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# Effect of disorder on magnetic ordering of $La_{0.5}Gd_{0.2}Sr_{0.3}MnO_3$ manganite

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**Abstract**

We have undertaken dc magnetization and linear and nonlinear ac susceptibility measurements for the $La_{0.5}Gd_{0.2}Sr_{0.3}MnO_3$ (LGSMO) bulk system. Our experimental results provide evidence of a glassy phase associated with the system. Doping of Gd on La sites and its antiferromagnetic (AFM) coupling with Mn lattices are expected to induce random magnetic disorder in the magnetic lattice of the LGSMO system. This random disorder in an otherwise long range ordered ferromagnetic (FM) host divides the system into finite size FM clusters, which undergo cluster-glass-like freezing. Further, we have found a non-monotonic dependence of freezing temperature on the applied magnetic field. We have attributed this behavior to the non-monotonic variation of the cluster size or concentration with the external field, which in turn is related to the modulation of inherent randomness of AFM Gd–Mn exchange interaction in the system. We suppose that the increase in coherence in response of the random AFM Gd spins with increase in applied field initially increases spin disorder in the FM host and then decreases it. This, in turn, modulates the cluster size and accordingly the collective glassy state of the system.

(Some figures in this article are in colour only in the electronic version)

## 1. Introduction

It is well known that interesting correlated electron materials are unusually sensitive to disorder [1, 2]. In fact, the ordered state or the phase behavior of different condensed matter systems is unstable against arbitrarily small defects and impurities [3]. A strong effect of small disorder on material properties is basically one of many examples of sensitivity of

materials properties to perturbations. Very recently the effect of random disorder has been shown to be responsible for several unusual electronic, structural, thermal and magnetic properties in the diversified field of condensed matter, as well as soft matter physics, such as the specific heat problem [4], superconductivity [5, 6], manganites [7], ferroelectricity [8], directed polymers [9], liquid crystals [10] etc. Magnetism with the effects of random disorder is one of the areas that receives continuous interest [11–17]. For example, suppression of ferromagnetism by a relatively small number of sites with local negative field in diluted magnetic semiconductors [18], relaxor-like ferromagnetism in $Nd_{0.5}Ca_{0.5}Mn_{0.98}Cr_{0.02}O_3$ manganites [19] with Cr doping, suppression of the magnetic phase transition close to the metal–insulator crossover in manganites [20], the glassy transport phenomenon in a phase separated perovskite cobaltite [21] etc. Sensitivity of measured properties towards random disorder may be an indication of some interesting and potentially useful properties of the material. Understanding and control of this phenomenon is an important open issue in materials physics.

Colossal magnetoresistance (CMR) manganites—a strongly correlated electronic system having correlated degrees of freedom—offer a surprising new paradigm for strong effects of weak disorder [1, 2, 22]. An interesting point is that the CMR effect itself is an example of a generically extreme sensitivity to perturbations. An intriguing example of this phenomenon is a sharp sensitivity of electrical resistivity to changes in chemical composition as shown by Sawaki *et al* [23] for Al doping into the electrically active Mn site. Doped perovskite manganites, with the general formula $La_{0.7-x}R_xD_{0.3}MnO_3$ (R = smaller rare earth (RE) cations, D = divalent alkaline earth cations such as $Sr^{2+}$, $Ca^{2+}$, $Ba^{2+}$, $Pb^{2+}$), have attracted enormous research interest during the last decade due to exotic modulation of their electronic and magnetic properties with the doping of smaller RE ions (R) on the La site, while maintaining an optimum $D^{2+}$ doping level of 30% [24–31]. As a result of replacement of La by other RE elements, perovskite-based structures ($ABO_3$) occasionally show lattice distortions as modifications from the cubic structure [24, 31]. Such a lattice distortion is governed by the so-called tolerance factor, $t = \langle A\text{–}O\rangle/\sqrt{2}\langle Mn\text{–}O\rangle$, where $\langle A\text{–}O\rangle$ and $\langle Mn\text{–}O\rangle$ are the mean cation–oxygen bond lengths of A ($La^{3+}$ and $D^{2+}$) and Mn sites, respectively. In this context, we should mention the systematic studies as documented by Hwang *et al* on the tolerance factor $t$ dependence of the electronic and magnetic properties and a para–ferromagnetic Curie temperature ($T_C$)–$t$ phase diagram of $La_{0.7-x}R_xCa_{0.3}MnO_3$ (R = $Pr^{3+}$, $Y^{3+}$) manganites [31]. Additionally, there exists a plethora of reports on the heavy RE doped prototype manganite system $La_{0.7-x}R_xD_{0.3}MnO_3$, as contributed by Sun *et al* [24] for R = $Gd^{3+}$, Terashita *et al* [25] for R = $Gd^{3+}$, Terai *et al* [26] for R = $Dy^{3+}$ etc. Due to the much smaller ionic radius and high magnetic moment, replacement of $La^{3+}$ by a heavy RE element will not only influence the lattice distortion, i.e. $t$, but also introduce extra magnetic coupling [24–30]. Apart from having smaller ionic radius (1.107 Å) of $Gd^{3+}$ than $La^{3+}$ (1.216 Å) and the largest magnetic moment (7/2 $\hbar$) of all REs, progressive substitution of $La^{3+}$ for heavy RE $Gd^{3+}$ is a particularly favorable case among other heavy REs, since the $Gd^{3+}$ ion has orbital angular momentum $L = 0$ and therefore no complications would be caused by the crystalline field. As generally recognized from previous studies, doping of Gd on manganites, besides decreasing the tolerance factor $t$ [24, 30], promotes random character of distribution of A site cations ($La^{3+}$, $Sr^{2+}$, $Gd^{3+}$). As a result, there is random distribution of hopping of conduction electrons as well as exchange between localized spins that induces random disorder in the magnetic lattice of the system [24]. Additionally, several groups [24, 25, 27, 28, 30] have established from different magnetic measurements that in case of the Gd doped manganite system there exists an antiferromagnetic (AFM) exchange coupling between Gd and Mn moments, resulting in $Gd^{3+}$ spins being polarized antiparallel to the ferromagnetic (FM) component of the Mn

