

Static and dynamic response of cluster glass in $\text{La}_{0.5}\text{Sr}_{0.5}\text{CoO}_3$

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The static and dynamic response of cluster glass in $\text{La}_{0.5}\text{Sr}_{0.5}\text{CoO}_3$ has been experimentally investigated through linear and nonlinear ac susceptibility measurements in the frequency range 137 Hz to 1370 Hz; ac field (1–10 Oe), dc field (0–40 Oe). Field cooled and zero field cooled dc magnetization data have also been reported. An attempt has been made to compare the dynamic response of cluster glass with spin glass, reentrant spin glass, and canted spin systems. The interesting observation is that in the cluster glass system, the coercivity at low fields under the zero field cooled condition does not increase rapidly, as one approaches the so-called freezing temperature (T_f), unlike that in typical reentrant spin glass systems. We also observed a linear relation of field cooled coercivity, H_c^{FC} (i.e., the coercive force to bring the thermoremanent magnetization to zero) with cooling fields (H_{FC}) at different temperatures. The variation of $dH_c^{\text{FC}}/dH_{\text{FC}}$ with $T(K)$ shows an exponential behavior for $T \leq T_c$. We believe that this is a significant observation for cluster glass systems. [S0163-1829(96)00834-X]

I. INTRODUCTION

Spin glass (SG) states have been found in a wide variety of systems including the magnetic insulators or amorphous alloys with the following common features: (1) frozen in magnetic moments below some freezing temperature T_f , (2) lack of periodic long range magnetic order, and (3) remanence and magnetic relaxation over macroscopic time scales below T_f when there are changes of magnetic field. From the macroscopic point of view, these properties have been characterized by the temperature dependence of dc magnetization, ac susceptibility in the field cooled (FC) and zero field cooled (ZFC) conditions, hysteresis effect and magnetic relaxation experiments. In a certain number of alloys like AuFe (beyond 12 at. % Fe), one encounters a domain of reentrant behavior which shares the characteristics of both SG and ferromagnetic orderings—the so-called reentrant spin glass (RSG). These systems undergo a PM-FM transition at a Curie temperature T_c and have a lower freezing temperature, characterized by a strong decrease in the initial susceptibility with thermomagnetic irreversibility.^{1,2} However, it is important to mention that the macroscopic properties of many systems (e.g., canted, RSG systems) undergoing multiple magnetic phase transitions may show common features, although the microscopic natures and underlying physics could be quite different. To differentiate between these systems, a careful ac susceptibility study (with frequency and field dependence) with static biasing field in the FC and ZFC condition, etc., has been suggested by us.³

A modified version of the SG system, termed “cluster glass” (CG) can be considered to be a set of clusters, formed due to short range ordering at some temperature near the Curie-like temperature T_c . Below T_c , the cluster glass system is expected to show SG-like behavior with increased magnetic spin density (instead of considering sets of isolated atomic moments).^{2,4} On the other hand, due to the finite

range ferromagnetic ordering below T_c , the system also shows some features similar to those found in RSG systems. Systems like $\text{Cu}_{75}\text{Mn}_{25}$, YFeAl_2 , and Fe_xTiS_2 have been considered to belong to the family of cluster glass.⁵ Therefore, it is interesting to study the static and dynamic response of a well-defined CG system in comparison with SG, RSG and canted spin systems.

According to the phase diagram suggested for $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$ on the basis of magnetization measurements, $\text{La}_{0.5}\text{Sr}_{0.5}\text{CoO}_3$ belongs to the cluster glass phase. In this system the double-exchange interaction between trivalent and tetravalent cobalt spins and the exchange interaction between high spin Co^{4+} and low spin Co^{III} is considered to be ferromagnetic whereas the superexchange interactions between high spins ($\text{Co}^{3+}-\text{Co}^{3+}$, $\text{Co}^{4+}-\text{Co}^{4+}$) are antiferromagnetic. The competition between the ferromagnetic and the antiferromagnetic interactions along with the randomness lead to SG states observed for $0 < x \leq 0.18$. When the ferromagnetic exchange interactions just overcome the antiferromagnetic one, the cluster glass phase appears with short range ferromagnetic ordering ($0.18 \leq x \leq 0.50$).⁶ In this paper, we experimentally investigate the dynamic and static response of the metallic system $\text{La}_{0.5}\text{Sr}_{0.5}\text{CoO}_3$,⁷ through the study of linear and nonlinear components of $\chi(\omega_{\text{ac}}, H_{\text{dc}})$, dc magnetization and hysteresis experiments under ZFC and FC mode to compare with SG, RSG, and canted spin systems.

II. EXPERIMENT

The sample ($\text{La}_{0.5}\text{Sr}_{0.5}\text{CoO}_3$) was prepared by the high temperature ceramic method as reported earlier.^{7,8} Appropriate molar ratios of La_2O_3 which was preheated at 1000 °C, cobalt oxalate, $\text{CoC}_2\text{O}_4 \cdot 2\text{H}_2\text{O}$, and SrCO_3 were thoroughly mixed in an agate mortar and initially fired at 900 °C for the decomposition of oxalate and carbonate. The

