMO single crystal showed less than that of the free electron ($g_e = 2.0023$) value, which is shown in Figure 3.11. The ferromagnetic material shows the g value more than free electron g value (Ji et al 1998) and the lower value may be due to the magnetic inhomogeneity. The Asymmetric Parameter (AP) serves as a clue to determine the homogeneity in the sample (Dyson 1955).
Figure 3.10  Temperature dependence of $H_r$ and $\Delta H_{pp}$ below and above the Curie temperature.

Figure 3.11  Temperature dependence of $g_{\text{eff}}$ and AP (asymmetry parameter) below and above the Curie temperature.
The lower value of AP confirms the inhomogeneity of the sample due to the random distribution of ion throughout its volume and is shown in the Figure 3.11. The intensity of FMR was used to identify the nature of the magnetic ions contributing to the resonance which is proportional to the number of the spins. Double integrated intensity decreases gradually with increase in temperature because part of FM clusters transform to PM phase (Yang et al 2007) and is shown in the Figure 3.12. Increase of line width below $T_c$ due to the local magnetic inhomogeneity and an increase with the decreasing temperature can be attributed to the random shape and distribution (Ding et al 2005). The splitting of the ESR line below and above the Curie temperature region is due to some of the Mn$^{3+}$ ions, which may be surrounded by Mn$^{4+}$ to form spin clusters and associated local distortion below the paramagnetic to ferromagnetic phase transition temperature.

![Figure 3.12 Double integrated intensity as function of temperature](image)

Figure 3.12 Double integrated intensity as function of temperature
The interaction between the localized magnetic moments becomes more significant, which contributes to the broadening of the resonance line (Narsinga Rao et al 2002). Glassy magnetism in LPMO material reported earlier (Chau et al 2003, Shi et al 2002) is consistent with our experimental investigations.

3.5 CONCLUSION

Hysteresis measurement on LPMO single crystal shows the easy axis of magnetization along the ‘ab’ plane and difference in saturation magnetization due magneto-crystalline anisotropy. Paramagnetic to ferromagnetic transition occurs at 252 K for the application of different field value. Low field ZFC and FC magnetic measurements show that the irreversibility may be due to glassy magnetism and magnetic inhomogeneity. In particular, a cluster glass magnetic state was observed in low magnetic field below the Curie temperature and destroyed in higher applied field value of 500 Oe. Inverse susceptibility results clearly indicate the presence of ferromagnetic cluster in the paramagnetic region above the Curie temperature. Frequency dependent AC susceptibility showed cluster glass behavior and consistent with DC magnetization measurements. Temperature dependent electron spin resonance studies show broadening of peaks below the Curie temperature. Increase of line width below $T_c$ due to the local magnetic inhomogeneity and an increase with the decreasing temperature can be attributed to the random shape and distribution. The lower value of AP confirms the inhomogeneity of the sample due to the random distribution of ion throughout its volume.