lattice. This AFM coupling between Gd and Mn moments would also contribute to the random disorder in the FM host of the Gd doped manganite system [3, 24]. Furthermore, all these reports, especially those on bulk polycrystalline samples [24], explore the possibility of external field induced tuning of this extra AFM coupling between Gd and Mn moments. This also modulates the microscopic magnetic structure of the Gd doped manganite system. All these above mentioned studies show that Gd doped manganites are a relevant system to study the effect of A-site disorder on magnetic properties of manganites, and prompted us to probe the nature of magnetic ordering in such a system. We have carried out dc magnetization and linear and non-linear ac susceptibility ($\chi_{ac}$) measurements on the $La_{0.7-x}Gd_xSr_{0.3}MnO_3$ bulk polycrystalline system for a lower doping level of Gd ($x \leqslant 0.3$). In this paper, we present evidence for a spin-glass-like frozen phase or cluster glass phase associated with the system $La_{0.5}Gd_{0.2}Sr_{0.3}MnO_3$, i.e. for $x = 0.2$. We have observed a non-monotonic variation of freezing temperature with applied ac, as well as dc, magnetic field, where freezing temperature first increases with an increase in applied field up to a certain limit, beyond which it decreases with further increase in field. We understand this observed field dependent behavior of freezing temperature in terms of tuning of the disorder or glassiness associated with this system with an applied magnetic field. We suppose that the possible explanation for this non-monotonic change in the freezing temperature, i.e. the glassiness of the system, arises from the non-monotonic change in the cluster size with the external magnetic field, which in turn is related to the modulation of inherent randomness of the AFM Gd–Mn exchange interaction in the system. The increase in coherence, in response to the random AFM impurity Gd spins with an increase in applied field, initially increases spin disorder in the FM host and then decreases it. This, in turn, modulates the cluster size and accordingly the glassy phase of our system.

## 2. Experimental details

Polycrystalline bulk samples of $La_{0.5}Gd_{0.2}Sr_{0.3}MnO_3$ (LGSMO) were prepared using a standard solid-state reaction route. We employed stoichiometric mixtures of high purity $La_2O_3$(99.99%), $Gd_2O_3$(99.99 + %), $SrCO_3$(99.9 + %) and $MnO_2$(99.99%), which were first heated at 800 °C for 12 h, then at 1000 °C for 12 h and at 1200 °C for another 12 h, with intermediate grinding. After final grinding and pelletization, the pelletized sample was sintered at 1400 °C for 36 h. The structure and phase purity of the as-prepared sample were checked by powder x-ray diffraction (XRD) using a Rigaku Rotoflex RTC 300 RC powder diffractometer, with monochromatic Cu K$\alpha$ radiation ($\lambda \sim 1.542$ Å). Our XRD data established a single phase of the sample (within the detection limit) formed in the orthorhombic structure [24]. We carried out a detailed magnetic study using dc magnetization and linear and nonlinear $\chi_{ac}$ measurements. Low field DC magnetization measurements were made using a home-built vibrating sample magnetometer [32], where a loudspeaker was used as the vibrating system and a lock-in amplifier as the detection system (Stanford Research, SR 530). During the measurements, the sensitivity of the set-up was better than $10^{-6}$ emu in the measured field range (up to 1 kOe). The phase resolved linear and nonlinear $\chi_{ac}$ as a function of field, frequency and temperature were measured using a home made susceptometer [33] in combination with a lock-in amplifier (Stanford Research, SR 830) and a temperature controller (Lakeshore, DRC-93C). During the measurements, the sensitivity of the set-up was better than $10^{-7}$ emu in the measured field range (up to 60 Oe). Temperature measurements were carried out in the temperature range of 80–310 K in a liquid-nitrogen variable temperature cryostat by a calibrated platinum resistance thermometer (Pt-100, Lakeshore). The Pt-100 sensor was attached with the sample holder using a special varnish (GE), which is a very effective low temperature adhesive, especially useful for good thermal contact. The sample holder was a sapphire plate attached to a

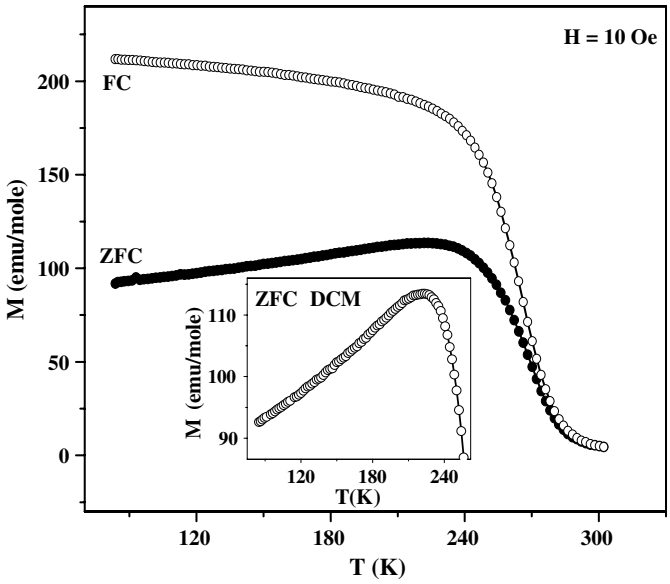


**Figure 1.** FC–ZFC DCM at a measuring magnetic field of 10 Oe. The inset shows the same ZFC DCM in a narrow temperature scale.