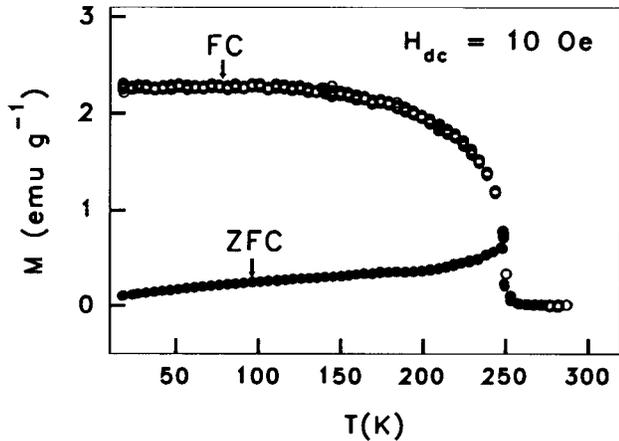


FIG. 1. dc magnetization data under zero field cooled and field cooled conditions as a function of temperature at a field of 10 Oe.

resulting mixture was heated to 1000 °C for 3 days with intermediate grindings and finally the sample was heated at 1150 °C for 24 h with one intermediate grinding. Formation of the single phase compound was established by powder x-ray diffraction on a Philips PW 1730 x-ray diffractometer. Lattice parameters were calculated by a least-squares method. The calculated lattice parameters were comparable to those reported earlier.⁷ The oxygen stoichiometry of the compound was determined by iodometric titrations. The composition of the final compound thus determined was $\text{La}_{0.5}\text{Sr}_{0.5}\text{CoO}_{2.96 \pm 0.02}$.

The magnetization m is expressed as

$$m = m_0 + \chi_1 h + \chi_2 h^2 + \chi_3 h^3 + \dots, \quad (1)$$

where h is an applied field, m_0 is the spontaneous magnetization, χ_1 is the linear susceptibility, and χ_2 and χ_3 are the nonlinear susceptibilities. Using an ac field $h = h_0 \sin \omega t$, the ω and 3ω components of m are measured.

The ac susceptibilities χ_1' and χ_1'' (linear components) are measured using a L'ATNE mutual inductance bridge. The details of the apparatus are given in Ref. 9. The details of the measurements of the 3ω component $[(3/4)\chi_3' h_0^2]$ along with the effect of the dc field in the measurements of higher harmonics have been discussed in Ref. 10. The low field magnetization and hysteresis experiments have been performed using an automated flux-type magnetometer¹¹ and the high field magnetization data have been taken using the commercial PAR EG & G vibrating sample magnetometer.

III. RESULTS AND DISCUSSION

Figure 1 shows the temperature dependence of dc magnetization for $\text{La}_{0.5}\text{Sr}_{0.5}\text{CoO}_3$ in the zero field cooled (ZFC) and field cooled (FC) conditions. On lowering the temperature, the ZFC magnetization rises sharply at around 250 K and then undergoes a less rapid drop down to 15 K. It may also be noted that the rate of decrease of magnetization with respect to temperature, i.e., dM/dT in the region 75–185 K is comparatively smaller than in the region 15–75 K. The FC and ZFC magnetizations have the same value around and above 250 K. Below 250 K, FC magnetization rises rapidly

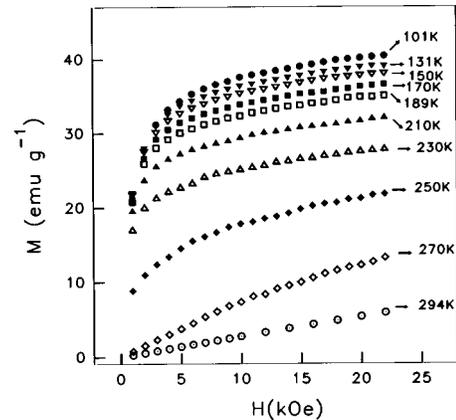


FIG. 2. dc magnetization data under the zero field cooled condition as a function of field at different temperatures (101–294 K).

with lowering of temperature down to 185 K and then undergoing a comparatively less rapid change, it finally levels off. For $M(T)$, our observation is similar to the results found in Ref. 6. We associate the sharp rise in M with the appearance of the finite range ferromagnetic coupling, forming the clusters around a quasicritical temperature T_c . The absence of true long range order is apparent from the large difference between M_{FC} and M_{ZFC} below T_c (≈ 245 K). The change in dM/dT around 75 K may be associated with the freezing of the clusters in the static field that will be discussed subsequently. In the SG systems, the irreversibility starts below T_f with the FC curve almost flat whereas the ZFC magnetization drops at lower temperature. The difference between M_{FC} and M_{ZFC} (i.e., remanence) is much higher in the cluster glass phase compared to the spin glass phase (corresponding to $\text{La}_{1-x}\text{Sr}_x\text{CoO}_3$ for $x \leq 0.2$) at sufficiently low temperatures, indicating the presence of short range ferromagnetic ordering within a cluster. It may also be noted that in a typical RSG system (i.e., PM-FM-SG phase) the irreversibility occurs at low temperatures far below T_c whereas in CG irreversibility occurs just at $T \leq T_c$.

Figure 2 shows the measurement of $M(H)$ under the ZFC condition at several temperatures using the vibrating sample magnetometer in the field range ≤ 22 kOe. The data are in good agreement with those found in Ref. 6. Even at 101 K no saturation has been observed for fields of the order of 2.2 T. No S-shaped curve characteristic of SG systems has been obtained. For temperatures below the freezing temperature, in both canonical SG as well as insulating SG systems, ZFC magnetization shows an S-shaped behavior, i.e., the initial slope of the curve is smaller than the slope at an inflection point for nonzero H . The linear relation can be obtained only for field > 1 T in Arrot's plot similar to Ref. 6 suggesting the field induced FM behavior.

The measurement of ac susceptibility (ACS) gives us a detailed insight into the dynamics of freezing. Preliminary ACS data for different Sr concentrations have been reported.⁷ In an assembly of clusters formed out of short range interactions, the freezing appears through the probability of each cluster to overcome the energy barrier E , induced by the local anisotropy, expressed in terms of relaxation time $\tau = \tau_0 \exp(E/kT)$. On lowering the temperature, the relaxation time increases and an anomaly in ac susceptibility will ap-

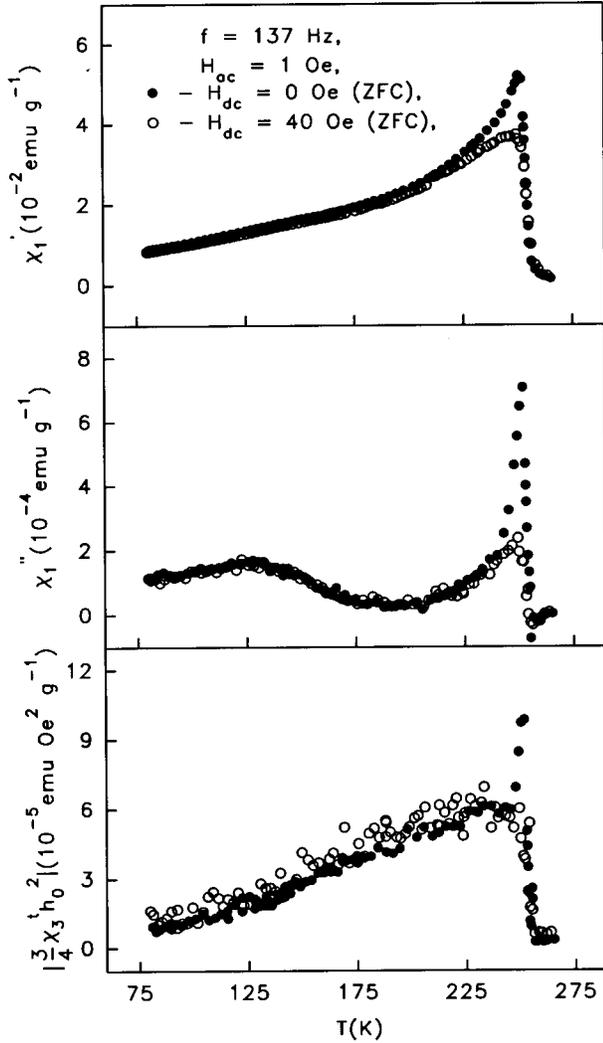


FIG. 3. The linear and nonlinear ac susceptibility data measured at an ac field of 1 Oe, 137 Hz with or without a dc field of 40 Oe.

pear at T_f when τ becomes larger than the characteristic measuring time. On the other hand, if the relaxation time spectrum is very broad, this anomaly in ac susceptibility can be associated with the maximum or some average of the largest relaxation times of the distribution. Figure 3 shows the temperature dependence of the real part χ_1' , imaginary part χ_1'' , and the nonlinear susceptibility $|(3/4)\chi_3' h_0^2|$ under the ZFC condition at a fundamental frequency ω (≈ 137 Hz) and ac field of 1 Oe with or without a superimposed dc field (40 Oe). The rise in $\chi_1'(T)$, accompanied by a peak in $\chi_1''(T)$ and the nonlinear component at a temperature around $T_c \approx 250$ K, can be associated with the appearance of short range FM ordering, i.e., the formation of clusters. At a lower temperature $T_f \approx 138$ K, the kink in $\chi_1'(T)$ and a peak in $\chi_1''(T)$ can be associated with the freezing of clusters in our time scale of observation. In this context, it is important to point out that no low temperature nonlinear anomaly has been observed for a low ac field amplitude $H_{ac}^{rms} \approx 1$ Oe. This is due to a very small signal compared to the noise level for the nonlinear measurements at low fields, as described in Ref. 12. The dynamic magnetic response gets modified dras-

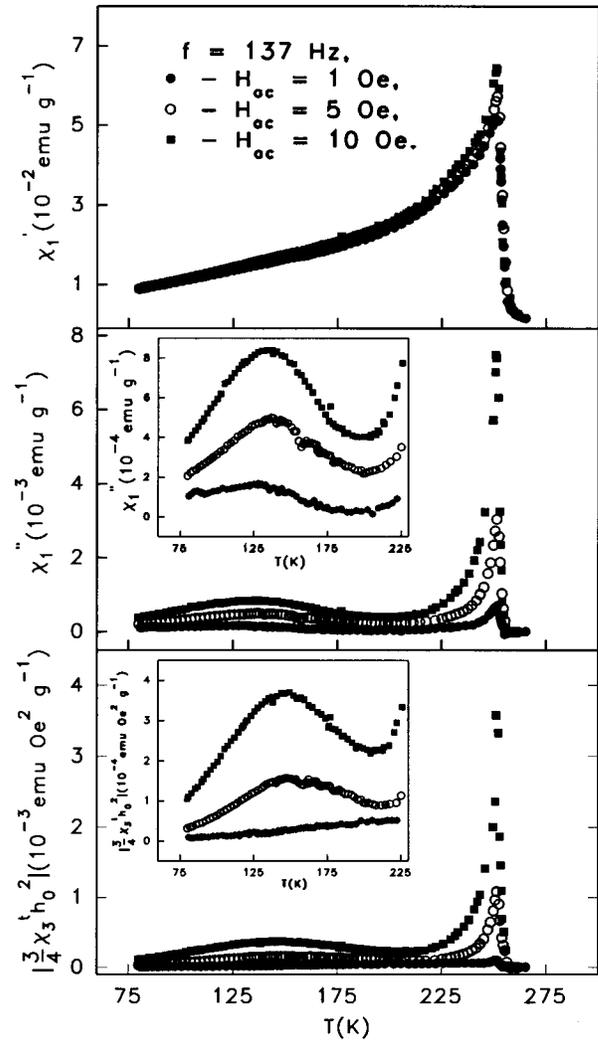


FIG. 4. The linear and nonlinear ac susceptibility data measured at different ac fields (1–10 Oe) at a frequency of 137 Hz. The insets show the low temperature region.