hollow stainless steel rod. A heater (phosphor–bronze) was wound with non-inducting winding just above the sample position. The stability of the temperature during the measurements was better than $\pm 10$ mK. The details of the temperature measurements have been described in previous literature [32, 33]. Both set-ups were computer controlled through a general purpose interface bus (GPIB). Both ac and dc measurements were made on the same pellet of our samples. All the measurements were performed with the applied field parallel to the long edge of the samples to minimize the effect of the demagnetization field.

## 3. Results and discussion

We have investigated the temperature dependence of field-cooled (FC) and zero-field-cooled (ZFC) DC magnetization (DCM) for the LGSMO bulk sample (figure 1). A strong irreversibility in DCM at $H = 10$ Oe was observed, as indicated by the large bifurcation in the FC and ZFC curves. Moreover, this irreversibility starts at a temperature very close to the paramagnetic–FM transition temperature ($T_C$). The absence of true long range ordering is apparent from this strong history dependence of FC and ZFC DCM below $T_C$.

AC susceptibility ($\chi_{ac}$) measurement is a compatible technique to probe the associated magnetic phase and its sensitivity towards several influencing factors of a system. Investigation of the nonlinear $\chi_{ac}$ is one of the most important tools to understand any magnetic phase transition [34–39]. In general, the nonlinearity of magnetization ($M$) in the presence of a magnetic field ($H$) is given by the series expansion

$$M = M_0 + \chi_1 H + \chi_2 H^2 + \chi_3 H^3 + \cdots \tag{1}$$

where $M_0$ is the spontaneous magnetization, $\chi_1$ is the linear susceptibility and $\chi_2$, $\chi_3$, ... are the nonlinear susceptibilities. Linear $\chi_{ac}$, measured at a frequency ($f$) of 131 Hz and at an ac magnetic field ($H_{ac}$) of 4 Oe, shows a reasonably sharp peak at the transition temperature for its out-of-phase component or imaginary part ($\chi_1^I$) (figure 2(a)), whereas its in-phase counterpart

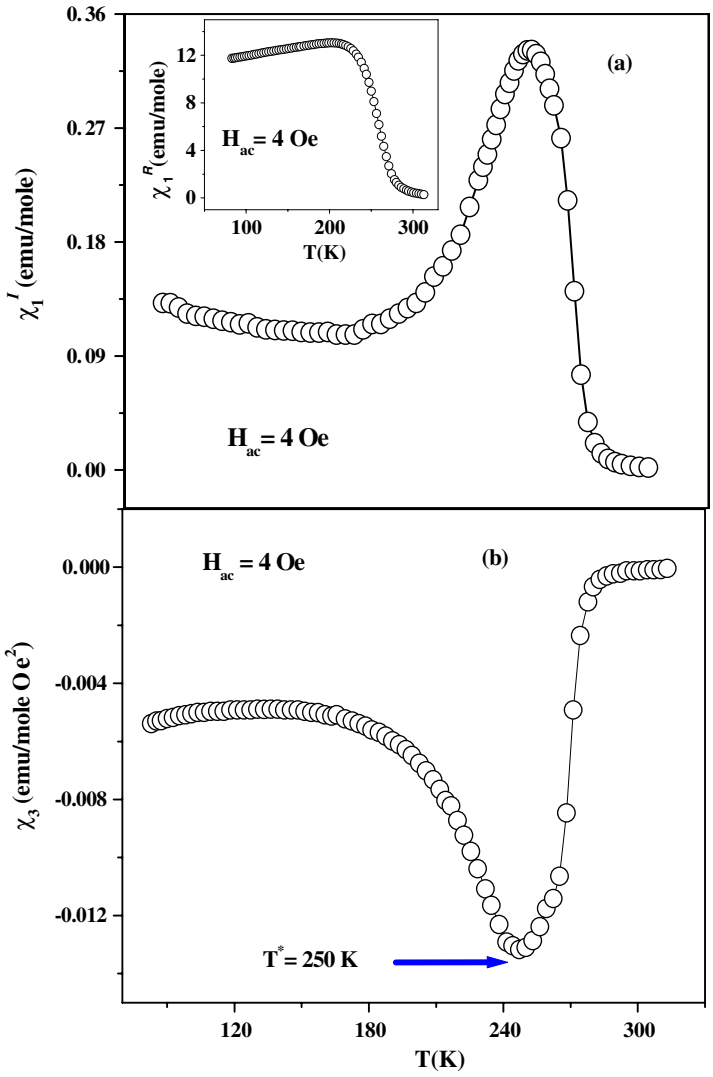


**Figure 2.** (a) Out-of-phase linear $\chi_{ac}$, $\chi_1^I$ versus temperature curve measured at $H_{ac} = 4$ Oe. The inset shows the same plot for its in-phase counterpart $\chi_1^R$. (b) $\chi_3$ versus temperature curve measured at $H_{ac} = 4$ Oe, where the arrow indicates the freezing temperature $T^*$.