tically with the application of a superimposed dc field (40 Oe). Around T_c , the suppression of $\chi_1''(T)$ and χ_3' peaks may be considered to be the signature of the partial suppression of the magnetic fluctuation with the application of a dc field.¹³ Below T_c , the modification in the presence of a dc field including the suppression of the low temperature peak in $\chi_1''(T)$ is due to the absence of true long range order as well as the slow response of the clusters in the presence of a dc field. Moreover, in the case of true long range FM ordering, in the ZFC condition, the maximum in χ_1' has been found to be much sharper compared to the rounded maximum in the case of finite clusters.

Figure 4 and the inset (in the low temperature region) show the ac field amplitude dependence of $\chi_1'(T)$, $\chi_1''(T)$, and $|(3/4)\chi_3' h_0^2|$ as a function of temperature. The small signal-to-noise ratio in the measurement of higher harmonics and the contribution from other higher harmonics have already been mentioned. When the ac field amplitude is increased from 1 Oe to 5 Oe and 10 Oe, a broad nondivergent peak appears in χ_1'' and χ_3' at a lower temperature. The higher temperature maxima in χ_1' , χ_1'' , and χ_3' shifts to lower tem-

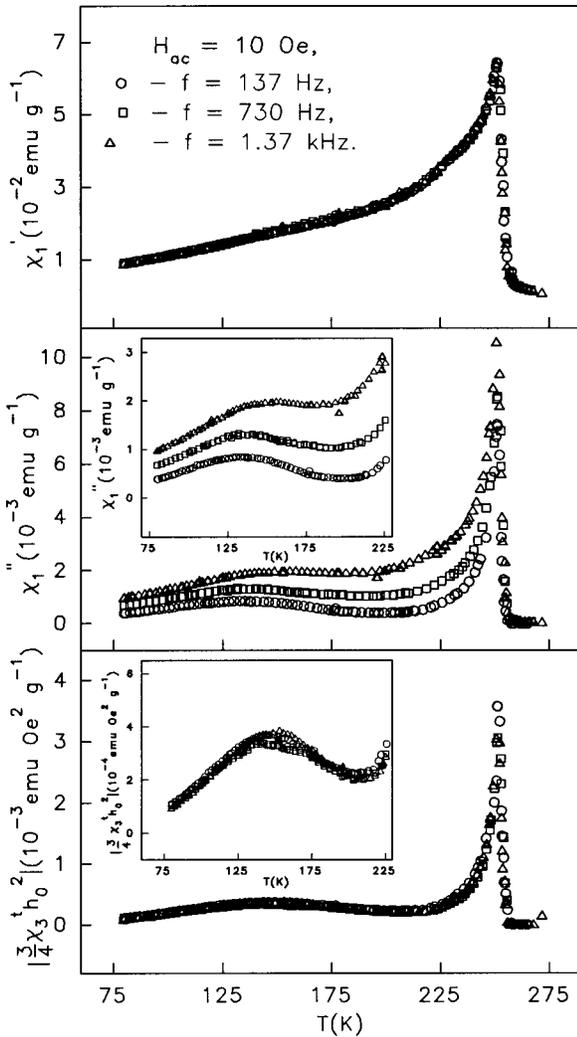


FIG. 5. The linear and nonlinear ac susceptibility data measured at different frequencies (137 Hz–1.37 kHz) for an ac field of 10 Oe.

perature (251 K for $h_{ac}=1$ Oe, 249 K for $h_{ac}=5$ Oe, 247 K for $h_{ac}=10$ Oe) with the increase of ac field amplitude. Regarding the nonlinear susceptibility, the mean field theory for SG suggests a divergent behavior, i.e., χ_{2n+1} diverges at T_f according to the power law, $\varepsilon^{-n(\gamma+\beta)+\beta}$ for $n \geq 1$, where ε is the reduced temperature equal to $(T-T_f)/T_f$, considering the SG freezing as a cooperative phenomenon.^{1,2} The value of γ is in good agreement for canonical SG systems.² For RSG systems, the study of the nonlinear behavior is very important to decide whether the low temperature SG order is associated with a cooperative phase transition or not. The analyses of susceptibility isotherms based on $M(H,T)$ data show the presence of a weak, nondivergent, anomalous peak in T_f in a number of systems NiMn, PdMn, FeZr. This has been attributed to a longitudinal response to transverse spin freezing and is consistent with Mossbauer data. Moreover, Katori and Suzuki predicted theoretically that the longitudinal susceptibility in vector spin systems should exhibit a complementary nonlinear anomaly in response to transverse spin freezing.¹⁴ Similarly, for the canted spin system $\text{Ce}(\text{Fe}_{0.96}\text{Al}_{0.04})_2$ with a well-defined PM-FM transition, a peak in χ_3' has been observed at a lower temperature with the

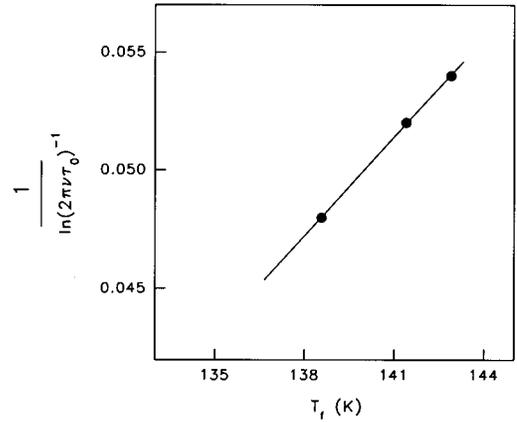


FIG. 6. Fit of the experimental data of $\chi_1''(T)$ of Fig. 5 with the Vogel-Fulcher law.

appearance of canting of moments in the absence of any glassy behavior.³ In the present case, the nonlinear term shows a very broad peak at a lower temperature $T_f \approx 146$ K, associated with the freezing of clusters. The maxima in χ_3' data appears to be at a temperature different from the χ_1'' maxima.

In the SG systems, the temperature associated with the maxima of χ_1' , corresponding to the PM-SG transition varies slowly with the measuring frequency ω . The strong frequency dependence along with large $(T_f-T_0)/T_f (\approx 0.5)$ has been suggested to be signature of cluster freezing.^{15,16} Moreover, the susceptibility itself is frequency dependent even above T_f for superparamagnetic clusters, whereas it is slowly frequency dependent below T_f only for SG systems. It is important to point out that the study of the frequency dependence may be useful to compare a number of systems¹⁶ but nothing can be definitely concluded about the existence or nonexistence of a finite temperature phase transition.² Figure 5 and the inset (in the low temperature region) shows the results of the measurements of $\chi_1'(T)$, $\chi_1''(T)$, and χ_3' for different frequencies in the range 137 Hz to 1.37 kHz at $H_{ac} \approx 10$ Oe. There is no frequency dependence at higher temperature (≈ 250 K) maxima in $\chi_1'(T)$, $\chi_1''(T)$, and χ_3' . As in the inset in Fig. 5, there is a strong frequency dependence of the low temperature peak in $\chi_1''(T)$ (138.6 K for 137 Hz, 141.4 K for 730 Hz, 142.9 K for 1.37 kHz) and χ_3' (146.2 K for 137 Hz, 143 K for 730 Hz, 148.6 K for 1.37 kHz). χ_1'' peaks shift to higher temperatures and become progressively rounded with increase of frequency whereas no systematic relationship is obtained for the χ_3' peak. The anomaly in ACS at a particular temperature appears when the maximum of the relaxation time τ_{max} is of the order of $1/\omega$ where ω is the measuring frequency. This τ_{max} varies considerably with temperature ($\tau_{max} \approx 0.0012$ at $T=138.57$ K and $\tau_{max} \approx 0.00012$ at $T=142.9$ K) as has been found in other CG freezing. In the present system the shifts in peak temperature of $\chi_1''(T)$ with frequency are found to satisfy the phenomenological Vogel-Fulcher law, $\tau = \tau_0 \exp[E_a/k(T_f-T_0)]$ for $0 < T_0 < T_f$ (shown in Fig. 6) with $E_a/k = 719.64$, $\tau_0 = 10^{-12}$ sec and $T_0 = 104.01$. Here, the quantity $(T_f-T_0)/T_f \approx 0.25$ is an order of magnitude higher than those reported for canonical spin glasses (e.g.,

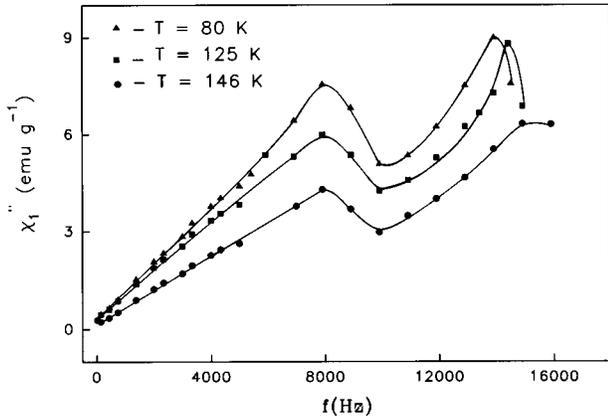


FIG. 7. The absorption part of the linear component of the ac susceptibility as a function of frequency for different temperatures at 10 Oe. The solid lines are a guide to the eyes.

for $\text{CuMn} \approx 0.07$) while it is comparable to values obtained for systems, described in terms of progressive freezing of clusters, for example, $\text{ZnCr}_{1.6}\text{Ga}_{0.4}\text{O}_4$.¹⁵ If instead of a single relaxation time, there exists a distribution of relaxation times then the variation of χ_1'' with ω can be written as¹

$$\chi_1''(\omega) \propto \int_{\tau_{\min}}^{\tau_{\max}} \frac{\omega \tau g(\tau, T)}{1 + \omega^2 \tau^2} d\tau, \quad (2)$$