or real part ($\chi_1^R$) shows a broad rounded maximum (inset in figure 2(a)). It is evident from the inset in figure 2(a) that for $\chi_1^R$ the exact position of the shoulder as well as of its $H_{ac}$ and $f$ dependence is rather difficult to determine. In contrast, since the nonlinear $\chi_{ac}$ becomes more pronounced in the vicinity of the phase transition, the hump/peak as well as their $H_{ac}$ and $f$ dependence becomes much clearer. In fact, several subtle features not discernible using linear $\chi_{ac}$ can be picked up using nonlinear $\chi_{ac}$. It has already been established how nonlinear $\chi_{ac}$ can be used to unravel the magnetism of interesting metastable systems [34–38] as well as to effectively probe the critical behavior of systems with long range magnetic order [38, 39]. In this work, we have concentrated mainly on the behavior of second ($\chi_2$) and third ($\chi_3$) order $\chi_{ac}$. Third order susceptibility ($\chi_3$), measured at $H_{ac} \sim 4$ Oe, shows a reasonably broad peak

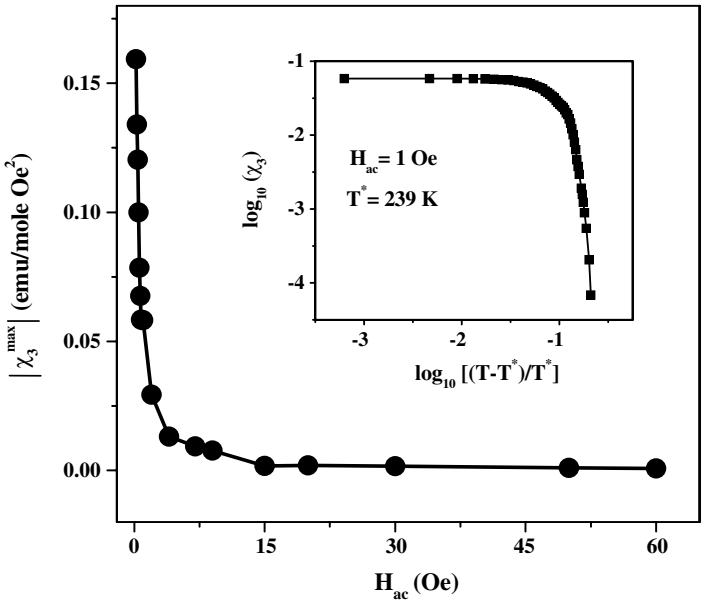


**Figure 3.** $|\chi_3^{max}|$ plotted as a function of $H_{ac}$ clearly shows the divergent nature of $\chi_3$ at $H_{ac} \rightarrow 0$ Oe, thus indicating the cooperative freezing phenomenon. The inset shows a log–log plot of $\chi_3$ against reduced temperature $(T - T^*)/T^*$ at $H_{ac} = 1$ Oe.

with a large transition width as shown in figure 2(b). In fact, $\chi_3$ has long been used as a direct probe of the divergence of Edward–Anderson order parameter, signifying the onset of a spin glass (SG) transition [40], where theoretically $\chi_3$ is expected to have a negative divergence in the limit of $H_{ac} \rightarrow 0$ at glass freezing temperature [41]. Generally, this is demonstrated by plotting the magnitude of the peak value of $\chi_3$ ($|\chi_3^{max}|$) as a function of $H_{ac}$. Figure 3 shows the divergence of $|\chi_3^{max}|$ in the limit of $H_{ac} \rightarrow 0$ for the sample LGSMO, indicating that the magnetic phase occurs due to a cooperative freezing phenomenon, whether it constitutes a classical SG or the behavior is introduced by random dipolar intercluster interactions among magnetic clusters. It should be mentioned in this context that such divergence of $|\chi_3^{max}|$ at $H_{ac} \rightarrow 0$ cannot be obtained for other metastable or long range ordered magnetic systems. For instance, for a superparamagnetic system, $|\chi_3^{max}|$ is clearly found to saturate in the limit of $H_{ac} \rightarrow 0$ [42, 43]. On the other hand, for a long range ordered FM or ferrimagnetic system, higher order susceptibilities would not show a divergence in this limit of $H_{ac} \rightarrow 0$. In contrast, as the magnetization becomes linear in the limit of $H_{ac} \rightarrow 0$, $|\chi_3^{max}|$ approaches zero in that limit of $H_{ac}$. Even anisotropy-driven systems or domain-wall dynamics in a long range ordered FM system are not expected to show such negative divergence (like SG) of $|\chi_3^{max}|$ at $H_{ac} \rightarrow 0$ [44]. We have also shown (inset in figure 3) a log–log plot of $\chi_3$ against reduced temperature $(T - T^*)/T^*$ ($T^*$ is the freezing temperature as found from the peak in $\chi_3$) at $H_{ac} = 1$ Oe for our system. In this analysis, $\chi_3$ is found to get flattened at $T \rightarrow T^*$ instead of being divergent, as can be clearly seen from the inset in figure 3. We attribute this flattening of $\chi_3$ near the critical temperature to the inherent inhomogeneity of this sample. Even for confirmed canonical SGs, e.g. AuFe with 0.97 at.% Fe, similar flattening of $\chi_3$ close to the critical temperature was observed [45]. This was attributed to the fluctuations induced by inhomogeneities in real systems on large scales close to the critical temperature. This is why we have not characterized the phase transition by quoting any critical exponent, since taking