where $g(\tau, T)$ is the distribution function of relaxation times. Thus the variation of χ_1'' with ω gives us information about $g(\tau, T)$ at a particular temperature. In our case, in the frequency range 137 Hz–1.37 kHz (Fig. 5), χ_1'' increases with frequency for all temperatures below T_c . In Fig. 7 the variation of χ_1'' with frequency over a wide range (15 Hz–15 kHz) has been shown for three temperatures (80, 125, 146 K) below and around T_f (≈ 140 K), the so-called CG freezing temperature at 10 Oe. The following conclusions can be made from this figure. Even at temperatures $T \ll T_f$, the distribution function $g(\tau)$ varies considerably with frequency compared to the slowly varying function as has been seen for SG systems like (EuSr)S. It is difficult to get an exact form of $g(\tau)$.¹⁷ There are two peaks in the $g(\omega, T)$ versus ω curve, as has been expected for cluster glass samples (Fe_xTiS_2 , cobalt aluminosilicate glass).^{5,17} The maximum in the $g(\omega, \tau)$ versus ω curve corresponds to the most probable distribution of relaxation time with $\tau_{\text{av}} = 1/\omega_{\text{max}}$, where ω_{max} is the frequency corresponding to the maximum of $\chi_1''(\omega)$. For the temperature range investigated in the present study, τ_{av} is small ($\approx 10^{-5}$) compared to the long spin relaxation times ($> 10^{-3}$) observed in case of spin glass freezing. Moreover, the present study suggests that the variation of τ_{av} with temperature is small as observed in case of cluster glass freezing.¹⁷

Figure 8 and the inset (in the low temperature region) show the field cooled behavior of the linear and nonlinear components. Here the irreversibility starts at a temperature very close to T_c with the appearance of blocking of these finite size clusters arising out of anisotropy. In the RSG systems having a well-defined domain structure below T_c , the Heisenberg mean field model suggests a crossover

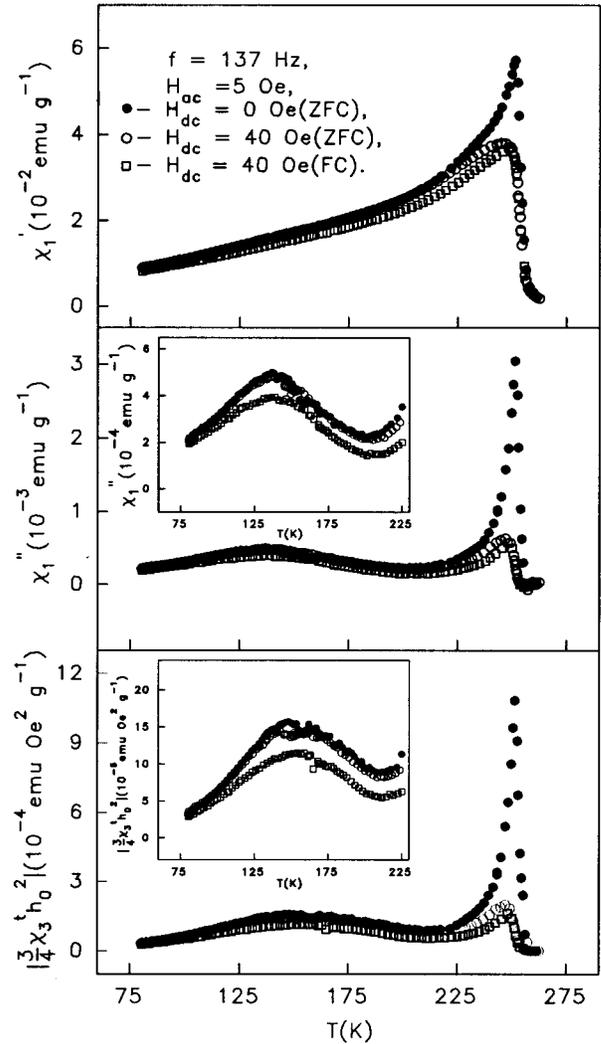


FIG. 8. The linear and nonlinear ac Susceptibility data measured at an ac field of 5 Oe, 137 Hz in the presence of a superimposed dc field 40 Oe under zero field cooled and field cooled (40 Oe) conditions.

from weak to strong irreversibility at a lower temperature $T_{\text{AT}} (< T_{\text{GT}})$. For a number of systems like AuFe, $(\text{Fe}_x\text{Ni}_{(1-x)})_{(1-y)}\text{Mn}_y$, NiMn the presence of an anomalous double-peaked structure in $\chi_1'(T)$, $\chi_1''(T)$ at a temperature much below T_c is considered to be reminiscent of the two reentrant phase boundaries associated with weak (GT) and strong irreversibility (AT) predicted by the isotropic vector spin model. There it has been found from ACS experiments, performed under ZFC and FC conditions that the real and imaginary components χ_1' , χ_1'' show strong irreversibility below T_{AT} .¹⁴ In the present observation there is no such behavior; however, it is similar to that obtained in the canted spin system.³

In the SG systems, hysteresis phenomena have been observed in the region of irreversibility below the freezing temperature T_f both under the ZFC and FC conditions.¹ In the case of $\text{La}_{0.5}\text{Sr}_{0.5}\text{CoO}_3$, the irreversibility starts at a temperature close to T_c (Figs. 1 and 8) and in this region the hysteresis loop opens even for a very small (\approx few Oe) field. In the ZFC condition the loops are drawn for three tempera-

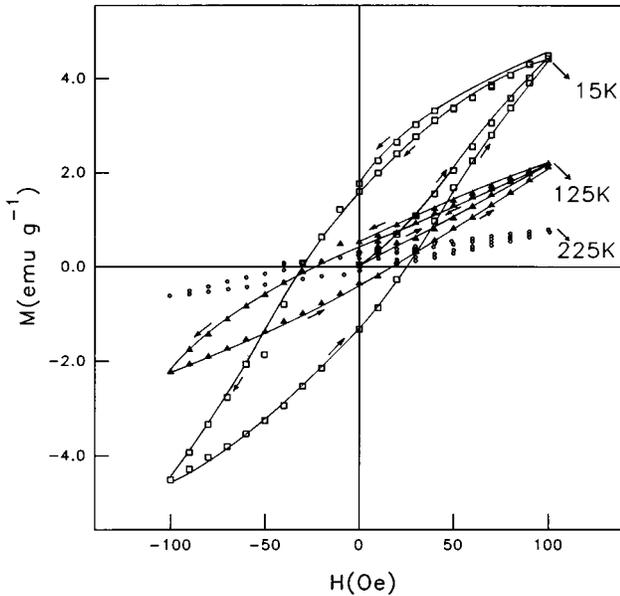


FIG. 9. Hysteresis loops drawn under the zero field cooled condition at three temperatures 15, 125, 225 K. The solid lines are a guide to the eyes.

tures; 15 K (much below T_f), 125 K (around T_f), and 225 K (below T_c) where the limiting value of the cycling field has been set to ± 100 G (Fig. 9). The loops within the limiting value of the cycling field are quite smooth. So we can conclude at least within this limiting value of the cycling field, there is no coherent reversal of magnetization of the clusters. Coherent reversal of magnetization at a sharp value of the magnetic field (showing rectangular hysteresis loops) suggests cooperative behavior among a large number of interacting particles.¹⁸

Hysteresis loops drawn under the FC condition show various types of structures in SG and RSG systems. In the SG systems like CuMn, displaced hysteresis loops have been observed in the FC initial state in contrast to the ZFC initial state.¹ For the RSG system $\text{Au}_{81}\text{Fe}_{19}$,¹⁹ after cooling in a field (25 kOe) down to 1.2 K, the field has been reduced to zero and then increased in the negative ($-H$) direction. The curve obtained intersects the negative H axis at a value which is twice the coercive field obtained after ZFC. The same kind of magnetic behavior has been observed for AuFe alloys in the SG region. This incremental coercivity in the field cooled case has been found to drop with increasing temperature until it disappears at around 10 K which corresponds to the center of the AT transitional region, i.e., the region of crossover from weak to strong irreversibility in RSG systems. The same temperature behavior has been obtained for the coercive field, obtained from experiments under the ZFC condition in $\text{Au}_{81}\text{Fe}_{19}$ (Ref. 20) and $(\text{Fe}_{78}\text{Mn}_{22})\text{P}_{16}\text{B}_6\text{Al}_3$.²¹ For another RSG system $\text{Ni}_{79}\text{Mn}_{21}$ (Ref. 19) also the displaced hysteresis loop has been observed in the FC condition. There, in contrast to the AuFe system, the loop is extremely reproducible even after cycling the field isothermally between upper limits of experimental fields.

In our experiment, under the FC condition, we cool the sample down to 14 K with a field ≤ 8 Oe. Then the sample is

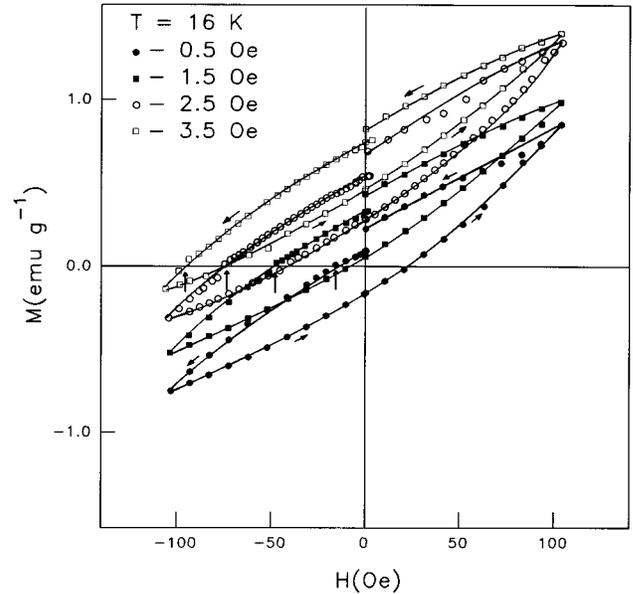


FIG. 10. Hysteresis loops drawn under different cooling fields at 16 K. The solid lines are a guide to the eyes.

warmed to different selected temperatures $T \leq T_c$ (15 K, 45 K, 65 K, 100 K, 135 K, 165 K, 210 K, 235 K), at which the field is first reduced to zero and then made to follow the cycle $0 \rightarrow -100 \text{ Oe} \rightarrow +100 \text{ Oe} \rightarrow 0 \text{ Oe}$. In this way, the observed hysteresis loop under different cooling fields at 15 K are as shown in Fig. 10. After the removal of the cooling field, some TRM exists which is higher for higher cooling fields. We also see in our case a displaced hysteresis loop. At the time of increasing the field in the negative ($-H$) direction, the negative field at which the $M-H$ curve cuts the H axis increases with the cooling field. This field may be called FC coercivity (H_c^{FC} as marked by the arrow in Fig. 10), i.e., the field required to bring the TRM to zero. The plot of the cooling field (H_{FC}) versus the FC coercivity (H_c^{FC}) at the corresponding cooling field gives a straight line at a given temperature [Fig. 11(a)]. Moreover, except at two very high temperatures close to T_c , all these straight lines intersect the cooling field axis at a value -0.5 ± 0.2 Oe. We think this is due to the uncompensation of the Earth's field (≈ 0.3 Oe) during the cooling process. So with the assumption that the straight lines pass through the ideal (0,0) of the coordinate system [which is (0, Earth's magnetic field) in the present coordinate system] the slope of these curves at different temperatures gives the FC coercivity in the unit field at the corresponding temperatures. This assumption is quite reasonable as it supports zero TRM and hence zero H_c^{FC} in an ideal ZFC condition. The slopes $dH_c^{\text{FC}}/dH_{\text{FC}}$ estimated from these curves [Fig. 11(a)] at various temperatures are plotted in Fig. 11(b). We get an exponential curve such that the slope is proportional to $\exp(-\alpha T)$ with $\alpha = 0.01$. This exponential temperature dependence of H_c^{FC} indicates that the blocking of these clusters decreases exponentially with increasing temperature due to thermal activation. The present study shows that in the CG system, under ZFC condition, the coercivity (H_c) does not increase rapidly as one approaches T_f unlike RSG systems ($T \leq T_f$).^{20,21} On the other hand, the