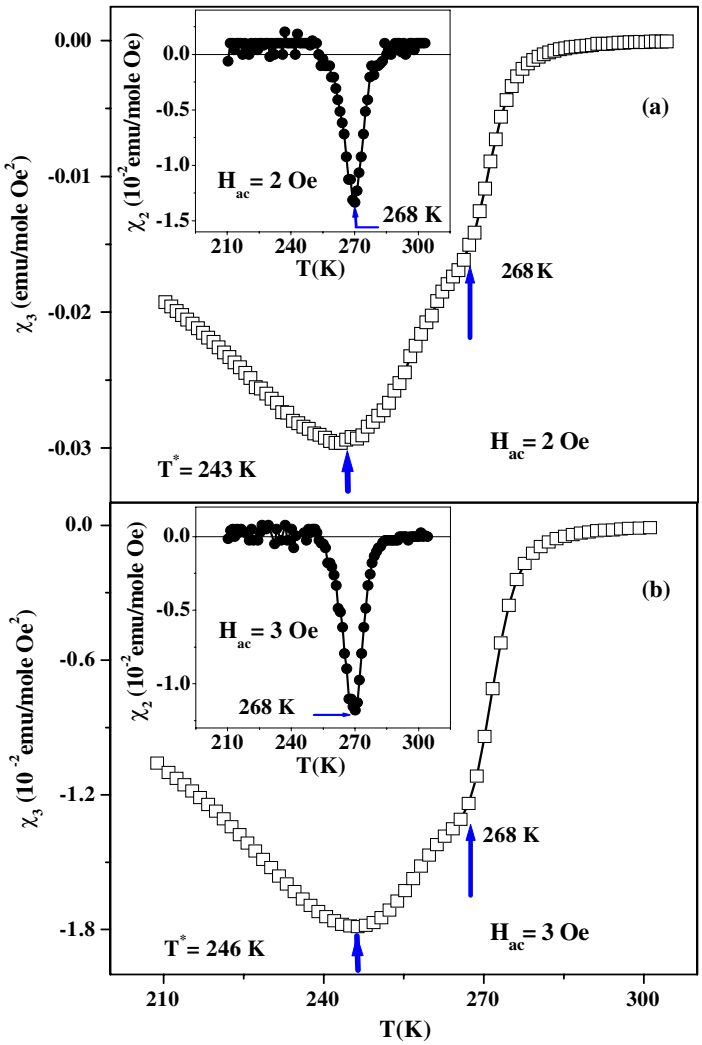


**Figure 4.** $\chi_3$ versus temperature curves measured at (a) $H_{ac} = 2$ Oe and (b) $H_{ac} = 3$ Oe. The insets in (a) and (b) show the corresponding $\chi_2$ versus temperature curves at $H_{ac} = 2$ and 3 Oe, respectively.

any value of the exponent $\gamma$ from data of this kind will give the mean-field value, not the asymptotic critical exponent [40]. Furthermore, we can compare our results with that obtained for $(La_{0.25}Nd_{0.75})_{0.7}Ca_{0.3}MnO_3$ manganites by Rivadulla *et al* [46], where, similar to our case, the negative peak in $\chi_3$ was very broad compared to that of a typical SG. Although in their study they showed the divergent character of nonlinear susceptibility, they found it difficult to ascribe the critical exponents to any existing universality class of SGs and have concluded that the glassiness of the system arises due to an interacting assembly of magnetic clusters. Likewise, we would like to point out that our system, although it does not constitute a conventional canonical SG, shows an important feature (i.e. divergence of $\chi_3$) of SG and thus we have designated the observed phase as the glass-like phase.

Figure 4(a) and its inset show $\chi_3$ and $\chi_2$ at $H_{ac} = 2$ Oe, respectively. A distinct kink, as indicated by arrow in figure 4(a), is clearly noticed in $\chi_3$ at 268 K, higher than the temperature

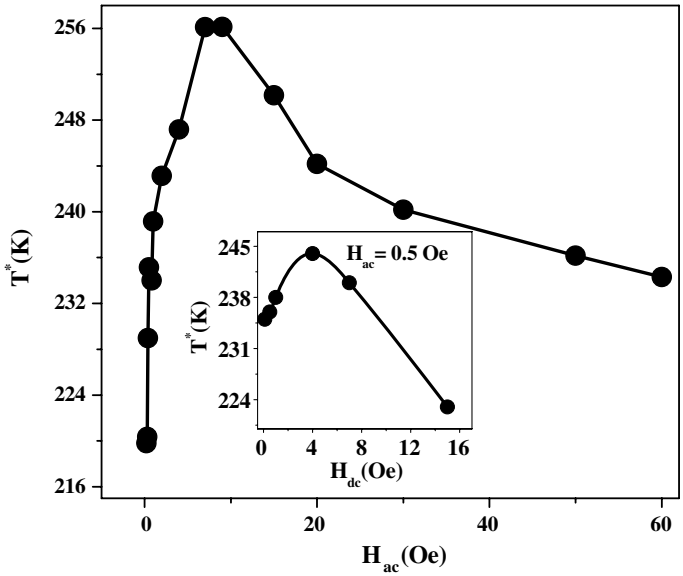


**Figure 5.** $H_{ac}$ dependence of freezing temperatures $T^*$, estimated from the peaks of the $\chi_3$ versus temperature curves. The inset shows the $H_{dc}$ dependence of $T^*$ at $H_{ac} = 0.5$ Oe.