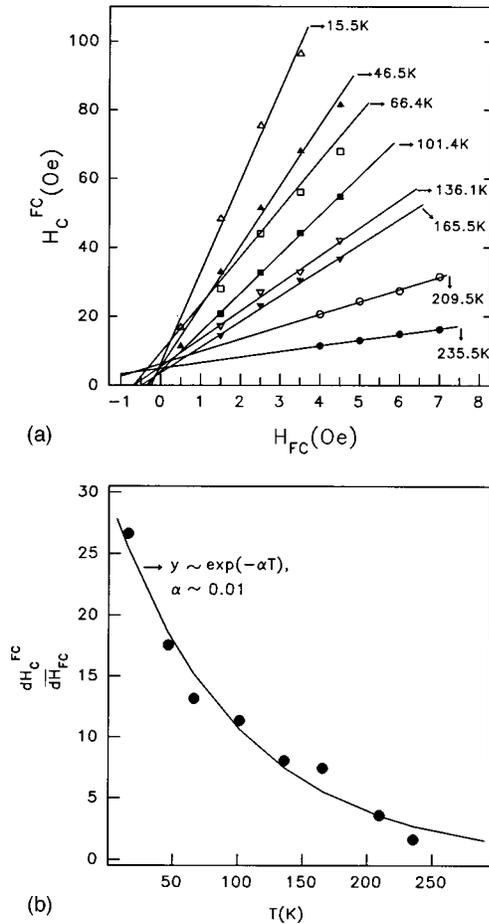


FIG. 11. (a) Plot of the cooling field (H_{FC}) vs the field cooled coercivity (H_c^{FC}) at the corresponding cooling field at different temperatures. The solid lines show the linear fit. (b) Plot of dH_c^{FC}/dH_{FC} as a function of temperature. The solid line shows the exponential fit.

FC coercivity (i.e., the coercive force to bring the TRM to zero) shows an exponential increase with lowering of temperature just at $T \leq T_c$ (250 K) rather than at $T \leq T_f$ (≈ 135 K). We believe that this is a significant observation in a typical CG system.

IV. CONCLUSION

We measured the static and dynamic response of cluster glass in the $\text{La}_{0.5}\text{Sr}_{0.5}\text{CoO}_3$ sample. Our ac susceptibility

data show that the freezing of the cluster occurs at $T_f \approx 138.6$ K for 137 Hz, 10 Oe below the Curie-like temperature ≈ 250 K. The frequency dependence study (137–1370 Hz) shows that it satisfies the Vogel-Fulcher law with $E_a/k = 719.64$, $\tau_0 = 10^{-12}$, $T_0 = 104.01$ K. $(T_f - T_0)/T_0 \approx 0.25$ is an order of magnitude higher than those of canonical SG ($\text{CuMn} \approx 0.07$) while it is comparable to values obtained in the case of progressing freezing of clusters. We observed two peaks in $\chi_1''(\omega, T)$ versus ω curve, as is expected for cluster glass samples (Fe_xTiS_2 , cobalt aluminosilicate glass). Here, the τ_{av} corresponding to the most probable distribution of relaxation time is small ($\approx 10^{-5}$) compared to the long spin relaxation times ($> 10^{-3}$) observed in case of spin glass freezing. The variation of τ_{av} with temperature is small as seen in the case of CG freezing. Like SG and RSG systems, a peak occurs in the nonlinear χ_3^t data around T_f , for which the physical origin is not clear. It may be noted that in the absence of glassy behavior, a canted spin system also exhibits such a nonlinear anomaly. Focusing on the dc magnetization results, the difference between M_{FC} and M_{ZFC} is much higher in the cluster glass phase compared to the spin glass phase (corresponding to $\text{La}_x\text{Sr}_{1-x}\text{CoO}_3$ for $x \leq 0.2$) at sufficiently low temperatures indicating the presence of short range ferromagnetic ordering within a cluster. Moreover, in this case the irreversibility starts just at $T \leq T_c$ whereas in typical RSG systems it occurs at low temperatures far below T_c and in the SG case it occurs below T_f . We believe that a significant result is obtained in the low field cooled (FC) hysteresis measurements. There, for low cooling fields we observed a linear relation between the FC coercivity (H_c^{FC}) and the cooling field (H_{FC}) for $T \leq T_c$. The slopes dH_c^{FC}/dH_{FC} of these linear curves at different temperatures show an exponential decrease with the increase of temperature. This exponential dependence suggests that the blocking of the clusters decreases exponentially with increasing temperature due to thermal activation. Such types of experiments are useful to compare SG, RSG, CG, and canted spin systems from the macroscopic point of view.

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