($T^* = 243$ K) of the peak arising as a result of the cooperative freezing phenomenon, as has already been established previously. A similar kink at almost the same temperature of 268 K is also visible in $\chi_1^I$ (not shown here). A more crucial point is that these kinks in $\chi_3$ and $\chi_1^I$ at 268 K are accompanied by a very distinct and sharp peak in $\chi_2$ at exactly the same temperature of 268 K (inset in figure 4(a)). It is well known that even order $\chi_{ac}$ ($\chi_2$, $\chi_4 \ldots$) can be observed only if the system exhibits spontaneous magnetization ($M_0$), which breaks the inversion symmetry of the magnetization with respect to $H_{ac}$. Hence, we may correlate the peak in $\chi_2$ as well as the kinks in $\chi_3$ and $\chi_1^I$ at 268 K to the formation of FM clusters. To explain the cooperative freezing phenomenon associated with the distinct peak in $\chi_3$ at a lower temperature $T^*$ (∼243 K), we suppose that these FM clusters (formed around 268 K) subsequently undergo random dipolar intercluster interactions and finally freeze to give rise to a peak at a lower temperature ($T^* = 243$ K). Thus it emerges that the observed glassiness of our system arises due to interacting assembly of magnetic clusters, which constitutes a cluster glass (CG) phase. In a previous report, Sun *et al* [24] also attributed a similar CG phase to the bulk $La_{0.5}Gd_{0.2}Sr_{0.3}MnO_3$ system. In fact, for strong random dipolar intercluster interaction in the system the individual energy barrier of FM clusters can no longer be identified and only the energy of the cluster assembly is significant. This, in turn, establishes a collective state with magnetic properties similar to that of an SG. Significantly, as can be clearly seen from figure 4(a) and its inset, corresponding to the freezing temperature of $T^* = 243$ K at $\chi_3$ there is no response of $\chi_2$ at all, further substantiating its glassy origin [47]. Similar features of $\chi_{ac}$ are also seen in case of other low $H_{ac}$ values (figure 4(b) and its inset, at $H_{ac} = 3$ Oe).

The temperature associated with the peak in $\chi_3$, showing divergence in the limit of $H_{ac} \rightarrow 0$, is the corresponding freezing temperature ($T^*$) [41]. Figure 5 shows unusual dependence of $T^*$ on $H_{ac}$. This clearly indicates an initial sharp rise in $T^*$ with increase in $H_{ac}$ up to a certain value, beyond which $T^*$ decreases with further increase in $H_{ac}$. $T^*$ also exhibits similar non-monotonic behavior with superimposed dc field ($H_{dc}$) at the same $H_{ac}$

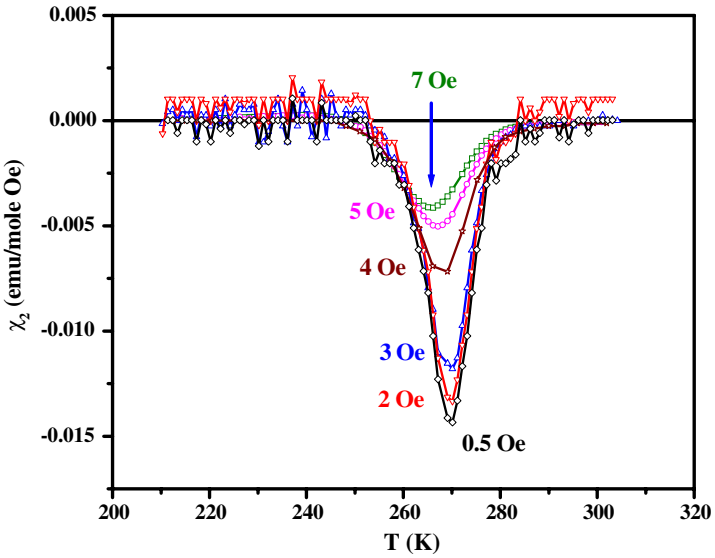


**Figure 6.** $\chi_2$ versus temperature curves measured at different $H_{ac}$ values from 0.5 to 7 Oe showing a decrease in absolute value of $\chi_2$ with an increase in $H_{ac}$.

(inset in figure 5). We believe that this observed field dependence of $T^*$ is in contradiction with the established literature of any kind of glassy system [40]. It is well established that increase in cluster concentration causes enhancement of random dipolar intercluster interactions. This boosts frustration and collectivity in the relaxation of the system, which, in turn, shifts the freezing temperature towards the higher temperature side [48, 49]. Here our primary conjecture to address the rise in $T^*$ with field (figure 5) is the increase in cluster concentration with field in the system. Likewise, the subsequent fall in $T^*$ with field (figure 5) can be correlated with the decrease in cluster concentration. Since our parent compound $La_{0.7}Sr_{0.3}MnO_3$ is a long range ordered double exchange ferromagnet and the formation of finite size FM clusters occurs due to Gd doping on the La site, modulation of number/concentration of those FM clusters in a realistic specimen can take place only at the cost of size of these clusters. Thus the rise in cluster concentration in our system with $H_{ac}$ up to a certain limit takes place as a result of the decrease in cluster size. This can be substantiated by the corresponding decrease in absolute value of $\chi_2$ with increase in $H_{ac}$ (figure 6). It can be understood that reduction in cluster size can occur as a results of a decline in magnetic correlation length ($\xi$). This implies a reduction in $M_0$ in the system that expectedly causes the corresponding decrease in absolute value of $\chi_2$. But from this point one cannot conclusively assert whether this decline in $\chi_2$ with an increase in $H_{ac}$ is associated with a decrease in $M_0$ in the system or this is just an effect of nonlinearity of the system with a rise in $H_{ac}$. To avoid this controversy, we have investigated the effect of $H_{dc}$ on $\chi_2$ at the same $H_{ac} = 0.5$ Oe for different well separated values of $H_{dc} = 0.1$, 4, 7 and 15 Oe. Figure 7(a) shows that the sharp peak in $\chi_2$ at 268 K, without any superimposed $H_{dc}$ (black symbol and line), decreases considerably when $H_{dc} = 0.1$ Oe is superimposed (red symbol and line). This peak disappears when $H_{dc}$ is increased to 4 Oe (figure 7(b)). However, with the application of $H_{dc} = 7$ Oe, the same peak reappears and further grows with the application of a much higher field of $H_{dc} = 15$ Oe (figure 8). Moreover, figures 7 and 8 exhibit the appearance of a broad low temperature peak in $\chi_2$ in the presence of superimposed finite $H_{dc}$. This can be understood in terms of the biasing effect of spin clusters on the glassy background

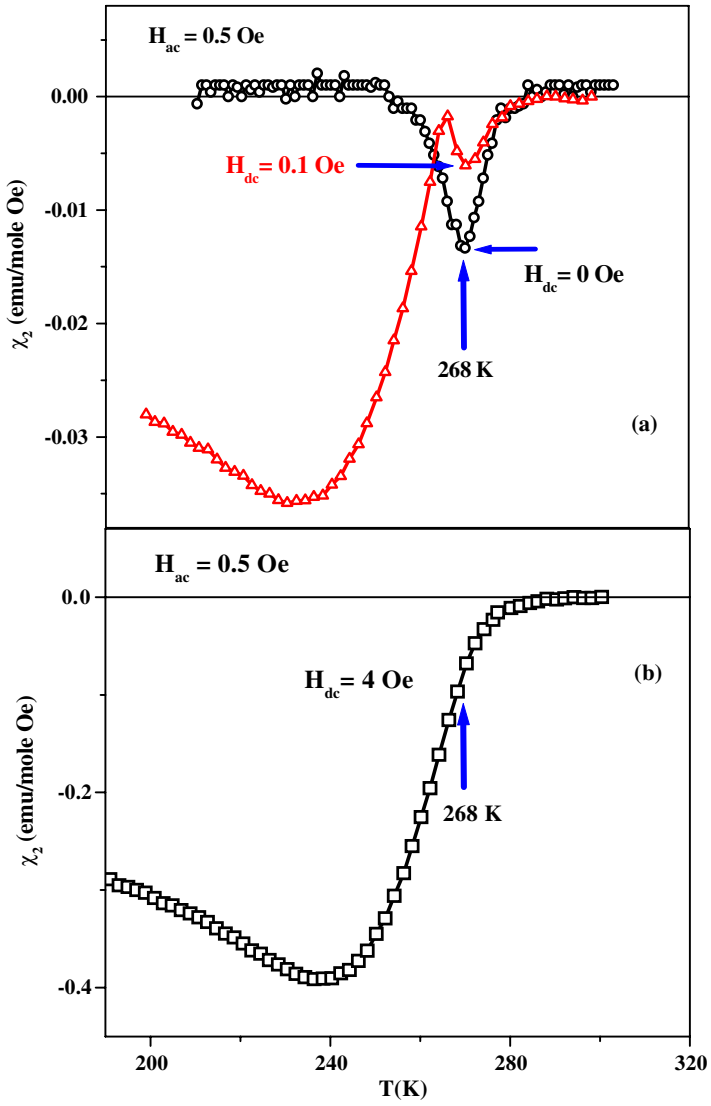


**Figure 7.** (a) $\chi_2$ versus temperature curves measured at $H_{ac} = 0.5$ Oe with $H_{dc} = 0$ Oe (unfilled circle, black symbol and line) and 0.1 Oe (unfilled triangle, red symbol and line). (b) $\chi_2$ versus temperature curve measured at $H_{ac} = 0.5$ Oe for $H_{dc} = 4$ Oe.

in the presence of $H_{dc}$, resulting in a strong component of symmetry breaking field inside the system. This broad peak cannot be related to the freezing phenomenon, since for this $\chi_2$ would be essentially absent [47]. These results (figures 7 and 8) support our conjecture that with the increase in applied field up to a certain limit there is a gradual destruction of long range FM ordering ($\xi$ becomes smaller) in this system. This results in decrease in cluster size, as indicated by the corresponding suppression and finally disappearance of $\chi_2$ up to $H_{dc} \sim 4$ Oe (figures 7(a) and (b)). However, beyond this certain limit of field ($H_{dc} \sim 4$ Oe) there is again a rise in cluster size, as indicated by the reappearance of $\chi_2$ (figure 8). This crossover field ($H_{dc} \sim 4$ Oe) is consistent with that $H_{dc}$ (∼4 Oe) where $T^*$ also shows similar crossover

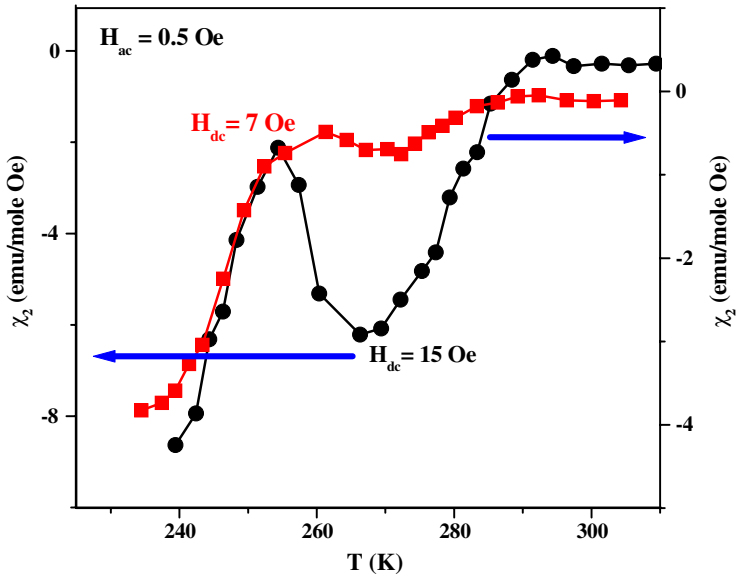


**Figure 8.** $\chi_2$ versus temperature curves measured at $H_{ac} = 0.5$ Oe for $H_{dc} = 7$ Oe (filled square, red symbol and line) and 15 Oe (filled circle, black symbol and line).

behavior (inset of figure 5). Thus the variation of $T^*$ with $H_{dc}$, as well as with $H_{ac}$ (figure 5), can be attributed to the modulation of cluster size with the applied field. This modulates cluster concentration, that tunes the frustration and collectivity in the relaxation of the system and consequently tunes $T^*$.

In order to explain these experimental results first we should consider the role of Gd doping in manganites. At the outset, doping of Gd, besides decreasing the tolerance factor $t$ [24, 30], promotes the random character of distribution of A site cations ($La^{3+}$, $Sr^{2+}$, $Gd^{3+}$). As a result, there is a random distribution of hopping of conduction electrons as well as an exchange between localized spins, thus introducing a random disorder in the magnetic lattice of the system [24]. Additionally, as generally recognized from previous studies [24, 25, 27, 28, 30], there exists an AFM exchange coupling between Gd and Mn moments in the Gd doped manganite system. This AFM coupling between Gd and Mn moments would also contribute to the random disorder in the FM host of our system. On account of these considerations, the experimental results reported here may be well interpreted in the following terms. We suppose that this random disorder in an otherwise long range ordered FM system prevents the magnetic correlation length $\xi$ from diverging at the transition temperature. This possibly forms 'finite size' FM clusters, where $\xi$ becomes restrained within the size of these FM clusters [3, 34, 46]. In fact, these finite size clusters are only magnetic entities formed because of finite $\xi$, which defines their size. These clusters are found to freeze at a lower temperature, showing CG-like behavior. Sun *et al* observed a similar CG-like feature for the $La_{0.5}Gd_{0.2}Sr_{0.3}MnO_3$ system and have attributed this glass-like phase to the competing AFM exchange coupling between Gd and Mn moments [24].

Based on this scenario, we may suppose that after the magnetic phase transition from para to FM phase, via the AFM coupling of the Gd spins with the Mn moments, Gd spins become polarized antiparallel with respect to the FM component of the Mn moments. However, as already discussed, because of the random disorder in the magnetic lattice due to the doping of Gd on La sites, all types of magnetic exchange interaction, e.g. Mn–Mn,

Gd–Mn interactions, have random distributions. Following the generic feature of any Gd–transition metal compound [50], for low values of magnetic field Gd and Mn moments are antiferromagnetically coupled. However, beyond a certain field, because of the higher spin moment of $Gd^{3+}$ ($7/2\ \hbar$), they would like to align towards the field. In our case, increase in applied magnetic field could possibly cause a torque on the Gd moments, and because of the higher spin moment of Gd they would like to align towards the field. However, due to the inherent randomness in the magnetic lattice, the AFM Gd–Mn exchange interaction has a broad distribution. Consequently, the external magnetic field needed for flipping these Gd spins would also have a rather broad distribution. This means that with increase in applied magnetic field one may suppose a fraction of Gd spins tending to flip in the direction of field [50]. This introduces a high level of spin disorder in the FM host, resulting in a possible decrease in cluster size in the system. With further increase in field, a coherence in the response of Gd spins to the field is expected, that would decrease the spin disorder in the FM host, causing possibly an increase in cluster size. Our study thus shows that the possible tuning of random AFM Gd–Mn exchange interaction by external field can tune the spin disorder in the FM host, that in turn tunes the cluster size and accordingly the glassy phase of our system. It is noteworthy that a non-monotonic variation in the size of the 'finite size' cluster, formed in a long range order system in the presence of random field, was theoretically predicted earlier [51]. In our system also, an indirect correlation among those randomly distributed $Gd^{3+}$ spins, induced by the FM Mn lattice through AFM Gd–Mn interaction, can be supposed, possibly giving rise to a random AFM field [3]. Based on this, an intuitive picture of external field induced tuning of random field, causing non-monotonic modulation of cluster size, can also be framed, which has strong theoretical support [51]. Thus further study can be initiated to test the justification of our experimental findings on the basis of the non-monotonic variation of the cluster size with the modulation of random field and relate it to the theoretical argument given in [51].

## 4. Conclusions

We have shown experimental results providing evidence of a glassy phase associated with the LGSMO system. We have observed a non-monotonic variation of freezing temperature with applied magnetic field, where the freezing temperature first increases with an increase in applied field up to a certain limit, beyond which it decreases with further increase in field. We understand this observed field dependent behavior of freezing temperature in term of tuning of disorder or glassiness associated with this system with applied magnetic field. From our experimental results, we suppose that the possible explanation of this non-monotonic change in the freezing temperature arises from the non-monotonic change in the cluster size with external magnetic field, which in turn is related to the modulation of inherent randomness of AFM Gd–Mn exchange interaction in the system. We consider that the increase in coherence in response of the random AFM impurity Gd spins with an increase in applied field initially increases spin disorder in the FM host and then decreases it. This modulates the cluster size and accordingly the glassy phase of our system. We have presented our work as a working hypothesis and have given a possible physical explanation of our observed experimental results. Direct experimental evidence might further support our discussion.

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